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Gist, Grant Alan

DISORDERED MAGNETISM IN DILUTE MAGNETIC ALLOYS STUDIED BY MUON SPIN RELAXATION

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Disordered Magnetism in Dilute Magnetic Alloys

Studied by Muon Spin Relaxation

by

Grant Alan Gist

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DOCTOR OF PHILOSOPHY

Approved, Thesis Committee:

S. A. Dodds
Associate Professor of Physics
Chairman

T. L. Estle
Professor of Physics

W. L. Wilson, Jr.
Professor of Electrical Engineering

Houston, Texas

January, 1986
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ABSTRACT

Muon spin relaxation ($\mu$SR) has been used to study the magnetic ordering in \textit{PdMn} and \textit{AgMn} dilute alloys representing disordered ferromagnetism, reentrant ferromagnetism, and spin glass order. A theory of static linewidths for both transverse applied field and zero applied field $\mu$SR is described for all types of spin order. The temperature dependence of the static linewidth in the spin glass and ferromagnetic phases is consistent with both a mean field theory of disordered magnetism and an inhomogeneous percolation model. Linewidths in paramagnetic AgMn provide the first direct observation of a conduction electron mediated RKKY component to the muon-local moment interaction. In PdMn, muons are excluded from occupying octahedral interstitial sites directly adjacent to Mn impurities. A theory relating muon spin-lattice relaxation to impurity spin correlation times in the paramagnetic and ordered phases is described. All types of magnetic ordering show a rapid slowing down of impurity spin fluctuations just above the ordering temperature. Muon spin-lattice relaxation in both the ferromagnetic and spin glass state is expected to be insensitive to
both long wavelength spin waves and to higher frequency localized spin wave-like excitations. The observed spin glass state relaxation is consistent with a phenomenological theory of low-frequency barrier mode excitations.
ACKNOWLEDGEMENTS

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Finally, I wish to thank my advisor, Dr. S. A. Dodds for his continual dedication, advice, and support.
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I. Introduction

This thesis reviews results of muon spin relaxation (μSR) experiments on dilute magnetic alloys exhibiting a variety of magnetic ordering phenomena. The materials chosen for study are representative of disordered ferromagnetism, spin glass order, and reentrant ferromagnetism. A description of these types of order and the model magnetic systems studied by μSR will be presented below.

Muon spin relaxation is analogous to nuclear magnetic resonance (NMR), in providing information on the static and dynamic properties of the microscopic local field. Unlike NMR, μSR experiments can be performed in zero applied field, an important capability in studies of spin glasses, since applied fields drastically modify the character of spin glass order. Furthermore, the time scale of μSR ($10^{-11}-10^{-5}$ sec) is well matched to the time scale of important dynamical processes in dilute magnetic alloys. Because of these characteristics, and the lack of an appropriate nucleus for NMR in many alloys, μSR provides microscopic information on magnetic ordering in dilute alloys unavailable from other techniques.

The primary conclusions of this work can be summarized as follows.
(1). The static linewidth in both the spin glass state and the disordered ferromagnetic state agrees reasonably well with a mean field theory of disordered magnetism.

(2). The dynamics of a spin glass above the transition temperature are strikingly similar to the dynamics of a disordered ferromagnet known to exhibit true critical phenomena. This is evidence for an extremely sharp spin glass transition. It is unknown whether this is a thermodynamic phase transition, since the experiments are unable to enter a true critical region.

(3). Muon spin-lattice relaxation in both spin glasses and disordered ferromagnets is expected to be insensitive to limited amplitude spin wave-like excitations. Observed relaxation is instead attributed to large amplitude barrier mode excitations. Relaxation rates in spin glasses are in quantitative agreement with calculations from a phenomenological theory of barrier modes.

(4). Novel muon-Mn interaction effects have been observed in AgMn and PdMn. High temperature linewidths in AgMn provide the first direct observation of a conduction electron mediated RKKY component to the muon-local moment interaction. In PdMn, muons appear to be excluded from occupying octahedral interstitial sites directly adjacent to Mn impurities, possibly due to an electrostatic repulsion enhanced by the large susceptibility of the Pd d-band electrons.
The remainder of this introduction defines the types of magnetic order in dilute alloys, and their realization in the particular materials chosen for this study. Chapter 2 describes experimental methods and sample characterization. The next four chapters are devoted to the measurement and interpretation of the static field distribution width, or linewidth. Chapter 3 presents theoretical results on the calculation of linewidths for all types of spin ordering. Chapter 4 describes mean field theories of disordered ferromagnetism and the spin glass state, and shows how μSR linewidths can be calculated from mean field theory. Chapters 5 and 6 present experimental results of the static linewidth in the paramagnetic and ordered state. The next three chapters are devoted to the measurement and interpretation of dynamic properties of the muon local field. Chapter 7 presents the theory of dynamic spin-lattice depolarization, and chapter 8 describes experimental results. Chapter 9 describes possible interpretations of spin-lattice relaxation results in the ordered state. Conclusions regarding the nature of magnetic ordering in spin glasses and disordered ferromagnets are summarized in chapter 10.

**dilute alloys**

A dilute magnetic alloy is a solid solution of a few percent of magnetic impurity atoms (e.g. Mn or Fe) dissolved into a non-magnetic host lattice (e.g. Pd, Ag or Cu). The impurities are
fixed at random on the sites of the solvent lattice, ideally without correlations in the atomic positions. Impurity spins separated by a distance $r$ interact via an indirect range-dependent exchange interaction $J(r)$, mediated by the host lattice conduction electrons. The random distribution of impurities on the lattice produces a distribution in the impurity separations, which in turn produces a distribution $p(J)$ of exchange energies. The distribution $p(J)$ determines the character of the low temperature magnetic ordering. The varieties of magnetic order in dilute alloys are reviewed by Hurd (1982). Ferromagnetism and spin glass order are described briefly below, along with the examples studied experimentally.

spin glass order

A spin glass is a state where localized moments (spins) are frozen into random orientations which do not vary with time, and with no net magnetization or long-range order (for reviews of spin glass properties, see Mydosh, 1981; Chowdhury and Mookerjee 1984; van Hemmen and Morgenstern, 1983). The randomness in the spin orientations is reminiscent of the structural randomness in an ordinary glass; hence the name spin glass. The spin glass state exists below the glass temperature $T_g$, which is defined by a cusp in the low field ac susceptibility. Although some experimental techniques show a sharp transition at $T_g$ (e.g. ac susceptibility,
Mössbauer effect, μSR, and the onset of magnetization irreversibilities and remanence), other techniques expected to show anomalies at a magnetic phase transition are featureless at $T_g$ (e.g. specific heat, resistivity, and ESR). The spin glass state below $T_g$ is characterized by severe thermomagnetic history effects, remanent magnetization, and relaxation phenomena with a broad distributions of relaxation times.

An important question in spin glass research concerns the nature of the transition at $T_g$. Is it a true thermodynamic phase transition, with associated critical phenomena, or is it a continuous dynamical freezing analogous to the rapid but continuous change in viscosity at the transition from a liquid to a structural glass? The customary definition of the spin glass state describes the spins as thermodynamically frozen in random orientations, but this condition can be replaced by 'apparently frozen over long time scales'. The thermodynamic condition is required for a true ordered phase, whereas the apparently frozen condition is reminiscent of the slow, non-equilibrium relaxation effects in a structural glass. Experimentally, it is very hard to distinguish these two conditions, and the nature of the transition is still an open problem.

This thesis presents experimental results on a typical spin glass, AgMn. In AgMn and the other so-called canonical spin glasses CuMn and AuFe (Mydosh, 1981), spins interact via the
conduction electrons by the RKKY interaction (Ruderman and Kittel, 1954),

\[ J(r) = A \frac{\cos(2k_F r + \phi)}{r^3} \quad (k_F r >> 1), \tag{1.1} \]

where \( k_F \) is the Fermi wavevector, and \( A \) is an amplitude parameter which contains the host density of states and the s-d exchange energy. The oscillations in \( J(r) \) produce a distribution in both magnitude and sign of \( J(r) \) between spins. The presence of both ferromagnetic and anti-ferromagnetic interactions means that some spins will be unable to satisfy competing ordering instructions, an effect known as 'frustration' (Toulouse, 1977). Frustration is thought to be essential for spin glass order.

\underline{disordered ferromagnetism}

Dilute ferromagnetic alloys are seen only in Pd and Pt hosts, where the large exchange enhancement of the d-band susceptibility makes \( J(r) \) ferromagnetic for large impurity separations (Nieuwenhuys, 1975; Mydosh and Nieuwenhuys, 1980). An example is \( \text{PdMn} \), where at low concentrations the Mn form 'giant moments', consisting of a localized moment on the Mn, along with a polarization cloud induced in the surrounding Pd d-band conduction electrons. The total giant moment of Mn in Pd is 7.8\( \mu_B \), described
by a spin $S=5/2$, and an effective g-factor $g_{\text{eff}}=2.7$ (Star et al., 1975). The indirect, long-ranged ferromagnetic exchange between giant moments competes in PdMn with a direct, short-ranged anti-ferromagnetic exchange (Moriya, 1967). The presence of competing interactions introduces frustration to the spin system; thus PdMn is an example of a disordered ferromagnet. The spin state disorder is evident in the spontaneous magnetization of PdMn(2.45 at.%), which is considerably smaller than the high field saturation magnetization (Star et al., 1975). The difference is attributed to anti-ferromagnetically coupled spins.

The disordered ferromagnetic phase is limited to concentrations $\leq 3$ at.% In this concentration range, critical phenomena are observed at the Curie temperature $T_C$ (Boerstoel et al., 1972; Ho et al., 1981a; Verbeek et al., 1980). A typical example, PdMn(2 at.%), has been chosen for study by $\mu$SR.

At higher concentrations, the average Mn spacing decreases, so that direct anti-ferromagnetic interactions are increasingly important. For $\geq 5$ at.%, the ordering appears similar to that in canonical spin glasses (Coles et al., 1975; Ho et al., 1981b; Ho et al., 1981c). PdMn(7 at.%) has been chosen for study by $\mu$SR for comparison with the disordered ferromagnetic phase, and as an example of a non-RKKY spin glass. The competing interactions in PdMn come not from the oscillations in the RKKY interaction, but from a competition between the indirect ferromagnetic and direct anti-ferromagnetic couplings.
A complicated, 'mixed' region, with no well-defined ordering temperature, is found for concentrations between 3 and 5 at.% (Zweers and van den Berg, 1975; Coles et al, 1975).

**Reentrant ferromagnetism**

Reentrant ferromagnetism refers to materials with two transitions, an upper transition from the paramagnetic to ferromagnetic state at $T_c$, and a lower reentrant transition from the ferromagnetic to a spin glass state at $T_g$. The possibility of a double transition was first predicted theoretically by a mean field theory of systems with random exchange (Sherrington and Kirkpatrick, 1975). Since then, a wide variety of materials have been found to have a double transition, suggesting that reentrant ferromagnetism is a common feature of magnetic systems with competing interactions (Nieuwenhuys et al, 1979).

Early measurements (Howarth et al, 1979) on PdMn in the mixed ordering region between ferromagnetic and spin glass order suggested reentrance, but later ac susceptibility data were interpreted as evidence against a double transition (Ho et al, 1981c). Verbeek et al (1978) discovered that the addition of a small amount of Fe to PdMn increases the ferromagnetic transition temperature while leaving the spin glass transition line of the phase diagram unchanged, and thereby produces a system with a clear double transition. This PdFeMn alloy has been studied in this work.
by μSR to investigate the reentrant transition and its effect on properties of the ferromagnetic state.

**Spin offs of spin glass research**

Spin glasses and the related disordered ferromagnets are of interest as model systems for the study of disorder effects in magnetism, and more generally, for the development of theoretical approaches to the effects of disorder in condensed matter physics. Spin glass research has stimulated the development of new theoretical concepts in statistical mechanics, which are beginning to find applications in a wide range of subjects. The most important of these is a generalization of the standard mean field theory to include systems with random exchange bonds (Chap. 4). The numerical technique of simulated annealing was first used to 'cool' a system of spins with random exchange couplings, but has also been applied to computer design and the traveling salesman problem (Kirkpatrick et al., 1983). Spin glass theory has been applied to the study of Ising random field systems (Richards, 1984). The spin glass ground state is known to have a large degeneracy of metastable states, due to the frustration of bonds. This has led to a spin glass model for neural networks, where the metastable state structure provides a content-addressable memory (Hopfield, 1982; Amit et al., 1985). A frustrated Hamiltonian has been proposed as a model for the surface of a crystal in the presence of
screw dislocations, with the possibility of comparing typical spin
glass phenomena with experiments on surfaces (Sachas, 1985). These
examples show the wide range of application of the spin glass state
as a model system for the study of disorder in complex, many body
systems.
Chapter II. Experimental Technique

A. The muon in a solid

The technique of muon spin relaxation (or \( \mu \text{SR} \)) is based on the implantation of positively charged muons into a solid. Relevant muon properties are listed in Table 2.1. From the standpoint of \( \mu \text{SR} \), the muon is a light isotope of the proton, with spin 1/2 and a magnetic moment. However, the muon is unstable, and decays with a lifetime of 2.2\( \mu \text{s} \) to a positron and two neutrinos.

Muons are produced from the decay of pions, which are in turn produced by high energy proton collisions. The details of muon production have been recently reviewed by Karlsson (1982) and Grynszpan (1984). The important characteristic of the pion decay is that it produces a spin polarized muon beam, with a typical momentum of 30-80MeV/c. As muons enter the sample, they lose energy by producing vacancies (radiation damage) in the sample, and by muon-electron scattering. The muon energy will eventually drop below a threshold for vacancy production. Brice (1978) estimated that after dropping below this threshold, the muon will continue to travel 100-600A before stopping. Thus the muon is unlikely to interact with its own radiation damage track. The entire thermalization process takes place in a time on the order of 10\(^{-10}\)s, and preserves the muon polarization (Eisenstein et al, 1966).


**TABLE 2.1. Muon properties.**

<table>
<thead>
<tr>
<th>Property</th>
<th>Value</th>
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<tbody>
<tr>
<td>Mass</td>
<td>105.7 MeV/c²</td>
</tr>
<tr>
<td>Spin</td>
<td>1/2</td>
</tr>
<tr>
<td>Magnetic moment</td>
<td>$4.84 \times 10^{-3} \mu_B$</td>
</tr>
<tr>
<td>Lifetime</td>
<td>2.2 μs</td>
</tr>
<tr>
<td>Gyromagnetic ratio</td>
<td>13.554 kHz/G</td>
</tr>
</tbody>
</table>

**TABLE 2.2. Muon diffusion parameters in Ag and Pd.**

<table>
<thead>
<tr>
<th></th>
<th>Ag</th>
<th>Pd</th>
</tr>
</thead>
<tbody>
<tr>
<td>ν₀ (sec⁻¹)</td>
<td>$10^{13.7}$</td>
<td>$10^{9.8}$</td>
</tr>
<tr>
<td>Δ (K)</td>
<td>3400</td>
<td>850</td>
</tr>
<tr>
<td>$\tau_\mu$ ψhop(150K)</td>
<td>.016</td>
<td>48</td>
</tr>
<tr>
<td>$\tau_\mu$ ψhop(80K)</td>
<td>$3.8 \times 10^{-11}$</td>
<td>0.34</td>
</tr>
</tbody>
</table>

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*b* Dodds et al (1982), Stein (1982)
When the muon has slowed to thermal velocity, its subsequent state depends largely on the electronic structure of the material. In an insulator or semiconductor, the muon will typically capture an electron into a 1s state, forming muonium, an analog of the hydrogen atom with the muon replacing the proton. In a metal, the Coulomb potential of the muon is screened by the conduction electrons, and can form only virtual bound states.

The potential energy of the muon in simple metals has been calculated from a pseudopotential method (Estreicher and Meier, 1983). For the case of Ag, these calculations show that the muon can localize in the potential energy minimum of the octahedral site, but not in the tetrahedral site. The same conclusions follow from a much simpler model for the potential energy $U(\hat{r})$, based on an electrostatic repulsion of the muon from the Ag$^+$ ion cores, screened by the conduction electrons:

$$U(\hat{r}) = \sum_j \frac{Ze^2}{|\hat{r} - \hat{R}_j|} \exp\left(-\frac{|\hat{r} - \hat{R}_j|}{r_s}\right). \quad (2.1)$$

The screening length $r_s$ is given by the Thomas-Fermi theory for free electrons (Mott, 1936), and $Z=1$ for Ag. The resulting energy profiles for several symmetry directions are shown in Fig. 2.1. The potential energy minimum at the octahedral site can be approximated by a parabolic potential. The ground state of the muon in this harmonic oscillator potential has a zero point of energy
FIG. 2.1. Muon potential energy profiles in a simple metal, fcc lattice. 0 marks octahedral sites, T marks tetrahedral sites. The horizontal line is the estimated muon ground state energy in the octahedral energy well.
$E_0=0.28$ eV, and is indicated on Fig.2.1. The expectation value $<r>$ gives a spatial extent of the muon wavefunction, $<r>=0.31$ Å. The general shape of the energy profiles in Fig.2.1 and the muon zero point energy are similar to the pseudo-potential results of Estreicher and Meier.

Muon localization in the octahedral site has been confirmed experimentally in Cu (Camani et al, 1977), but not in Ag or Pd. The calculation of Fig.2.1 suggests that the tetrahedral site cannot localize the muon in any fcc lattice with a simple metal electronic structure. We therefore assume the muon occupies an octahedral site in the Ag and Pd based alloys.

The muon can diffuse through the lattice by thermally activated hopping through tetrahedral sites. The hopping rate can be written in the Arrhenius form, $v_{\text{hop}}=v_0 \exp(-\Delta/T)$. The parameters $v_0$ and $\Delta$ are listed in Table 2.2 for Ag and Pd, along with the average number of hops in the muon lifetime at 80K and 150K. The small number of average hops shows that the effect of muon diffusion is negligible below 150K in Ag and 80K in Pd, which is the temperature range of interest in this work.

B. $\mu$SR measurements

The experimental technique of $\mu$SR has been discussed in several reviews (Karlsson, 1982; Chappert, 1984). A brief summary of the time-differential $\mu$SR technique will be presented here,
along with details of the apparatus and data analysis techniques. In Sec. A, we saw how spin polarized muons are implanted into interstitial sites in a metal. To measure the subsequent interaction of the muon spin with the local magnetic field, we need to detect the muon polarization as a function of time. This is achieved by detecting the angular distribution of the positrons produced by the muon decay. Suppose the decay positron momentum makes an angle $\theta$ with the muon spin at the time of decay. Then the probability that the positron momentum will lie in a solid angle $d\Omega$ at $\theta$ is $dN = (1 + A \cos \theta) d\Omega$, where $A$ is the asymmetry of the decay. The asymmetry has a maximum of one for the most energetic positrons, but since the positron detectors average over a range of energies, the average asymmetry is less, typically 0.2. In summary, three muon characteristics make the measurement of muon spin relaxation possible: (1) the muon beam is spin polarized, (2) the muon has a magnetic moment, and (3) the time decay of the muon polarization can be monitored from the asymmetry of the decay positron momentum distribution.

**spectrometer description**

\textit{uSR} experiments were performed at the stopped muon channel of the Los Alamos Meson Physics Facility in Los Alamos, New Mexico. The transverse field spectrometer configuration is indicated schematically in Fig.2.2. The sample is surrounded on four sides
FIG. 2.2. Schematic view of transverse field configuration $\mu$SR spectrometer.

1. Sample
2. BE (beam) counter
3. M (muon) counter
4, 5, 6. Three counters of the forward positron telescope.
by three successive boxes of plastic scintillator, connected by light pipes to photomultiplier tubes. The three scintillators in each of four directions from the sample form a 'positron telescope'; they are connected by a coincidence requirement so that only particles originating in the target and passing through all three scintillators are accepted as decay positrons. The coincidence requirement greatly reduces the background due to particles not originating in the sample.

The incoming muon beam is detected by two plastic scintillators (labeled BE and M). Muons detected in these counters, but not in the positron telescopes, are assumed to have stopped in the sample. This event sends a start signal to a time-to-amplitude converter (TAC), which is stopped when the decay positron is detected in a positron telescope. The TAC output is digitized and histogrammed for each telescope in a PDP 11/45 computer for many muon decay events. The result is a histogram of the time difference from a muon arrival to its decay. Pileup rejection is used to ensure that each positron can be associated with a particular parent muon. This is done by rejecting any event where a muon arrives less than 10μs (many muon lifetimes) before or after the arrival of another muon.

The Helmholtz magnet produces a static field up to 5kG, homogeneous to within 10ppm over the sample volume. Sample temperature is controlled from room temperature to 2.5K with a Helitran cold finger cryostat. Temperature is measured by a
carbon-glass resistor in good thermal contact with the sample, and is kept constant to better than 1% during data acquisition. Experiments below 2.5K in one sample (PdFeMn) were performed in a $^3$He-$^4$He dilution refrigerator, designed and constructed at Los Alamos National Laboratory, to be described in detail in a forthcoming publication (Cooke et al., 1985).

**transverse field $\mu$SR**

In transverse field $\mu$SR, the applied field is perpendicular to the initial muon polarization. The four positron telescopes give four time differential histograms, which represent the time distribution of the probability of observing a decay positron in each telescope. The asymmetry of the muon decay with respect to the precessing muon spin orientation superimposes a sinusoidal variation on the exponential decay of the muon. The number of events $N(t)$ in the histograms has the form

$$N(t) = BG + N_0 \exp(-t/\tau) \left[ 1 + A f(t) \cos(\omega t + \phi) \right], \quad (2.2)$$

where BG is a time independent background, $N_0/BG$ is the signal to noise ratio (typically ~50), and $A$ is the asymmetry of the muon decay at $t=0$. The muon Larmor precession frequency is $\omega = \gamma_\mu H_0$, where $H_0$ is the applied field, and $\phi$ is a phase which indicates the
angle between the initial muon polarization and the axis of the positron telescope.

The function \( f(t) \) is the transverse relaxation function, and is identical to the free induction decay function in an NMR experiment. The measurement of \( f(t) \) in a transverse field is identical to an NMR measurement of the spin-spin relaxation time \( T_2 \). We will often use an exponential form \( f(t) = \exp(-t/T_2) \). The \( T_2 \) time and the other unknowns (BG, \( N_0 \), \( A \), \( \omega \), and \( \phi \)) are determined by fitting Eq.(2.2) to the observed \( N(t) \) for each histogram using a non-linear least squares fitting procedure.

**longitudinal and zero field \( \mu \)SR**

In the longitudinal field configuration, a field may be applied parallel to the initial muon polarization, but such a field is not necessary to observe the muon depolarization. The positron time differential spectrum is recorded only for the forward and backward positron telescopes (0° and 180° to the initial muon polarization direction). The forward spectrum \( F(t) \) and the backward spectrum \( B(t) \) have the form

\[
F(t) = BG_1 + N_1 \exp(-t/\tau_\mu) \left[ 1 + A_1 \ P(t) \right], \quad (2.3)
\]
\[
B(t) = BG_2 + N_2 \exp(-t/\tau_\mu) \left[ 1 - A_2 \ P(t) \right], \quad (2.4)
\]

where \( P(t) \) is the longitudinal depolarization function. The
measurement of \( P(t) \) in a longitudinal field is identical to an NMR measurement of the spin-lattice relaxation time \( T_1 \). The capability of a zero field measurement of \( P(t) \) is unique to \( \mu \text{SR} \). Throughout this thesis we will use \( P(t) \) for the depolarization function in zero or longitudinal applied field, and \( f(t) \) for the transverse relaxation function (or free induction decay function) measured in a transverse applied field.

Two analysis procedures are possible to fit a trial form of \( P(t) \) to the data. If \( P(t) \) is relaxed within \( \approx 2\mu \text{s} \), then \( P(t) \) can be determined by a 'separate histogram' fit. In this method, \( P(t) \) and \( B(t) \) are separately but simultaneously fit to the expressions above for the six parameters \( B_{g1,2}, N_{1,2}, A_{1,2} \), and for the parameters of \( P(t) \). If \( P(t) \) relaxes more slowly, then this procedure becomes inaccurate, due to the difficulty in separating a slow relaxation from the constant background. For this case additional information is required; namely, reliable estimates of \( A_{1,2} \), and of the ratio \( \alpha = N_1/N_2 \). We note that these quantities are substantially independent of the depolarization \( P(t) \); they depend on such factors as the geometrical alignment of the sample and counters, and the muon stopping distribution in the sample. An experimental estimate of these quantities is made by a transverse field \( \mu \text{SR} \) measurement in 100G, using small auxiliary field coils. The resulting values of \( A_{1,2} \) and \( \alpha \) are assumed to be constant over an ensuing series of longitudinal measurements of \( P(t) \) and \( B(t) \). The experimental depolarization function is then defined as
\[ A \, P(t) = \frac{F(t) - \alpha \, B(t)}{A_2 \, F(t) + \alpha \, A_1 \, B(t)} \]  

(2.5)

where \( A \) is the average of \( A_1 \) and \( A_2 \), and constant backgrounds \( B G_1 \) and \( B G_2 \) have been subtracted from \( F(t) \) and \( B(t) \). The experimental \( A \, P(t) \) can be fit to various forms to obtain the relaxation rate. Finally, \( A_{1,2} \) and \( \alpha \) are again measured with a transverse field after a series of longitudinal experiments, to gauge their variation. To minimize systematic errors, the error in the relaxation rate due to deviations in \( A_{1,2} \) and \( N_{1,2} \) should be less than the statistical error in fitting \( P(t) \). This condition is satisfied for the data reported in this thesis.

C. Sample preparation and analysis

The Pd based samples studied in this work were prepared by collaborators at the Kamerlingh Onnes Laboratory of the University of Leiden, The Netherlands. After arc-melting ultrapure materials in an Ar atmosphere, the samples were rolled and spark cut into polycrystalline discs of typical size 3mm thick and 30mm diameter. (The PdFeMn(5 at.\%) sample was a square of comparable dimensions). The PdMn samples were annealed at 800°C for 1 hour, then returned to room temperature within 15 min. The PdFeMn sample was initially annealed for 22 hours at 1000°C. Since this sample was prepared in 1978 (six years before these experiments) an additional annealing
of several hours at 1000°C followed by a quench was made just before the measurements, to reduce any atomic clustering due to aging.

The Ag based samples were prepared by collaborators at the Los Alamos National Laboratory. Ultrapure materials (6N Ag and >4N Mn) were arc melted, annealed at 800°C for 1 hour, then rapidly quenched. Sample sizes were similar to the Pd samples.

