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Spin-polarized metastable de-excitation spectroscopy: A new probe of alkali chemisorption on surfaces

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SPIN-POLARIZED METASTABLE DE-EXCITATION SPECTROSCOPY:
A NEW PROBE OF ALKALI CHEMISORPTION ON SURFACES

by

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Abstract

Spin-Polarized Metastable De-excitation Spectroscopy: A New Probe of Alkali Chemisorption on Surfaces

by

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Metastable (Atom) De-excitation Spectroscopy (MDS) provides a powerful technique with which to investigate surface electronic structure with extreme surface specificity. In this technique a thermal energy beam of noble-gas metastable atoms is directed at the surface under study and the kinetic energy distribution of ejected electrons that result from metastable atom de-excitation is measured. Although the measured distribution contains information about the electronic structure of the outermost surface layer, its detailed analysis requires knowledge of the dynamics of the metastable atom-surface interaction. In the present work, these dynamics have been investigated directly by use of spin-labeling techniques. The electron spins of the incident metastable atoms are polarized and the spin-polarization of the ejected electrons is measured with a Mott polarimeter. Energy resolved electron spin-polarization measurements are reported for a variety of sub-monolayer coverages of cesium on a Cu(100) surface, and for oxygen and cesium co-adsorption on a Cu(100) surface. The Cs/Cu(100) system exhibits a large (~2.8 eV) change in the surface work function. The results of the current work suggest that MDS interactions in both high and low work function regimes are more complex than has previously been supposed. Several additional interactions are suggested to explain the data acquired. The question of the occupancy of the adsorbed cesium valence level at various coverages is also addressed.
Acknowledgments

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In a more general way, I would like to thank the Jr. High kids at First Presbyterian Church for their friendship, and for providing an often humorous diversion from what could have been grueling years. I would also like to thank God for making the universe as marvelously complex as it is, and my family for encouraging me to understand it more completely throughout my education.
A wise man has great power, and a man of knowledge increases strength.

It is the glory of God to conceal a matter; to search out a matter is the glory of kings.

King Solomon, in Proverbs 24:5, 25:2

For since the creation of the world God's invisible qualities--his eternal power and divine nature--have been clearly seen from what has been made, so that men are without excuse.

Paul the Apostle, in Romans 1:20
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Chapter I
Introduction

A. Metastable De-excitation Spectroscopy

Metastable De-excitation Spectroscopy (MDS) provides a powerful probe of the electronic structure of a surface. The probe uses a thermal energy beam of long-lived excited metastable atoms (usually rare gas atoms) which is directed at the surface under study. The stored internal electronic energy in the metastable atoms is deposited in the surface, and may cause electron ejection from the atom-surface system. Analysis of the ejected electron energy distribution yields information about the electronic structure of the surface and the nature of the metastable-surface interaction. Because the metastable atoms have only thermal energies, de-excitation occurs before the atoms are able to penetrate the surface, giving the probe extreme surface specificity. MDS was first investigated in the 1920's, and early work with thermal rare gas atoms was done in the 60's and early 70's. More recently, MDS has been used to study adsorbates on surfaces, free atoms and molecules, and organic substances.

In order to understand the information which can be gained from MDS, one must first understand the dynamics involved in metastable atom de-excitation. Consider a metastable helium atom (He*) approaching a clean metal surface (Fig. 1). Two de-excitation channels are possible. In the first, Auger De-excitation (AD), an electron from the metal fills the 1s vacancy in the He*. The energy released is transferred to the 2s electron, which is ejected. In the second channel, Resonance Ionization followed by Auger Neutralization (RI+AN), the He* 2s electron first tunnels into a vacant surface state above the metal's fermi energy,
from the surface which tunnels into the 1s hole. The energy liberated is transferred to another electron in the surface, which may be ejected. These processes are commonly

called the one-step and the two-step processes, respectively. Obviously, the energetics of the two mechanisms are quite different. With the one-step process, the energy of the ejected electron directly reflects the energy of the electron which fills the 1s hole. The ejected electron energy distribution therefore corresponds to the surface electron density of

Figure 1 - Two de-excitation mechanisms for He*(2^3S) at a surface: a) Auger De-excitation, b) Resonance Ionization followed by Auger Neutralization.
states, to a first approximation. The two-step process, however, involves two surface electrons, and in fact one could imagine many possible surface electron pairs which would yield the same ejected electron energy (Fig. 2). Therefore, the ejected electron energy distribution, to a first approximation, is a self-convolution of the surface density of states. It is critical, then to know which de-excitation mechanism is operative before one can begin to analyze the data.

Figure 2 - Representative kinetic energy distributions of electrons ejected by the one- or two-step processes. In a) the distribution closely mirrors the surface density of states; while in b) the distribution is a self-convolution of the surface density of states.

Generally, the two-step process is believed to dominate in any situation where it is allowed to take place at all. The idea is that RI should occur rapidly when the 2s excited
electron cloud overlaps significantly with vacant states above the fermi level in the surface. The one-step process, on the other hand, requires overlap between the inner 1s vacancy and the tails of the surface electron wavefunctions extending into the vacuum, and is expected to become important only at smaller atom-surface separations than those characteristic of RI. The two cases where RI is inhibited, allowing the one-step process to occur, are when the surface is covered with a molecular adsorbate, or when the surface has an extremely low work function. In the first case, the adsorbed molecules physically prevent the He* atom from approaching close enough to the surface for RI to take place (Fig. 3a). In the second case, the surface states in resonance with the He* 2s electron are all filled (Fig. 3b).

Figure 3 - Two cases where RI is inhibited. a) Adsorbed molecules prevent the 2s electron from overlapping significantly with empty surface states. b) All surface states in resonance with the 2s electron are filled.
In conventional MDS, the operative mechanism is inferred from knowledge of the state of the surface, and/or from observations of the ejected electron energy distribution. The two-step process, which involves a self-convolution of surface state densities, generally yields broad, featureless, distributions, even if the surface density of states contains sharp features. The one-step process, on the other hand, produces to first order an image of the surface density of states, and so may produce sharp features in the ejected electron energy distribution. Additional information regarding the operative process may be gained by spin-labeling the electrons in the He*, and measuring the polarization of the ejected electrons. As shown in Fig. 4, if both He* electrons are polarized spin up, then the ejected electron due to the one-step process must be spin up. If the two-step process is operative, the neutralizing electron must be spin down, but the ejected electron could be either spin up or down. There may be some correlation between the electron spins, so that one spin or the other may be preferentially ejected, but the effect should not be 100%, as in the one-step case. Some experimental evidence from spin polarized auger electron spectroscopy of a ferromagnetic surface suggests that such correlation might occur at the tens of percent level. Measurement of the ejected electron spin can therefore reveal which ejection mechanism occurs under given conditions. When spin information is utilized, the probe is called Spin-Polarized MDS or SPMDS.

