RICE UNIVERSITY

Nonequilibrium Dynamics of Quantum-Degenerate Fermionic and Bosonic Gases in Semiconductors Probed by Coherent Terahertz Magneto-optics

by

Qi Zhang

A THESIS SUBMITTED IN PARTIAL FULFILLMENT OF THE REQUIREMENTS FOR THE DEGREE Doctor of Philosophy

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ABSTRACT

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Quantum-confined semiconductor structures are ideal systems in which to study non-equilibrium and coherent dynamics of interacting many particles in a highly controllable fashion. In particular, two-dimensional (2D) semiconductor systems in a strong perpendicular magnetic field provide one of the cleanest condensed matter systems with ultralong coherence times, allowing us to excite and control macroscopic coherent phenomena. When doped, either electrically or optically, such systems can accommodate quantum-degenerate fermions (electrons) and/or bosons (excitons). In this dissertation, we studied the coherent terahertz (THz) dynamics of 2D gases of electrons and excitons in GaAs quantum wells in magnetic fields with time-domain THz magneto-spectroscopy. In high-mobility 2D electron gases, we made the first observation of collective radiative decay, or superradiance, of cyclotron resonance (CR). The decay rate of coherent CR oscillations increased linearly with the electron density in a wide range, which is a hallmark of superradiant damping. Our fully quantum mechanical theory provided a universal formula for the decay rate. We further achieved ultrastrong coupling of coherent CR with THz photons in a high quality factor 1D photonic crystal cavity. We directly observed time-domain vacuum Rabi oscillations, and the square root of N dependence of collective Rabi splitting with
respect to the carrier density. Superradiance decay of CR was significantly suppressed in the cavity, and an intrinsic CR linewidth as sharp as 5.6 GHz was resolved. In undoped GaAs quantum wells, we systematically investigated the nonequilibrium dynamics of electron-hole pairs using ultrafast optical-pump THz-probe spectroscopy. We simultaneously monitored the intraexcitonic 1s-2p transition, which splits into the 1s-2p$_+$ and 1s-2p$_-$ transitions in a magnetic field, and the CR of unbound carriers as a function of pair density, temperature, magnetic field, and probe delay time. We found that the 1s-2p$_-$ feature is robust at high magnetic fields even under high excitation fluences, indicating magnetically enhanced stability of excitons. While mimicking some of the well-known phenomena in quantum optics of atomic and molecular gases, these results highlight some of the unique features of condensed matter systems due to strong many-body Coulomb interactions among carriers and open a door to the novel physics of THz many-body electrodynamics.
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# Contents

Abstract .................................................. ii
List of Illustrations ................................... viii

1 Introduction ............................................. 1
   1.1 Scope of the thesis .................................. 3

2 Experimental Methods and Samples .................. 5
   2.1 Time-domain THz magneto-spectroscopy .......... 5
   2.2 Time-resolved THz magneto-spectroscopy ........ 9
   2.3 GaAs quantum wells ................................ 11
   2.4 THz 1D photonic-crystal cavities ................. 16

3 Superradiant Decay of Electron Cyclotron Resonance in
   Two-Dimensional Electron Gases .................... 21
   3.1 Introduction ....................................... 21
   3.2 Experiments and results .......................... 23
   3.3 Theoretical analysis .............................. 28
   3.4 Conclusion ....................................... 33

4 Strong Coupling of Collective Electron Cyclotron Reso-
   nance and THz Cavity Photons ....................... 34
   4.1 Introduction ....................................... 34
   4.2 Experiments and results .......................... 38
   4.3 Quantum Mechanical Picture ....................... 48
4.4 Classical Electrodynamical Picture .......................... 55
4.5 Conclusion and prospects .................................... 59

5 Ultrafast Dynamics of Excitons and Magneto-excitons in GaAs Quantum Wells

5.1 Introduction .................................................. 62
5.2 Continuous Wave Spectroscopy .............................. 68
5.3 Exciton Dynamics at Zero Magnetic Field ............... 71
5.4 Exciton Dynamics at Finite Magnetic Fields ............ 76
5.5 Conclusion ..................................................... 81