The Mn concentration in AgMn and PdMn samples was measured by atomic absorption spectroscopy and electron microprobe analysis at Los Alamos. The Mn and Fe concentration in the ternary PdFeMn alloy was measured by atomic absorption spectroscopy at the Kamerlingh Onnes Laboratory. The analyzed concentrations shown in Table 2.3 are in good agreement with the nominal concentrations in all cases. No Mn inclusions were found in the microprobe scan of the PdMn(2 at.%) sample surface. Two such inclusions were found in the AgMn(1.6 at.%) sample, each a few μm in diameter. The inclusion volume is estimated to be <10⁻⁶ of the sample volume, and so has a negligible effect on the Mn concentration and the μSR results.

D. ac susceptibility measurements

ac susceptibility measurements on the AgMn samples were performed by D. E. MacLaughlin at the University of California, Riverside. The temperature of the susceptibility cusp, T_g, is
<table>
<thead>
<tr>
<th></th>
<th>AgMn</th>
<th>PdMn</th>
<th>PdFeMn</th>
</tr>
</thead>
<tbody>
<tr>
<td>nominal c (at.%)</td>
<td>1.6</td>
<td>3.0</td>
<td>2.0</td>
</tr>
<tr>
<td></td>
<td></td>
<td>7.0</td>
<td>0.35 Fe</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>5.0 Mn</td>
</tr>
<tr>
<td>c (at.%) atomic absorption analysis</td>
<td>1.57</td>
<td>2.04</td>
<td>0.37</td>
</tr>
<tr>
<td>c (at.%) electron microprobe analysis</td>
<td>1.71(3)</td>
<td>2.05(6)</td>
<td></td>
</tr>
<tr>
<td>ac susceptibility cusp Tg (K)</td>
<td>7.6(1)</td>
<td>11.8(2)</td>
<td>4.8</td>
</tr>
<tr>
<td>c (at.%) from Tg</td>
<td>1.6^a</td>
<td>2.8^a</td>
<td>7.3^b</td>
</tr>
</tbody>
</table>

^a Cannella and Mydosh, (1974)
^b Ho et al, (1981c)
shown in Table 2.3, along with the corresponding concentration from the literature. The PdFeMn ac susceptibility was measured by J. A. Mydosh at the Kamerlingh Onnes Laboratory. Results will be shown in Chap.8.

Additional measurements were made with a susceptibility apparatus constructed at Rice University. A measuring coil, consisting of a primary with 1800 turns and two secondaries, each with 3650 turns was wound by M. Murphy. The primary was driven at 23Hz by a PAR HR-8 lock-in amplifier. The ac field produced by the primary at the sample location was estimated to be 2G. The two secondaries were connected in opposition and the induced voltage measured by a mutual inductance bridge built from a design by Corson (1982), with the lock-in used as a null detector. The bridge circuit was calibrated with a paramagnetic salt, and provides a sensitivity of 5x10^{-7} emu. The maximum sample volume is 0.5 cm³.

E. X-ray measurements

The lattice parameter of Pd based alloys was measured with a Debye-Scherrer camera. A carbide file was used to make filings from the µSR samples. The filings were sieved to #320 mesh and annealed for one hour at 800°C under vacuum. The samples were mounted in the camera on the outside of a glass fiber lightly coated with Vaseline. Exposures were made with filtered Mo Kα
radiation. Plots of lattice parameter vs. the Nelson-Riley extrapolation function for the back angle diffraction lines showed negligible slope, indicating very small systematic errors. Results will be presented in Chap. 5.
Chapter III. Theory of the dilute alloy static linewidth

A. Nuclear dipolar broadening

Spin-lattice relaxation times of nuclear moments in Ag, Pd and Cu are very much longer than the muon lifetime. For example, \( T_1 = 10.8/T(K) \) in Ag (Carter et al, 1977). The addition of Mn local moments will shorten \( T_1 \) due to the coupling of local moments to nuclei, but \( T_1 \) is still much longer than the muon lifetime (McHenry et al, 1972). During the muon lifetime, the nuclear moments produce a distribution of quasistatic dipole fields at the muon sites. Following Hayano et al (1979), the zero applied field muon depolarization rate due to nuclear dipoles in Ag, Pd and Cu are .022 \( \mu s^{-1} \), .043 \( \mu s^{-1} \), and .44 \( \mu s^{-1} \), respectively. The rate in Ag and Pd is too small to cause significant depolarization within the lifetime of the muon, whereas the rate in Cu is large enough to observe the quasistatic nuclear depolarization. The extra complication of nuclear depolarization makes CuMn dilute alloys less suitable than AgMn and PdMn for \( \mu SR \) studies of magnetic ordering.

The spin-lattice relaxation time of the Mn nuclear moment is very short, due to the strong hyperfine coupling to the fluctuating electronic moment. In zero field (Walstedt and Narath, 1972),
\[ \frac{1}{(T_1)_{\text{nucl}}} = \frac{2}{3} S(S+1) \gamma^2 H_{hf}^2 \tau_e, \quad (3.1) \]

where \( \tau_e \) is the electronic spin correlation time, \( \gamma = 1.050 \text{kHz/G} \) for \(^{55}\text{Mn} \), and the hyperfine field \( H_{hf} \) can be determined from nuclear orientation data. In \( \text{AgMn} \), \( H_{hf} = 313 \text{kG} \) (Cameron et al, 1966), and in \( \text{PdMn} \), \( H_{hf} = 380 \text{kG} \) (Khoi et al, 1976). The Korringa time, as measured in \( \text{AgMn} \) by neutron scattering (Murani, 1981) gives a lower limit of \( \tau_e = 0.17 \text{ns-K/T} \) for the correlation time. (The correlation time can be longer due to cooperative effects near a transition). The resulting nuclear \( T_1 \) is very short, \( T_1 = 0.23 \text{ns-K}^{-1} \text{T} \). These rapid fluctuations in the nuclear moments will cause an exponential depolarization of the muon with rate

\[ \frac{1}{(T_1)_{\mu}} = 2 \Delta^2 (T_1)_{\text{nucl}} \quad (3.2) \]

where \( \Delta \) is the calculated zero field relaxation rate for quasistatic Mn moments. As a typical example, in \( \text{AgMn}(5 \text{ at.\%}) \), \( \Delta = 0.10 \mu \text{s}^{-1} \), giving \( (T_1)_{\mu} = 0.22 \text{ s-K/T} \), an extremely long relaxation time. Even if \( \tau_e \) were \( 10^3 \) times longer than the Korringa estimate, the relaxation due to Mn nuclear moments would still be negligible, and would in fact be dominated by dipolar coupling to the much larger electronic moments. Thus the data presented here are unaffected by either impurity or host nuclear moments.
B.Muon-impurity coupling

The dominant static broadening mechanism in the dilute alloys studied here is due to the random distribution of impurity electronic moments. Suppose a single impurity has a normalized spin polarization ̂\hat{s} = <\hat{s}>/S, and is located at ̂\hat{r} from the muon. The muon can couple to this impurity through both a direct dipolar interaction and an indirect RKKY interaction (Ruderman and Kittel, 1954; Yosida, 1957). To calculate the strength of the RKKY contribution to the coupling, it is necessary to make a number of assumptions. We assume that the host metal can be described by a free electron approximation, and that the muon-impurity separation r is large enough that we can use the simple cosine asymptotic form for the RKKY field. Then the local field \hat{H}_μ at the muon is

\[ \hat{H}_μ = \{ B \ [ 3\hat{s} - 3\hat{s}(\hat{s} \cdot \hat{r}) \] + A \cos(k_F r + \phi) \hat{s} \} S/r^3. \]  (3.3)

The constant B = \gamma_μ g μ_B is the strength of the dipolar coupling, where \gamma_μ is the muon gyromagnetic ratio, g is the impurity spin g-factor, and μ_B is the Bohr magneton. The constant A is the strength of the RKKY interaction, and k_F is the Fermi wavevector for the host metal.

The RKKY interaction strength can be calculated in the s-d exchange model (Yosida, 1957). In this model, the constant A can be expressed in terms of an exchange energy J(0), which expresses
the coupling strength between the impurity moment and the conduction electrons, and the muon Knight shift $K_{\mu}$, which expresses the strength of the contact interaction between the muon and the conduction electrons. A modification of Yosida's form to include an exchange enhanced host band susceptibility is discussed by Walstedt and Walker (1975). If the uniform electronic susceptibility is enhanced by a factor $(1-\alpha)^{-1}$, then

$$A = \frac{K_{\mu} Y_{\mu} J(0)}{\frac{1}{8} \pi \rho \mu_B} \frac{1-\alpha}{(1-0.295\alpha)^2}$$  \hspace{1cm} (3.4)$$

where $\rho$ is the density of lattice sites in the host. Muon Knight shifts in Ag, Pd and Cu are presented in Table 3.1, along with estimates of the exchange enhancement factor $\alpha$.

The $s$-$d$ exchange energy can be estimated by a first principles calculation from the integral definition (Yosida, 1957)

$$J(0) = 2N \int d^3r_1 \int d^3r_2 \psi^*(0,r_2) \psi^*(3d,r_1) \psi(3d,r_2) \phi(0,r_1)$$

$$x \frac{e^2}{|r_1-r_2|} \psi(3d,r_2) \phi(0,r_1),$$  \hspace{1cm} (3.5)$$

where $N$ is the number of lattice sites, $\phi(k,r)$ is a free electron wave function for the conduction electrons, and $\psi(3d,r)$ is the wave function for the Mn 3d orbital. We can approximate $\psi(3d,r)$ with the calculated 3d Hartree-Fock orbital for neutral atomic Mn.
TABLE 3.1. Mn s-d exchange energies in Ag, Pd and Cu, and the mixed to dipolar linewidth ratio for the paramagnetic and spin glass states.

<table>
<thead>
<tr>
<th></th>
<th>Ag</th>
<th>Pd</th>
<th>Cu</th>
</tr>
</thead>
<tbody>
<tr>
<td>Hartree-Fock J(0) (eV)&lt;sup&gt;a&lt;/sup&gt;</td>
<td>1.5</td>
<td>1.7</td>
<td>2.1</td>
</tr>
<tr>
<td>NMR J(0) (eV)&lt;sup&gt;b&lt;/sup&gt;</td>
<td>1.5</td>
<td></td>
<td>2.0</td>
</tr>
<tr>
<td>μSR J(0) (eV)&lt;sup&gt;a&lt;/sup&gt;</td>
<td>2.9(1)</td>
<td></td>
<td></td>
</tr>
<tr>
<td>$K_\mu$ (ppm)&lt;sup&gt;c&lt;/sup&gt;</td>
<td>94</td>
<td>200</td>
<td>60</td>
</tr>
<tr>
<td>α</td>
<td>0.&lt;sup&gt;d&lt;/sup&gt;</td>
<td>.95&lt;sup&gt;e&lt;/sup&gt;</td>
<td>.12&lt;sup&gt;b&lt;/sup&gt;</td>
</tr>
<tr>
<td>Hartree-Fock A/B</td>
<td>0.89</td>
<td>0.18</td>
<td>0.52</td>
</tr>
<tr>
<td>$(T_2^*)^{-1}$ mixed : paramagnet</td>
<td>1.2&lt;sup&gt;h&lt;/sup&gt;</td>
<td>1.01</td>
<td>1.05</td>
</tr>
<tr>
<td>$(T_2^*)^{-1}$ dipolar spin glass</td>
<td>1.06</td>
<td>1.005</td>
<td>1.02</td>
</tr>
</tbody>
</table>

<sup>a</sup> this work

<sup>b</sup> Walstedt and Walker (1975)

<sup>c</sup> Schenck (1981)

<sup>d</sup> Vier et al (1984)

<sup>e</sup> Parra and Medina (1980)
(Fischer, 1972). A numerical evaluation of the integral gives \( J(0) = 100. \text{ eV/a}^3(A) \), where \( a \) is the fcc lattice parameter. Results for Ag, Pd and Cu are presented in Table 3.1.

Experimental values of \( J(0) \) from host NMR linewidths, as interpreted by Walstedt and Walker (1975) are shown in Table 3.1. A Korringa \( s-d \) exchange energy \( J_K \) can be determined from quasielastic neutron scattering or ESR linewidth measurements of the Korringa relaxation time \( \tau_0 = \hbar / T \). If \( \rho(E_F) \) is the density of states per spin at the Fermi surface, then

\[
b^{-1} = \pi \frac{k_B}{\hbar} \rho^2(E_F) J_K^2. \tag{3.6}
\]

The exchange energies \( J(0) \) and \( J_K \) are related through the potential scattering phase shifts, which are not reliably known (Walstedt and Walker, 1975). Alternatively, we can assume that \( l=2 \) partial wave scattering dominates the exchange interaction and approximate \( J(0) = \sqrt{5} J_K \) (Murani, 1981), but this relationship is not satisfied in AgMn (Heffner et al, 1985). We conclude that Korringa relaxation time measurements do not give adequate predictions of \( J(0) \).

The table shows good agreement between the NMR estimates of \( J(0) \), and the simple Hartree-Fock calculation for Ag and Cu. Using the Hartree-Fock \( J(0) \), \( K_H \), and \( \alpha \), we calculate the ratio of the RKKY to dipolar interaction strength \( A/B \) in Table 3.1. The
resulting RKKY contribution to the muon linewidth is calculated in the next section.

C. Transverse applied field $\mu$SR - paramagnetic state

In a dilute magnetic alloy the measured transverse field relaxation rate $T_2^{-1}$ has two contributions: (a) relaxation produced by a coupling of the muon to the static spin polarization induced on the impurity moments by the applied field, and (b) relaxation produced by the finite fluctuation time of the impurity moments. In this section we calculate the relaxation from contribution (a) only, which we will write as $(T_2^a)^{-1}$ to emphasize that it is the static contribution to the relaxation. Contribution (b) can be ignored if the impurity spin correlation time $\tau_0$ is very much shorter than the muon Larmor period. Since this condition is almost always satisfied, the relaxation rate $T_2^{-1}$ is generally dominated by $(T_2^a)^{-1}$. In Sec.D we calculate the relaxation when both contributions are significant.

The static relaxation rate $(T_2^a)^{-1}$ was calculated by Walstedt and Walker (WW - Walstedt and Walker, 1974) for both dipolar and RKKY couplings. They assumed the impurities to be randomly distributed in the host lattice at a concentration $c$, specifically excluding any short ranged atomic ordering. The impurity spin polarization is oriented along the external applied field with magnitude $g \cdot \langle S_z \rangle / S$ chosen independently for each site from a
distribution function \( p(\phi) \). The spins cannot have a polarization transverse to the applied field \( \langle S_x \rangle = \langle S_y \rangle = 0 \). This restriction will require an important modification to the theory for the spin glass state. The external field \( H_0 \) is assumed to be much larger than the local fields at the muon produced by the surrounding impurities. With this assumption, we can ignore the components of the local field perpendicular to the applied field, and consider only the local field parallel to the applied field. We write this local field as a frequency shift \( \omega \) relative to the resonance frequency \( \gamma \mu H_0 \). The contribution to the frequency shift from an impurity located at \( \mathbf{r} \) from the muon is

\[
\omega = \{ A \cos(2k_Fr+\phi) + B \left[ 1 - 3(2\cdot\mathbf{r})^2 \right] \} S \sigma/r^3, \quad (3.7)
\]

where the applied field is in the \( z \) direction. The free induction decay function \( f(t) \) is then (WW)

\[
f(t) = \prod_j \left[ 1 - c + c \int d\phi p(\phi) \exp( i\gamma \mu H_0 t ) \right], \quad (3.8)
\]

where the index \( j \) runs over all sites in the host lattice. This equation is the starting point for the numerical calculations presented below.

In the dilute limit \( (c \ll 1) \), WW evaluated Eq. (3.8) with a continuum approximation for the lattice sum. The result is an exponential \( f(t) = \exp(-t/T_2) \) (Lorentzian lineshape \( g(\omega) \)), and for
pure dipolar muon-impurity coupling the relaxation rate (or linewidth) is

$$\frac{1}{T_2^*} = 5.065 \rho c B S \langle |\alpha| \rangle, \quad (3.9)$$

where \(\rho\) is the density of lattice sites in the host (inverse atomic volume) and \(\langle \rangle\) represents an average over the distribution \(p(\alpha)\). For purely paramagnetic spins, \(\langle S_z \rangle\) is the same at every site, so \(\langle |\alpha| \rangle = \langle S_z \rangle / S\), and the linewidth is proportional to the macroscopic magnetization.

The relaxation rate for mixed dipolar plus RKKY coupling requires a numerical evaluation of an integral, as discussed by WW. We can use their result, and the predicted strength of the RKKY interaction from Sec. B above to calculate the mixed linewidth. The result is expressed in terms of a ratio to the pure dipolar linewidth [Eq. (3.9)], and is given in Table 3.1. The RKKY interaction is a small correction to the dominant dipolar linewidth in \(\text{PdMn}\) and \(\text{CuMn}\), but is significant in \(\text{AgMn}\) (Heffner et al, 1985).

Several assumptions used to derive the analytic result of Eq. (3.9) may not be valid in a real dilute alloy. The importance of these assumptions has been investigated in a number of numerical calculations. In each of these calculations, we have assumed the muon occupies an octahedral site of an fcc lattice, as discussed in Chap. 2.
finite concentration

The effect on $f(t)$ of assuming $c \ll 1$ can be evaluated using Eq. (3.8), which is valid for any concentration. The product over lattice sites is performed over a large spherical volume around the muon for a particular orientation of the applied field with respect to the crystal axes. We obtain a polycrystalline average by averaging $f(t)$ from several carefully chosen field orientations which take into account the cubic symmetry. (Hasselbach and Spiering, 1980). The result is in good agreement with the dilute limit exponential [Eq. (3.9)] at concentrations up to $c = 0.03$. In Fig. 3.1 we compare the lattice sum calculation for $c = 0.07$ with the dilute limit exponential. At this high concentration, $f(t)$ differs from the dilute limit result by no more than 10-15%.

The lineshape $g(\omega)$ is obtained by a Monte Carlo procedure. Impurities are distributed at random on a lattice surrounding a central muon site. The total dipole field at the muon (parallel to the applied field) is calculated as a sum of the dipole field due to each impurity moment. The muon local field is calculated for many spatial configurations of the impurities, and the resulting values are histogrammed to obtain the lineshape function $g(\omega)$. A polycrystalline average is obtained by varying the field orientation as in calculating $f(t)$. The resulting $g(\omega)$ is in good agreement with the dilute limit Lorentzian for $c = 0.03$. Fig. 3.2 shows $g(\omega)$ for a much higher concentration of $c = 0.07$ (corresponding
FIG. 3.1. Paramagnetic state transverse field relaxation function for $c = 0.07$. The dashed line is the dilute limit exponential with rate $(T_2^*)^{-1}$ given by Eq. (3.9). The solid line is a polycrystalline average of Eq. (3.8).
FIG. 3.2. Paramagnetic state transverse field lineshape function for $c = 0.07$. The dashed line is the dilute limit Lorentzian with linewidth $(T_2)^{-1}$ given by Eq. (3.9). The solid line is calculated from a Monte Carlo procedure as described in the text with a polycrystalline average.
to \( f(t) \) from Fig. 3.1) and the Lorentzian lineshape predicted by the
dilute limit theory. Small satellite peaks are seen in the wings
of the lineshape, but the overall shape and width is in good
agreement with the WW prediction. We conclude that the dilute
limit theory is accurate up to 3 at.\%, and is even applicable to
7 at.\% to within an error of approximately 10%.

**muon-giant moment coupling**

Mn and Fe impurities in the exchange enhanced Pd host induce a
giant moment polarization cloud localized around the impurity. If
these polarization clouds are well separated in space (low
concentrations), then we would expect the muon coupling to the
polarization cloud to make a contribution to the relaxation rate.
At higher concentrations, the polarization clouds will tend to
overlap and merge into a quasi-uniform background of spin
polarization. The muon coupling to a uniform spin polarization
does not contribute to the relaxation rate since it is the same at
every muon site (it produces a frequency shift). This argument was
used in Dodds et al (1983) to argue against muon-giant moment
coupling in \( \text{PdMn}(2 \text{ at.\%}) \).

The possibility of a giant moment contribution to the muon \( T_2 \)
has been investigated numerically for \( \text{PdFe} \), where there is detailed
information on the spatial extent of the polarization cloud from
neutron diffraction experiments (Hicks et al, 1968). Medina and
Parra (1982), developed a model for the Fe giant moment (in agreement with the neutron data), which assumes that the polarization cloud is due to localized moments induced on the Pd atoms surrounding the Fe impurity. This is an alternative to the more conventional description in terms of an exchange-enhanced RKKY interaction (Verbeek et al, 1980; Verbeek, 1979), and is more convenient for a dipole field calculation. We calculate the muon free induction decay function from Eq.(3.8) where now \( \omega_j \) is a dipole coupling to both the bare Fe impurity moment, and to the moment induced on each surrounding Pd atom (to a distance \( \approx 8\AA \) from the Fe). At a concentration of 0.3 at.% Fe, the result is not significantly different from the (exponential) free induction decay function due to bare Fe moments only. A second calculation with the Medina and Parra model shows that the moments induced on the Pd have a broad distribution in magnitude, in contradiction to the idea of a near-uniform giant moment polarization. The absence of a giant moment contribution to \( \mathcal{T}_2 \) is instead due to the extremely small moment \( <0.1\mu_B \), induced at each Pd site (Low and Holden, 1966). Since the dipole field falls off as \( r^{-3} \), only a small fraction of all the moments in the polarization cloud contribute significantly to the dipole field at the muon, and this small field is dominated by the field from the single large Fe moment. The total giant moment is significant only in a macroscopic measurement such as magnetization, which samples equally all of the many Pd sites which make up the polarization cloud. In view of the weaker
giant moment polarization for PdMn (Nieuwenhuys, 1975), we assume the muon-giant moment coupling is negligible in both PdFe and PdMn.

**impurity-impurity near neighbor exchange**

In the dilute limit theory, the spin polarization $\sigma = \langle S_z \rangle / S$ is allowed to vary from site to site according to a distribution $p(\sigma)$, but the values of $\sigma$ must be uncorrelated (independent spins assumption). This condition is violated if we allow an exchange coupling between near-neighbor (nn) impurity spins. In PdMn, the nn exchange is anti-ferromagnetic (Star et al, 1975; Smit et al 1979), and is important in creating the frustration which leads to the spin glass state. The magnitude of the nn exchange has been estimated from magnetization studies to be $J_{nn} = -25 \pm 10K$ (Smit et al, 1979), which can cause significant short range order in the paramagnetic state.

The possible effect of this short ranged magnetic order on the lineshape $g(\omega)$ in PdMn has been investigated with a Monte Carlo procedure similar to that discussed above under the effect of finite concentration. The Mn spins are distributed in a cubic volume of 10 lattice constants on a side for concentrations of up to 7 at.%. In each configuration, the Mn spins are separated into nn clusters. The spin polarization of each Mn is calculated by an exact summation of the cluster partition function, including a nn Ising coupling of strength $J_{nn} = -25K$. (At the highest
concentrations, spin 1/2 was used instead of spin 5/2 to avoid excessive computational time for the largest clusters.) The resulting correlated magnetization is used to calculate the dipole field at the muon site. The calculation is repeated for different Mn spin configurations, and the resulting values for the muon local field are histogrammed to give the lineshape. This procedure is a form of high temperature expansion, since in ignoring next near-neighbor and further correlations we cannot simulate the cooperative effects that become important near Tg.

The introduction of antiferromagnetic short-range order in this model produces a distribution in the local magnetization p(σ). The calculated linewidth is then found to be proportional to $|σ|$ according to Eq.(3.9), even in the presence of significant short-ranged magnetic order. Thus the presence of nn correlations in the spin polarization does not effect the dilute limit linewidth prediction of Eq.(3.9).

atomic short range order

Atomic short range order (SRO) describes the degree of correlation in the positions of impurity atoms on the lattice. Atomic SRO is known to exist in many dilute alloy systems, and can strongly effect the character of the magnetic order (Morgownik and Mydosh, 1983a) and the Curie-Weiss temperature, (Morgownik and Mydosh, 1983b). The degree of SRO is described by Cowley
parameters (Cowley, 1950) \( \alpha_i = (p_i - c) / (1 - c) \), where \( c \) is the impurity concentration and \( p_i \) is the probability that a lattice site in the \( i^{th} \) near-neighbor shell from an impurity at the origin is also occupied by an impurity. Positive values of \( \alpha_i \) mean that the probability for an impurity to be in the \( i^{th} \) shell from another impurity is greater than for a purely random distribution (clustering), and negative \( \alpha_i \) mean that this probability is smaller (anti-clustering).

\( \alpha_i \) can be determined experimentally by diffuse neutron or X-ray diffraction. The nn Cowley parameter in \( \text{PdMn}(10 \text{ at.}\%) \) is \( \alpha_1 = -0.058(2) \) (Ahmed and Hicks, 1974; Verbeek, 1979), indicating nn anti-clustering. X-ray diffraction measurements detected no SRO in \( \text{AgMn} \) for concentrations below 8 at.% (Bouchiat et al., 1981). Thus of the alloys studied in this work by \( \mu \text{SR} \), only the most concentrated \( \text{PdMn} \) alloy (7 at.%) is expected to have non-negligible atomic SRO.

A Monte Carlo procedure has been used to investigate the effect of atomic SRO on the linewidth in \( \text{PdMn}(7 \text{ at.}\%) \). Impurities were distributed at random in a cubic volume with periodic boundary conditions. The configuration energy of the impurities is described by a Hamiltonian,

\[
H = -\sum_{\langle i,j \rangle} V_{ij} s_i s_j \tag{3.10}
\]

where the sum is over all pairs of nn lattice sites, \( s_i \) is an
operator describing whether site i is occupied by an impurity, and $V_1$ is the atomic interaction energy for near neighbor impurity atoms. The configuration energy can be minimized by the Metropolis Monte Carlo procedure (Binder, 1979); at each Monte Carlo step, we choose an impurity at random, then move that impurity to an adjacent site. The new configuration is accepted with a Boltzmann probability $\exp(-\Delta E/T)$, where $\Delta E$ is the change in configuration energy in moving the impurity. We can vary $V_1/T$ and the number of Monte Carlo steps to produce the desired nn Cowley parameter $\alpha_1$. After the Monte Carlo procedure produces an impurity configuration with SRO, we calculate the dipole field at the muon from the impurity moments. The entire procedure is repeated many times, and the resulting field values are histogrammed to give the lineshape $g(\omega)$.