Since RI is thought to occur at clean metal surfaces with high work function, and to be inhibited at lower work function surfaces, it is interesting to study a surface system where the work function is a controllable parameter. Such systems can be obtained by deposition of adsorbates on a surface. In particular, deposition of elements of the alkali and alkaline earth families cause the typically high work function of transition metal surfaces to decrease. In the current work we study a Cu(100) surface with varying sub-
monolayer amounts of adsorbed cesium. These surfaces vary in work function from 4.6 eV (a clean Cu(100) surface where RI should occur) to less than 2.0 eV (a \sim 50\% cesium covered surface where RI should be inhibited).

\begin{figure}[h]
\centering
\includegraphics[width=\textwidth]{figure4.png}
\caption{Two de-excitation mechanisms with spin labels}
\end{figure}

B. Alkali deposition on surfaces

Deposition of even a small amount of an alkali adsorbate onto a metal substrate causes a dramatic lowering of its work function. This effect was first discovered by Kingdon and Langmuir, who found that thermionic emission from a tungsten filament increased dramatically with the deposition of cesium.\textsuperscript{10} The theoretical foundation describing this phenomena was laid by Gurney in 1934,\textsuperscript{11} and extended quantitatively by Muscat and
Newns in 1978, Studies of alkali adsorption are currently being actively pursued, both experimentally and theoretically, and the Gurney/Muscat-Newns model is by no means universally accepted. However, it is at least the model to which most work is compared, whether in support or disagreement. Therefore a brief explanation of this picture follows.

![Diagram](image)

**Figure 5 - Shifts and broadening of atomic energy levels as an atom approaching a surface**

As a single alkali atom approaches a clean metal surface, its single occupied valence state rises and broadens (Fig. 5). The rise occurs due to effects of the image charges induced in the surface by the core-electron pair of the alkali atom. The additional electronic interactions between the electron and its image and the electron and the core's image cause a net increase in the energy of the alkali valence state. The lifetime of the state decreases since the otherwise stable alkali valence electron can tunnel into the surface as the atom
approaches the surface. Since this lifetime, $\Delta t$, decreases, the Heisenberg uncertainty relation $\Delta E \Delta t \geq \hbar / 2$ requires that the width of the state, $\Delta E$, increase.

Since the ionization energy of a typical alkali atom ($\sim 4$ eV) is less than the work function of a typical metal surface ($\sim 5$ eV), the electron is donated at least partially into the surface, filling a vacant state just above the Fermi energy. The lower edge of the energy-broadened alkali valence state may extend below the surface Fermi level, causing the state to be partially filled, but the alkali-surface bond is ionic, forming a dipole at the surface. As more alkali atoms adsorb on the surface, these dipoles repel each other and distribute themselves evenly over the surface. This creates a dipole layer whose moment per unit area increases linearly with alkali coverage (at low coverages), and lowers the work function of the surface. As the alkali coverage increases, the dipole density increases, and individual dipoles are forced closer together. It therefore becomes energetically favorable for the individual dipoles to decrease their charge separation. The number of dipoles continues to increase, but each of their moments diminishes. Thus, at intermediate coverages, electrons begin to return to the alkali atoms, filling more of the broadened valence orbitals. In this regime the work function continues to decrease, but less quickly than before. Finally, at very high coverages (approaching one close-packed layer), the alkali valence orbitals become metallized (not localized to a single adatom), and the adlayer forms a true metallic overlayer. Further alkali deposition from this point causes no further change in the work function, which is now that of the alkali metal. Quantitative calculations using jellium for both the substrate and the adlayer also predict a minimum in the work function at some coverage short of one close packed layer, with a subsequent increase in work function up to the alkali metal value.\(^{14}\) This characteristic variation in work function is seen for virtually every combination of alkali and metal substrate,\(^{15}\) and in fact is seen for semi-
conducting substrates as well.\textsuperscript{16} A typical work-function versus coverage curve is shown in Fig. 6.

Figure 6 - Work function vs. cesium deposition time on Cu(100) surface. LEED pattern calibrate close-packed and half close-packed coverages.
C. Measurement of electron energies

Generally in an MDS experiment, electrons are ejected into the vacuum with a continuous spectrum of energies and directions. Some electrons are also directed into the sample, where they may be lost, may backscatter elastically or inelastically, or may scatter one or more "secondary" electrons of lower energy into the vacuum. In the current work, electron energy distributions are measured by the "retarding grid" method. An electron detector is situated behind a double grid which may be biased at some arbitrary potential. These grids create between them a field free region at this potential. Electrons with insufficient energy to overcome this barrier will be rejected, so the detector will count all electrons with energies greater than some threshold. By ramping the voltage applied to the grids through the range of interest, and taking the first derivative of the resulting curve, the energy distribution of the electrons is obtained. This method actually measures the electron energy perpendicular to the grids, since a very energetic electron travelling nearly parallel to the retarding grids could still be rejected. However, the total acceptance angle of the energy analyzer is $\sim 20^\circ$, which means that the perpendicular energy will differ from the total energy by no more than 4%. The Mott electron spin polarimeter, is also equipped with retarding grids. The performance of these grids is the subject of a more extensive study described in Appendix A. The upshot of the study is that the Mott analyzer has an energy resolution of $\sim 1$ eV.

Next we must consider how the work function of the sample and of the grids affect the data which is collected, especially in light of the fact that the sample work function will change. If the grids are biased at some voltage $V_g$ with respect to the sample, it is the Fermi levels of the grids and the sample, not necessarily the vacuum levels, which have the
Auger De-excitation

Figure 7 - Energy diagram showing the effects of a changing work function on the ejected electron energy (as measured with a retarding grid analyzer) for the two de-excitation mechanisms.
potential difference $V_f$ between them. As a He* atom approaches the sample, however, its vacuum level references itself to the local vacuum level, which is that of the sample. Fig. 7 shows how the one- and two-step processes are affected by a changing sample work function. If the one-step process is operative, the surface electron falls further to fill the 1s He vacancy as the work function of the sample decreases, but the 2s electron also begins with less energy (relative to the sample Fermi level). These two effects exactly cancel, so that the energy of an electron ejected via the one-step process and measured by a retarding grid method is unaffected by changes in the sample work function. Note that if the electron energy were measured by a time-of-flight method, where the electron's average speed were measured, an effect would be seen. The kinetic energy (and hence the velocity) of an ejected electron would be greater just outside the surface of a low work function surface than outside a high work function surface. However, relative to the sample Fermi level (and therefore to the retarding grids), the electron energy does not depend on the sample work function. Note that a change in work function may change other parameters of the interaction, such as the atom-surface distance at which de-excitation takes place, which could in turn affect the kinetic energy of electrons ejected into the vacuum. Therefore, the ejected electron energy may depend slightly on changes in the sample work function.