6 Conclusions .................................................... 82

Bibliography ...................................................... 85
Illustrations

2.1 Schematic diagram of the time-domain THz magneto-spectroscopy system used in this work. .............................................. 7

2.2 Typical (a) time-domain THz waveform and (b) the power spectrum in the frequency domain (log scale). ................................. 8

2.3 Schematic diagram of the time-resolved THz magneto-spectroscopy system (TR-TMS) used in this work. ................................. 10

2.4 Structure diagrams of a modulation doped high-mobility 2DEG (left) and an undoped GaAs multiple quantum well sample (right). .... 12

2.5 Preparation procedures for a 2DEG sample on a thin Si wafer for inclusion in a THz photonic-crystal cavity. .............................. 14

2.6 Preparation procedures of undoped GaAs quantum well samples for investigating exciton dynamics. ................................. 15

2.7 Schematic diagram of (a) a 1D photonic-crystal and (b) a 1D photonic-crystal cavity. ................................................................. 17

2.8 1D photonic crystal cavity constructed by stacking thin silicon wafers and copper spacers. (a) Schematic structure diagram. The thin 2DEG film (blue) is transferred onto the center Si wafer. Air spacing is created by the thin copper films with holes in the middle. (b) Photographs of silicon wafer and copper spacer. (c) Side view and (d) front view of a THz cavity sample on the aluminum sample holder. 19
2.9 (a) Experimental transmission spectrum of a two-layer Si 1D PCC without a 2DEG. (b), (c) and (d) show, respectively, the first, second, and third cavity modes together with Lorentzian fits. The FWHM of the three modes were 2.3 GHz, 2.8 GHz and 2.6 GHz, respectively.

3.1 Coherent THz pulse creates a superposition of adjacent Landau levels with massive degeneracy. In the figure, the Landau level spacing equals $\hbar \omega_{CR}$, where $\omega_{CR}$ is the cyclotron frequency. The free induction decay of such a superposition state can be observed after the excitation pulse.

3.2 (a) A schematic of the polarization-resolved THz magneto-transmission experiment in the Faraday geometry. (b) Coherent cyclotron resonance oscillations in the time domain. Each blue dot represents the tip of the THz electric field at a given time. The red traces are the projections of the waveforms onto the $E_x$-$t$ and $E_x$-$E_y$ planes. The bottom trace is the difference between the top (0 T) and middle (2.5 T) traces.

3.3 (a) Magnetic field dependence of CR oscillations, showing peaks (blue) and valleys (red). (b) The frequency-domain version of (a). Black dashed line: linear fit with a cyclotron mass of $0.069m_e$. (c) Magnetic field dependence of $\tau_{CR}$ at 3 K. (d) Temperature dependence of $\tau_{CR}$ at 2.5 T. All the data are for Sample 1.
3.4 (a) Low-density sample (Sample 2) exhibiting the longest $\tau_{CR}$ of $\sim$40 ps. (b) CR oscillations in Sample 1 with different densities by controlling the illumination time. (c) Decay rate as a function of density. Blue solid circle: Sample 2. Red solid circles: Sample 1. The blue dashed line represents Eq. (3.11) with no adjustable parameter, where $n_{GaAs} = 3.6$ and $m^* = 0.069m_e$ are used. (d) Incident $E_i$, reflected $E_r$ and transmitted $E_t$ THz pulses at the 2DEG. (e) The decay of the THz-pulse-excited energy in the 2DEG. The red dashed line is an exponential fit. About 80% of the energy relaxes through CR superradiance.

3.5 (a) The measured values of $\Gamma_{CR} - \Gamma_{DC}$ versus $\Gamma_{SR}$ given by Eq. (3.11) for four representative data points in the present study and values from Refs. [1,2]. The solid line has a slope of 1. (b) $\Gamma_{scatt.} \equiv \Gamma_{CR} - \Gamma_{SR}$ as a function of $\sqrt{B}$. Red solid line: linear fit. Blue dashed line: prediction based on short-range scattering [3].