The lineshape for $c=0.07$ is shown in Fig.3.3. The dotted line is the dilute limit Lorentzian (no SRO). The solid line has $\alpha_1=-0.071$, similar to the anti-clustering observed in $\text{PdMn}$. The dashed line has $\alpha_1=+0.093$, a corresponding degree of clustering. The width of the central peak is clearly independent of the degree of SRO, and is close to the width of the dilute limit Lorentzian (dotted line). The satellite peak at $\omega T_2=5.5$ is due to muons adjacent to impurities. The intensity of this peak is dependent on the SRO, suggesting that as the impurities cluster, fewer muon sites are adjacent to impurities, and the intensity of this peak decreases. Since the observed $T_2$ is not significantly affected by
FIG. 3.3. Paramagnetic state transverse field lineshapes for $c = 0.07$ with atomic short ranged order. The dotted line is the dilute limit Lorentzian (no clustering). The solid and dashed lines are for anti-clustering and clustering, respectively.
satellite lines, we conclude that the linewidths are independent of
the degree of SRO, and are accurately calculated by the dilute
limit theory.

D. Impurity fluctuations and the static linewidth

The WW theory assumes that impurity spins fluctuate rapidly
enough that relaxation is dominated by the static linewidth \((T_2^\text{S})^{-1}\).
This rapid fluctuation assumption is generally satisfied in
paramagnets. An analysis of the relaxation when this condition is
not satisfied opens the possibility of directly observing the
impurity spin correlation time with transverse field µSR.

Suppose we have an ensemble of muons, all stopping at a
position \(\vec{r}\) from a single impurity (all stopping at the same site).
Under the rapid fluctuation assumption, the ensemble polarization
will precess in the total (applied plus dipole) static field, with
no dephasing. If we now allow the impurity spin to have a
correlation time \(\tau_c > 0\), then the ensemble polarization will dephase
with a rate given by the motional narrowing expression (Slichter,
1980);

\[
\frac{1}{\tau_2} = \gamma_\mu^2 \left\{ \left[ (H_{\text{dip}})_z + (H_{\text{RKKY}})_z \right]^2 + (H_{\text{dip}})_y^2 \right\} \tau_c, \quad (3.11)
\]

where we have assumed \(\gamma_\mu H_0 \tau_c \ll 1\), and
\[ \gamma_{\mu} (H_{\text{dip}})_{z} = B S \frac{1-3(\rho \cdot 2)^2}{r^3} \]

\[ \gamma_{\mu} (H_{\text{dip}})_{y} = -B S \frac{3(\rho \cdot 2)(\rho \cdot \nu)}{r^3} \]  

(3.12)

\[ \gamma_{\mu} (H_{\text{RKKY}})_{z} = A S \frac{\cos(2k_{F} r)}{r^3} \]

The contribution to \( f(t) \) from an impurity at site \( j \) now has both a precession term, described by the frequency shift \( \omega_j \) [Eq. (3.7)], and a damping term, described by \( \tau_2 \) [Eq. (3.11)]. Thus \( f(t) \) from Eq. (3.8) is generalized to

\[ f(t) = \prod_{j} \left[ 1 - c + c \exp\left(-\frac{t}{\tau_2 \omega_j}ight) + i\omega_j t \right]. \]  

(3.13)

The dilute limit continuum approximation to Eq. (3.13) does not have an analytic form. Instead, \( f(t) \) can be calculated numerically from Eqs. (3.11)-(3.13). Results are shown in Fig. 3.4 for \( c=0.01 \). The dashed line is the dilute limit exponential for \( \tau_2 = 0 \) (with relaxation time \( T_2^* \)), which is indistinguishable from the numerical calculation for \( \tau_2/T_2^*<10^{-4} \). As \( \tau_2/T_2^* \) increases, \( f(t) \) becomes non-exponential, and decays more rapidly than the \( \tau_2 = 0 \) exponential. We now consider two cases where a finite correlation time can have an observable effect on \( f(t) \).
FIG. 3.4. Paramagnetic state transverse field relaxation functions for $c=0.01$ with various impurity spin correlation times. The dashed line is the dilute limit exponential ($\tau_c=0.$), with relaxation rate $(T_2^*)^{-1}$. The solid curves from top to bottom have increasing correlation times: $\tau_c/T_2^* = 10^{-3}, 10^{-2}$ and $10^{-1}$. 
First consider the possibility of observing the non-exponential $f(t)$. For $\text{AgMn}$, we use the Korrina expression for the correlation time, $\tau_c = b/T$, and calculate the paramagnetic state $T_2^*$ from Eq. (3.9) assuming a Curie law for $<S_z>$, and the RKKY contribution from Table 3.1. Then

$$\frac{\tau_c}{T_2^*} = \frac{c b H_0}{T^2} 1.83 \times 10^5 \text{ K}^{-1} \text{ s}^{-1} \text{ cm}^{-1}.$$  (3.14)

The Korrina expression is only valid well above the spin glass transition temperature $T_g$. The largest values of $c/T_g^2$ are at low concentrations, so we use $c = 0.03$, $T = 3T_g = 6K$ (Cannella and Mydosh, 1974), and the largest available field of $H_0 = 5kG$. The Korrina rate as measured by neutron scattering is $b = 0.175 \text{ ns} \cdot \text{K}$ (Murani, 1981), which gives $\tau_c/T_2^* = 1.3 \times 10^{-5}$, which according to Fig. 3.4 is much too small to observe the non-exponential $f(t)$. A similar result holds for $\text{PdMn}$. The larger values of $b$ in rare earth-noble metal alloys are more favorable for observing the predicted effect. For example, in $\text{AuGd}$ the EPR linewidth slope gives $b = 7 \text{ ns} \cdot \text{K}$ (Rettori, Davidov and Kim, 1973). The spin glass transition temperatures in rare earth alloys are typically less than 1K for $c = 0.01$ (Aarts et al., 1980), so we use $c = 0.01$ and $T = 3K$, and assume pure dipolar coupling. Then at 5kG we have $\tau_c/T_2^* = 0.013$, which according to Fig. 3.4 will cause a significant correction to the exponential $\tau_c = 0$ theory. This prediction has not been tested with
μSR on a rare earth alloy. We note that this method of measuring \( r_C \) by μSR is not affected by the presence of a bottleneck in the EPR signal, thus providing an alternative method of measuring \( r_C \) in bottlenecked systems.

A finite correlation time can also have important consequences in the case of a small applied field. In the lattice sum of Eq.(3.13), the radius \( r_j \) has some minimum value \( r_0 \) which corresponds to a maximum in the relaxation rate \((r_2)^{-1}\). Now suppose \( r_C \) is small enough that \( \max(t/r_2)\ll 1 \). Then the lattice sum of Eq.(3.13) is approximately

\[
f(t) = \exp \left\{ - \sum_j \left[ t - \exp(i\omega t) \right] - c \sum_j \frac{t}{(r_2)_j} \exp(i\omega t) \right\} \quad (3.15)
\]

Since \( \omega_j (r_2)_j = <S_z^2> r_j^3 / B r_0 \) (ignoring factors of order one), this condition can generally be satisfied in a small field. To first order in \( \max(t/r_2) \), \( f(t) = \exp(-t/T_2) \), where

\[
\frac{1}{T_2} = \frac{1}{T_2} + \frac{1}{T_2'} \quad (3.16)
\]

and

\[
\frac{1}{T_2'} = c \sum_j \frac{1}{(r_2)_j} \quad (3.17)
\]

Since the static linewidth \((T_2)^{-1}\) is proportional to the applied field, Eq.(3.16) predicts that for small fields \( H \) a plot of \((T_2)^{-1}\)
vs. H will have a non-zero intercept given by Eq. (3.17), and a slope given by the WW theory for the static linewidth. This prediction is compared with AgMn data in Chap. 5.

E. Ordered state zero applied field \( \mu SR \)

In the ordered state of a dilute alloy, the impurity spins are polarized in the absence of an applied field. The resulting distribution of static fields at the muon causes relaxation of the zero field muon polarization. The formalism for treating zero field static depolarization in dilute alloys was first developed by Mikaelian and Smilga (MS - 1981). They generalized the WW method to calculate the distribution function \( g(\tilde{z}) \) for all components of the local field. (WW calculate the local field distribution function \( g(\omega_z) \) along the direction of the applied field.) The zero field muon depolarization function \( P(t) \) is determined directly from \( g(\tilde{z}) \).

\[
P(t) = \int d^3\omega \ g(\tilde{z}) \left[ \omega_2^2 + (\omega_x^2 + \omega_y^2) \cos \omega t \right] / \omega^2, \quad (3.18)
\]

where the initial muon polarization defines the \( \tilde{z} \) direction.

We now review the basic results from MS. As in the WW theory, we assume the impurities to be distributed on the lattice at random at a concentration \( c \ll 1 \). For convenience we consider only dipolar couplings; the generalization to include the RKKY interaction can
be found in MS. The impurity dipole moment is written as $g\nu B \hat{s}$, where $\hat{s} = \langle \hat{S} \rangle / S$ at each site is chosen independently from a distribution function $p(\hat{s})$, which will be chosen to reflect the type of magnetic ordering. The procedure in MS gives the Fourier transform of $g(\omega)$.

$$A(\omega) = \int d^3 \omega \; g(\omega) \; e^{i \omega \cdot \hat{s}},$$

$$A(\omega) = \exp\left\{ - \frac{\pi}{6} \rho c B S \int d\omega \; \left[ \langle \hat{\omega} \cdot \hat{\omega} \rangle - 3 \langle \hat{\omega} \cdot \hat{\omega} \rangle \langle \hat{\omega} \rangle \langle \hat{\omega} \rangle \right] \right\},$$

where $\langle \cdot \rangle$ is an average over the distribution $p(\hat{s})$. Equation (3.20) will be used to calculate $P(t)$ for three types of magnetic ordering.

**spin glass order**

This case was considered by MS. We model the spin glass state by assuming that for zero applied field the spins are randomly oriented, and have an arbitrary distribution of magnitudes $p(o)$. Then $A(\omega)$ depends only on the magnitude $q$, $A(\omega) = \exp(-a_s q)$, where $a_s$ is the zero field static linewidth.

$$a_s = K_{sg} \rho c B S \langle |\hat{\omega}| \rangle.$$  

(3.21)

Here $\langle \cdot \rangle$ is an average over the distribution $p(o)$, and the constant
\[
K_{sg} = \frac{\pi}{6} \int d\Omega \int \frac{d\Omega'}{4\pi} \left| \cos\theta' - 3\cos\theta \right| \\
x[\cos\theta\cos\theta' + \sin\theta\sin\theta'\cos(\phi - \phi')]  \tag{3.22}
\]

was evaluated analytically by MS (1981). After correcting a typographical error in their Eq.(12) we find \( K_{sg} = 4.5406 \), in agreement with a numerical evaluation of \( K_{sg} \) by Fiory (1981). The exponential \( A(q) \) gives a Lorentzian local field distribution

\[
g(\hat{\omega}) = \frac{a_s}{\pi^2(a_s^2 + \hat{\omega}^2)^2} \tag{3.23}
\]

and from Eq.(3.18),

\[
P(t) = \frac{1}{3} + \frac{2}{3} \left( 1 - a_s t \right) \exp(-a_s t). \tag{3.24}
\]

This expression for \( P(t) \) is known as the spin glass form, and is found to describe \( \mu \)SR data in a number of spin glasses (Heffner et al, 1982; Uemura et al, 1980; Uemura and Yamazaki, 1983; Heffner et al, 1984).

The spin glass \( P(t) \) was first derived by Uemura (1980), but with a different expression for \( a_s \). In the \( T=0 \) limit, his result can be written \( a_s = K'pcBS \), where

\[
K' = \frac{1}{\rho} \left( \frac{\pi}{3} \sum_{j} r_j^{-6} \right)^{1/2}. \tag{3.25}
\]
The sum extends over all lattice sites, and \( r_j \) is the distance from the muon site to lattice site \( j \). For the muon in the octahedral site of the fcc lattice, \( K' = 5.255 \), and for the interstitial site of a simple cubic lattice, \( K' = 4.644 \). These values of \( K' \) are considerably higher than the continuum theory result \( K_{sg} = 4.541 \), and \( K' \) is clearly dependent on the lattice structure. This result is surprising, since in the dilute limit, we would expect \( a_s \) to be independent of the underlying lattice structure.

The discrepancy between the continuum theory and Uemura's theory based on a lattice sum has been investigated using a Monte Carlo procedure. Impurities are distributed at random in a spherical volume of fcc lattice around an octahedral muon site, and each Mn moment is randomly oriented. The total (vector) dipole field at the muon site is calculated, and the procedure is repeated for many realizations of the impurity positions and spin orientations. If \( \mathbf{H}_i \) is the dipole field for the \( i \)th configuration, then the total observed depolarization is

\[
P(t) = \frac{1}{N} \sum_i \cos^2 \theta_i + \sin^2 \theta_i \cos(\chi_i \mathbf{H}_i t),
\]

where \( N \) is the total number of configurations, and \( \theta_i \) is the angle between \( \mathbf{H}_i \) and the muon polarization direction. For low concentrations, \((c = 0.01)\), \( P(t) \) is identical to the spin glass form, with \( a_s \) given by the continuum theory [Eq. (3.21)]. If Uemura's theory were correct, we would have found \( a_s \) to be 16% higher.
Fiory (1981) applied an analogous Monte Carlo procedure to the case of Mn distributed at random in a spherical volume (with no underlying lattice structure), and also found both excellent agreement with the spin glass form for \( P(t) \) and with the continuum theory result for \( a_s \). We will use the continuum theory of MS (\( K_{sg} = 4.541 \)) in calculating \( a_s \).

ferromagnet

The zero field \( P(t) \) for a dilute ferromagnet was first derived by Gist and Dodds (1984). We assume the spins are all aligned in a fixed direction \( \hat{\omega} \), the direction of the spontaneous magnetization within a single domain. Then \( A(\hat{\xi}, \hat{\delta}) \) depends on both the magnitude \( q \) and on the angle \( \psi \) between \( \hat{\xi} \) and \( \hat{\delta} \).

\[
A(q, \psi) = \exp\{- K(\psi) \rho c B S [\hat{\omega}] q \}, \tag{3.27}
\]

\[
K(\psi) = \frac{\pi}{\delta} \int d\eta \left| \cos \psi - 3 \cos(\cos \eta \cos \psi + \sin \eta \sin \psi \cos \phi) \right|. \tag{3.28}
\]

A numerical calculation of \( K(\psi) \) is shown in Fig.3.5. Note that \( K_{sg} \) defined by Eq.(3.22) is the average of \( K(\psi) \) over the polar angle \( \psi \).

The local field distribution function \( g(\hat{\omega}) \) depends on both the magnitude \( \omega \) and on the angle \( \theta \) between \( \hat{\omega} \) and \( \hat{\delta} \). A numerical calculation of \( g(\omega, \theta) \) for three particular angles is shown in Fig.3.6, where the scale factor \( \lambda \) is defined as
FIG. 3.5. Plot of $K(\psi)$, defined by Eq. (3.28). Note that the zero is suppressed.
FIG. 3.6. Zero applied field muon local field distribution function $g(\omega, \Theta)$ in a ferromagnet for three values of $\Theta$ as shown. The scale parameter $\lambda$ is defined in Eq. (3.29). The straight lines are linear least-square fits to the calculated values.
\[ \lambda = K(0) \rho \cos B S \{ |\phi| \}. \quad (3.29) \]

and \( K(0) = 5.065 \).

We can now use \( g(\omega) \) from Fig. 3.6 to calculate the depolarization function \( P(t) \), given the dipole orientation \( \hat{e} \) with respect to the muon polarization direction \( \hat{z} \). To make this procedure computationally feasible, it is necessary to find a parametrization of \( g(\omega, \theta) \). The straight lines in Fig. 3.6 are linear least-square fits to the calculated values, showing a Lorentzian dependence at each \( \theta \). The simplest angular dependence meeting the symmetry requirements of \( g(\omega, \theta) \) is to use a \( \cos^2 \theta \) dependence to interpolate between the \( \theta = 0 \) and \( \theta = \pi/2 \) values. This gives a parametrization

\[
g(\omega, \theta) = \frac{1.143\lambda}{\pi^2(\lambda^2 + 1.236\omega^2)^2} \sin^2 \theta + \frac{1.151\lambda}{\pi^2(\lambda^2 + 0.869\omega^2)^2} \cos^2 \theta. \quad (3.30)
\]

Eq. (3.30) gives \( g(\omega, \theta) \) accurately for all \( \omega \) when \( \theta \) is near 0 or \( \pi/2 \), and gives the full angular dependence accurately for small \( \omega \). However, it is not very accurate for the angular dependence at large \( \omega \), particularly near \( \pi/4 \).

Using Eq. (3.30), we can calculate \( P(t) \) from Eq. (3.18) for a given domain orientation \( \hat{e} \). Results are shown in Fig. 3.7 for domains oriented parallel (solid line) and perpendicular (dashed line) to the initial muon polarization (\( \hat{z} \)). The numerical evaluation of Eq. (3.18) requires a cut-off at large \( \omega \) in the
FIG. 3.7. Zero applied field muon depolarization functions in a ferromagnet when impurity spins are aligned parallel (solid line) and perpendicular (dashed line) to the initial muon polarization direction. $\lambda$ is defined by Eq. (3.29).
integration. The error introduced by this cut-off is less than 4% at all times, as determined by calculating \( P(t) \) with the Lorentzian \( g(\omega) \) [Eq.(3.23)], and comparing with the analytic spin glass form [Eq.(3.24)]. The inaccuracy of the parametrization at large \( \omega \) is also a source of error, and is reflected in the short time behavior of \( P(t) \). In particular, we find \( P(0) = 1.04 \) for both curves in Fig.3.7, where we have corrected for this error by plotting \( P(t)/P(0) \).

The angular dependence of \( g(\omega,\theta) \) has a simple physical interpretation. If a particular muon site sees a large field, Fig.3.6 shows that it is most likely to occur along the domain orientation \( \hat{\theta} \), whereas a small field is equally likely in all directions. When \( g(\omega) \) is isotropic, as for a spin glass, the muon polarization must tend to 1/3 at long times, since \( \omega_2^2/\omega^2 \) averages to 1/3. But for the anisotropic dilute ferromagnet, \( \omega_2^2/\omega^2 \) averages to more than 1/3 for the domain in the \( \hat{2} \) direction, and less than 1/3 for the domain perpendicular to the \( \hat{2} \) direction. Therefore, \( P(t) \) in Fig.3.7 tends to values greater than and less than 1/3 for these two cases.

For a ferromagnetic sample in zero applied field, it is physically appropriate to perform an average of the local field distribution function over a random distribution of domain orientations \( \theta \). Using the parametrization of Eq.(3.30), this can be performed analytically,
\[
\overline{g}(\omega) = \frac{1}{4\pi} \int d\Omega \, g(\omega, \theta)
\]

(3.31)

\[
\overline{g}(\omega) = \frac{0.76201}{\pi^2(\lambda^2+1.236\omega^2)^2} + \frac{0.3837\lambda}{\pi^2(\lambda^2+0.8169\omega^2)^2}.
\]

(3.32)

Using \( \overline{g}(\omega) \) in Eq. (3.18), we get an approximate domain averaged \( P(t) \),

\[
P(t) = 0.5545\left[ \frac{1}{3} + \frac{2}{3}(1-0.8895\lambda t)\exp(-0.8895\lambda t) \right] 
\]

\[
+ 0.5196\left[ \frac{1}{3} + \frac{2}{3}(1-1.106\lambda t)\exp(-1.106\lambda t) \right].
\]

(3.33)

As in the discussion of Fig. 3.7, the errors in \( g(\omega, \theta) \) at large \( \omega \) cause errors in the short time behavior of \( P(t) \), since \( P(0)=1.074 \). If we normalize by \( P(0) \), then a comparison of \( P(t)/P(0) \) with the spin glass form (with \( \lambda=a_g \)) shows the two expressions to differ by no more than 1% for all times. We conclude that the domain averaged ferromagnetic depolarization function is not significantly different from the spin glass form, but with a different numerical value for the depolarization rate \( (K(0)=5.065 \) instead of \( K_{sg}=4.541 \).
Canted ferromagnet

Some ferromagnetic dilute alloys may not have a complete alignment of spin orientations. Instead, spins may be canted away from the direction of the total magnetization. The canting angle $\theta'$ of each spin with respect to the direction of the total magnetization within a domain $\theta_o$ is a random variable from some distribution $p(\theta')$. As an illustration, we will use a Gaussian distribution,

$$p(\theta') = N(\delta) \exp\left(-\frac{\theta'^2}{2\delta^2}\right),$$

(3.34)

where $\delta$ is the width of the distribution, and $N(\delta)$ is a normalization constant. A distribution $p(\theta')$ based on a mean field theory will be given in the next chapter. The width $\delta$ plays the role of an alignment parameter, since we recover a uniform angular distribution for $\delta \gg 1$, and complete alignment in the $\theta_o$ direction for $\delta \ll 1$. Using this distribution, $A(\xi)$ depends on both $q$ and on the angle $\psi$ between $\xi$ and $\theta_o$:

$$A(q, \psi) = \exp(-K(\psi, \delta) p_{CBS}[|q|] q).$$

(3.35)

$$K(\psi, \delta) = \frac{\pi}{6} \int d\theta' \int d\theta'' p(\theta') [\cos \psi \cos \theta' + \sin \psi \sin \theta' \cos \theta']$$

$$-3[\cos \psi \cos \theta + \sin \psi \sin \theta \cos \theta']\cos(\psi - \theta').$$

(3.36)
If we are interested in a depolarization function averaged over a random distribution of the average domain orientation \( \mathbf{\delta}_0 \), then according to the analysis for a ferromagnet, the form of \( P(t) \) will not differ significantly from the spin glass form. The linewidth \( a_\delta \) is then found by setting \( \psi=0 \) in Eq. (3.36),

\[
a_\delta = K(0, \delta) \rho \, \cos B \, S \, |\mathbf{\delta}|. \tag{3.37}
\]

In Fig. 3.8, we plot \( K(\psi=0, \delta) \) and the average angle of canting with the \( \mathbf{\delta}_0 \) direction within a domain,

\[
\bar{\theta} = \int \theta \, p(\theta) \, d\theta, \tag{3.38}
\]

as a function of the alignment parameter \( \delta \). This Gaussian canting model clearly interpolates between the spin glass case (\( \delta \gg 1 \), \( K=4.541 \)) and the ferromagnetic case (\( \delta=0 \), \( K=5.065 \)). We conclude that for any degree of alignment of spins within a domain, the domain averaged depolarization function is given by the spin glass form with a linewidth given by Eq. (3.37), which varies by only 10% between the two extreme cases.

F. Transverse applied field \( \mu\text{SR} \) - ordered state

In a transverse applied field, the relaxation function \( f(t) \) is given by the Fourier transform of the lineshape \( g(\omega) \) in the direction of the applied field. Using the theory of \( \mu\text{SR} \), \( f(t) \) is
FIG. 3.8. Plot of $K(\psi=0, \delta)$, defined by Eq.(3.36), and the average canting angle $\bar{\theta}$ as a function of the alignment parameter $\delta$ using the Gaussian canted model.
given by $A(qz)$, where $A(q)$ is given by Eq. (3.20), $2$ is the direction of the applied field, and $q = t$. This implies an exponential $f(t)$, with linewidth $(T_2)^{-1}$ equal to the zero field static linewidth $a_s$.

The correspondence between $(T_2)^{-1}$ in transverse field and $a_s$ in zero field was noted by MS for the case of spin glass ordering, but it is also true for ferromagnetic ordering. In a dilute ferromagnet, the impurity spins are all oriented in the same direction within each domain. In an applied field much larger than the coercive force $H_c$ of the hysteresis loop, the domains will all align with the external field. (If the applied field were smaller than $H_c$, the magnetization in some domains would not be parallel to the applied field.) In ferromagnetic PdMn, the coercive force is very small, $(H_c < 0.5G$, Star et al, 1975), so all spins are aligned with the external field. This is the same situation as in the paramagnetic state (Sec. C), so the linewidth is given by Eq. (3.9). This expression for $(T_2)^{-1}$ is identical to Eq. (3.29) for the ferromagnetic state zero field $a_s$.

The ratio of the mixed coupling linewidth to the pure dipolar linewidth is different in the paramagnetic state (where there are no transverse spin components) and in the spin glass state (where there are frozen transverse spin components). The expected mixed to dipolar linewidth ratio for the spin glass state can be calculated from the MS theory; results are shown in Table 3.1 for
Ag, Pd, and Cu. The mixed to dipolar linewidth ratio is smaller in the spin glass state than in the paramagnetic state.

G. Muons adjacent to impurities and the static linewidth

Two kinds of complications can arise when the muon has a near-neighbor impurity moment. The first is a modification of the muon-impurity coupling from the dipole plus asymptotic RKKY form of Eq.(3.7). For example, the RKKY interaction can be drastically modified at short distances (Jena and Geldart, 1973), or the muon can couple to the Mn d-orbital by a direct contact interaction. A second kind of complication can arise if the muon is excluded from occupying octahedral sites adjacent to impurities. We will consider the effect of these two complications in turn.

To understand the effect of a modification of the coupling strength, consider the role of muons adjacent to impurities for pure dipolar coupling. The dipole field at the muon is of order $H_{nn} = \mu B <\mathbf{S_z}>/r_{nn}^3$, where the muon-impurity separation is $r_{nn} = 0.5a$. Comparing this with the linewidth $\Delta H$ from Eq.(3.9) we find $H_{nn}/\Delta H = 0.395/c$. This shows that muons adjacent to impurities do not contribute to the central peak in $g(\omega)$ even at concentrations as high as $c = .10$, where $H_{nn}/\Delta H = 4$. Instead, these muons contribute 'satellite' lines to $g(\omega)$, well separated from the main resonance line. The precise position of the satellite lines depends on the angle of the muon-impurity separation vector to the applied field.
so in a polycrystalline average, the satellites average out to a broad background in comparison with the main line. If we now modify the coupling, the same results apply; these muons will still contribute only a broad background in comparison with the main resonance line, so the observed relaxation rate is insensitive to details of the muon-near neighbor impurity interaction.

The muon potential energy can be modified by an adjacent substitutional impurity. Suppose $\Delta U(\text{host})$ is the depth of the octahedral site potential energy well in the host lattice, which was estimated for a simple metal in Fig. 2.2, and suppose $\Delta U(\text{imp})$ is the well depth at sites adjacent to impurities. If $\Delta U(\text{imp}) < \Delta U(\text{host})$, impurity sites become less favorable for muon localization relative to the host sites, and if $\Delta U(\text{imp})$ is less than the muon zero point energy, these sites cannot localize the muon at all. In this limit, the muon is excluded from occupying sites adjacent to impurities. Two specific mechanisms for changing $\Delta U(\text{imp})$ will be discussed in Sec. 5.A; in the remainder of this section we calculate the effect on the linewidth of excluding the muon from sites adjacent to impurities.