Similarly, it can be seen that if the two-step process is operative, a lower work function surface allows the neutralizing electron to release more energy. This energy is transferred to another surface electron, whose energy (relative to the Fermi level) is unaffected by any work function change. Thus, there is no cancellation, and low work function surfaces will eject higher energy electrons than high work function surfaces. Therefore, the high energy cutoff due to the one step process should not change as the work function changes, but the cutoff due to the two-step process should. The low energy
cutoff due to either process results from those electrons with just enough energy to get into the vacuum, and will decrease as the work function decreases. This cutoff can even be observed at a reverse grid bias, if the grid work function is greater than the sample work function.
Chapter II
Apparatus

The apparatus used in the present work is the result of several years of development. Many of the construction and design considerations, and detailed performance parameters are contained in the theses of previous students.\textsuperscript{17-21} The following description of the apparatus will cover only those aspects which pertain to the current experiment. Basically, the apparatus is designed to produce a thermal beam of pure $^{23}$S\textsubscript{1} metastable helium atoms, and to allow these atoms to impact the surface under study. Fig. 8, a schematic of the apparatus, shows the helium gas source, the metastable excitation and polarization regions, and the sample chamber with attendant measurement devices. Each of these parts will be described in some detail.

Ground state helium gas flows from a compressed gas tank through a liquid nitrogen cold trap and a multi-channel array, into the source region. The cold trap removes many potential gas impurities, such as H\textsubscript{2}O and O\textsubscript{2}. These impurities could inhibit the creation of the helium metastable atoms, and could contaminate the surface under study. The multi-channel array consists of a large number of long, narrow tubes which give the atomic beam some initial collimation. Once in the source region, the helium atoms are bombarded by a co-axial electron beam, causing excitation or ionization of the atoms while not imparting significant transverse momentum to the atoms. The electrons are produced by a hot tungsten filament (7 mil diameter) wound twice helically around the beam line. The electrons are extracted by a Pierce geometry electrode,\textsuperscript{22} and injected into a field-free region. The electrons are collimated by an axial magnetic field of about 400 gauss imposed
Figure 8 - Schematic of the apparatus.
on the entire source region. Fig. 9 shows clearly each of these elements of the metastable atom source.

![Diagram of the He* (2^3S) source](image)

**Figure 9 - Schematic of the He* (2^3S) source**

In addition to He*(2^3S) metastable atoms, several other kinds of particles are created within the source region which may be capable of ejecting electrons from a surface (e.g. the sample under study or a channeltron electron detector surface), and producing spurious signals. These signals must be identified, and if possible, their attendant parent particles eliminated from the beam line while data taking occurs.
Particles which are created in the source region and which are capable of producing spurious signals are:

i) He$^+$ ions

ii) Other excited He states - the only ones to survive the $\sim 400\mu s$ travel time to the target are the metastable $2^1S_0$ state and Rydberg states above $n=20$

iii) Fast neutral helium atoms arising from the reaction

$$\text{He}^+ \text{ (fast)} + \text{He (thermal)} \rightarrow \text{He}^+ \text{ (thermal)} + \text{He (fast)}$$

iv) Photons - these arise both from the glowing source filament and from cascades from high lying excited helium states created by electron impact

The He$^+$ ions are readily removed by an electrostatic deflector plate in the middle chamber. The $2^1S$ atoms are removed by exciting $2^1S \rightarrow n^1P \rightarrow 1^1S$ transitions with a helium discharge "quench" lamp. The lamp also causes transitions $2^3S \rightarrow n^3P$, but the $n^3P$ states can only decay to the lowest state of the triplet spin system, which is the $2^3S$ state. The cross section for producing $n>20$ Rydberg atoms is much smaller than the cross-section for producing $2^3S_1$ atoms,$^{23}$ so there is no significant contamination with Rydberg atoms.

The electric fields present in the source region are designed to minimize the generation of fast neutral particles that move toward the sample chamber. Any helium ions which are created in the first half of the (semi-) field-free region will be accelerated back toward the Pierce electrode, since it is biased less positively than the field-free region. Ions created in the latter half of the region, however, will be accelerated forward by the grounded elements further down the beam line, and could charge-exchange to produce forward moving fast neutral atoms.

Previous studies with neon in the present apparatus show that even the slowest of the fast neutrals have approximately one hundred times more kinetic energy than the metastable
atoms.24 One would expect that this energy ratio would remain about the same for helium, meaning that the fast neutrals move at least ten times faster than the metastables. Thus, the fast neutrals (and the photons) are easily discriminated against by a time-of-flight chopping/gating technique. The atomic beam is physically chopped by an in-vacuum chopper wheel, and the data collection cycle begins only after a brief delay after the atomic beam is blocked, so that the fast particles have already traversed the beam line, but the slower metastable atoms are still in transit. Fig. 10 shows a schematic of this data-taking method. The chopper wheel used in the present work has eight teeth, such that the teeth and the space between the teeth is the same size, creating a 50% duty cycle. The wheel is spun by a vacuum-compatible synchronous motor whose speed is controlled by its driving frequency. It is spun at 9400 rpm, so that each tooth/hole cycle is 800μs long. Thus, a 400μs burst of particles is followed by 400μs during which all particles are blocked. The data cycle is triggered by a light-photodiode detector which signals the blocking of the beam. Data collection begins 20μs after the photodiode signal and ends 360μs later, for a usable duty cycle of 42.5%. Since the beam line is about a meter long, particles with velocities $1.3 \cdot 10^5 < v < 5 \cdot 10^6$ cm/sec will contribute to the collected signal. By comparison, thermal energy helium has a velocity of $\sim 2.5 \cdot 10^5$ cm/sec. If the very slowest fast neutrals traverse the beam line in 40μs (ten times faster than the metastables), only those slowest of the fast neutrals which pass the chopper in the last 20μs (or 5%) of the cycle will arrive during the counting time. Since the source is operated under conditions such that even without chopping the fast neutral flux is <5% of the metastable atom flux, and since the vast majority of fast neutrals travel more than 20 times faster than the metastable atoms (and so will not be counted in the chopped data at all), the fast neutral contribution to the collected signal will be negligible (<0.1%).
Figure 10 - Schematic of chop/gate data taking method
Thus the apparatus is capable of measuring effects due virtually exclusively to interactions of $2^3S_1$ metastable helium atoms. The background thermal energy ground-state helium atoms (which make up the vast majority of the beam) have too little energy (<0.1 eV) to eject electrons from a surface, and so play no role in the experiment.