The qualitative effect of the exclusion from near-neighbor sites on $g(\omega)$ is clear from the discussion of the satellite lines. The exclusion removes weight from $g(\omega)$ at large $\omega$. Since $\int g(\omega) d\omega$ is a constant, the central resonance line must increase in weight, reducing the linewidth. Detailed calculations of $g(\omega)$ in the paramagnetic state have been carried out using the Monte Carlo
simulation described in Sec.C, where we exclude muon sites adjacent to impurities. Results for $c=.07$ are shown as the solid line in Fig.3.9, where $(T_g^2)^{-1}$ is the WW linewidth without exclusion, and the dotted line is the WW Lorentzian. The exclusion has removed all contributions to $g(\omega)=0$ for $|\omega T_g|>4$ (cf. Fig.3.2). The dashed line is a Lorentzian with a width $0.70(T_g^2)^{-1}$. Figure 3.10 shows the same calculation for $c=.02$, where exclusion has a much smaller effect. The dashed line is a Lorentzian with a width $0.88(T_g^2)^{-1}$. These calculations show that an exclusion of muons from sites adjacent to impurities will cause a narrower line than predicted by the WW theory, and that the deviation from the WW theory will be concentration dependent.

The zero field static linewidth in the spin glass state will also be narrowed by the exclusion mechanism, but the degree of narrowing is not necessarily the same as in the paramagnetic state. Note that the ratio of the mixed (RKKY plus dipolar) coupling linewidth to the pure dipolar linewidth is different for paramagnetic and spin glass order. The effect of exclusion has been calculated from the Monte Carlo procedure described above in Sec.E, by excluding from the sum in Eq.(3.26) all sites where the muon is adjacent to an impurity. Results for spin glass order are shown in Fig.3.11 for three different concentrations. The solid line is the result for no exclusion, with $t$ scaled by the spin glass continuum theory $a_g$ [Eq.(3.21)]. The dotted, dashed, and dot-dashed lines include exclusion for $c=.02,.05$, and .07.
FIG. 3.9. Paramagnetic state transverse field lineshape for \( c = 0.07 \) when the muon is excluded from octahedral sites adjacent to impurities (solid line). The dotted line is the dilute limit Lorentzian (including all sites) with linewidth \( (T_2^*)^{-1} \). The dashed line is a Lorentzian with linewidth \( 0.70(T_2^*)^{-1} \) chosen to approximate the solid line.
FIG. 3.10. Paramagnetic state transverse field lineshape for c = 0.02 when the muon is excluded from octahedral sites adjacent to impurities (solid line). The dotted line is the dilute limit Lorentzian (including all sites) with linewidth \((T_2)\)\(^{-1}\). The dashed line is a Lorentzian with linewidth \(0.88(T_2)\)\(^{-1}\) chosen to approximate the solid line.
FIG. 3.11. Zero applied field spin glass depolarization functions when the muon is excluded from octahedral sites adjacent to impurities. Solid line, c=.02; dashed line, c=.05; dot-dashed line, c=.07. The solid line includes all octahedral sites, and is identical to the spin glass depolarization function [Eq. (3.24)] and $a_3$ is given by Eq. (3.21).
respectively. There is very little effect for c=.02, but at higher concentrations, P(t) decays more slowly, and the minimum is smaller before reaching the limiting value of 1/3. For c=.07, the linewidth narrows by approximately 50%. The Monte Carlo procedure has also been used to calculate P(t) for ferromagnetic order, using a random distribution of domain orientations. Results for c=.02 and .05 are very close to the P(t) curves in Fig.3.11, if t is scaled by the ferromagnetic a_3 with no exclusion (K=5.065). We conclude that if Fig.3.11 describes P(t) data in the spin glass or ferromagnetic state with exclusion of muons from sites adjacent to impurities, then a fit of the spin glass P(t) [Eq.(3.24)] will yield linewidths smaller than a_3, with a strong concentration dependence to the deviation. The degree of linewidth narrowing is different for the zero field spin glass static linewidth and the transverse field paramagnetic linewidth.
Chapter IV. Mean field theory and the static linewidth

In this chapter we will describe a mean field theory of disordered magnetism for both Ising and Heisenberg spins, and use this theory to calculate the temperature and field dependence of the static linewidth in a dilute alloy. The main result is the calculation of a new order parameter, which is proportional to the measured zero field static linewidth $a_s$, defined in Sec.3.E. The relationship of the linewidth order parameter to the usual thermodynamic order parameters will be described.

A. Disordered mean field theory

The development of a mean field theory for spin glasses and disordered ferromagnets has been a subject of much recent activity in theoretical statistical mechanics. (For reviews, see Fischer, 1983; Chowdhury and Mookerjee, 1984.) The most successful results have been obtained on a model first studied by Sherrington and Kirkpatrick, known as the SK model (Sherrington and Kirkpatrick, 1975; Kirkpatrick and Sherrington, 1978). In the SK model, $N$ Ising spins are coupled by infinite-ranged interaction, with exchange energies $J_{i,j}$ chosen as Gaussian random variables with mean $\overline{J}_0$ and standard deviation $\overline{J}$. Following the solution method of Mookerjee (1978), we write the (normalized) thermodynamic average of spin $i$ as
\[ a_i = B_S \left\{ \frac{S}{kT} ( g \mu_B H_0 + \sum J_{ij} a_j ) \right\}, \quad (4.1) \]

where \( B_S(x) \) is the spin \( S \) Brillouin function and \( H_0 \) is an external field. The term involving \( J_{ij} \) is the exchange field on spin \( i \) from the random exchange couplings with all other spins. The average over all spins (configuration average) is defined as an integral over a distribution function \( p(a) \), which we write as \([ \cdot ]\). The thermodynamics of the SK model can then be described in terms of two order parameters; the magnetization \( m = \langle a \rangle \), and a new order parameter \( q = \langle a^2 \rangle \), usually called the Edwards-Anderson order parameter (Edwards and Anderson, 1975). Note that \( q^{1/2} \) can be interpreted as a root-mean-squared frozen moment per site. The order parameters \( m \) and \( q \) are determined by two coupled equations,

\[ m = \int dh \ p(h) \ B_S(h/kT) \quad (4.2) \]
\[ q = \int dh \ p(h) \ B_S^2(h/kT) \quad (4.3) \]

\[ p(h) = (2\pi J^2 q)^{-1/2} \exp\left[ -\frac{(h - J_0 m - g \mu_B H_0)^2}{2J^2 q} \right]. \quad (4.4) \]

where \( J_0 = NJ_0 \) and \( J^2 = NJ^2 \) are intensive variables for the mean and variance of the exchange energy distribution. Alternatively, we can think of these equations as an implicit definition of the temperature dependence of the distribution function \( p(a) \). The
phase diagram as a function of $\frac{J_o}{J}$ is shown in Fig. 4.1. The spin glass phase is defined by the condition $q > 0$ and $m = 0$ (no long range order) for $H_o = 0$. The paramagnet-spin glass transition is a second order phase transition in the order parameter $q$. For $\frac{J_o}{J} > J$, there is a ferromagnetic phase with a spontaneous magnetization $m > 0$. The temperature dependence of the order parameters $m$ and $q$ in zero field is shown in Fig. 4.2 for three values of $\frac{J_o}{J}$. Note that in a restricted range of $\frac{J_o}{J}$, the SK model predicts two second order transitions; a transition at $T_C$ from a paramagnet to a ferromagnetic state, and a transition at $T_g$ to a spin glass state.

The SK model for Ising spins can be generalized to the case of Heisenberg spins (Gabay and Toulouse, 1981; Mookerjee, 1980; Mookerjee and Roy, 1983). Suppose the applied field (or spontaneous magnetization) is in the $z$ direction. Then the magnetization and frozen order for Heisenberg spins are described by three order parameters:

$$m = [a_z], \quad q_z = [a_z^2], \quad q_t = [a_z^2 + a_y^2], \quad (4.5)$$

where $[ \ ]$ is an average over the distribution $p(\vec{a})$, and $q_t$ and $q_z$ describe the frozen order transverse and parallel to the applied field. The order parameters are determined by three coupled equations,

$$m = \int d^3 h \ p(\vec{a}) \ \left( \frac{h_z}{h} \right) B_s(h/kT) \quad (4.6)$$
FIG. 4.1 Phase diagram of the Ising SK mean field theory (solid lines): \( J_0 \) and \( J \) are defined in the text. The dashed line is the de Almeida-Thouless instability line.
FIG. 4.2 Order parameters in the Ising (left) and Heisenberg (right) SK mean field theory for $S=5/2$ and $\frac{J_0}{J}=0$ (top), 1.2 (middle) and 1.6 (bottom). Solid line=$m$, dashed line=$q^{1/2}$ or $q_z^{1/2}$, dotted line=$q_b^{1/2}$, dot-dashed line=$r$. 
\[ q_z = \int d^3 h \ p(\hat{h}) \left( \frac{h_z}{h} \right)^2 B_S^2 \left( \frac{h}{kT} \right) \] (4.7)

\[ q_t = \int d^3 h \ p(\hat{h}) \left( \frac{h_x + h_y}{h} \right)^2 B_S^2 \left( \frac{h}{kT} \right) \] (4.8)

\[ p(\hat{h}) = \left[ \frac{1}{\pi} \right]^{3/2} \frac{1}{2J^2 q_t J_q^1} \] (4.9)

\[ x \ \exp \left[ -\frac{h_x^2 + h_y^2}{2J^2 q_t} - \frac{(h_z - J_0 m - S g B H_0)^2}{2J^2 q_z} \right]. \]

Again, we can view these equations as defining the temperature dependence of the distribution function \( p(\hat{h}) \). Note that when \( q_t = 0 \), \( p(\hat{h}) = \delta(h_x) \delta(h_y) p(h_z) \), where the distribution for the \( z \)-component is given by the Ising \( p(h) \) [Eq. (4.4)] with \( q_z = q \), and \( \delta(x) \) is the Dirac delta function. Thus the Heisenberg model reduces to the Ising model when \( q_t = 0 \).

The temperature dependence of \( q_t \) and \( q_z \) for a Heisenberg spin glass with \( J_0 = 0 \) is shown in Fig. 4.2, where the isotropy of the spin orientations in the spin glass state implies \( q_t = 2q_z \), and the \( T = 0 \) limiting value \( q_z = 1/3 \). The phase diagram for the Heisenberg SK model is shown in Fig. 4.3. There are two phases with spontaneous magnetization in the Heisenberg model, which differ in the transverse frozen order \( q_t \). The usual ferromagnetic phase (co-linear) has \( q_t = 0 \), so the spins are confined to the direction of the magnetization. The mixed (canted) phase has \( q_t > 0 \), so spin glass freezing in the transverse spin components coexists with a
FIG. 4.3 Phase diagram of the Heisenberg SK mean field theory (solid lines). $\frac{J_0}{J}$ and $\frac{1}{J}$ are defined in the text. The dashed line is the de Almeida-Thouless instability line. The mixed phase is defined in the text.
spontaneous magnetization. Unlike the Ising model, the Heisenberg phase diagram does not have a reentrant transition from a phase with m > 0 to the m = 0 spin glass phase. Instead, the reentrant transition is a canting transition, where the spins become canted to the magnetization direction by random angles. This transition is also called transverse freezing. In an applied magnetic field there is no transition in m or q_z, as these order parameters are finite for all T. However, even in an applied field, there is a second order transverse freezing transition in q_t.

The mean field equations given above are known as the replica symmetric theory, and they are only approximate solutions to the Ising and Heisenberg SK model. In this solution it is assumed that exact copies of the interacting spin system will have identical equilibrium configurations, hence the term replica symmetric. The correct solution of the SK model requires that this symmetry be broken at low temperatures (Parisi, 1979; de Dominicis, 1985). The temperature where the replica symmetry is broken is determined by certain pathologies of the replica symmetry solution (de Almeida and Thouless, 1978), and corresponds physically to the onset of irreversibilities in the macroscopic behavior. This temperature is indicated as a dashed line on the phase diagrams. The replica symmetric theory which we will use for comparisons with data is correct above this line, and is an approximation to the proper solution below this line.
The replica symmetric solution to the SK model gives a theoretical framework for calculating many experimental properties of dilute alloys, such as the susceptibility, the frozen moment $q^{1/2}$, and the existence of the reentrant transition. Conspicuous among the failings of the SK model is the prediction of a cusp in the specific heat at $T_g$, which is never observed. The more fundamental prediction of the SK model, that $T_g$ is a second order phase transition, is difficult to address directly with experiment. The current evidence in favor of a phase transition is indirect; from Monte Carlo computer simulations (Ogielski and Morgenstern, 1985), and from the general success of the SK Heisenberg model in describing experimental quantities (Krey, 1983).

B. Comparing mean field theory with experiment

Two assumptions are inherent in comparisons of the SK mean field theory with experimental results in dilute alloys. First, the exchange interaction must be long-ranged, so that each spin has a large effective coordination number. This condition is satisfied in RKKY coupled systems such as AgMn, where $J_{1,j}$ falls off slowly ($-r_{1,j}^{-3}$), and the mean free path is much longer than the typical Mn separation. Second, the distribution of exchange energies $J_{1,j}$ must be Gaussian. Mookerjee (1978) showed that for RKKY exchange, the distribution $p(J_{1,j})$ is very close to a Gaussian, so that typical RKKY spin glasses such as AgMn and CuMn satisfy both
criteria for comparison with the SK mean field theory. However, the exchange interaction $J(r)$ in PdMn is not RKKY; it has a short-ranged anti-ferromagnetic component representing direct exchange and a long-ranged exchange-enhanced ferromagnetic component. Niewenhuys et al (1977) assumed a particular form for $J(r)$:

$$J(r) = \begin{cases} 
-120K + r \frac{25K}{A}, & r < 5.25A \\
\frac{220K-A}{r} \exp(-r/5A) & r > 5.25A
\end{cases} \quad (4.10)$$

The parameters of Eq.(4.10) give good agreement with transition temperatures, specific heat, and magnetic susceptibility for concentrations up to $c=0.05$. Fig.4.4 shows the distribution of $J_{i,j}$ for the fcc lattice. The large anti-ferromagnetic $J_{i,j}$ for nearest neighbors and next-nearest neighbors is off the scale of the figure. Since the distribution is clearly not a Gaussian, we expect problems in applying the SK theory to PdMn.

To make comparisons of the SK mean field theory with experimental results, we must specify the ratio $\overline{J_0}/\overline{J}$ for each sample. For a ferromagnet, such as PdMn(2 at.%), $\overline{J_0} = (3S/(S+1))T_c$, and $\overline{J}$ can be determined by matching the temperature dependence of $m$ to the measured spontaneous magnetization. Fig.4.5 shows the spontaneous magnetization of PdMn(2.45 at.%) (Star et al, 1975) for
FIG. 4.4 Distribution of exchange energies in PdMn, with $J_{i,j} = J(r_{i,j})$ defined by Eq.(4.10).
FIG. 4.5 Normalized spontaneous magnetization in PdMn (2.5 at.%) from Star et al. (1975). The dot-dashed line is the ordinary mean field theory, (J=0), adjusted to give the high field saturation magnetization at T=0. The solid and dashed lines are the Ising and Heisenberg SK mean field theories, respectively, for J_o/J=1.2.
comparison with \( m \). The dot-dashed line is the homogeneous mean field theory \( (J_0=0) \), where the \( T=0 \) limit is the observed high field saturation magnetization. The data for \( T=0.7-1.0T_C \) are described very well by the SK theory for \( J_0/J=1.2 \). The dashed line is for Heisenberg spins, and the solid line for Ising spins. The Heisenberg model fails below \( 0.7T_C \), where there is a transverse freezing transition, and \( m \) reaches a maximum (cf. Fig. 4.2). The Ising model fits the data down to \( 0.3T_C \), but there is no experimental evidence for the predicted decrease in \( m \) leading to a reentrant transition. The lack of a double transition can be attributed to the asymmetric exchange energy distribution. The additional ferromagnetic exchange will tend to stabilize the ferromagnetic state, opposing the decrease in \( m \) predicted for a Gaussian distribution. For purposes of calculation, it is clear from Fig. 4.5 that we can approximate the magnetic order in \( \text{PdMn}(2 \text{ at.\%}) \) by the Ising model with \( J_0/J=1.2 \) provided \( T>0.3T_C \).

For a spin glass, \( J_0/J=\theta/T_g \), where \( \theta \) is the paramagnetic Curie-Weiss temperature, provided that \( |\theta|<T_g \). The condition \( |\theta|<T_g \) is satisfied in \( \text{AgMn} \) (see Chap. 5), but not in spin glass \( \text{PdMn}(7 \text{ at.\%}) \), where \( \theta \) is slightly larger than \( T_g \). This is easily understood by the asymmetric distribution \( p(J_{i,j}) \) (Fig. 4.4), which is skewed towards ferromagnetic exchange. We will assume the main features of \( \text{PdMn}(7 \text{ at.\%}) \) data can be described by the spin glass Heisenberg SK model \( (J_0=0) \). Finally, for a sample with two
transitions, the phase diagram gives $J_0/J$ from the ratio of the transition temperatures.

C. Mean field static linewidths

The zero field depolarization function was calculated in Sec. 3.6 for any degree of spin alignment, where it was found that $P(t)$ is always well represented by the spin glass form [Eq. (3.24)], provided we consider a random distribution of the spontaneous magnetization directions (domain orientations). The mean field expression for the zero field static linewidth $\alpha_s$ follows directly from Eqs. (3.36)-(3.37). For pure dipolar coupling,

$$ a_s = \frac{\pi}{6} \rho c B S \int d\Omega \int d^3 h \, p(\mathbf{\hat{h}}) \, |B_S(h/kT)| $$

$$ x \left[ \frac{h_z}{h} (1 - 3\cos^2 \phi) - 3\sin \phi \cos \phi \left( \frac{h_x}{h} \cos \phi + \frac{h_y}{h} \sin \phi \right) \right]. \quad (4.11) $$

For Heisenberg spins, $p(\mathbf{\hat{h}})$ is the distribution in Eq. (4.9). For Ising spins, $p(\mathbf{\hat{h}}) = \delta(h_x) \delta(h_y) p(h_z)$, where $p(h_z)$ is the distribution in Eq. (4.4), and $q_z = q$.

Equation (4.11) is used to define the linewidth order parameter $\alpha_s$,

$$ a_s = a_0 \, \alpha_s, \quad (4.12) $$
where \(a_o\) is defined as the \(T=0\) limit of Eq. (4.11). The angular integrals in Eq. (4.11) can be performed in two special cases, yielding simplified expressions for the order parameter \(r\).

For a Heisenberg spin glass, where \(q_t=2q_z\), the distribution \(p(\hat{n})\) depends only on the magnitude \(h\), and the angular integrals become identical to the definition of \(K_{sg}\) in Eq. (3.22). In this case

\[
r = [\mathcal{O}] = \int d^3h \ p(\hat{n}) \ |B_S(h/kT)|.
\]

(4.13)

For the Ising model,

\[
r = [\mathcal{O}] = \int dh \ p(h) \ |B_S(h/kT)|.
\]

(4.14)

In the general case of Heisenberg spins with \(q_t \neq 2q_z\), the angular integrals in Eq. (4.11) must be calculated numerically.

The \(T=0\) limit of Eq. (4.11) can be written \(a_o = K_PcBS\), where the constant \(K\) depends on the degree of transverse freezing. For the Ising model, \(K=5.065\), and for an isotropic distribution \((q_t=2q_z)\), \(K=K_{sg}=4.541\). Intermediate values of \(K\) were described in Sec. 3.E by an ad hoc Gaussian distribution. In the Heisenberg SK model, we can calculate intermediate values of \(K\) as a function of \(\overline{J}_o/\overline{J}\) as shown in Fig. 4.6. The spin glass phase has \(K_{sg}=4.541\), and \(K\) increases in the mixed phase toward the \(q_t=0\) value \(K(0)=5.065\). Also shown in Fig. 4.6 is the average canting angle \(\overline{\theta}\), defined as in Eq. (3.38).
FIG. 4.6. Plot of $K$, which determines the $T=0$ linewidth $\omega_0$ from Eq. (3.37), and the average canting angle, defined as in Eq. (3.38), as a function of $J_0/J$ in the Heisenberg SK mean field theory.
The linewidth order parameter $r$ can be calculated as a function of $T$ and $\overline{J}_0/\overline{J}$, with typical results shown in Fig. 4.2. For the Heisenberg case, the total frozen moment is $q = [s^2] = q_z + q_t$. For a spin glass ($\overline{J}_0=0$), $r$ agrees with $q^{1/2}$ to within 5% for all temperatures. This important result shows that within the mean field theory, the linewidth $a_s$ gives a zero field measurement of the order parameter $q$, which is the quantity usually calculated in spin glass theories. In the Ising case, we find a much larger disagreement of up to 20% between $r$ and $q^{1/2}$. A second important result is that the transverse freezing transition has no effect on the linewidth $r$. This is apparent in Fig. 4.2 for $\overline{J}_0/\overline{J}=1.2$ and 1.6, where the transverse freezing transition temperature is not reflected in the temperature dependence of $r$.

In Sec.3.F we found that the transverse applied field linewidth is the same as $a_s$, so mean field values of $T_2^{-1}$ can be calculated from Eq. (4.11), provided we include the applied field in $p(\hat{n})$. 
Chapter V. Transverse applied field results

Transverse field μSR experiments have been performed on AgMn and PdMn samples. An exponential function was fit to the relaxation f(t) in all cases, as suggested by the calculations of Chap. 3. Relaxation from impurity fluctuations (lifetime broadening) contributes to the measured $T_2$ only at low fields and high concentrations, so $T_2$ at fields of 1kG to 5kG reflects only the static broadening $T_2^s$. More direct evidence for an absence of lifetime broadening corrections will be presented in Chap. 8, where $T_1$ is seen to be very much longer than the measured $T_2$ at high field. The mean field theory of Chap. 4 will be compared with the data where appropriate.

A. PdMn paramagnetic state

Results of μSR experiments on PdMn(2 at.%) and PdMn(7 at.%) have been published in Dodds, et al (1983), and Heffner, et al (1984). For a paramagnet, we expect $(T_2^s)^{-1}$ to be proportional to the impurity spin polarization $\langle S_z \rangle$ and to the concentration $c$. Since the high temperature magnetization should follow a Curie-Weiss behavior we can write the transverse relaxation rate as

$$\frac{1}{T_2} = A \frac{c \cdot H}{T - \theta} \quad (5.1)$$

90
where $\theta$ is the paramagnetic Curie-Weiss temperature, and $A$ can be calculated from the theory of Chap. 3. Figure 5.1 shows a plot of $T_2H$ vs. $T$ for ferromagnetic $PdMn(2 \text{ at.} \%)$ above $T_c=5.75K$. The data are well described by a straight line, which confirms that lifetime broadening is negligible, and that the static broadening follows the form of Eq. (5.1). A fit to the data gives $\theta=7.2(3)K$, and $A=0.242(4) \text{ K}^{-1}\mu\text{s}^{-1}$. In Fig. 5.2 we show data with a wider range of $H$ and $T$ by plotting $\log T_2^{-1}$ vs. $\log H/(T-\theta)$. The solid line is from the fit value of $A$, and is in good agreement with the data over two orders of magnitude.

High temperature susceptibility of $PdMn(2.45 \text{ at.} \%)$ (Star et al, 1975), gives $\theta=7.1K$ in excellent agreement with $\theta$ determined from the $\mu$SR linewidth. Assuming a spin 5/2, the Curie constant gives an enhanced effective $g$-factor $g_{\text{eff}}=2.65$ (Star et al, 1975). As discussed in Chap. 3, we neglect the dipolar coupling of the muon to the giant moment polarization cloud, so we use the bare $g$-factor $g=2$ in calculating the dipolar strength $B=\gamma_{\mu}g_{\text{eff}}B$. The WW formula then gives $A_{\text{WW}}=0.291 \text{ K}^{-1}\mu\text{s}^{-1}$, which is plotted as the dashed line on Fig. 5.2. The data lie below the predicted linewidth from the WW theory including only the bare impurity moments, confirming that the giant moment polarization cloud makes a negligible contribution to the linewidth, as predicted in Sec. 3.C. The experimental value of $A$ from Fig. 5.1 is smaller than the WW prediction: $A=0.83A_{\text{WW}}$. 
FIG. 5.1 PdMn(2 at.%) paramagnetic state transverse field relaxation time scaled by the applied field as a function of temperature. Squares, $H=5kG$; triangles, $H=1kG$; circles, $H=200G$. The solid line is a linear least-squares fit to the data.
FIG. 5.2 $\text{PdMn}(2\text{ at.\%})$ paramagnetic state transverse field linewidth, as a function of $H/(T-\Theta)$, with $\Theta$ determined from Fig. 5.1. Symbols are the same as in Fig. 5.1. The solid line is calculated from Eq. (5.1), with $A$ determined from Fig. 5.1. The dashed line is the WW dilute limit prediction.
In Fig. 5.3 we plot $T_2H$ vs. $T$ above the glass temperature $T_g = 4.8K$ for PdMn(7 at.%). The straight line fit demonstrates Curie-Weiss behavior from $T=15K$ to $80K$, with deviations just above $T_g$. The fit gives $\theta=7.9(7)K$ and $\Lambda=0.087(3) K^{-1} \mu s^{-1}$. A wider range of $T$ and $H$ is shown in Fig. 5.4, where we plot $\log T_2^{-1}$ vs. $\log H/(T-\theta)$. The solid line is from the fit value of $\Lambda$, and is in good agreement with the data over nearly two orders of magnitude.

Susceptibility data on PdMn(6.5 at.%) (Coles et al., 1975) do not show a giant moment, so we use $\mathcal{E}_{eff}=2$ for PdMn(7 at.%). The WW formula then gives $A_{WW}=0.213 K^{-1} \mu s^{-1}$, which is plotted as the dashed line on Fig. 5.4. The experimental value is less than half the WW prediction, $A=0.41 A_{WW}$.

In Sec. 3.C, a number of deviations from the WW prediction were considered theoretically, none of which produced the degree of line narrowing observed in PdMn(7 at.%). Since the deviation of the exponential rate $T_2^{-1}$ from the WW prediction is by a constant factor over two orders of magnitude in $H/(T-\theta)$, we attribute the deviation to a diminished average coupling strength between the muon and the impurity spins. Since the dipolar coupling strength is accurately known, the dipolar coupling can only be reduced by moving the muon away from the impurity. We therefore hypothesize that the muon is excluded from occupying octahedral sites adjacent to Mn impurities in Pd.
FIG. 5.3 PdMn(7 at.%) paramagnetic state transverse field relaxation time scaled by the applied field as a function of temperature. Symbols are the same as in Fig. 5.1. The solid line is a linear least-squares fit to the data for T > 15K.
FIG. 5.4 PdMn(7 at.%) paramagnetic state transverse field linewidth, as a function of $H/(T-\Theta)$, with $\Theta$ determined from Fig. 5.3. Symbols are the same as in Fig. 5.1. The solid line is calculated from Eq. (5.1), with A determined from Fig. 5.3. The dashed line is the WW dilute limit prediction.
The effect of this exclusion on the paramagnetic linewidth was calculated in Sec. 3.6. For $c = .02$, the linewidth with exclusion is $0.88A_{WW}$, in good agreement with the observed linewidth reduction in PdMn(2 at.%). For $c = .07$, the linewidth with exclusion is $0.70A_{WW}$, which is not as large as the observed reduction in PdMn(7 at.%).

The failure to predict the magnitude of the line narrowing in PdMn(7 at.%) can be attributed to effects left out of this simple calculation. As discussed in Sec. 3.C, the expected anti-clustering (atomic SRO) in PdMn(7 at.%) increases the fraction of octahedral sites that are adjacent to an impurity, as seen in the intensity of the satellite lines. This will increase the effect of excluding those sites, and thus further narrow the line. The line narrowing due to exclusion including anti-clustering has been estimated with the Monte Carlo procedure described in Sec. 3.C. As expected, the result is a line narrower than without anti-clustering, with a linewidth $0.60A_{WW}$, which is still broader than the experimental linewidth $0.41A_{WW}$. The remaining discrepancy between the Monte Carlo calculation and experiment may be due to Mn clusters excluding muons beyond the near-neighbor octahedral sites. Furthermore, the dilute limit theory applied to $c = .07$ is only accurate to about 10%. We emphasize that of all the linewidth calculations discussed, only the exclusion of muons from octahedral sites adjacent to impurities can cause a significant degree of concentration dependent line narrowing. Further evidence for
exclusion of muons from sites adjacent to Mn will be presented in Chaps. 6 and 8.

One possible mechanism for this exclusion is a contraction of the host lattice around the impurity. The effect of a lattice distortion can be understood qualitatively with the potential energy calculations described in Sec. 2.A. If the lattice contracts, the octahedral potential wells adjacent to impurities become shallower, and displace away from the impurity. If the contraction is large enough, these sites cannot localize the muon, and are thus excluded. This mechanism can be tested with X-ray diffraction by measuring the lattice parameter as a function of concentration. Suppose the host atoms have a radius \( r_1 \), and the impurities a radius \( r_2 \). Then a continuum theory of the lattice distortion due to the mismatch in impurity size gives the fractional change in lattice parameter (Eshelby, 1956),

\[
\frac{\Delta a}{a} = c \frac{r_2-r_1}{r_1}.
\]  

Lattice parameters in PdMn have been measured with the Debye-Sherrer method, with results shown in Table 5.1. Results on the pure Pd sample agree fairly well with the literature value 3.8907A (Pearson, 1967). There is clearly no change in lattice parameter with concentration. The PdFeMn(5 at.% ) lattice parameter is \( a = 3.8926(7) \)A, suggesting a slight lattice expansion with the
TABLE 5.1. Lattice parameters in Angstroms of PdMn and AgMn alloys.

<table>
<thead>
<tr>
<th>c(at.%)</th>
<th>PdMn&lt;sup&gt;a&lt;/sup&gt;</th>
<th>AgMn&lt;sup&gt;b&lt;/sup&gt;</th>
</tr>
</thead>
<tbody>
<tr>
<td>0</td>
<td>3.8918(7)</td>
<td>4.0862</td>
</tr>
<tr>
<td>2</td>
<td>3.8901(7)</td>
<td>4.0861</td>
</tr>
<tr>
<td>5</td>
<td></td>
<td>4.0866</td>
</tr>
<tr>
<td>7</td>
<td>3.8907(7)</td>
<td></td>
</tr>
<tr>
<td>8</td>
<td></td>
<td>4.0866</td>
</tr>
<tr>
<td>10</td>
<td>3.8917(7)</td>
<td>4.0864</td>
</tr>
</tbody>
</table>

<sup>a</sup> this work

<sup>b</sup> Pearson (1967)
addition of Fe. Applying Eq.(5.2) to the data in the table gives $(r_2-r_1)/r_1<0.2\%$, which is far too small to cause significant changes in the adjacent octahedral potential wells. Published lattice parameters in AgMn are shown in the table, and also show negligible lattice distortion.

A second mechanism for excluding the muon from sites adjacent to impurities is the possibility of significant charge density oscillations in the surrounding interstitial region. Since the muon also produces charge density oscillations, there may be a complicated repulsive interaction when the muon and impurity are adjacent. Estreicher and Meier (1983) calculated this effect for impurities in Al, and found that different impurities can raise or lower the depth of adjacent muon potential energy wells. A complete calculation of the muon-Mn interaction in Pd or Ag is unavailable. The repulsive muon-Mn interaction in Pd (if present) will likely be more pronounced than in Ag or Cu due to the large exchange enhancement in Pd, suggesting that the exclusion may only be significant in Pd.

B. PdMn ordered state

A small number of transverse field μSR experiments have been performed on PdMn samples in the ordered state. Figure 5.5(a) shows $T_2^{-1}$ at high fields as a function of $T/T_g$ in spin glass PdMn(7 at.%), which shows that the linewidth is independent of the
FIG. 5.5 (a). PdMn (7 at.%) spin glass state transverse field linewidth as a function of temperature scaled by $T_c = 4.8K$. Symbols are defined in Fig. 5.1. The solid and dashed lines are the spin glass Heisenberg SK theory prediction for $H=5kG$ and a $T=0$ linewidth of $85 \mu s^{-1}$ and $43 \mu s^{-1}$, respectively. (b). PdMn (2 at.%) ferromagnetic state transverse field linewidth as a function of temperature scaled by $T_c = 5.75K$. Symbols are defined in Fig. 5.1. The solid and dashed lines are the Ising SK theory prediction for $H=5kG$ and $H=1kG$ respectively, and a $T=0$ linewidth of $22.6 \mu s^{-1}$. 
applied field from 1kG to 5kG. This follows from the SK theory for a spin glass (Sec.4.A) where the order parameters below $T_g$ have a negligible field dependence unless $S_g \mu_B H > kT_g$, which for $\text{PdMn}(7 \text{ at.\%})$ corresponds to a field $H > 15kG$.

The temperature dependence of the linewidth is given by $T_2^{-1} = a_0 r$, where $a_0$ is the zero temperature linewidth. For $\text{PdMn}(7 \text{ at.\%})$, we use $K = 4.541$ to get $a_0 = 85 \mu s^{-1}$. The Heisenberg mean field theory then gives the solid curve in Fig.5.5(a) for $\overline{J}_o = 0$ and $H = 5kG$. The linewidth in $\text{PdMn}(7 \text{ at.\%})$ clearly does not agree with the mean field theory. However, the results from the paramagnetic state show that the WW prediction is too large, which we attribute to an exclusion of the muon from sites adjacent to Mn atoms. The dashed line in Fig.5.5(a) plots $0.5a_0 r$, and thus shows that a linewidth reduction of 0.5 gives good agreement with the mean field theory. This is consistent with the Monte Carlo simulation of Sec.3.6 for the spin glass state, where the linewidth is reduced by about 0.5 for $c = 0.07$ (Fig.3.11). Note that the spin glass state linewidth reduction is not necessarily the same as the observed paramagnetic state linewidth reduction of $0.41\Delta_{WW}$ in $\text{PdMn}(7 \text{ at.\%})$.

Figure 5.5(b) shows the linewidth in ferromagnetic $\text{PdMn}(2 \text{ at.\%})$ as a function of $T/T_C$. In mean field theory, the linewidth is $T_2^{-1} = a_0 r$, where $r$ is given by the Ising SK theory for $\overline{J}_o / J = 1.2$, and $a_0 = 27.2 \mu s^{-1}$ is the zero temperature linewidth calculated from Eq.(3.29). As described in Sec.3.F, the applied
fields in Fig. 5.5(b) align the domains, and thus the polarization of every spin, along the applied field. The local spin orientations are therefore the same as in the paramagnetic phase, and the degree of line narrowing due to the exclusion mechanism is the same as in the paramagnetic phase. Using $0.83 A_{\text{WW}}$, we find $a_0 = 22.6 \mu s^{-1}$, and calculate the solid line in Fig. 5.5(b) for $H_0 = 5kG$, and the dashed line for $H_0 = 1kG$. The Ising SK model for this value of $a_0$ gives a reasonable agreement with the ferromagnetic state linewidth.

C. **AgMn** paramagnetic state

Transverse applied field $\mu$SR results on AgMn(1.6 at.%) have been published in Brown et al (1981), and Heffner et al (1982). Results on other AgMn samples are described in Heffner et al (1985). Figure 5.6 shows the linewidth $T_2^{-1}$ vs. $H$ in AgMn(3 at.%) at $T=40K$. The linear dependence with non-zero intercept was predicted in Sec. 3.D; the H=0 intercept gives the lifetime broadening $(T_2^*)^{-1} = 0.066(3) \mu s^{-1}$, which makes a significant contribution to the observed $T_2^{-1}$ at low fields. In Fig. 5.7 we plot $T_2H$ vs. $T$, where to minimize the contribution from $T_2$ we use only high fields. The point at $T=40K$ was determined from the slope of Fig. 5.6. The solid line in Fig. 5.7 is a fit to the data, and shows that like PdMn, the static broadening follows a Curie-Weiss
FIG. 5.6. AgMn(3 at.%) paramagnetic state transverse field linewidth as a function of applied field for T=40K. The solid line is a linear least-squares fit to the data.
FIG. 5.7. AgMn(3 at.%) paramagnetic state transverse field relaxation time scaled by the applied field as a function of temperature. Squares, $H=5kG$; triangles, $H=1.7kG$; circle, from slope of Fig. 5.6, $H=50-800G$. The solid line is a linear least-squares fit to the data.
form. Here \( \theta = 11.0(5) \text{K} \), consistent with the susceptibility data of Majumdar et al (1983), and \( A = 0.206(5) \text{ K}^{-1}\mu \text{s}^{-1} \).

Very similar results are seen in AgMn(1.6 at.\%). Figure 5.8 shows \( T_2^{-1} \) vs. \( H \) at \( T = 15 \text{K} \) and \( 10 \text{K} \), which is quite close to the transition temperature \( T_g = 7.6 \text{K} \). At both temperatures there is noticeable curvature. The linear portion of the 15K data gives a small lifetime broadening contribution, \( T_2^{-1} = 0.020(4) \mu \text{s}^{-1} \), which is quite negligible at higher fields. Figure 5.9 shows \( T_2H \) vs. \( T \), which again shows reasonable agreement with the Curie-Weiss form. The solid line gives \( \theta = 5.4(3) \text{K} \) and \( A = 0.256(6) \text{ K}^{-1}\mu \text{s}^{-1} \).

We have also made measurements in a very dilute sample, AgMn(0.3 at.\%). Figure 5.10 shows \( T_2^{-1} \) vs. \( H/T \) for two temperatures well above the glass transition at \( T_g = 2 \text{K} \) (Cannella and Mydoosh, 1974). At this low concentration and large reduced temperature \( T/T_g \), we expect a negligible \( \theta \) and lifetime broadening. The solid line is a fit to the data ignoring these effects, which gives \( A = 0.270(7) \text{ K}^{-1}\mu \text{s}^{-1} \).

The coupling strength \( A \) is essentially constant from \( c = 0.003 \) to \( c = 0.016 \), a factor of over 5 in concentration. This is strong evidence against the existence of an exclusion mechanism like that in PdMn, where there was significant concentration dependence to \( A \). The value of \( A \) at the highest concentration (\( c = 0.03 \)) is about 20\% smaller. We can average the two most dilute samples to find \( A = 0.262(4) \text{ K}^{-1}\mu \text{s}^{-1} \).
FIG. 5.8. AgMn(1.6 at.%) paramagnetic state transverse field linewidth as a function of applied field. Open circles, $T=10K$; closed circles, $T=15K$. The solid line is a linear least-squares fit to the $T=15K$ data for $H=2kG$. The dashed lines are calculated from the Heiseberg SK mean field theory, with parameters given in the text.
FIG. 5.9. AgMn(1.6 at.%) paramagnetic state transverse field relaxation time scaled by the applied field as a function of temperature. Squares, H=5kG; triangles, H=1kG. The solid line is a linear least-squares fit to the data.
FIG. 5.10. AgMn(0.3 at.%) paramagnetic state transverse field linewidth as a function of applied field over temperature. Open circles, T=18K; closed circles, T=25K. The solid line is a fit to the data constrained to pass through the origin.
Magnetization data on the $\mu$SR $\text{AgMn}(1.6 \text{ at.} \%)$ sample (Azevedo) give $\theta=6.2K$, in fair agreement with $\theta$ determined from the linewidth. The Curie constant gives an effective moment $P_{\text{eff}}=g[S(S+1)]^{1/2}=5.53$, in good agreement with literature values (Mydosh and Nieuwenhuys, 1980), but significantly smaller than for a free Mn$^{++}$ ion. This reduction can be understood in terms of the Anderson model, where the spin value has been reduced from $S=2.5$ to $S=2.31$ due to part of the minority spin band being below the Fermi energy (Podloucky et al., 1980). The $g$-factor is unchanged, so the dipolar coupling strength $B=\gamma_{\mu}\mu_{B}$ is unchanged. We can express the WW linewidth formula (for pure dipolar coupling) in terms of $A$ as defined in Eq. (5.1), and in a form convenient for comparison with magnetization data:

$$A = 5.065 \rho \gamma_{\mu} \frac{(P_{\text{eff}} \mu_{B})^2}{3k_{B}}$$  \hspace{1cm} (5.3)

We use $P_{\text{eff}}=5.53$ to obtain the dipolar $A_{\text{dip}}=0.160 \text{ K}-\text{G}^{-1} \cdot \text{ms}^{-1}$. The total $A$ must include the RKKY contribution, which was estimated in Sec. 3.B. The total, mixed coupling linewidth is then $A_{\text{WW}}=1.24 A_{\text{dip}}=0.198 \text{ K}-\text{G}^{-1} \cdot \text{ms}^{-1}$.

The experimental value of $A$ is larger than $A_{\text{WW}}$ by 32%, the opposite situation to that in PdMn. It is reasonable to attribute this discrepancy to an inaccurate estimate of the RKKY contribution to the muon-impurity coupling (Heffner et al., 1985). We can use
the observed ratio $A/A_{dip} = 1.64(2)$ to determine $J(0)$ from the theory of Chap.3, which gives $J(0) = 2.9(1)$ eV. This value of $J(0)$ is considerably larger than obtained by NMR (Table 3.1), but the NMR data may be affected by metallurgical problems (Walstedt and Walker, 1975). AgMn is the only known case of a significant RKKY contribution to the muon-local moment coupling. In all other materials studied by μSR, only dipolar coupling is considered significant (Karlsson, 1982).

Figure 5.11 shows detailed high field measurements of $T_2^{-1}$ above and below $T_g$ in AgMn (1.6 at.%). The dashed line is the Curie-Weiss form [Eq.(5.1)], with parameters determined by the high temperature data in Fig.5.9. The data are not well described by the high temperature Curie-Weiss form near $T_g$. We can try to improve on the simple Curie-Weiss form with the SK mean field theory of Chap.4. We use the observed RKKY coupling strength from the paramagnetic Curie-Weiss plot (Fig.5.9) $A/B = 1.74$, and $S = 2.3$ to calculate the $T = 0$ linewidth $a_0 = 19.3$ μs$^{-1}$. Then $T_2^{-1} = a_0 r$, where $r$ is given by the Heisenberg SK model with $J_0/J = 8/T_g$ and $T_g = J(S+1)/(3S)$. The result is the solid line in Fig.5.11, which shows good agreement with the data through the transition.

A second comparison with the SK model can be made using the field dependence of the linewidth at 10K and 15K in Fig.5.8. The dashed lines are calculated from the SK model, with the same parameters as above. The SK model reproduces the general features
FIG. 5.11. Ag$_{100}$Mn$_{1.6}$ at. % paramagnetic and spin glass state transverse field linewidth as a function of temperature. H=5 kG. The dashed line is Eq. (5.1) with $\theta$ and $A$ determined from Fig. 5.9. The solid line is the Heisenberg SK theory with parameters given in the text.
of the data (in particular the non-linearity at high fields), but is only accurate to 15-30%.

In conclusion, the transverse field linewidth in AgMn follows a Curie-Weiss law at high temperatures, in agreement with the theory of Chap.3, and the magnitude of the linewidth can be interpreted in terms of an unusually large RKKY contribution to the muon-impurity coupling. There is a drastic increase in the linewidth at the glass transition $T_g$, which is well described by the Heisenberg SK mean field model, both in the temperature dependence above and below $T_g$, and the field dependence just above $T_g$. 
Chapter VI. Zero applied field static linewidth results

A. Depolarization functions

Muon depolarization in both spin glasses and dilute ferromagnets is expected to follow the spin glass form, Eq. (3.24). The decay of the long-time 1/3 component of \( P(t) \) due to spin-lattice relaxation will have different forms depending on the specific dynamic model used (Uemura, 1981; Heffner et al, 1983; Uemura et al, 1985). If the correlation time \( \tau_c \) of the local field fluctuations satisfies \( a_3 \tau_c >> 1 \), then the dynamic effects will not significantly affect the short time relaxation. We will see in Chap. 8 that this condition is satisfied except very near the transition, so fits of the spin glass form to zero field \( P(t) \) data give a direct measurement of the temperature dependence of \( a_3 \). Figure 6.1 shows a typical example of zero field relaxation in an ordered state (for this case a ferromagnet). The solid line is a fit of the data to the spin glass form, where the 1/3 component is allowed to decay as an exponential.

B. Spin glass state linewidths

The identification of \( a_3(T)/a_0 \) as the order parameter \( r = |\varphi| \) is sensitive to the value of \( a_0 \). Parameters used to calculate \( a_0 \) from Eq. (3.21) are summarized in Table 6.1. In each case we have
FIG. 6.1. PdMn (2 at.%) ferromagnetic state zero applied field depolarization function at T = 5.75K. The solid line is a fit to the data of the spin glass form, Eq. (3.24), with the 1/3 component allowed to decay as an exponential.
TABLE 6.1. Calculation of the zero field T=0 linewidth $a_0$ for spin glasses. The spin glass temperature $T_g$ is determined by ac susceptibility.

<table>
<thead>
<tr>
<th></th>
<th>AgMn</th>
<th>CuMn</th>
<th>AuFe</th>
<th>PdMn</th>
</tr>
</thead>
<tbody>
<tr>
<td>$c$ (at.%)</td>
<td>1.6</td>
<td>1.1</td>
<td>1.0</td>
<td>7.0</td>
</tr>
<tr>
<td>$S$</td>
<td>$2.31^a$</td>
<td>$2.0^b$</td>
<td>$1.83^c$</td>
<td>$2.5^d$</td>
</tr>
<tr>
<td>$T_g$</td>
<td>$7.6^e$</td>
<td>$10.8^f$</td>
<td>$9.1^g$</td>
<td>$4.8^h$</td>
</tr>
<tr>
<td>$a_0$ (dipolar) ($\mu$s$^{-1}$)</td>
<td>15.5</td>
<td>13.4</td>
<td>7.7</td>
<td>85.3</td>
</tr>
<tr>
<td>$a_0$ (mixed) ($\mu$s$^{-1}$)</td>
<td>19.2</td>
<td>13.4</td>
<td>7.7</td>
<td>85.3</td>
</tr>
</tbody>
</table>

$^a$Azevedo  
$^b$Cohen and Slichter (1980)  
$^c$Morgownik and Mydosh (1983a)  
$^d$Star et al (1975)  
$^e$see Table 2.3  
$^f$Uemura et al (1985)  
$^g$Uemura and Yamazaki (1983)
used g=2, and the spin value determined from the effective moment. The RKKY contribution to $a_0$ is significant only in AgMn, where we have used the s-d exchange energy determined in Chap.5 from the uSR linewidth. Figure 6.2 shows $a_s/a_0$ vs. $T/T_g$ for AgMn(1.5 at.%), CuMn(1.1 at.%) (Uemura et al, 1985) and AuFe(1.0 at.%) (Uemura and Yamazaki, 1983). The scaled plot shows the spin freezing in all three spin glasses to have the same temperature dependence, and $a_s$ appears to approach the calculated value of $a_0$. The solid line in Fig.6.2 is the linewidth order parameter $r$ calculated from the Heisenberg mean field theory for $S=2$. The difference in spin values for the three systems has a negligible effect on $r$ in comparison with the errors in the data.

The measured linewidth in PdMn(7 at.%) just below $T_g$ is shown in Fig.6.3. The solid line is the Heisenberg mean field theory for $J_0=0$ and $a_0=85.\mu s^{-1}$ from Table 6.1. Unlike the spin glasses in Fig.6.2, the mean field theory strongly disagrees with the data, suggesting that we have overestimated $a_0$. As in the transverse field data, the unexpectedly small linewidth can be explained by the exclusion of muons from octahedral sites adjacent to Mn atoms. As described in Sec.5.B, $a_0$ should be reduced by a factor 0.5 to account approximately for the exclusion mechanism. The dashed line in Fig.6.3 shows the Heisenberg mean field theory for the reduced value $a_0=42.5\mu s^{-1}$. The same data are plotted in Fig.6.2 scaled by the reduced value of $a_0$, and are seen to be in good agreement with the temperature dependence of $a_s$ in the other spin glasses.
FIG. 6.2. Zero applied field spin glass static linewidths as a function of scaled temperature. Closed circles, AgMn(1.6 at.%); open circles, CuMn(1.1 at.%); triangles, AuFe(1.0 at.%); squares, PdMn(7 at.%). Scale parameters $a_0$ and $T_g$ are given in Table 6.1 and in the text. The solid line is the Heisenberg SK theory linewidth parameter $r$ for $S=2$ and $J_0=0$. The dashed line is the percolation model order parameter $\phi(t)$. 

FIG. 6.3. PdMn(7 at.%) zero applied field spin glass static linewidth as a function of temperature. The solid and dashed lines are the Heisenberg SK theory predictions for $a_0 = 85\mu$s$^{-1}$ and $43\mu$s$^{-1}$, respectively.
C. Homogeneous vs. inhomogeneous freezing in spin glasses

In the SK mean field theory, all spins develop some static spin polarization $c$ at a single temperature $T_g$, in that the probability for any spin to have $c=0$ is one above $T_g$ and zero below $T_g$. We define this situation as homogeneous spin freezing, and any mean field model with infinite ranged interactions will be homogeneous. Fig.6.2 shows that the homogeneous SK theory with its prediction of a second order phase transition at $T_g$ gives a reasonable description of the zero field static linewidth.

As an alternative to the mean field theory, we now describe an inhomogeneous model of the spin glass freezing, based on percolation theory, and ask to what extent this model is consistent with the data of Fig.6.2. In this model the freezing process is not a new kind of phase transition, but rather is similar to a large class of percolative transitions, such as polymer gelation, the metal-insulator transition in composite materials, and the (structural) glass transition (Zallen, 1983).

The percolation concept is often applied to systems where magnetic atoms with near-neighbor interactions are distributed at random on a lattice. Long range order is possible only above the critical concentration, where the magnetic atoms can form an unbounded (infinite) cluster of near-neighbor atoms. There are two difficulties in applying the usual percolation ideas to spin glasses: how to define near-neighbor clusters of atoms, and how to
get a percolative transition as a function of temperature. These problems were resolved by Smith (1974) and later extended by Mookerjee and Chowdhury (1983). Consider two spins, separated by \( r_{i,j} \), with an RKKY exchange energy \( J(r_{i,j}) = R \cos(2k_F r_{i,j})/(2k_F r_{i,j})^3 \). If \( kT < |J(r_{i,j})| \), then the two spins are highly correlated and will form a pair of locked spins. If \( kT > |J(r_{i,j})| \), then the two spins are uncorrelated. This suggests that a magnetic cluster can be defined as a group of spins coupled by exchange bonds at least as strong as the thermal energy. This definition does not necessarily mean that all spins in the cluster will be aligned. For a spin glass, we expect a random but static orientation of spins within each cluster.

With this definition of a magnetic cluster, the spin glass freezing can be described as a percolation transition. At \( T=0 \), all spins will be locked together in a single infinite cluster. As \( T \) is raised, finite spin clusters will break away from the infinite cluster, until at some critical temperature which we identify as \( T_g \), the single infinite cluster is broken up into finite clusters. The percolation transition is described by a geometrical order parameter \( \phi(T) \), defined as the probability that a spin is part of the infinite cluster. \( \phi(T) \) can be calculated with a Monte Carlo simulation program by distributing spins at random in a cubic volume with periodic boundary conditions. An algorithm identifies clusters of spins linked by exchange energies with \( |J(r_{i,j})| > kT \). For \( T>T_g \), \( \phi(T) \) is approximately equal to the fraction of spins in
the largest cluster. An average of several simulations of $\phi(T)$ is plotted as the dashed line in Fig.6.2, where we have used $c=0.1$, and values of $k_F$ and $a_o$ appropriate for Ag. The effect of a finite sample volume must be taken into account in estimating $\phi(T)$ just below $T_g$. The estimated critical temperature is $T_g=4.9(4) \times 10^{-5} R$, where $R$ is the Mn-Mn RKXY amplitude.

The percolation model is clearly inhomogeneous, since a fraction $1-\phi(T)$ of all spins (in finite clusters) fluctuate rapidly, and have no static spin polarization. If the remaining spins in the infinite cluster have a distribution of static spin polarization $P_m(\sigma)$, then the order parameters $q$ and $r$ can be calculated from the overall distribution

$$p(\sigma) = (1 - \phi(T)) \delta(\sigma) + \phi(T) P_m(\sigma), \quad (6.1)$$

where the direction of the spin polarization is random at each site. We will use two plausible assumptions for the distribution $P_m(\sigma)$ and calculate the temperature dependence of the order parameters in each case.