The metastable atoms are polarized by optical pumping to preferentially populate the $m_J = +1$ or $-1$ sublevel. This is accomplished by illuminating the beam with 1.08 $\mu$m circularly polarized light from an RF discharge helium lamp. This process induces transitions $2^3S \rightarrow 2^3P$ with $\Delta m_J = +1$ for right-hand circularly polarized (RHCP) light, and $\Delta m_J = -1$ for LHCP light. The subsequent decay $2^3P \rightarrow 2^3S$ has no preferred $\Delta m_J$, so after a number of cycles the atoms accumulate in either the $m_J = +1$ or $-1$ sublevel of the $3^1S_1$ state, depending on whether RHCP or LHCP light is used. The RF lamp is capable of producing ~40% polarization of the $2^3S$ metastables, where the polarization is defined as

$$P = \frac{N_{+1} - N_{-1}}{N_{+1} + N_0 + N_{-1}}$$

and $N_i$ is the number of atoms in an $m_J = i$ sublevel.

The polarization of the He* beam may be measured by allowing the beam to enter a Stern-Gerlach apparatus on the opposite side of the sample chamber. This device is shown in Fig. 11. It produces a large inhomogeneous "two wire" magnetic field with a constant vertical gradient of ~4500 gauss/cm over a horizontal length of 8 cm, which serves to split up the He* atoms according to their $m_J$ value. The atoms are detected by a movable channeltron, and the various $m_J$ populations can be calculated by suitable integrations. Sample Stern-Gerlach profiles, showing integrated populations, are shown in Fig. 12.
Figure 11 - Stern-Gerlach apparatus

Most of the measurements in the current work take place in the sample chamber. This chamber is designed for UHV conditions, and is separated from the source chamber by a differentially pumped middle chamber (where the optical pumping takes place) which has small entrance and exit apertures (these apertures also define the He* beam collimation). The sample chamber is composed of two levels, one containing the He* beam line and the SPMDS probes, and the other containing sample preparation and analysis equipment. It is pumped by a 450 l/s turbo pump, a 400 l/s ion pump, and a titanium sublimation pump surrounded by a cryoshroud which may be cooled to liquid nitrogen temperatures (77K). During an experimental run, the ion pump is turned off, since pumping helium will sputter the ion pump surfaces and cause the pump to regurgitate previously pumped gases. The sublimation pump was found not to have a large effect, and so was rarely used, although the cryoshroud was usually maintained at 77K. After the chamber is baked at 180°C for several days, a base pressure < 2⋅10^{-10} torr is achievable, pumping with either the turbo or ion pumps alone. Even with the He* source running, and the turbo pump on, the pressure only increases to 2⋅10^{-9} torr. The sample under study (in this case a Cu(100)
Figure 12 - Stern-Gerlach profiles: a) no optical pumping, b) RHCP optical pumping, c) LHCP optical pumping. Numbers indicate relative peak areas.
single-crystal) is mounted on an XYZΩ manipulator which in turn is mounted on a long Z drive which can move the sample between the upper and lower levels of the sample chamber. The main chamber also contains a Bayard-Alpert ionization pressure gauge,27 an RF quadrupole residual gas analyzer,28 and a gas handling system which allows the introduction of small, controlled quantities of known gases into the chamber.

The top level of the chamber contains the He* beam line, a retarding grid electron energy analyzer and a Mott polarimeter29 (also equipped with retarding grids) to measure electron polarizations. An excellent discussion of the principles of Mott polarimetry can be found in a previous student's thesis,30 and a brief description follows.

In the late 1920's, Mott first predicted that the trajectory of electrons scattered elastically from atomic nuclei would be spin-dependent.31,32 From the electron's point of view, the positive nucleus moving toward it creates a current which produces a magnetic field. This field will interact with the electron's spin and cause the electron to be more or less deflected than it would have been in the absence of this spin-orbit interaction. This scattering asymmetry is particularly large for scattering angles near 120°, and increases rapidly with Z, the charge of the nucleus. It also depends on the velocity of the electron, higher velocities producing greater asymmetries.

In the current detector design, which is illustrated in Fig. 13, electrons are accelerated to high velocities (~20kV) within the detector body and are scattered from a high-Z material (in our case, gold). Electrons which scatter through 120° (either to the left or to the right) with <900eV of energy loss are collected. The polarization of the incident electrons may be determined from the relation
Figure 13 - Micro-mott polarimeter

\[ P = \frac{1}{S_{\text{eff}}} \left[ \frac{X-1}{X+1} \right] \quad (\text{II.2}) \]

where \( X = \frac{R_l}{R_r} \), and \( R_{l,r} \) are the left and right scattering rates, respectively.

There generally exists some instrumental asymmetry, so that \( \frac{R_l}{R_r} \neq 1 \) even for an unpolarized beam of electrons. In the current apparatus, this asymmetry ratio, \( \delta \), is \( \approx 1.05 \). To eliminate this error, polarization data is taken with the incident He* beam polarized first in one direction, then in the opposite direction. If one then uses \( X = \left[ \frac{R_l R_r}{R_r R_l} \right]^{1/2} \), where the primed rates are measured with opposite incident beam polarization, relation II.2 gives the
electron polarization free of any instrumental asymmetry. Note that this method assumes that reversal of the polarization of the incident He* beam exactly reverses the polarization of the ejected electrons. This will not be the case for a magnetic surface, but does hold for the surfaces studied in this work.

The bottom level of the chamber contains an argon-ion sputter gun, a cesium filled oven which emits Cs when heated (~0.1 monolayer/sec at 65°C), and a LEED/Auger analysis system. These devices were used to prepare the surface and check its order, cleanliness, and alkali overlayer coverage.

The sample surface was prepared initially by chemical washes with 1,1,1 trichloroethane, ethanol, and acetone. Once in the evacuated chamber, the surface was further cleaned by successive cycles of bombardment with 400eV normal incidence argon ions (5μA/cm² for 30 min) followed by annealing to 500°C for 5 minutes. The sample would then radiatively cool to ~100°C over the next hour. These cycles were repeated until Auger analysis showed that the surface contamination was no longer decreasing, which required ~10 cycles, and resulted in a contamination level at the surface of 8% (1% oxygen, 7% carbon). An Auger scan of the cleaned surface is shown in Fig. 14. After this initial cleaning, a single sputter/anneal cycle was performed before each alkali deposition. A LEED pattern was occasionally examined for the surface, which confirmed the presence of a well-ordered (100) FCC crystal structure surface.