(1). Frozen infinite cluster. In this model we suppose that each spin in the infinite cluster is randomly oriented and completely static ($\sigma=1$). The definitions of the order parameters as averages over the distribution $p(\sigma)$ give immediately $r=q=\phi(T)$. This is significantly different from the SK Heisenberg theory, where to a good approximation, $r=q^{1/2}$. 
(2). Uniform spin polarization in the infinite cluster. In this model we suppose that all spins in the infinite cluster are randomly oriented and have the same temperature dependent spin polarization $a_a(T)$, so $p_a(a) = \delta(a - a_a(T))$. The order parameters are $q = \phi(a_a(T))^2$ and $r = \phi(a_a(T))$, which implies that $r$ and $q$ are related by $r^* = (\phi q)^{1/2}$.

The dashed line in Fig.6.2 shows $r = \phi(T)$ for comparison with $a_a(T)/a_0$. The poor agreement suggests that the frozen infinite cluster model is inadequate for describing the spin glass order. Since $\phi(T)$ is larger than the data, we can have good agreement with the uniform spin polarization model with a reasonable $a_a(T)$.

In the absence of a theory for the infinite cluster spin polarization $a_a(T)$, we can further test the percolation model from the predicted relationship between $r$ and $q$. Given an independent measurement of $q(T)$, we can distinguish the various models by calculating $r$ from $q$ and comparing with $a_a(T)/a_0$. We will consider two methods for measuring $q(T)$: the first from the low field susceptibility, the second from Monte Carlo simulation.

A method for determining $q(T)$ from low field susceptibility was developed by Edwards and Anderson (1975), and extended by Fischer (1975). The derivation assumes that the exchange energy distribution $p(J_{ij})$ is symmetric, and that the susceptibility follows a Curie-Weiss Law, $\chi^0 = C/(T - \theta)$ for all $T > T_c$. The measured low field susceptibility is related to $q(T)$ by (Kirkpatrick and Sherrington, 1978)
\[ \chi(T) = C \frac{1 - q(T)}{T - \theta \left[ 1 - q(T) \right]} \]  

(6.2)

Unfortunately, \( \chi(T) \) in AgMn (Majumdar et al, 1983) and CuMn (Morgownik Mydosh, 1981) shows significant deviations from Curie-Weiss behavior between \( T_g \) and 3\( T_g \), which has been attributed to the presence of short ranged correlations in small clusters of spins (Morgownik and Mydosh, 1983a). If we apply Eq.(6.2) to the data using \( C \) and \( \theta \) from the high temperature susceptibility, we find a substantial non-zero \( q \) in the temperature range from \( T_g \) to 3\( T_g \). This is in contradiction to the \( \mu \)SR linewidth, which is zero in this temperature range. As an alternative ad hoc procedure, we can fit a Curie-Weiss law to \( \chi(T) \) just above \( T_g \), and use the resulting \( C \) and \( \theta \) to calculate \( q(T) \) from Eq.(6.2). This is an attempt to allow for the effect of short ranged correlations in calculating \( q(T) \), but is not rigorously justified. Using susceptibility data on AgMn(2 at.\%) from Cannella and Mydosh (1974), we obtain \( q^{1/2}(T) \), plotted as the solid line in Fig.6.4. Uemura et al (1985) applied the same procedure to CuMn(1.1 at.\%) susceptibility, giving the dashed line in Fig.6.4. The results in the two spin glasses are essentially identical, as expected from the identical behavior of \( a_s(T)/a_o \).

The dotted line in Fig.6.4 shows \( q^{1/2}(T) \) from detailed Monte Carlo simulations of CuMn(0.9 at.\%) by Walstedt and Walker (1982). Their results lie 15\% to 40\% lower than \( q^{1/2} \) determined by Uemura.
FIG. 6.4. Spin glass frozen moment order parameter $q^{1/2}$ as a function of temperature. The solid and dashed lines are calculated from Eq. (6.2) using susceptibility data for AgMn(2 at.%) and CuMn(1.1 at.%), respectively. The dotted line is from a Monte Carlo simulation.
et al from CuMn(1.1 at.%) susceptibility data. The Monte Carlo simulation is reliable only in confirming the general trend of $q^{1/2}(T)$, since there is uncertainty regarding the role of anisotropy in stabilizing the spin glass phase (Walstedt, 1983).

For comparison with $a_0(T)/a_o$ data, assume that $q^{1/2}(T)$ is given by the solid line in Fig.6.4, obtained from AgMn(2 at.%) susceptibility data. We plot in Fig.6.5 the same linewidth data points as in Fig.6.2. The solid line is $r=q^{1/2}$ as predicted by the SK mean field theory. The dashed line is $r=(\phi q)^{1/2}$, as predicted by the percolation theory for uniform spin polarization in the infinite cluster. The dotted line is $r=q$, as predicted by the percolation model for a frozen infinite cluster. The data clearly contradict this model as expected from the comparison in Fig.6.2 of $r=\phi(T)$ with the data. Uemura et al (1985) first pointed out the strong disagreement between $r$ and $q$ as calculated from the susceptibility, but then concluded that the spin glass order must be homogeneous, as predicted by the mean field theory. This conclusion is not justified, since the uniform spin polarization model is inhomogeneous, and is in reasonable agreement with the data. Given the uncertainty in $q(T)$, the zero field linewidth does not distinguish between homogeneous order and inhomogeneous order where the spin polarization of the infinite cluster is allowed to have some temperature dependence $a_0(T)$. 

FIG. 6.5. Comparison of zero applied field static linewidth data of Fig. 6.2 with calculations using $q$ from the solid line of Fig. 6.4. The dotted line is $r=q$ (frozen infinite cluster model). The dashed line is $r=(qq)^{1/2}$ (uniform infinite cluster spin polarization model). The solid line is $r=q^{1/2}$ (approximate mean field theory).
D. Ferromagnetic state linewidths

The T=0 zero field static linewidth in ferromagnetic PdMn(2 at.%), is $a_0 = 27 \mu s^{-1}$, calculated from Eq. (3.37) using $K = 5.065$, $S = 5/2$, and $g = 2$, which neglects coupling of the muon to the giant moment. Excluding muons from sites adjacent to Mn will reduce $a_0$, but not necessarily by the same amount in zero applied field below $T_c$ as observed in the paramagnetic state. We can estimate the reduction from the Monte Carlo calculation of Sec. 3.C, which indicates a negligible effect for $c = 0.02$, so we ignore the exclusion effect for the zero field results in PdMn(2 at.%). The static linewidth scaled by $a_0$ is shown in Fig. 6.6 as a function of $T/T_c$, where $T_c = 5.75K$ is determined from the muon spin-lattice relaxation rate (Chap. 8). The solid line is $r$ calculated from the Ising SK model with $J_0/\bar{J} = 1.2$. The data are in good agreement with the mean field prediction. We emphasize that the mean field calculation does not have an adjustable parameter, since $J_0/\bar{J}$ was determined independently from the spontaneous magnetization (Sec. 4.B).

The dot-dashed line in Fig. 6.6 is the mean field theory prediction for ferromagnetic order with no frustration (i.e. ordinary mean field theory with $\bar{J} = 0$). The obvious disagreement with experiment shows that frustration coexists with ferromagnetism, and must be included in calculating mean field
FIG. 6.6. PdMn(2 at.%) ferromagnetic zero applied field static linewidth as a function of scaled temperature. T_c=5.75K, a_o=27. μs^-1. The dot-dashed line is the ordinary mean field theory prediction (J=0). The solid line is the Ising SK mean field theory linewidth parameter r with S=5/2 and J_o/J=1.2.
order parameters. This conclusion also follows from the spontaneous magnetization data in Fig. 4.5.

We can develop a percolation theory description of the ferromagnetic ordering in PdMn, analogous to that for spin glass ordering. As in the spin glass case, the percolation model can only be compared with linewidth data given an assumption for the spin polarization in the infinite cluster. We can then test the assumptions by comparing the predicted relationship between r and the spontaneous magnetization m (there is no separate measurement of q(T) for a ferromagnet).

(1). Unfrustrated infinite cluster. In this model we assume uniform ferromagnetic order in the infinite cluster, i.e., all spins have the same polarization \( c_\infty(T) \) and are oriented in the same direction. The order parameters are \( r = m - ^c \phi_\infty(T) \), in strong disagreement with experiment, since \( r \) is approximately twice as large as \( m \) at \( 0.5T_c \). We conclude that inhomogeneous ordering without frustration is insufficient to describe ferromagnetism in PdMn.

(2). Frustrated infinite cluster. As in case (1), assume that all spins in the infinite cluster have the same spin polarization \( c_\infty(T) \). In a crude attempt to incorporate frustration into the infinite cluster, assume that a fraction \( f \) of the spins in the infinite cluster are randomly oriented (frustrated), with the remaining fraction \( 1-f \) oriented in a single direction. The order parameters are \( r = ^c \phi_\infty(T) \) and \( m = ^c \phi(1-f)c_\infty(T) = (1-f)r \). We can calculate
from the magnetization and linewidth data, with the result shown in Fig. 6.7. The frustrated fraction \( f = 0.35 \) is temperature independent from \( 0.6T_g \) to \( T_g \), but appears to increase at lower temperatures.

In summary, we have presented two theoretical approaches to the zero field static order in \( \text{PdMn}(2 \text{ at.\%}) \). The Ising SK mean field model provides an excellent description of both the linewidth order parameter \( r \) and the spontaneous magnetization \( m \). An inhomogeneous percolation model of the ferromagnetic order is consistent with \( r \) and \( m \) only if we assume that some fraction of spins in the infinite cluster are fully frustrated. The frustrated fraction is approximately 0.35.

E. Reentrant ferromagnetic state linewidths

For two dilute atomic species, the calculation of \( a_s(T) \) separates into a sum of contributions from the two species. In the present case we write explicitly for \( \text{Pd}_{1-x-y}\text{Fe}_x\text{Mn}_y \)

\[
a_s(T) = a_s(\text{Pd}_{1-x}\text{Fe}_x,T) + a_s(\text{Pd}_{1-y}\text{Mn}_y,T).
\]  

The \( T=0 \) limits for ferromagnetic order are \( a_0(\text{Pd}_{1-x}\text{Fe}_x)=3.3\mu\text{s}^{-1} \), using \( S=1.75 \) as determined by neutron scattering in \( \text{PdFe} \) (Low and Holden, 1966), and \( a_0(\text{Pd}_{1-y}\text{Mn}_y)=68\mu\text{s}^{-1} \), using \( S=5/2 \). The giant moment contribution is neglected in both cases.
FIG. 6.7. Fraction of frustrated spins in the infinite cluster for PdMn(2 at.%) as a function of scaled temperature, calculated from $m$ (Fig. 4.5) and $r$ (Fig. 6.6).
The zero field linewidth $a_s$ was measured at several temperatures below the Curie temperature $T_C=8.25K$, (determined from the muon spin-lattice relaxation rate in Chap.8). At $T=7.5K$ the data can be fit to the spin glass form for $P(t)$ to get $a_s=8.7\pm1.5\ \mu s^{-1}$, which is plotted in Fig.6.8. At $T=6K$, $a_s$ becomes too large to observe within the dead time of the apparatus. This sets a lower limit to $a_s$, indicated by an error bar in the figure. The temperature dependence expected in two different models will be discussed below.

We first assume that the Ising SK theory describes the magnetic ordering. Since the SK model is infinite ranged, the Fe and Mn linewidths will have the same temperature dependence, and $a_s(T)=a_0 r$ where $r$ is the linewidth order parameter and $a_0=a_0(Pd_{1-x}Fe_x)+a_0(Pd_{1-y}Mn_y)=71\ \mu s^{-1}$. The Ising SK phase diagram and the observed ratio $T_g/T_C=0.23$ (see Sec.8.B) gives $J_0/\overline{J}=1.12$. The result for $a_s(T)$ is shown as the solid line in Fig.6.8, which appears to be in rough agreement with the observed rapidly increasing rates. If muons are excluded from sites adjacent to Mn atoms in $PdFeMn$, then $a_s$ should be smaller by roughly 30%. This does not affect the conclusion that $a_s$ increases rapidly below $T_C$, as predicted by the SK theory.

An alternative model has been suggested (Senoussi et al, 1980; Kettschau et al, 1983) in which only the Fe spins develop a spontaneous magnetization at $T_C$. Below $T_C$, Mn spin clusters would remain loosely coupled to each other and to the Fe spins, finally
FIG. 6.8 Temperature dependence of the zero field static linewidth in PdFeMn (5 at.%). Solid line, SK mean field theory. Dashed line, SK mean field theory for separate transitions in the Fe and Mn spin systems. Data point is a zero field measurement. The arrow indicates a temperature where $a_s(T)$ is greater than 30 $\mu$s$^{-1}$. 
freezing at \( T_g \). This separate transition model is supported by the similarity in transition temperatures between \( \text{PdFeMn}(5 \text{ at.\%}) \) with \( T_c = 8.25\text{K} \), \( T_g = 2.\text{K} \), (see Chap.8) and the binary alloys with the same Fe and Mn concentrations, ferromagnetic \( \text{PdFe}(0.35 \text{ at.\%}) \) with \( T_c = 9.\text{K} \), (Verbeek et al, 1978) and spin glass \( \text{PdMn}(5 \text{ at.\%}) \) with \( T_g = 3.\text{K} \) (Ho et al, 1981c). The transition temperatures in the binary alloys are only slightly above the corresponding transition temperatures in \( \text{PdFeMn} \). In this model the zero field linewidth would be expected to saturate at the Fe only limit \( a_0 = 3.3\mu s^{-1} \) between \( T_g \) and \( T_c \) because the Mn spins would not be static in this temperature range. Below \( T_g \), the Mn spins would freeze, and \( a_g(T) \) should rise dramatically. This temperature dependence is shown as the dashed line in Fig.6.8, calculated from the Ising SK theory for the two uncoupled spins systems using parameters determined from \( T_c \) and \( T_g \). The datum at 7.5K and the observed lower limit of the linewidth at lower temperatures are clearly inconsistent with this picture indicating that the Mn spins do participate in the ordering at \( T_c \).
VII. Theory of muon spin-lattice relaxation

This chapter describes the theory of muon depolarization in longitudinal and zero applied field due to fluctuations in the muon's local field. The goal is to develop a theory which extracts an effective correlation time of the impurity moments from the observed muon depolarization. Section A describes the theory for the paramagnetic state, and Sec.B describes various models that have been used to describe ordered state relaxation.

A. Paramagnetic state

Suppose each impurity fluctuates rapidly, so that the muon depolarization from a single paramagnetic impurity is exponential. Then a muon at site j has an exponential depolarization

$$P_j(t) = \exp\left[-t \sum_i \frac{1}{T_1(\vec{r}_{i,j})}\right].$$  \hspace{1cm} (7.1)

where the sum is restricted to the occupied impurity sites surrounding the muon site j, and $T_1^{-1}(\vec{r}_{i,j})$ is the relaxation rate due to a single impurity at $\vec{r}_{i,j}$ from the muon. For pure dipolar coupling,
\[
\frac{1}{T_1(\hat{r}_{ij})} = B^2 S(S+1) \frac{\tau_c}{1 + \gamma_\mu \frac{2H_0}{\tau_c} \frac{2}{3}} \\
\times [3\sin^2 \theta \cos^2 \theta + \frac{1}{6}(1-3\cos^2 \theta)^2 + \frac{3}{2}\sin^4 \theta] r_{ij}^{-6},
\]

(7.2)

where \(B = \gamma_\mu E_B\) is the dipolar coupling strength, \(\tau_c\) is the impurity correlation time, \(H_0\) is the applied longitudinal field (possibly zero) and \(\theta\) is the angle between \(\hat{r}_{ij}\) and the muon polarization direction. The overall depolarization function \(P(t)\) is an average of the \(P_j(t)\) over all muon sites. For a single crystal sample, \(P(t)\) will be anisotropic through the angular dependence of the dipolar coupling. For a polycrystalline sample, the observed \(P(t)\) is an average of the anisotropic \(P(t)\) in each crystallite. This averaging procedure is commonly approximated by taking an angular average of \(1/T_1(\hat{r})\) before calculating \(P_j(t)\), which gives an isotropic \(P(t)\). The resulting error is smaller than typical errors in the experimental data (Seymour and Scholl, 1985). The angular averaged relaxation rate can be written \(1/T_1(r) = K/r^6\), where \(K\) depends on the correlation time and the muon-impurity coupling strength. For mixed dipolar and RKKY coupling (McHenry et al., 1972),

\[
K = \frac{4}{3} (B^2 + 4A^2) S(S+1) \frac{\tau_c}{1 + \gamma_\mu \frac{2H_0}{\tau_c} \frac{2}{3}}.
\]

(7.3)
A simplified expression for the muon depolarization from a random distribution of paramagnetic impurities can be derived from assumptions identical to those used by Walstedt and Walker (1974) in their derivation of the transverse field linewidth (sect. 3.C). With $1/T_1 = K/r^6$, the depolarization function is (McHenry et al., 1972),

$$P(t) = \prod_j \left[ 1 - c + c \exp(-tK/r_j^6) \right]. \quad (7.4)$$

We can evaluate Eq. (7.4) analytically in the dilute limit by replacing the lattice sum with a continuum approximation integral, having an inner cut-off radius $r_0$. The result is (McHenry et al., 1972; Seymour and Scholl, 1985),

$$P(t) = \exp\left\{ -\frac{4\pi}{3} r_0^3 \rho c \right\}$$

$$\left[ \exp\left( -\frac{t}{t_0} \right) - 1 + \frac{t}{t_0} \right]^{1/2} \text{erf}\left[ \frac{t}{t_0} \right], \quad (7.5)$$

where $t_0 = r_0^6/K$.

Of particular interest are the limiting forms of $P(t)$ for very long times and very short times. For $t \gg t_0$, $P(t)$ has the 'root-exponential' form, $P(t) = \exp[-(\lambda t)^{1/2}]$, with relaxation rate $\lambda$ independent of the cut-off radius $r_0$. 
\[ \lambda = \frac{16\pi^3}{9} \rho^2 c^2 K. \]  

For \( t \ll t_0 \), \( P(t) \) is an exponential, with relaxation rate \( (T_1)^{-1} \) dependent on \( r_o \):

\[ (T_1)^{-1} = \frac{4\pi \rho}{3} \frac{c}{r_o^3} K. \]

The short time exponential regime may not be observable if little depolarization takes place in a time \( t_0 \). From Eq. (7.5) we find \( P(t_o) = \exp(-3.60r_o^3\rho c) \), which is close to one at low concentrations for typical values of \( r_o \).

Figure 7.1 compares the calculation of \( P(t) \) from the lattice sum result valid for any concentration, with the continuum approximation for \( c = 0.05 \). The dashed line is calculated from Eq. (7.4) where the sum is over all impurity sites, while the dotted line excludes sites adjacent to the muon. The corresponding solid lines are the continuum theory, with \( r_o \) chosen to fit the lattice sum results. The lower curve has \( r_o = 0 \), so if all sites are included in the lattice sum, \( P(t) \) is given by the root-exponential. The upper curve has \( r_o/a = 0.7 \) where \( a \) is the fcc lattice parameter, and shows exponential decay for small \( t \). This value of \( r_o \) provides a good approximation to the lattice sum when the muon is excluded from sites adjacent to impurities. The same conclusions hold for lower concentrations.
FIG. 7.1 Comparison of \( P(t) \) from the lattice sum and continuum expressions for \( c=0.05 \). The dashed line is calculated from Eq. (7.4) including all the octahedral interstitial sites, while the dotted line excludes sites adjacent to impurities. The corresponding solid lines are calculated from Eq. (7.5) with \( \Gamma_0/a=0.0 \) and 0.7, chosen to give agreement with the lattice sum calculation. The time has been scaled by the root-exponential relaxation rate \( \lambda \), given by Eq. (7.6).
A basic assumption of this theory is that the depolarization due to a single fluctuating paramagnetic impurity is exponential, which is true in the limit of rapid fluctuations. The range of correlation times for which this is true can be evaluated analytically for zero applied field in the paramagnetic state. Suppose the correlation function between the $\alpha$ and $\beta$ components of the local field is $G_{\alpha\beta}(t)=\langle H_\alpha(t)H_\beta(0) \rangle$. Assume that different components of the local field are uncorrelated, so $G_{\alpha\beta}(t)=0$ if $\alpha\neq\beta$, and assume that the fluctuations are isotropic, so $G_{\alpha\alpha}(t)=G(t)$. With these assumptions for the local field correlation function we can derive an exact expression for the depolarization function $P(t)$ from the equation of motion of the muon density matrix. The result is

$$\frac{dP(t)}{dt} = -2\gamma \mu^2 \int_0^t G(t-t') P(t') \, dt'. \tag{7.8}$$

If the correlation function is exponential, $G(t)=\langle H^2 \rangle \exp(-t/\tau_0)$, then Eq. (7.8) can be solved analytically by a Laplace transform,

$$P(t) = \exp\left( \frac{-t}{2\tau_0} \right) \left[ \cosh\left( \frac{X}{2\tau_0} t \right) + \frac{1}{X} \sinh\left( \frac{X}{2\tau_0} t \right) \right], \tag{7.9}$$

where

$$X = (1 - 4\omega^2\tau_0^2)^{1/2}, \tag{7.10}$$

and
\[ \omega^2 = 2 \gamma_\mu^2 \langle H^2 \rangle. \] \hspace{1cm} (7.11)

To first order in \( \omega \tau_c \), Eq.(7.9) reduces to an exponential depolarization,

\[ P(t) = \exp \left( -\frac{t}{\tau_1} \right), \quad \frac{1}{\tau_1} = \omega^2 \	au_c, \] \hspace{1cm} (7.12)

which is the standard expression for rapid fluctuations in a small applied field (Slichter, 1970).

We can now evaluate the validity of the rapid fluctuation limit as an approximation to the exact analytic form of Eq.(7.9) for a typical value of the local field strength \( \omega \). Given typical error bars in the measurement of \( P(t) \), we can assume the two forms are distinguishable only if for some range of \( t \),

\[ |P(t) - \exp(-t/\tau_1)| > 0.01, \] \hspace{1cm} where \( P(t) \) is given by Eq.(7.9), and \( \tau_1 \) by Eq.(7.12). This condition can only be satisfied if \( \omega \tau_c > 0.2 \). Using \( a_0 \) as an average value of the fluctuating local field, we find that the depolarization at a typical muon site is exponential unless

\[ \tau_c > 0.2/a_0 = 10^{-8} \text{sec for AgMn(1.6 at.\%)}. \] \hspace{1cm} This is very much longer than the observed correlation times for \( T \gg T_g \) (Chap.8). We conclude that depolarization at each muon site follows the rapid fluctuation limit exponential.
B. Ordered state

The depolarization function $P(t)$ in both the ferromagnetic and spin glass state with static local fields is given by the spin glass form, Eq. (3.24). Fluctuations in the muon local field are reflected in the decay of the long time limit of $1/3$ in Eq. (3.24). We now review several theories which relate the relaxation rate of the $1/3$ component to the correlation time of the local field. Although developed to describe spin glass depolarization, these theories apply equally well to dilute ferromagnets, since the form of the local field correlation function is expected to be the same in both cases. Fluctuations in the muon local field are described by a single effective correlation time, which is equivalent to assuming an exponential correlation function. A simple generalization to treat the case of non-exponential spin correlation functions is described.

Full reorientation models

Uemura (1981) and Leon (1981) developed the first simple models for $P(t)$ in a spin glass. The local field at a site is chosen from a Lorentzian distribution of width $a_0$, which gives the correct static spin glass $P(t)$ for $T=0$. For $T>0$, the local field is allowed to reorient fully at each fluctuation with a temperature dependent correlation time $\tau_c$. In the limit of slow fluctuations,
\( a_0 \tau_c \gg 1 \), both models predict that the 1/3 component will decay as an exponential independent of \( a_0 \),

\[
P(t) = \frac{1}{3} \exp(-2t/3\tau_c) + \frac{2}{3} (1 - a_0 t) \exp(-a_0 t). \tag{7.13}
\]

The short time, 2/3 component is unaffected since the field does not change appreciably in a time \( 1/a_0 \). In the limit of fast fluctuations, \( a_0 \tau_c \ll 1 \), both models give the motional narrowing limit root-exponential,

\[
P(t) = \exp\left[ -(4 a_0^2 \tau_c t)^{1/2} \right]. \tag{7.14}
\]

The two models differ in the detailed behavior in the intermediate regime, where \( a_0 \tau_c \approx 1 \).

Heffner et al (1982) pointed out the major flaw in comparing the full reorientation theory with \( P(t) \) data in the spin glass phase; the model does not allow for a temperature dependent static linewidth. As shown in Chap.6, the static linewidth has a dramatic temperature dependence, which cannot be reproduced with a complete reorientation of the field at each fluctuation (Heffner et al, 1983).
**limited amplitude fluctuation models**

Heffner et al (1982) argued that the temperature dependence of 
$\alpha_s$ implies the existence of rapid, limited amplitude fluctuations.
Uemura and Yamazaki (1983) developed an approximate treatment of 
$P(t)$ for limited amplitude fluctuations, where the static linewidth 
can have an arbitrary temperature dependence described by the order 
parameter $r=\alpha_s/\alpha_o$. Uemura divided the local field at a muon site 
into a static field and a rapidly fluctuating dynamic field, chosen 
independently from Lorentzian distributions of widths $\alpha_s$ and $\alpha_d$ 
respectively, with the constraint $\alpha_s^2 + \alpha_d^2 = \alpha_o^2$. Fluctuations in the 
dynamic field are assumed to satisfy $\alpha_o \tau_c << 1$, so that the 
depolarization at each muon site can be approximated by a product 
of static and dynamic relaxation functions. Then

$$P(t) = \frac{1}{3} \exp\left( -\left[ 4\alpha_0^2 (1-r^2) \tau_c t \right]^{1/2} \right)$$

$$+ \frac{2}{3} \left[ 1 - x(t) \tau_0 t \right] \exp\left( -\tau_0 t/x(t) \right), \quad (7.15)$$

where

$$x(t) = \left[ 1 + 4(r^2-1) \tau_c / t \right]^{-1/2}. \quad (7.16)$$

If we restrict the application of $P(t)$ to measuring times $t>0.1\mu s$, 
a $T=0$ linewidth $\alpha_o>20\mu s^{-1}$, and temperatures not too close to $T_g$ so 
that $r>0.3$, then the condition that $\alpha_o \tau_c << 1$ implies $x(t)=1$, and the 
short time part of Eq. (7.15) is the same as the static spin glass
form. These assumptions are equivalent to saying fluctuations are so rapid that the rapidly fluctuating part of the local field induces very little depolarization within a time $1/a_s$. In Uemura's model, the correlation time can be determined by fitting the $1/3$ component of $P(t)$ to the root exponential rate $\lambda$.

$$\lambda = 4 \ a_o^2 \ (1 - r^2) \ \tau_0. \quad (7.17)$$

Leon (1984) and Heffner et al (1983) developed a more detailed model of limited amplitude fluctuations, called the quasimagnon model, in analogy with spin wave excitations in a ferromagnet. They divided the local field at a site into a static field $H_0$ distributed as a Lorentzian with width $a_s$, and a fluctuating field distributed as a Gaussian with width $\Delta = \Delta H_0$. The constant $\alpha$ is the characteristic fluctuation angle of the total field, and is related to the static linewidth order parameter by $r = (1 + 3 \alpha^2)^{-1/2}$ (Leon, 1983). The condition $\Delta = \Delta H_0$ at each site imposes a correlation between the amplitude of the static and fluctuating fields at each site, which is absent in Uemura's model. The depolarization function $P(t)$ can be derived analytically in the limit of rapid fluctuations ($a_s \tau_s << 1$) and for small $\alpha$ (temperatures not too close to $T_g$). The long time ($a_s t >> 1$) $1/3$ component of $P(t)$ decays as

$$P(t) = \frac{1}{3} \ \frac{d^2}{dz^2} \left[ \exp(z^2/2) \ \text{Erfc}(z/\sqrt{2}) \right] \bigg|_{z^2 = (4a_o^2a_s^2\tau_0)} 1/2. \quad (7.18)$$
Expanding to lowest order in $a_s \tau_c$ gives

$$p(t) = \frac{1}{3} \left[ 1 - \left( \frac{16}{\pi} a_s^2 a_s^2 \tau_c \right)^{1/2} + \ldots \right], \quad (7.19)$$

which is the first term in an expansion of the root exponential, so like the Uemura model, the $1/3$ component decays approximately as a root exponential with relaxation rate

$$\lambda = 1.70 a_0^2 \left( 1 - r^2 \right) \tau_c, \quad (7.20)$$

which differs from Uemura's model [Eq. (7.17)] by a factor of over two in the constant. This difference is presumably due to the correlation imposed by a constant $\alpha$ at all sites.