The LEED gun was also used to measure the sample work function by setting the gun to emit 100eV electrons, then ramping the sample bias through −100V, and observing where the electron current to the sample cut off. A block diagram showing the data taking method is shown in Fig. 15. The importance of the voltage on the hemispherical grids, which produce a radial electric field centered near the sample surface focusing the
Figure 14 - Sample Auger scan of cleaned Cu(100) surface. Impurity signatures of oxygen and carbon are indicated.
Work function measurement schematic

Figure 15 - Schematic of the work function measurement system
electrons onto the sample, cannot be overemphasized. Without this voltage, as the sample voltage approaches the gun voltage, electrons which are not directed very nearly normally toward the surface are deflected away from the surface. The result (Fig. 16a) is an ill-defined current cut-off which appears to occur at a much lower voltage than expected. The grid voltage was set empirically, by optimizing the cut-off clarity. This optimization produced cut-offs such as is illustrated in Fig. 16b. It was found that changes of more than 10 volts from the optimized value significantly affected the cut-off quality (but not position), but that cut-off position changes due to sample work function changes (~3 volts) were not large enough to require changing the grid voltage for different sample work functions.

The cesium oven used in the present work was designed to overcome difficulties experienced in the past with the more common SAES alkali dispensers. These difficulties included contamination with O$_2$ and CO, poor reproducibility, and limited lifetime. The current design is illustrated in Fig. 17. It consists of cesium metal inside several inches of 3/8" OFE ("oxygen-free electronic" grade) copper tubing. The tubing is sealed on one end by a cold weld pinch-off, and the other end is connected via a copper gasket/knife edge seal to an all-metal bakable valve. This entire assembly is suspended in vacuum by two support brackets which are welded to the valve and to a 6" conflat flange. A stainless steel U shaped tube whose ends extend through the flange into the air outside is welded onto the valve body. Hot gas may be flowed through the inside of the tube, heating the valve, and, by conduction, the alkali reservoir. A hot wire detector and physical beam block are mounted on a rotatable support which may be moved in front of the valve to measure or block the alkali beam. The principle and operation of the hot wire detector is described in Appendix B.
Figure 16 - Sample current cut-off curves under a) poor focussing conditions, b) optimized focussing conditions
Figure 17 - Views of the alkali oven from the a) side, b) front, c) bottom
The intended operation procedure of the oven is simple. Dry nitrogen (from an LN$_2$ dewar) would be flowed through a heated copper block, then into the U-tube welded to the valve. The valve and reservoir would warm, with the valve always being warmer than the reservoir. After the proper temperature had been reached (which would be determined by an initial calibration phase) and the sample surface was ready for the alkali deposition, the valve would be opened for a predetermined length of time. Ideally, alkali would begin effusing out of the valve as soon as it was opened, and cease as soon as it was closed. Since the valve was always the hottest part of the apparatus, no alkali should stick to it during this time.

The actual operation of the oven was somewhat different. After the very first opening of the valve, the alkali flux was independent of whether the valve was open or closed, and only varied with the valve temperature. Apparently enough alkali did stick to the outside of the valve to provide a good source for many days. The entire experiment, after the oven calibration phase, was performed without opening the valve a single time. Deposition amounts were controlled using the beam block on the adjacent rotatable feedthrough.
Chapter III
Results & Discussion

There are two objectives of the current work. The first objective is to obtain a better understanding of the He* de-excitation mechanisms by examining a range of surfaces (of different work functions) through which the de-excitation mechanism should switch from the two-step mechanism to the one-step mechanism. The second objective is to address the issue of the alkali-surface bond, and the location of the alkali valence electron for various alkali coverages. This chapter is divided into four sections, the first two reporting the results, the third summarizing and discussing the results in light of the above objectives, and the last looking forward to future experiments which could be done with the apparatus.

A. Cs/Cu(100) Results

The purpose of this phase of the experiment was to measure the MDS electron energy distribution, and the polarization of broad segments of that distribution, for a series of known cesium coverages on a clean Cu(100) sample. Because of the relatively high background pressure (4·10⁻¹⁰ torr), and the extreme reactivity of the partially cesiated surface, problems were encountered in obtaining reproducible, clean surfaces. In fact, several months of cesium deposition and preliminary measurements were required before clean cesiated surfaces could be routinely obtained. During these months there apparently accumulated on the chamber inner walls enough fresh cesium to act as a getter for the more reactive gases (probably mainly O₂). Because of the relatively short time (~one hour) during which a cesiated surface would remain uncontaminated, it was not possible to
measure the surface work function (to obtain the cesium coverage), to acquire the MDS electron energy distribution, and to obtain polarization data while the surface remained clean. Therefore the dependence of the MDS energy spectrum on coverage was first studied, and a calibration developed, so that that spectrum could be used later to infer coverage for a particular polarization data set.

During the calibration part of the experiment, a correlation was developed between the surface work function (and hence the cesium coverage), and the MDS electron energy spectrum. The sample was first cleaned with a sputter and anneal cycle as described in the previous chapter. Cesium was then deposited on the surface, and the work function of the surface was measured. Then the MDS energy distribution was measured, and the work function was again measured. If these work function measurements differed by more than 0.1 eV (a rare occurrence), the MDS data was not used. The coverage was estimated from the work function, using the work function vs. coverage curve reported by Papageorgopoulos\textsuperscript{36}, which is shown in Fig. 6. In this way, a calibration was developed so that the coverage could be estimated from the MDS energy distribution alone. Figure 18 shows a representative set of these calibration distributions. Note that the coverage variable $\theta$ from the figure is defined to be unity when the number density of adatoms is equal to the number density of substrate atoms. Since cesium atoms are larger than copper atoms, one close packed adlayer corresponds to $\theta=0.27$. It was not possible to estimate coverage accurately from the deposition time and the cesium flux measured by the hot wire detector. The most likely explanation is that cesium's sticking coefficient varied widely over the range of sample temperatures (30°C - 100°C) at which depositions were performed from day to day.
Figure 18 - Representative set of coverage calibration energy distributions for cesium on Cu(100)
The amount of cesium deposited was controlled in one of two different ways. Either a short deposition time (<30 seconds) was used, so that full coverage was never reached, or a full layer was deposited by a minute or two of deposition, followed by controlled desorption by either flashing the sample to an appropriate temperature (between 150°C and 350°C), or by maintaining the sample at some elevated temperature. The thermal desorption methods both had drawbacks. If the sample were flashed, then it would take at least 30 minutes to cool back down enough so that its temperature was no longer changing rapidly (during which time the sample might become contaminated). If the sample were maintained at a certain temperature, low energy electrons from the heating filament would escape the sample holder enclosure and swamp the low energy part of the ejected electron spectrum. Therefore thermal desorption techniques were used only for some of the calibration data, where only the high energy peak was important. The polarization data was all taken following a timed, partial deposition with the sample temperature less than 100°C.

To obtain the polarization measurements, the sample was cleaned and annealed as before, and the cesium was deposited. Then an energy distribution was measured prior to and following a polarization measurement. The similarity of these two distributions assured a lack of contamination by background gases. These results are shown in Fig. 19, along with an estimate of the cesium coverage in each case.

Several interesting features of the data are:

i) The position of the upper edge of the high energy peak is constant, although the peak does grow in width. Its position is consistent with one-step de-excitation from the cesium 6s valence level.

ii) The low-energy cut-off occurs near zero volts for nearly-clean Cu(100), but quickly decreases to -2 volts for coverages as low as 0.06 monolayer, or 20% of a close packed layer.
Figure 19 - Energy and polarization distributions for different coverages of cesium on Cu(100)
iii) There is an intermediate coverage regime (Fig. 19b) where the broadened distribution characteristic of two-step energy process co-exists with the sharp high energy peak indicative of the one-step process.

iv) The polarization of electrons ejected from a nearly-clean Cu(100) surface is low, but is clearly greater than zero. The highest energy electrons also have the highest polarization.

v) The polarization of the low energy electrons drops, while the polarization of the high energy electrons rises with increasing cesium coverage.

vi) The polarization of the high energy peak is significantly less than 100% for all coverages.

Observations i) - iii) are consistent with previous work by several groups on similar systems\textsuperscript{37,38}. In particular, a very careful work by Hagstrum, et. al.\textsuperscript{39} with low energy He\textsuperscript{+} ions impacting on K/Cu(100) shows a smooth decrease of \~2.5 volts in the low energy cutoff as coverage is slowly increased. Observation iii) indicates that the local work function of the surface varies from point to point, since the two-step and one-step processes are occurring simultaneously at different sites. In fact, the two-step signature is still evident even at a surface with a global work function of 2.1 eV, indicating that bare sites still retain a locally much higher work function.

Observation iv) is not a new observation\textsuperscript{40}, but is significant in that it shows that at least one of the two usual model assumptions made in discussing metastable de-excitation at high work function surfaces is incorrect. The two assumptions are:

a) The two step process always dominates at clean, high work function metal surfaces

b) The spin-spin correlation between the neutralizing and ejected electrons participating in the two-step process is very small
If both of these assumptions were true, the polarization of electrons ejected from a clean copper surface should be small. Our data shows substantial polarization, possibly in excess of 50% at the highest energies.

If one assumes that a) is true, one must envision some mechanism that gives substantial spin-spin correlation between the two electrons participating in AN. There are currently no known theoretical models that predict such a correlation. As mentioned in the introduction, however, there is some experimental evidence suggesting such correlation could exist.9

If one assumes b) is true, one must envision some way to inhibit the two-step process, even at surfaces where it is energetically possible for it to occur. One possibility is the deformation of the 2s orbital due to its interaction with the surface. As the He* atom approaches the surface, both the nucleus and the two electrons induce image charges in the surface. Since the 1s electron is much more tightly bound than the 2s electron, one would expect any deformation of that orbital to be much smaller than the 2s orbital deformation. As a first approximation then, we can consider the nucleus and the 1s electron as a single +e core charge. This picture gives the following additional interactions - an attractive interaction between the 2s electron and its image, and between the core and its image; and a repulsive interaction between the 2s electron and the core's image, and between the core and the 2s electron's image. The Hamiltonian for the 2s electron can be written as the sum of the Hamiltonian for the free atom 2s electron, the 2s-2s image interaction, and the 2s-core image interaction:

\[
H = H_{2s} + V_{2s,2s'} + V_{2s,\text{core}'}
\] III.1
This Hamiltonian has been numerically modelled by Dr. Peter Nordlander for He (2^3S) atoms approaching a jellium surface of high work function. He finds significant hybridization between the 2s and 2p states as the atom approaches within 5Å of the surface (auger neutralization is thought to occur at ~3Å). The hybridization forms two new eigenstates, shown in Fig. 20. The energy of the orbital facing away from the surface is ~1 eV lower than the other, so the initial 2s orbital will adiabatically transform into that state. Since most of the excited electron probability density faces away from the surface, there is less overlap with the vacant surface states than for the unperturbed 2s state. This could reduce the electron tunnelling rate into the surface enough to allow the He* to approach close enough to the surface for the one-step process to occur before resonance ionization occurs.

![Diagram of He* near a metal surface](image)

Figure 20 - Hybridized 2s-2p states of He* near a metal surface
Observation v) is another confirmation of a change from two-step to one-step de-excitation as cesium is adsorbed onto the surface. The high polarization of the high energy feature indicates that these electrons are emitted via the one-step process from the partially filled cesium valence level. The drop in polarization of the low energy electrons can be explained by an understanding of their origin. Approximately half of the ejected primary electrons are directed into the surface, and may be lost, may scatter elastically or inelastically off other surface electrons and back into the vacuum (becoming re-diffused primaries), or may cause one or more other surface electrons (secondary electrons) to be ejected into the vacuum. Studies by another group at Rice indicate that electrons which scatter with little energy loss retain most of their polarization, so are believed to be mostly nearly elastically scattered primaries, with a few true secondaries. Larger energy primary electrons are capable of scattering more lower energy true secondary (unpolarized) electrons into the vacuum, which will dilute the polarization of the low energy ejected electrons. Therefore, larger primary-secondary energy losses generally produce electrons with more diluted (hence diminished) polarization. In the clean surface case, the primary electrons have 5-12 eV of energy, and the secondaries reflect about a 5 eV energy loss. In the fully covered case, however, the primary electrons have about 15 eV of energy, so the secondaries suffer 10-15 eV of energy loss, and hence have a lower polarization, even though the primary electrons begin with a higher polarization.