**Non-exponential spin correlation functions**

The limited amplitude fluctuation models are based on an exponential impurity spin auto-correlation function first considered by Edwards and Anderson (1975),

$$\langle S_i S_i(t) \rangle = q + (1 - q) \exp(-t/\tau_c). \quad (7.21)$$

where $q$ is the Heisenberg spin glass order parameter. An important assumption necessary to relate muon depolarization to spin correlations is that the local field correlation function has the
same form as the spin correlation function. This assumption is supported by the good agreement between the spin correlation time measured by neutron spin-echo experiments, and the muon local field correlation time above $T_g$ (Heffner and Maclaughlin, 1984). The power spectral density of the local field fluctuations is then taken to be the same as the power spectrum of $S_i S_j(t)$,

$$J(\omega) = \frac{1}{2\pi} \int_{-\infty}^{\infty} <S_i S_j(t)> e^{-i\omega t} dt, \quad (7.22)$$

$$J(\omega) = q \delta(\omega) + (1-q) G(\omega), \quad (7.23)$$

where $G(\omega)$ is the spectral density of the fluctuating part of the correlation function. For the exponential correlations of Eq.(7.21),

$$G(\omega) = \frac{1}{\pi} \frac{\tau_c}{1 + \omega^2 \tau_c^2}. \quad (7.24)$$

In general, the muon relaxation rate is proportional to $G(\omega)$ evaluated at the Larmor frequency $\omega_\mu$ in the local static field, provided that $G(\omega_\mu)$, which acts like an effective correlation time, is much shorter than the resulting muon relaxation time (Slichter, 1970). Since there is a distribution of static field strengths, we take $a_2$ as a measure of $\omega_\mu$, and the relaxation rate is proportional to $(1-q)G(a_2) = (1-q)\tau_c/\pi$. Approximating $q=\tau^2$ as found in the Heisenberg SK theory, we can generalize Eq.(7.20) for rapid
exponential correlations to the case of an arbitrary power spectral density $G(\omega)$.

$$\lambda = 5.33 \, a_\alpha^2 \,(1 - n^2) \, G(a_\alpha),$$  \hspace{1cm} (7.25)

This expression is valid provided $a_\alpha G(a_\alpha) \ll 1$, where $a_\alpha / \gamma$ is a measure of the average fluctuating field strength, and (like Eq. (7.20)) $\alpha \ll 1$ (temperatures not too close to $T_g$). In Chap. 9 we will use this equation to calculate $\lambda$ for various $G(\omega)$. 
VIII. Muon spin-lattice relaxation results

Muon spin-lattice relaxation has been measured in longitudinal and zero applied field for PdMn and AgMn alloys. Results on PdMn alloys have been published previously in Dodds et al, (1983) and Heffner et al (1984). Results on reentrant PdFeMn have been published in Gist et al (1985), and on AgMn in Heffner et al (1982) and MacLaughlin et al (1983). Section A will describe results in the paramagnetic state and the interpretation of calculated impurity correlation times. Sections B and C describe results at and below the transition temperature in spin glasses and ferromagnets. These results are discussed in detail in Chap. 9.

A. Paramagnetic state

1. Pd based alloys

Zero field muon depolarization data P(t) can be fit to either an exponential or root-exponential function with reasonable values of the goodness of fit parameter \( \chi^2 \). Data on PdMn(2 at.%) above \( T_C \) on average favor the exponential, so exponential rates were reported in the publication of this data (Dodds et al, 1983). Available data on PdMn(7 at.%) above \( T_g \) do not distinguish between the exponential and root-exponential, and root-exponential rates were published in Heffner et al (1984). The ambiguity between the
two functional forms can be resolved by accounting for the exclusion of the muon from sites adjacent to impurities, as was found in Chap. 5 and 6 from static linewidth results. Table 8.1 shows $\chi^2$ for fits of $P(t)$ to the exponential and root-exponential at three temperatures above $T_C$ in PdFeMn (5 at.%). The table demonstrates an apparent change from an exponential form at the highest temperature (lower relaxation rate) to a root-exponential form at the lowest temperature (higher relaxation rate). Neither function is adequate to fit the entire temperature range. The depolarization function with exclusion is given by Eq. (7.5) with $r_0$ considered an adjustable parameter. We can determine $r_0$ experimentally by fitting to Eq. (7.5) in a region where the data do not follow either limiting form. (The root-exponential limit is independent of $r_0$, and the exponential limit determines a combination of $K$ and $r_0$.) The data at $T=10K$ satisfy this requirement, and were fit to Eq. (7.5) adjusting both $K$ and $r_0$. The result is $r_0/a=0.72(8)$, where $a$ is the Pd fcc lattice parameter. As shown in Fig. 7.1, Eq. (7.5) for this value of $r_0$ is very close to $P(t)$ calculated from a lattice sum when the muon is excluded from sites adjacent to impurities. We will assume that the proper fitting function for both PdFeMn (5 at.%) and the binary PdMn alloys at all temperatures is not an exponential or root-exponential, but is Eq. (7.5) with $r_0/a$ fixed at 0.72. The $\chi^2$ values in the last column of Table 8.1 show that this function adequately describes
TABLE 8.1. Comparison of chi-squared per degree of freedom for three fitting functions at each of three temperatures above $T_C$ in PdFeMn(5 at.%). Eq. (7.5) uses fixed $r_0$ as discussed in the text.

<table>
<thead>
<tr>
<th>$T$(K)</th>
<th>$\chi^2$ exponential</th>
<th>$\chi^2$ root-exponential</th>
<th>$\chi^2$ Eq.(7.5)</th>
</tr>
</thead>
<tbody>
<tr>
<td>12</td>
<td>1.09</td>
<td>1.64</td>
<td>1.05</td>
</tr>
<tr>
<td>10</td>
<td>1.15</td>
<td>1.29</td>
<td>1.00</td>
</tr>
<tr>
<td>9</td>
<td>1.50</td>
<td>0.74</td>
<td>0.75</td>
</tr>
</tbody>
</table>
the data in the transition region between the exponential and root-exponential forms.

Figure 8.1 shows the parameter $K$ (scaled by $T/a^6$) for three Pd alloys obtained by fitting zero field data to Eq. (7.5) with $r_0$ fixed. In zero applied field $K$ is proportional to the impurity correlation time according to Eq. (7.3). Using $A/B=0.18$ from Table 3.1, the product $\tau_c T$ is given by the scale on the right side of Fig. 8.1. At high temperatures the correlation time should be given by the Korringa expression, $\tau_c = b/T$. This is demonstrated in Fig. 8.1, where $\tau_c T$ approaches the same constant at high temperatures for all three samples. The data give

$$\frac{T K}{a^6} = \frac{4}{3} \frac{B^2 + A^2}{a^6} S(S+1) b = 0.62 \pm 0.03 \text{ K } \mu\text{s}^{-1}, \quad (8.1)$$

which gives $b=0.065(3) \text{ ns-K}$. The Korringa product $\tau_c T = b$ is simply related to the slope of the paramagnetic EPR linewidth vs. temperature $d\Delta t/dT = (\gamma_e B)^{-1}$, which gives for Pd, $d\Delta t/dT = 875.4(40) \text{ G/K}$. Alquie et al (1978) measured a linewidth slope $d\Delta t/dT = 140 \text{ G/K}$ in PdMn. Comparison with the µSR value shows that the relaxation bottleneck was only partially broken in their experiments.

Ferromagnetic PdMn alloys are known to exhibit critical phenomena in the specific heat (Boerstoel et al., 1972), and susceptibility (Ho et al., 1981a). We therefore attribute the increase in relaxation rate above $T_c$ to critical slowing down of spin fluctuations. The temperature control in these experiments is not precise enough to enter the true critical region, so we do not
FIG. 8.1 Paramagnetic state spin-lattice relaxation rates in Pd alloys as a function of scaled temperature. Open circles, PdMn(2 at.%), T_C = 5.75K; Triangles, PdFeMn(5 at.%), T_C = 8.25K; Filled circles, PdMn(7 at.%), T_g = 4.8K. The right hand scale is calculated from Eq.(7.3).
attempt to derive a critical exponent from the data. Enhanced relaxation rates have also been observed by $\mu$SR near transitions in ordinary magnetic materials. In both anti-ferromagnetic MnF$_2$ (de Renzi et al, 1982) and ferromagnetic Ni (Nishiyama et al, 1983), the effect of critical slowing down on relaxation rates is observed over a temperature range $(T-T_c)/T_c<0.02$. In contrast, critical slowing down in PdMn(2 at.%) is observed over an order of magnitude wider range in temperature, $(T-T_c)/T_c<0.2$. This is in agreement with the broad temperature range of critical small angle neutron scattering observed in PdMn(2 at.%) by Verbeek et al (1980). The temperature range of slowing down of spin fluctuations clearly increases with increasing spin disorder from ferromagnetic PdMn(2 at.%) to reentrant PdFeMn(5 at.%) to spin glass PdMn(7 at.%). However, we cannot claim from Fig.8.1 that slowing down in the more disordered samples represents true critical phenomena.

2. AgMn(1.6 at.%)

Zero field relaxation data above $T_g$ in AgMn are on average better described by the root-exponential than the exponential. There is no evidence for a transition to exponential relaxation at lower relaxation rates as is seen in PdMn alloys. The parameter $K$ is determined by fitting the data to a root-exponential, and calculating $K$ from Eq.(7.6). Results are shown in Fig.8.2, scaled
FIG. 8.2 $\text{AgMn}(1.6 \text{ at.\%})$ paramagnetic state spin-lattice relaxation rates as a function of scaled temperature, $T/T_g=7.6K$. The solid and dashed lines are fits to the data of a power law [Eq.(8.2)] using $n=1$ and $n=2$, respectively. The dot-dashed line is a fit to the data of the Vogel-Fulcher law [Eq.(8.3)]. The right hand scale is calculated from Eq.(7.3).
by $T/a^6$, where $a$ is the Ag fcc lattice parameter. Using $A/B=1.72$ as determined by the static linewidth analysis of Chap.5, the product $\tau_0 T$ is given by the scale on the right side of Fig.8.2. Note that $4A^2/B^2=11.8>1$, so in AgMn, the RKKY coupling actually dominates the dipolar coupling in calculating the spin-lattice relaxation. The arrow shows $b=0.175\text{ns}\cdot\text{K}$ measured by neutron scattering (Murani, 1981). The EPR linewidth slope measured by Davidov et al. (1975) gives a much larger value of $b$, presumably due to bottlenecks in relaxation. The high temperature values of $\tau_0$ are considerably smaller than the neutron scattering prediction. This discrepancy may be due to an overestimate of the RKKY coupling strength, or due to unexpectedly large systematic errors in measuring the low muon relaxation rates far above $T_g$.

The form of $\tau_0$ as a function of temperature above $T_g$ is not very well constrained by the available data, so that comparisons with theoretical predictions are difficult. To illustrate, we will compare the data of Fig.8.2 with two forms for $\tau_0(T)$; the first from mean field theory, the second based on an analogy with viscous slowing down in structural glasses. Mean field theories predict a power law for $\tau_0(T)$,

$$\tau_0 = \tau_0 (1 - T_g/T)^{-n}. \quad (8.2)$$

Where $\tau_0=b/T$ is the Korringa relaxation rate in the limit of negligible exchange interactions. The mean field theory of Ma and
Rudnick (1978) predicts n=1, while the mean field theory and Monte Carlo calculations of Kikpatrick and Sherrington (1978) predict n=2. Correlation times in AgMn(1.6 at.%) from Fig.8.2 have been fit to Eq.(8.2) by adjusting b with n fixed at 1 and 2, yielding $b_1=0.020$ ns-K and $b_2=0.0032$ ns-K respectively. These fits are plotted on Fig.8.2 as a solid line for n=1 and a dashed line for n=2. The n=2 curve is a better fit to the data near $T_g$, but has an unphysically small value b, and is in strong disagreement with both the high temperature $\mu$SR data and the neutron scattering value of b. The n=1 curve is in fair agreement with the data near $T_g$ and gives a reasonable value for the high temperature limit. As a preliminary conclusion based on the available data, we conclude that a power law representation of $\tau_0(T)$ above $T_g$ is consistent with n=1 and not n=2. Uemura (1981) claimed the opposite; that Eq.(8.2) with n=2 described $\mu$SR correlation times better than n=1, but he used a constant for $\tau_0$ instead of the Korringa result $\tau_0=b/T$, which gives an unphysical high temperature limit.

An alternative form of $\tau_0(T)$ is the Fulcher law (Tholence, 1980; Ramal and Souletie, 1982),

$$\tau_c = \tau_0 \exp\left(\frac{E_a}{T-T_g}\right). \quad (8.3)$$

The Fulcher law was first used to describe the temperature dependent viscosity above the glass transition in some structural
glasses, and was used by Tholence (1980) to describe the frequency
dependence of the ac susceptibility maximum in some spin glasses.
A fit to the data is shown as the dot-dashed line in Fig. 8.2 with
b=0.028 ns-K and $E_a=0.20T_g$. These are to be compared with the
results of Tholence, who found $E_a>2T_g$ in CuMn (and other spin
glasses), and $\tau_0$ was assumed to be $10^{-13}$ sec universally. The mSR
data are consistent with the Fulcher law, but do not support any
definite conclusion on the proper functional form of $\tau_0(T)$.

Emmerich et al (1985) found that zero applied field
depolarization data $P(t)$ just above $T_g$ in CuMn(0.54 at.%) could not
be described by the root-exponential. They instead applied an
inhomogeneous ordering model where large regions of the sample have
different correlation times for temperatures close to $T_g$. The data
were shown to fit the form

$$P(t) = A + (1-A) \exp\left[-(\lambda t)^{1/2}\right]. \quad (8.4)$$

Here $A$ is the temperature dependent volume fraction of regions with
rapid fluctuations and negligible muon relaxation, and the fraction
$1-A$ fluctuates with a correlation time $\tau_0$ and contributes a
root-exponential to $P(t)$. We can make a simple estimate of the
size of the separate regions necessary for a two fraction model to
hold. The muon relaxation is dominated by the closest impurity,
which is on average at a distance $r/a=2.0$ from the muon for
c=0.0054 (Chandrasekhar, 1943). For the contribution to $P(t)$ of
muons at the boundaries of the two regions to be negligible, the separate regions must have spatial dimensions much larger than $r=2.0a$, suggesting that the regions are 70-100A across. Inhomogeneities at this length scale may be caused by fluctuations in Mn concentration in the slowly cooled single crystal sample used by Emmerich et al, and would not have been detected in their microprobe analysis. The temperature dependence of $\Delta$ then reflects a distribution in $T_g$ due to large regions with different concentrations, and is unrelated to the onset of spin glass order within a single region. This interpretation would imply a rounded ac susceptibility cusp in their sample, but susceptibility measurements were not reported.

B. Spin glass state

Spin-lattice relaxation below $T_g$ in zero applied field has been observed in both AgMn(1.6 at.%) and PdMn(7 at.%) from the decay of the 1/3 component of the spin glass depolarization function. The root-exponential function was shown in Sec.7.B to describe rapid, limited amplitude fluctuations, so we fit a root-exponential to the decay of the 1/3 component in AgMn(1.6 at.%). Results are shown in Fig.8.3, with the root-exponential rate $\lambda$ expressed in terms of $K/a^6$ using Eq.(7.6). For PdMn(7 at.%), we use Eq.(7.5) with $r_0/a$ fixed at 0.72 instead of the root-exponential to determine $K/a^6$. Results above $T_g$ are
FIG. 8.3 Spin glass spin-lattice relaxation rates above and below $T_g$ as a function of scaled temperature. Open circles, PdMn (7 at.%), $T_g = 4.8 \text{K}$; Filled circles, AgMn (1.6 at.%), $T_g = 7.6 \text{K}$. The solid line is a calculation of barrier mode relaxation described in Sec. 9.C. The dashed line is a fit to the data of relaxation from algebraic spin correlation decay, described in Sec. 9.D.
from Figs. 8.1 and 8.2. Temperatures in the figure are scaled by $T_g$ as measured by ac susceptibility (Table 2.3). The peak in relaxation rate clearly gives the same transition temperature as measured by susceptibility. Fits of a simple exponential to the decay of the $1/3$ component have comparable values of $\chi^2$ to the analyses shown in the figure, due to large statistical errors, so we have not demonstrated root-exponential decay below $T_g$.

The relaxation data of Fig. 8.3 can be converted to effective impurity correlation times with the limited amplitude fluctuation model described in Sec. 7.B. The spin-lattice relaxation parameter $K$ is related to $\tau_C$ by Eqs. (7.6) and (7.20). We use $a_o$ from Table 6.1, except that in PdMn(7 at.%) $a_o$ should be multiplied by 0.5 to account approximately for the exclusion of muons from sites adjacent to impurities (see the discussion of PdMn(7 at.%) $a_o$ in Secs. 5.B and 6.B). We will assume the Heisenberg SK mean field model gives an accurate measure of the static linewidth order parameter $r$. Results are shown in Fig. 8.4, where $\tau_C$ above $T_g$ is from Figs. 8.1 and 8.2. The error bars are due only to statistical errors in fitting the data for $K$; there is no attempt to estimate the error in $\tau_C$ due to either uncertainty in $a_o$ or in the mean field values of $r$. Note in particular that the lowest temperature value for $\tau_C$ in AgMn(1.6 at.%) has a very small value of $1-r^2$, so is very sensitive to the exact value of $r$. Despite these possible sources of error, the figure clearly indicates a rapid increase in
FIG. 8.4 Spin glass spin correlation times above and below $T_g$ as a function of scaled temperature. Open circles, PdMn(7 at.%), $T_g=4.8K$; Filled circles, AgMn(1.6 at.%), $T_g=7.6K$. Correlation times below $T_g$ are calculated assuming the limited amplitude fluctuation model [Eq.(7.20)].
correlation time at $T_g$, a maximum at $T_g$, and relatively long $\tau_c$ ($10^{-9}$ sec) persisting to the lowest temperatures measured.

C. Ferromagnetic state

Spin-lattice relaxation below $T_c$ in zero applied field has been observed in ferromagnetic Pd alloys in exactly the same manner as for spin glasses. The 1/3 component of the depolarization data in PdMn(2 at.%) and PdFeMn(5 at.%) have been fit to Eq.(7.5) with $\gamma_0/a$ fixed at 0.72 to determine the spin-lattice relaxation parameter $K$. As in the spin glasses, the $P(t)$ data are not accurate enough to determine the functional form of the depolarization directly. Results for $K/a^6$ in PdMn(2 at.%) are shown in Fig.8.5, with results above $T_c$ from Fig.8.1. The peak in relaxation rate is used to define the Curie temperature $T_c=5.75K$, which is consistent with $T_c$ determined by magnetization measurements (Star et al, 1975).

We use the same procedure as in the spin glasses to convert relaxation data to effective correlation times. Results are shown as open circles in Fig.8.6, where we have used $a_0=27.\mu s^{-1}$ (see Sec.6.D for the effect of muon exclusion on the zero field $a_0$) and $r$ given by the Ising SK mean field theory. Again, the error bars are due only to statistical errors in fitting the data for $K$ with no attempt to account for errors in $a_0$ or $r$. 
FIG. 8.5 PdMn(2 at.%) spin-lattice relaxation rates above and below $T_c=5.75K$ as a function of scaled temperature.
FIG. 8.6 Ferromagnet spin correlation times above and below $T_c$ as a function of scaled temperature. Open circles, PdMn (2 at.%), $T_c=5.75K$; Filled circles, PdFeMn (5 at.%), $T_c=8.25K$. Correlation times below $T_c$ are calculated assuming the limited amplitude fluctuation model [Eq. (7.20)]. The arrow indicates the reentrant ferromagnetic-spin glass transition temperature from Fig. 8.7(a).
The filled circles in Fig. 8.6(b) give $K/a^6$ in $\text{PdFeMn}(5 \text{ at.} \%)$. The peak in $K/a^6$ is used to define the Curie temperature, $T_c=8.25(10)K$. Two additional data points (triangles) were taken in an applied longitudinal field of 1kG. The solid line indicates relaxation rates in ferromagnetic $\text{PdMn}(2 \text{ at.} \%)$ from Fig. 8.5 and the dashed line relaxation rates in spin glass $\text{PdMn}(7 \text{ at.} \%)$ from Fig. 8.3.

Figure 8.7(a) shows the temperature dependence of the real and imaginary part of the low frequency ($21.7\text{Hz}$) ac susceptibility on a spherical piece cut from from the µSR sample, in agreement with susceptibility results published previously (Verbeek et al, 1978). The transition to a ferromagnetic state is seen in the rapid increase in $\chi'$, which becomes limited by the reciprocal demagnetizing factor, and by the peak in $\chi''$. It is not known why $T_c$ determined from the peak in $K/a^6$ is somewhat lower than the peak in $\chi''$. The second transition to a spin glass-like state is marked by the lower temperature peak in $\chi''$ at $T_g=1.9K$, and by the fall off in $\chi'$. There is no comparable signal in the muon relaxation rate, although $K/a^6$ gradually decreases below $T_g$.

The limited amplitude fluctuation model can be used to convert $\text{PdFeMn}(5 \text{ at.} \%)$ relaxation rates to effective correlation times, provided we have reasonable values of $a_0$ and $r$. We use the Ising SK model for $r$, since this model predicts a reentrant transition, and provides a good description of $r$ in $\text{PdMn}(2 \text{ at.} \%)$. The Ising SK phase diagram and the observed ratio $T_c/T_g=0.23$ gives $\Omega/\mathcal{J}=1.12$. 
FIG. 8.7 (a). PdFeMn(5 at.%) real $\chi'$ and imaginary $\chi''$ parts of the low field ac susceptibility as a function of scaled temperature. $T_c=8.25K$, determined from the peak in relaxation rate (b). Spin-lattice relaxation rates above and below $T_c$ as a function of scaled temperature. Circles: zero applied field. Triangles: 1 kOe longitudinal field. The solid line represents relaxation rates in PdMn(2 at.%) from Fig. 8.6. The dashed line represents relaxation rates in PdMn(7 at.%) from Fig. 8.3.
The resulting temperature dependence for $r$ is not significantly different from Fig. 6.6 for $J_0/J = 1.20$. Figure 3.11 suggests that the effect of excluding muons from sites adjacent to Mn should reduce $a_0$ by a factor of approximately 0.65, which gives from Eq. (6.3) a dipolar $a_0 = 46 \mu s^{-1}$. Results for $\tau_C$ are shown as solid circles in Fig. 6.6, and again the error bars do not reflect uncertainties in $a_0$ or $r$.

Correlation times above $T_C$ are similar in both ferromagnetic samples, but there are dramatic differences below $T_C$. The reentrant ferromagnetic phase has correlation times substantially longer than in $\text{PdMn}(2 \text{ at.\%})$ at comparable reduced temperatures. Unlike the ferromagnet, the correlation times are approximately temperature independent to very low temperatures. In addition, the ferromagnet to spin glass transition as determined by ac susceptibility (marked by the arrow) is not accompanied by critical fluctuations in this frequency range. We conclude that the dynamics of the reentrant ferromagnet are unlike the dynamics in either spin glasses or a disordered ferromagnet without a reentrant transition.

D. Longitudinal applied field

The dependence of the muon spin-lattice relaxation on applied field is qualitatively different for ferromagnetic and spin glass order. In ferromagnetic $\text{PdMn}(2 \text{ at.\%})$, an applied field of 5 kG
completely suppresses the peak in relaxation rate shown in Fig. 8.5 (Dodds et al., 1983), presumably by suppressing the critical fluctuations. The measured relaxation rate at $T_C$ in 5 kG is consistent with zero. Similar applied fields are sufficient to suppress the peak in the critical small angle neutron scattering intensity at $T_C$ (Verbeek et al., 1980). The same behavior is seen in the reentrant ferromagnet PdFeMn(5 at.%), where an applied field of 1 kG reduces the zero field relaxation rate by a factor of 17 just above $T_C$ (Fig. 8.7). A much weaker field dependence is seen in spin glass AgMn(1.6 at.%), where an applied field of 1 kG suppresses the zero field relaxation rate at $T_g$ by a factor of six (Heffner et al., 1982).