Observation vi) indicates that electron ejection during metastable de-excitation at a low work-function surface is not due simply to direct one-step AD. To obtain a more detailed picture, the polarization of only the high energy peak was studied carefully for various cesium coverages. The results are shown in Fig. 21. The polarization of the peak is highest at the lowest coverages at which the peak is observable. The polarization then
Figure 21 - Polarization of the high energy peak for various cesium coverages
decreases slightly (but significantly) as coverage is increased. As the high energy peak increases in height, it also increases in width. The polarization of just the highest energy tail of the peak is also significantly greater than polarization of the overall feature (Fig. 22). The highest energy electrons are the most highly polarized, but in no case was a polarization of 100% observed. One possible mechanism that might account for this depolarization is the partial resonant tunnelling exchange of electrons between the surface and the He* atom. This has been suggested by Kasai and Okiji,42 who model an effect which will depolarize the incident He* to values as low as 50%. This recent calculation is not undisputed, however, and more work is required to give a definitive answer.

Figure 22 - Polarization of two different high energy intervals of electrons ejected from a full monolayer of cesium on Cu(100)

It is also interesting to note from Fig. 21 that the high energy peak first increases in height, then decreases slightly at the highest coverages. The area of the peak exhibits the same behavior. The height (and area) of the peak correlates well with the work function of
the surface. That is, the largest peak occurs near the minimum surface work function. The size of the peak probably corresponds to the occupancy of the broadened 6s cesium valence level, and the width of the peak corresponds to energy breadth of the state which has been filled. One can thus see that the cesium valence orbital is the most filled at an intermediate coverage, and empties slightly at full close-packed coverage.

**B. Cs + O/Cu(100) Results**

During this phase of the experiment, the Cu(100) sample was first cleaned by a sputter-anneal cycle, then a full saturation coverage (one close-packed layer) of cesium was deposited on the surface. Oxygen was then leaked into the chamber to partial pressures of 5–10·10⁻⁹ torr for 25-100 seconds. Polarizations and energy distributions were measured for surfaces with oxygen exposures of 0.25L and 0.5L (1L=10⁻⁶ torr-sec). These results are shown in Fig. 23.

The following observations may be drawn from these data:

i) Deposition of even small amounts of oxygen causes drastic changes in the surface electronic structure. Specifically, the high energy peak disappears and a smaller peak around 10 eV appears.

ii) The low energy cutoff is unchanged.

iii) The electron polarization at all energies increases, but still remains less than 100%. The lowest energy electrons show the greatest polarization change, an increase of approximately a factor of four.

Observation i) agrees with the results of Woratschek, et. al.⁴³, who deposited oxygen onto a close packed single-layer Cs/Cu(110) surface. In their paper they identify the peak at 10 volts as being associated with the (atomic) oxygen 2p level.
Figure 23 - Energy and polarization distributions for different oxygen exposures on a full monolayer of cesium on Cu(100): a) no oxygen, b) 0.25L oxygen, c) 0.5L oxygen
A separate measurement of the global work function showed that it decreased slightly (~0.2 eV) with oxygen deposition. Observation ii) is consistent with this result, since work function shifts of 0.2 eV are too small to see in the low energy cutoff.

The increase in polarization of the low energy electrons is understandable if they are interpreted to be secondary electrons. The Cs+O/Cu(100) surface produces primary electrons (which are nearly fully polarized) having ~10 eV of energy, rather than ~15 eV, as in the Cs/Cu(100) surface. There is therefore less energy available to produce secondary electrons, resulting in fewer true unpolarized secondaries ejected for each primary electron. Re-diffused (polarized) primaries therefore make up a greater fraction of the low energy ejected electrons, so that those low energy electrons have a higher net polarization. It is interesting to note that the polarization of the low energy electrons ejected from the Cs+O/Cu(100) surface is about double that from the clean Cu(100) surface. This is reasonable, since the primary electrons in both cases have close to the same energies, but the polarization of the primary electrons from the Cs+O surface is about double that from the clean Cu(100) surface.

C. Summary

Six major conclusions may be drawn from this experiment:

i) The low coverage Cs/Cu(100) surface exhibits a local variation in work function, which varies from site to site on the surface. This allows metastable de-excitation to take place via both AD and RI+AN simultaneously. In fact, the two-step mechanism is still detected even when the global work function has decreased almost to its minimum value.
ii) The net polarization of inelastically scattered electrons (comprising both secondary and re-diffused primary electrons) depends strongly on the energy loss occurring in the scattering or re-diffusion interaction. As this energy loss increases, more energy is available to eject more unpolarized surface electrons, causing the net polarization of these low energy ejected electrons to be diluted.

iii) The simple picture that electron ejection at a low work-function surface results in AD is incomplete. Rather, the data suggest that either the incident metastable atoms are depolarized before the de-excitation occurs,\textsuperscript{42} or that a significant number of electrons suffer quasi-elastic spin-flip backscattering (a view which is not supported by other related experiments at Rice).\textsuperscript{41}

iv) At least one of two fundamental assumptions of metastable de-excitation at high work function surfaces is incorrect. At present the assumption that only RI+AN occurs at such a surface appears questionable. It is possible that some AD also occurs and produces the observed non-zero electron spin polarizations.

v) The greatest occupancy of the valence orbital of cesium seems to occur at about 50% coverage, where the surface work function is a minimum. At higher coverages, the size of the AD peak in the MDS energy distribution decreases, suggesting a lower occupancy. The peak does, however, become broader, suggesting that the valence level broadens further at high coverages.

vi) Oxygen has a dramatic effect on the Cs/Cu(100) electronic structure, even in small amounts. It possibly attaches the cesium valence electron, emptying that high energy surface state.
D. Future Directions

The present experiment suggests a number of future investigations and apparatus improvements:

1. The uncertainty in the polarization data in the current experiment is limited by counting statistics, which in turn are constrained by the limited time available to acquire data from the uncontaminated surface. The current optical pumping light source, an RF helium discharge lamp, is capable of producing a He \((2^3S)\) beam which is \(~40\%\) polarized. There is currently an effort under way to implement a titanium-sapphire laser system which would produce the 1.08\(\mu\)m light suitable for optical pumping. The laser should achieve near 100\% polarization, which would cut our data taking times by a factor of four. Conversely, with the same data taking times, our polarization uncertainties would be half what they are at present.

2. The question of the origin of the significant electron polarization observed at a clean copper surface could be resolved with a polarized ion experiment. Auger de-excitation cannot occur when an ion approaches a high work function surface, so any polarization of the ejected electrons would be caused by a correlation between the spins of the neutralizing and ejected electrons. Polarized ions could be formed by first polarizing metastable atoms, then ionizing the atoms via an energetic laser (not an easy task). The polarization of the ions could not be measured directly, but would have to be assumed to be the same as that of the metastable atom beam. However, even if some depolarization does occur, the experiment should still show whether the ejected electrons are polarized or not.
Some indication of the answer to this question may also be obtained by using faster or slower polarized metastable atom beams. A faster beam would allow even less time for RI to occur, so that one might expect increased ejected electron polarizations. Conversely, a slower beam might allow even the hybridized state enough time to ionize completely before AD occurred.

3. Although only one layer of cesium will stick to Cu(100) at room temperature, many layers will stick at cryogenic temperatures. In particular, Woratschek et. al.\textsuperscript{43} have done a careful study of thick layers of cesium and oxygen on Cu(110) at 140 K, and found a number of different structures, depending on the ratio of cesium to oxygen. These thick layers behave quite differently than the single cesium adlayer. It might be interesting to study these structures with a spin sensitive probe to see if the ejected electron polarizations remain high.
Appendix A

RETARDING POTENTIAL MOTT POLARIMETER

The Mott polarimeter used in the present work is equipped with a retarding potential energy analyzer. This allows one to measure the average polarization of electrons above some cutoff energy (by setting the retarding grids to repel any electrons with lower energy), and, by difference methods, to measure the polarization of electrons in some energy interval.

An ideal retarding grid set at $V_r$ will allow every electron with energy above $eV_r$ to pass, and will reject every electron with energy less than $eV_r$. The finite size of the grids (mesh size and overall dimension), the presence of a magnetic field of $\sim 0.75$ gauss (necessary to maintain a well defined quantization axis for the polarized He* atoms and the polarized electrons) in the main experimental region, and the different trajectories of electrons through the grids all influence the characteristics of the Mott polarization analyzer. To explore the transmission properties of the grids, the Mott polarimeter with grids was modelled with an electron trajectory simulation program. The program used was Simion v. 4.02 from the Idaho National Engineering Laboratory running on a Compaq deskpro 386/20e. The cylindrically symmetric model used is pictured in Fig. 24. A magnetic field of 1.5 gauss (double that which exists in the actual experimental chamber) directed into the figure was included in the model calculation. This doubled field will tend to decrease the energy resolution of the analyzer, so the results of the calculation should represent an upper limit of the resolution of the actual device. Electron trajectories from the sample were traced from five equi-spaced points along the sample surface, and for all ejection angles at
$5^\circ$ increments. This totals ($5$ positions) $\times (180^\circ/5^\circ) = 180$ trajectories for each parameter set modelled (most of those trajectories which did not pass through even the first aperture are not shown in the figure). The retarding grids were set at various voltages, and the number of electron trajectories which enabled an electron to pass through the grid region and arrive at the gold foil were counted.

These data were then graphed in Fig. 25. In the top figure, each line represents a different ejected electron energy. The horizontal axis represents retarding grid voltage. It is apparent that electrons of a given energy are transmitted through the energy analyzer with closely uniform efficiency until the retarding grids are biased to within a volt or so of the cutoff energy. In the bottom figure, each line represents a different retarding grid voltage and the horizontal axis represents ejected electron energy. Again, one can see that when the grids are set at a particular voltage, virtually all electrons with more than one volt more energy than the grid bias are passed with nearly uniform efficiency.
Figure 24 - Cylindrically symmetric model used to calculate electron trajectories into the Micro-mott polarimeter. Sample trajectories for 10eV electrons and -5V on the retarding grids are shown.
Figure 25 - Performance characteristics of the Micro-mott polarimeter. In the top graph, each line represents a different electron energy. In the bottom graph each line represents a retarding grid voltage, and shows the relative numbers of different energy electrons which are transmitted through the grids.
Appendix B
HOT WIRE ALKALI ATOM DETECTION

As an atom with ionization potential I approaches a metal surface of work function $\Phi$, there is some probability that an electron from the atom will transfer to the surface. This probability depends on the difference ($\Phi - I$) and on the surface temperature. It is given approximately (within a factor of two) by the relation

$$\frac{n_+}{n_a} = \exp\left[\frac{\Phi - I}{kT}\right]$$  \hspace{1cm} B.1

where $n_+$ and $n_a$ are the outgoing ionic and atomic species populations, respectively.

For a cesium atom approaching a warm (~900$^\circ$ C) tungsten wire, the relation becomes

$$\frac{n_+}{n_a} = \exp\left[\frac{4.5 \text{ eV} - 3.9 \text{ eV}}{0.12 \text{ eV}}\right] = 4160$$  \hspace{1cm} B.2

so that the incident atoms will be ionized with close to 100% efficiency. It has been found experimentally\textsuperscript{44} that a tungsten surface will ionize any atom having $I < 4.5$ eV (which includes K, Rb, Cs) with virtually 100% efficiency.

The detector used in the current work consists of a 7 mil tungsten wire 1/2" long which may be heated by passing a current through it. It is mounted on a rotatable feedthrough 1" from of the alkali oven. The current source used to heat the wire is well isolated from ground (we used two rechargeable D-cell high capacity Ni-Cd batteries), and current from the wire to ground (due to alkali valence electrons being captured by the wire) is measured with a Keithley electrometer. A schematic illustration is shown in Fig. 26.
The sole reason for heating the wire is to maintain a clean tungsten surface. At lower temperatures, cesium adsorbs to the wire, lowering the work function and rendering the detector inoperative. We found a broad plateau in the collected current between wire temperatures of ~800° C and 1500° C. Cesium boils (at atmospheric pressure) at 670° C. Apparently significant cesium adsorbed to the wire below 800° C. At temperatures above 1500° C, the collected current increased rapidly, probably caused by other impurities boiling off the wire as ions. The detector was generally operated somewhere in the middle of this plateau, where the wire just began to glow visibly.

![Hot wire schematic](image)

**Figure 26 - Schematic diagram of the hot wire detection electronics**

The alkali oven was typically operated at ~60° C, which created a flux of atoms corresponding to a collection current of 1.5·10⁻⁷A on the hot wire. The hot wire, being 7 mil in diameter and half an inch long, and suspended one inch from the valve, subtended a solid angle of 3.5·10⁻³sr. A current of 1.5·10⁻⁷A corresponds to ~10¹² atoms/sec, yielding a flux density of 2.8·10¹⁴ atoms/sec/sr. During depositions, the sample was
~4 cm from the valve, yielding a flux of $1.8 \times 10^{13}$ atoms/cm$^2$/sec on the sample surface. One close packed cesium layer corresponds to a surface number density of $4.0 \times 10^{14}$ atoms/cm$^2$. Thus, under typical operating conditions, one would expect to deposit one full layer in just over 20 seconds. Measurement of alkali coverage after deposition showed that between 15 and 30 seconds of deposition were actually required to obtain one full adlayer.
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