The field dependence of the relaxation rate in the paramagnetic phase can be calculated from Eq. (7.3), assuming that the applied field does not change $\tau_C$. In a field $H_0$, the zero field relaxation rate is reduced by a factor $\left[1 + (\gamma_0 H_0 \tau_C)^2\right]^{-1}$. Using $\tau_C = 10^{-9} \text{sec}$ near $T_g$ from Fig. 8.4, and $H_0 = 5 \text{kG}$, Eq. (7.3) implies the zero field relaxation rate at $T_g$ in AgMn(1.6 at.%) is reduced by a factor of 0.85, which is far less than the observed field dependence. For the shorter $\tau_C$ near $T_C$ in PdMn(2 at.%), Eq. (7.3) predicts an reduction of less than 1%. We conclude that $\tau_C$ is directly affected by the applied field. A theory of the field dependence of $\tau_C$ for comparison with experiment is unavailable.
The small longitudinal field relaxation rates in the paramagnetic phase make a negligible dynamic contribution to the transverse applied field linewidth for fields of 1 kG and above, and temperatures not too close to \( T_g \). This confirms the results of Chap. 5, where static broadening was assumed to dominate the dynamic contribution to the transverse field linewidth.
IX. Interpretation of ordered state muon spin-lattice relaxation

A. Spin wave excitations

1. Ferromagnetic state

Nuclear relaxation in ferromagnets is often attributed to local field fluctuation caused by spin wave excitations (Mitchell, 1957; Beeman and Pincus, 1968). The spin-lattice relaxation rate is largely determined by the spin wave dispersion relation, which for ferromagnets is quadratic in the wavevector $k$,

$$\omega_k = \Gamma + D k^2,$$  \hspace{1cm} (9.1)

where $D$ is the spin wave stiffness constant and $\Gamma$ is an anisotropy gap. In the absence of a uniaxial anisotropy in PdMn, $\Gamma$ is usually dominated by the dipolar anisotropy (Keffer, 1966). In an applied field $H_0$, a spontaneous magnetization $M_0$ produces a dipolar energy gap

$$\hbar \Gamma = g \mu_B | H_0 + (N_x - N_z) M_0 |,$$  \hspace{1cm} (9.2)

where $N_z$ and $N_x$ are the sample demagnetizing factors parallel and perpendicular to the applied field. In the longitudinal field configuration we estimate $N_x/4\pi = 0.1$ and $N_z/4\pi = 0.8$ by approximating the disc shaped $\mu$SR samples with an oblate spheroid (Osborn, 1945).
The spontaneous magnetization at $T=0.5T_C$ is $c_2=2.5\mu_B/\text{Mn atom}$ (Star et al, 1975), which gives a dipolar anisotropy field $H_A=|N_x-N_z|M_0=350\text{G}$. In the limit of a small applied field, $r=6.1\times10^9\text{s}^{-1}$. We will take this as a lower bound to the actual anisotropy gap, since additional mechanisms may be present.

The local static field in a ferromagnet is modulated by the limited amplitude spin wave excitations. The spin auto-correlation function due to spin wave excitations for $T<<T_C$ can be estimated from a first order Holstein-Primakoff expansion (Kéffer, 1966). The transverse correlation function is

$$\langle S_-^+(t)S_+(0)\rangle = \frac{2S}{N} \sum_k \frac{\exp(i\omega_k t)}{\exp(\frac{\omega_k}{kT})-1}$$  \hspace{1cm} (9.3)$$

and the longitudinal correlation function $\langle S_z(t)S_z(0)\rangle$ is constant. Converting the sum over $k$ to an integral over $\omega$ using the dispersion relation, we can calculate the power spectral density for the fluctuating part of the local field as in Eq.(7.22),

$$G(\omega) = \frac{1}{2\pi} \int_{-\infty}^{\infty} \langle S_-^+(t)S_+(0)\rangle e^{-i\omega t} \, dt.$$  \hspace{1cm} (9.4)$$

The result is
\[ G(\omega) = \begin{cases} 
0 & \omega < \Gamma \\
\frac{A}{2} \frac{(\omega - \Gamma)^{1/2} \exp(-\Gamma_0 \omega / kT)}{\exp(\Gamma_0 / kT) - 1} & \omega > \Gamma 
\end{cases} \]  
(9.5)

where \( A = S/(\pi^2 p c) \). Using the generalization of the limited amplitude fluctuation model of Sec.7.B, the root-exponential decay rate of the 1/3 component is proportional to \( G(a_g) \) [Eq.(7.25)]. Since \( a_g \ll \Gamma \), the corresponding relaxation rate is zero. We conclude that spin wave excitations make no contribution to the low frequency dynamics probed by \( \mu \)SR in the presence of a small anisotropy gap \( \Gamma \).

The anisotropy gap in disordered ferromagnets has not been measured, and is possibly smaller than calculated above. We now show that even if \( \Gamma = 0 \), spin-lattice relaxation due to spin waves in \( \text{PdMn}(2 \text{ at.\%}) \) is an order of magnitude smaller than the data. Star et al (1975) determined \( \tilde{\Gamma} = 0.7 kT_c a^2 \) from the spontaneous magnetization in \( \text{PdMn}(2.45 \text{ at.\%}) \). At \( T = 0.5T_c \), the zero field linewidth is \( a_g = 19.\mu s^{-1} \) (Fig.6.6), so \( k_B T \gg \tilde{\Gamma} a_g \), and the power spectral density at \( a_g \) of the spin wave excitations is found from Eq.(9.5),

\[ G(a_g) = 5.4 \times 10^{-13} \text{ s}^{-1} \text{ k}^{-1} \ \Gamma. \]  
(9.6)
Using $a_0 = 27 \, \mu s^{-1}$ (from Chap. 6), we use Eqs. (7.25) and (7.6) to express $G(a_s)$ in terms of the spin-lattice relaxation parameter $K$,

$$K/a^6 = 5.9 \times 10^{-3} \, \mu s^{-1} \cdot K^{-1} \cdot (1 - r^2) \, T,$$  \hspace{1cm} (9.7)

which is a factor of ten smaller than the observed relaxation in PdMn (2 at.%) using values of $r$ from Fig. 6.6. The spin wave excitations satisfy the rapid fluctuation condition of Eq. (7.25), since $G(a_s)K/a^6 < 1$. However, Eq. (7.25) is approximate, since we have evaluated $G(\omega)$ at the single average frequency $a_s$ instead of averaging $G(\omega)$ over a distribution of width $a_s$. With this uncertainty, the small rates in Eq. (9.7) suggest that spin waves cannot account for the observed relaxation, even for $r = 0$.

2. Spin glass state extended modes

Spin wave excitations are expected to propagate in a spin glass only for wavelengths longer than the average separation of impurity spins. In this long wavelength limit, the random distribution of impurities can be replaced by a continuum approximation, and excitations of the continuum are described by 'hydrodynamic' equations. Halperin and Saslow (1977) used a hydrodynamic theory to predict that long wavelength spin waves could propagate in a spin glass with a linear dispersion relation $\omega = \nu k$. The first direct observation of spin waves with linear
dispersion was made by Alloul and Mendels (1985), who found a stiffness constant \( v = 1.9 \times 10^5 \text{cm/s} \) in CuMn(9.6 at.\%). Saslow (1980) showed that the dispersion relation in the presence of anisotropy is \( \omega^2 = (vk)^2 + \Gamma^2 \), where \( \Gamma \) can be found from the anisotropy energy measured by transverse susceptibility experiments. Saslow estimated \( \Gamma = 2.8 \times 10^{10} \text{s}^{-1} \) for CuMn(1 at.%), which is very much larger than the typical muon Larmor frequency \( a_B \). As in the case of ferromagnetic spin waves, the large anisotropy gap implies muon relaxation cannot be attributed to spin waves.

The anisotropy gap \( \Gamma \) in the spin wave dispersion relation has not been observed directly, and may be considerably smaller than estimated by Saslow. However, the spin wave stiffness observed by Alloul and Mendels is large enough to make muon relaxation negligible even if \( \Gamma = 0 \). To show this, we assume \( v \) in AgMn is the same as in CuMn, but scaled as \( c^4/3 \) for the difference in concentration (Alloul and Mendels, 1985), which gives \( v = 1.7 \times 10^4 \text{cm/sec} \) for AgMn(1.6 at.%). Equation (9.3) for the spin auto-correlation function can be converted from a sum over \( k \) to integral over \( \omega \) using the dispersion relation \( \omega = vk \), then Eq. (9.4) used to calculate \( G(\omega) \),

\[
G(\omega) = \frac{A}{\sqrt{3}} \frac{\omega^2}{\exp(\hbar\omega/kT) - 1} \tag{9.8}
\]

where \( A \) is the same as in Eq.(9.5). With \( a_B = 19 \mu \text{s}^{-1} \), the available
temperatures satisfy $kT >> \hbar a$, so Eqs.(7.25) and (7.6) give the
spin-lattice relaxation rate due to spin glass spin wave
excitations,

$$K/a^6 = 1.13 \text{s}^{-1} \text{K}^{-1} (1-\tau^2) \text{T}. \quad (9.9)$$

This relaxation rate is negligible compared to the observed
relaxation in AgMn(1.6 at.%).

3. Spin glass state localized modes

Walker and Walstedt (1980) calculated the frequency spectrum
of small amplitude collective modes in a system of spins coupled by
RKKY exchange energies. They found a range of excitations from
localized, high energy modes involving excitations of a few spins
(a cluster of strongly interacting spins) to lower energy modes
pointed out that the peak in mode density of these excitations
corresponds to a correlation time on the order of $1.4 \times 10^{-12}$ sec,
which is three orders of magnitude shorter than the observed
correlation times in AgMn(1.6 at.%), and cannot be responsible for
the observed muon relaxation. Walstedt (1983) extended the earlier
calculations of the mode density to include a dipolar anisotropy
energy to the spin interaction. This introduces a gap in the
excitation spectrum at a substantial fraction of the frequency
$kT_g/\hbar$. This gap is much larger than $a_\delta$, reinforcing the conclusion of Heffner et al that the observed muon relaxation cannot be attributed to localized modes.

B. Ferromagnetic domain wall excitations

A ferromagnet in zero applied field below $T_C$ will form a large number of randomly oriented domains, separated by domain walls. The position of these walls can fluctuate, producing large fluctuating fields in the neighborhood of the walls. In ordinary ferromagnets, nuclei in domain walls can make a large contribution to the NMR spin-lattice relaxation (Weger, 1962). Winter (1961) examined the fluctuation spectrum of Bloch domain walls and found a minimum wall excitation energy (an anisotropy gap)

$$\hbar \Gamma = (8\pi K S g u_B M_0)^{1/2},$$

(9.10)

where $M_0$ is the spontaneous magnetization and $K$ is a uniaxial anisotropy energy. Since there is no uniaxial anisotropy in PdMn(2 at.%), we assume that dipolar anisotropy will also set a minimum wall excitation energy, which we estimate from Eq. (9.10) by setting $K$ equal to the average dipolar interaction energy of two Mn spins. The average separation of two Mn for $c=0.02$ is 1.3 fcc lattice parameters (Chandrasekhar, 1943), and at $T=0.5T_C$, $a_z=2.5u_B$/Mn atom (Star et al, 1975). We then estimate
\[ K = \frac{a_z^2}{(5.1A)^3} = 4.2 \times 10^{-18} \text{ erg.} \]  

(9.11)

Using \( M_0 = a_z c^4 / a^3 \approx 39G \), we find \( \Gamma = 1.3 \times 10^{10} \text{ s}^{-1} \). This corresponds to frequencies much higher than the typical muon Larmor frequency \( a_s \).

We conclude that as with spin waves, the inevitable existence of dipolar anisotropy raises the frequency of the usual collective excitations in the ordered state to frequencies much higher than probed by \( \mu \)SR.

C. Barrier modes

Villain (1980) identified two classes of excitations in spin glasses based on the idea that a spin glass has a large number of nearly degenerate equilibrium spin configurations, or ESC. Spin wave modes (both extended and localized) correspond to small amplitude excitations about a single ESC. Barrier modes are defined as large amplitude transitions over an energy barrier from one ESC to another. Walstedt (1983) classified a large number of ESC for a realistic computer model of a spin glass. Many of these ESC were found to differ only in the reversal of spins within a localized region or domain consisting of some 20–30 spins, suggesting that barrier modes correspond to reversals of spins within small domains. Walstedt estimated the energy barrier \( \Delta \)
required for transitions between various pairs of ESC, and found a
broad distribution of $\Delta$ from zero to around $kT_g$. Walstedt
concluded that a wide variety of over barrier transitions are
energetically possible in the spin glass phase.

The barrier mode transitions observed in computer experiments
were first predicted to exist by Anderson et al, (1972). They
described a barrier mode as the excitation of a two level system
(TLS) having a generalized coordinate corresponding to the
simultaneous rotation of all spins in a small domain. The observed
linear term in the low temperature specific heat of spin glasses is
consistent with a collection of TLS having a broad distribution of
barrier heights. Prejean and Souletie (1980) successfully modeled
the low temperature remanence and hysteresis in spin glasses with a
collection of TLS, each with a thermally activated relaxation time
$\tau = \tau_0 \exp(\Delta/T)$. Using a uniform distribution of energy barriers $\Delta$,
they estimated $\tau_0$ in a small applied field to be very long, on the
order of $10^{-6}$ sec, and estimated that each TLS in CuMn(1 at.%) involves
roughly 50 spins, in fair agreement with Walstedt's
computer simulations. A more direct experimental observation of
TLS in spin glasses is the enhanced thermal conductivity in large
applied field observed by Wasserman and Herlach (1984).

In chap.8 we attributed spin-lattice depolarization below $T_g$
to limited amplitude fluctuations, but the correlation times for
these excitations should be much shorter than observed. We can
resolve this discrepancy by attributing the observed depolarization
to barrier modes. As a model of spin glass dynamics, we will assume the spin system has limited amplitude spin wave excitations about each ESC with a correlation time on the order of $10^{-12}$ sec, and large amplitude barrier mode excitations as the spin system moves to different ESC. The limited amplitude fluctuations are too rapid to cause spin-lattice relaxation, but their temperature dependent amplitude explains the temperature dependence of the static linewidth $a_{s}$ (Heffner et al., 1982). Since barrier modes are large amplitude, we assume the local static fields reorient fully with a correlation time $\tau = \tau_0 \exp(\Delta/T)$. Using $\tau_0 = 10^{-6}$ sec (Prejean and Souletie, 1980), the fluctuations satisfy the slow fluctuation condition since $a_{s} \tau_0 \gg 1$ unless $T$ is very close to $T_c$. The depolarization due to a single TLS is given by Eq. (7.13). The overall depolarization is then found by averaging over a uniform distribution of energy barriers from zero to $\Delta_{\text{max}}$, and by assuming the local field at every muon site is modulated by TLS excitations. The long time $1/3$ component of the depolarization function is then

$$P(t) = \frac{1}{3} \int_{0}^{\Delta_{\text{max}}} \frac{d\Delta}{\Delta_{\text{max}}} \exp\left(-\frac{2t}{3\tau_0} e^{-\Delta/T}\right), \quad (9.12)$$

or

$$P(t) = \frac{1}{3} \frac{T}{\Delta_{\text{max}}} \left[ Ei\left(-\frac{2t}{3\tau_0}\right) - Ei\left(-\frac{2t}{3\tau_0} \exp(-\Delta_{\text{max}}/T)\right) \right], \quad (9.13)$$

where $Ei(x)$ is the exponential integral function. Finally, we set
$A_{\text{max}} = T_g$, as suggested by the simulations of Walstedt (1983), and calculate $P(t)$ as a function of $T/T_g$.

For $T = T_g$, $P(t)$ is approximately exponential, but at lower temperatures a plot of $\log P(t)$ vs. $t$ shows pronounced curvature. These curves can be approximated with a root-exponential by fitting a straight line to a plot of $\log P(t)$ vs. $t^{1/2}$. The resulting root-exponential rates $\lambda$ are compared with data on AgMn(1.6 at.%) in Fig. 8.3, where the solid line shows $\lambda$ expressed in terms of $\Delta / a^6$ using Eq. (7.6). There is reasonable agreement (with no adjustable parameters) demonstrating that barrier mode excitations can account for the observed relaxation in spin glasses.

In a disordered ferromagnet, barrier modes are presumably the reversal of domains of spins loosely coupled to the infinite cluster of ferromagnetically aligned spins. This suggests that the density of TLS per spin will be much reduced in a ferromagnet relative to a spin glass. Furthermore, the existence of a spontaneous magnetization will raise the energy barrier for some domain reversals. Both of these effects will tend to reduce the barrier mode relaxation rate in ferromagnets from that calculated above for spin glasses, in agreement with experiment. This also explains why relaxation rates in the ferromagnetic phase of PdFeMn(5 at.%) are considerably higher than in PdMn(2 at.%). The additional spin disorder in PdFeMn(5 at.%), which is manifest in the low temperature reentrant transition, presumably allows for a
higher density of TLS, and lower energy barriers than in PdMn(2 at.%).

D. Mean field dynamics and algebraic decay of spin correlations

Spin waves and barrier modes provide a microscopic description of the observed broad distribution of relaxation times in the spin glass phase (Mezei and Murani, 1979). An alternative approach is to calculate the spin auto-correlation function in a dynamical extension of the SK mean field theory. Several theories calculate an algebraic decay to the auto-correlation function at long times, $t^{-\nu}$ (Chowdhury and Mookerjee, 1984), which represents a broad distribution of relaxation times. Sompolinsky and Zippelius (1982) predicted algebraic decay with an exponent varying from $\nu=1/2$ at $T_g$ to smaller values at lower temperatures. The temperature dependence of $\nu$ is not clear, since Ma and Rudnick (1978) predicted $\nu=1/2$ for all $T$ below $T_g$. The Monte Carlo calculations of Kirkpatrick and Sherrington (1978) showed algebraic decay with $\nu=1/2$ for temperatures from $0.5T_g$ to $T_g$. Experimentally, MacLaughlin et al (1983) used the longitudinal field dependence of the muon relaxation rate to infer algebraic decay with an exponent close to $1/2$, and Heffner and MacLaughlin (1984) showed that the neutron spin-echo correlation function below $T_g$ is consistent with values of $\nu$ determined from $\mu$SR data.
Muon spin-lattice relaxation rates in AgMn(1.6 at.%) can be compared with the predicted algebraic decay of the spin auto-correlation function using the limited amplitude fluctuation model. We assume the spin auto-correlation function below $T_g$ is

$$\langle S_i(0)S_i(t) \rangle = q + (1-q) \left(1 + \omega_e t\right)^{-1/2}, \quad (9.14)$$

where $\omega_e$ is an exchange frequency on the order of magnitude of $kT_g/\hbar$. The power spectral density for $\omega \ll \omega_e$ is given by Eqs. (7.22) and (7.23),

$$G(\omega) = (2\pi \omega_e \omega)^{-1/2}. \quad (9.15)$$

From Eqs. (7.25) and (7.6), the spin-lattice relaxation parameter for $Q = 0.016$ is

$$\frac{K}{a^6} = 9.43 \frac{a_o^{3/2}}{\omega_e^{1/2}} \frac{1-r^2}{r^{1/2}}. \quad (9.16)$$

Using the Heisenberg mean field values of $r$ (Fig. 6.2), and $a_o = 19.2 \mu s^{-1}$ for AgMn(1.6 at.%), this equation has been fit to the data of Fig. 8.3 with $\omega_e$ as an adjustable parameter. The dashed line in the figure shows the result for $\omega_e = 3.9 \times 10^{10} s^{-1}$, about 20 times smaller than $kT_g/\hbar$. The rapid fluctuation condition of Eq. (7.25) is well satisfied, since $a_o \ll \omega_e$ implies $G(a_o)K/a^6 \ll 1$. 
The good agreement with data shows that an algebraic correlation decay provides an adequate description of the temperature dependence of the relaxation rate. The comparison cannot be taken as evidence for $v=1/2$, since the same conclusion holds for a range of $v$. 
X. Conclusions

A. Paramagnetism

The transverse applied field linewidth in the paramagnetic state of all the dilute alloys studied is proportional to the total magnetization, or in terms of the order parameters defined in Chap. 4, \(|\phi| - |\phi|_0\). This implies that there is no microscopic spin disorder or frustration except very close to the transition temperature, in qualitative agreement with the SK mean field theory. The zero field transition from the paramagnetic state to an ordered state with static spin polarization is seen in the rapid onset of a static linewidth. The temperature range of the transition is very narrow, in agreement with Mössbauer effect studies of spin glasses (Violet and Borg, 1966) and disordered ferromagnets (Takeda et al, 1985) which show a sharp onset of a static hyperfine field at the transition temperature.

Since the paramagnetic state linewidth does not reflect the complicated distribution of exchange energies, the magnitude of the paramagnetic linewidth provides easily interpreted information on the muon-local moment interaction. Novel effects have been found in both AgMn and PdMn. In AgMn, the muon-Mn coupling strength is approximately independent of concentration, and is larger than calculated theoretically, including an estimate of the RKKY coupling strength. This extra interaction is attributed to a
larger than expected RKKY contribution (Heffner et al., 1985), and is the first direct observation of RKKY muon-local moment coupling. In PdMn, the apparent muon-Mn coupling strength is concentration dependent, and is smaller than calculated theoretically. Calculations show that this decrease cannot be attributed to the effect of a finite concentration, atomic short-range order, or short-range spin correlations from near-neighbor exchange couplings. We instead attribute the decreased coupling strength to an exclusion of the muon from octahedral sites adjacent to Mn impurities. Measurements of PdMn lattice parameters rule out lattice distortion as a mechanism for this exclusion. We speculate that the effect is electronic in origin, and is present in the Pd host and not the Ag host due to the exchange enhanced d-band susceptibility in Pd.

The form of the zero applied field depolarization function \( P(t) \) above \( T_0 \) in \( \text{PdFeMn}(5 \text{ at.\%}) \) provides more direct evidence that the muon is excluded from octahedral sites adjacent to Mn. This form of \( P(t) \) resolves earlier ambiguities on the proper fitting function for PdMn data.

The high temperature single ion correlation time due to the Korringa process can be measured by \( \mu\text{SR} \). We find \( T_\tau = 0.065(3) \text{ ns-K} \) in PdMn. This is considerably shorter than measured by ESR (Alquie et al., 1978), suggesting that the ESR bottleneck has not been completely broken in PdMn. The Korringa product \( T_\tau \) is smaller in AgMn than measured by neutron scattering (Murani, 1981), but the
\mu SR data show considerable scatter, presumably due to systematic errors in measuring low relaxation rates far above \( T_g \). The temperature dependence of \( \tau_c \) above \( T_g \) is not well defined by the data, so comparisons with theoretical predictions for the slowing down of spin correlations are preliminary. The discrepancy with the neutron scattering Korringa rate and the temperature dependence \( \tau_c(T) \) may be resolved in the future with higher statistics data on more concentrated AgMn alloys.

Muon spin-lattice relaxation in zero applied field shows a rapid slowing down of impurity spin fluctuations above the transition temperature for all types of order. The temperature range of the slowing down increases with increasing disorder in the spin system.

B. Disordered ferromagnetism

Magnetization measurements on the disordered ferromagnet \( \text{PdMn}(2.5 \text{ at.\%}) \) (Star et al, 1975), show the spontaneous magnetization is considerably smaller than the high field saturation magnetization. Star et al attributed this to a short-ranged anti-ferromagnetic coupling between Mn which produces 'frustration' in the spin system. The 'short-fall' in the spontaneous magnetization can be modeled with the Ising SK mean field theory, except that a predicted low temperature reentrant transition is not observed. This failure of the SK mean field
theory can be attributed to a non-Gaussian distribution of exchange energies. The reduced spontaneous magnetization is reflected in the zero applied field static linewidth, which is in good agreement with the same mean field theory. An alternative to mean field theory for the development of static order is a percolation model. The data are consistent with a percolation model only if a fraction \( f = 0.35 \) of the frozen spins in the infinite cluster are randomly oriented (frustrated). Both the mean field and percolation descriptions imply considerable frustration and inhomogeneity in the spin system at low temperatures.

Spin-lattice relaxation in the ferromagnetic state is expected to be insensitive to spin waves and domain wall excitations, the usual mechanisms for nuclear spin-lattice relaxation in ferromagnets. A qualitative argument based on results in the spin glass phase suggests that the observed low-frequency excitations are localized barrier modes. The presence of barrier modes can be understood qualitatively in the percolation picture, where finite spin clusters exist below \( T_c \), loosely coupled to the infinite cluster. This qualitative picture was first introduced by Verbeek et al (1980) to describe small angle neutron scattering results in PdMn(2 at.%). They found the static spin polarization below \( T_c \) to be highly inhomogeneous, which they modeled by a collection of magnetic clusters having a distribution of sizes, with average radius 20A. We speculate that slow reorientations of these
magnetic clusters are responsible for the observed spin-lattice relaxation in PdMn(2 at.%).

C. Reentrant ferromagnetism

The large static linewidth in the ferromagnetic phase of PdFeMn(5 at.%) confirms that Mn spins are static below $T_c$. This is contrary to a recent suggestion (Senoussi, 1980; Kettschau et al., 1983) of separate transitions in the Fe and Mn spin systems. Dynamic spin-lattice relaxation below $T_c$ is more effective than in PdMn(2 at.%), and is approximately independent of temperature. This is consistent with a higher density of barrier mode excitations than in the disordered ferromagnet, due to additional disorder in the reentrant phase. Like PdMn(2 at.%), small angle neutron scattering shows a highly inhomogeneous magnetic state below $T_c$ in PdFeMn(5 at.%) (Shapiro, et al, 1980).

There is no change in the low-frequency dynamics at the ferromagnet-spin glass transition. This result may be of importance in evaluating theories of the ferromagnetic-spin glass transition.
D. Spin glass order

The temperature dependence of the static linewidth in zero applied field is consistent with the Heisenberg SK mean field theory for several spin glasses, and the linewidth approaches the calculated zero temperature limit. Two percolation models of the spin glass phase have been tested by comparing the linewidth with the Edwards-Anderson order parameter $q$ determined from ac susceptibility data. The data are inconsistent with a percolation model where all spins in the infinite cluster below $T_g$ have the full spin polarization $|\langle S \rangle| = S$. However, we cannot rule out a percolation model where the infinite cluster spin polarization follows a mean field-like temperature dependence. The difference between homogeneous (mean field) and inhomogeneous (percolation) development of static spin polarization probably cannot be resolved with linewidth measurements, as this involves the spatial distribution of spin polarization, which is unavailable with $\mu$SR.

Spin-lattice relaxation in spin glasses is expected to be insensitive to both long wavelength hydrodynamic spin wave modes and to localized spin wave-like excitations. We argue that the low frequencies probed by zero applied field $\mu$SR observe large amplitude barrier mode excitations. A phenomenological theory of barrier modes is consistent with observed relaxation rates with no adjustable parameters. Future data with smaller statistical errors may allow comparisons of data with detailed computer simulations of
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barrier mode excitations. This zero field dynamical information is unavailable from other experimental techniques, since it is at much lower frequencies than can be measured even with the neutron spin-echo technique (Mezei and Murani, 1979). An alternative to the barrier mode description of spin glass dynamics is dynamical mean field theory, which predicts slow (algebraic) decay of spin correlations. This description is also consistent with data.
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