4.1 Schematic diagram for a two-level matter system coupled with a cavity, where $g$ represents the coupling strength between the two-level system and the cavity field, while $\kappa$ and $\gamma$ are the cavity and matter decay rates, respectively.
4.2 1D THz photonic crystal cavity (PCC) with a high-mobility 2DEG (Cavity 1). (a) Schematic diagram for CR involving two adjacent Landau levels coupled with a THz cavity field. (b) 1D THz PCC structure. Two silicon layers are placed on each side of the center defect layer. The blue part is the transferred 2DEG thin film. (c), (d), and (e), calculated electric field amplitude distribution inside the cavity for the 1st, 2nd, and 3rd cavity modes, respectively. 2DEG is located at the field maximum for all three cavity modes. (f) Experimental power transmittance for Cavity 1. Three sharp cavity modes are clearly resolved in the middle of each stop-band. Zoom-in spectra for the three modes are shown in (g), (h), and (i), together with Lorentzian fitting. The FWHM is 2.6 GHz, 5 GHz, and 3.8 GHz for the 1st, 2nd, and 3rd mode, respectively.

4.3 Anticrossing of CR and the first cavity mode in Cavity 1. The residual cavity mode results from the CR inactive component of the linearly polarized THz beam. Transmission spectra at different magnetic fields are vertically offset for clarity. The magnetic field is increased from 0.4 T (bottom) to 1.4 T (top).

4.4 Anticrossing of CR with (a) the second cavity mode and (b) the third cavity mode in Cavity 1. In (a), the magnetic field is increased from 2.4 T (bottom) to 3.4 T (top). In (b), the magnetic field is increased from 4.6 T (bottom) to 5.2 T (top). (c) Summary of measured peak positions of the magneto-polariton branches for the three cavity modes, the value $\nu$ indicate the filling factor at the resonances.
4.5 Anticrossing of CR with (a) the fundamental cavity mode and (b) the second cavity mode in Cavity 2. All traces are vertically offset for clarity. In (a), the magnetic field is increased from 0.5 T (bottom) to 1.35 T (top) with a step size of 0.05 T. In (b), the magnetic field is increased from 2.1 T (bottom) to 3.2 T (top) with a step size of 0.05 T. The linewidth of both cavity modes and polaritons is limited by the 80 ps time window. (c) Peak positions for the magneto-polariton branches for the first (blue) and second (red) cavity modes.

4.6 (a) Vacuum Rabi splittings observed for 2DEGs with three different electron densities in Cavity 1. CR is resonant with the fundamental mode. (b) Square root of $n_e$ dependence evidencing the collective nature of vacuum Rabi splitting.

4.7 (a) Vacuum Rabi oscillations in the time domain. Cyclotron resonance is resonantly coupled with the second cavity mode at 2.975 T in Cavity 1. $\Delta E_y = E_y(+2.975 \text{T}) - E_y(-2.975 \text{T})$ is the measured difference between the transmitted THz waveforms taken at +2.975 T and -2.975 T in the $y$-polarization direction, as shown by the top black trace. More clear vacuum Rabi oscillations can be observed by removing the residual cavity mode due to the CR inactive THz component with a numerical notch filter (bottom blue trace). The beating nodes of two magneto-polaritons are indicated by arrows. (b) Frequency-domain spectrums for $\Delta E_y$ before and after the removal of residual cavity mode.
4.8 (a) Temperature dependence of magneto-polaritons in Cavity 1 at zero detuning with the fundamental cavity mode from 2 K to 80 K with Lorentzian fitting (black dashed lines). Traces are vertically offset for clarity. (b) Temperature dependence of CR decay time $\tau_{CR}$ in free space (red solid circles), in cavity (blue solid circles) and DC momentum scattering time $\tau_{DC}$ (black open circles).

4.9 The probe field strength dependence of the polariton splitting. (a) THz probe field The peak electric field from bottom to top is 5, 10, 20, 30, and 45 V/cm, respectively. The corresponding THz field at the cavity frequency is $\sim 10^{-3}$ times smaller than the peak field. (b) The corresponding polariton splitting for each case. The splitting remain unchanged from the case of $10^3$ averaged cavity photons to the few photons quantum limit.

4.10 (a) and (b) present the best fits to our experimental data of the two cavities using three different Hamiltonians— the full CR-cavity Hamiltonian (black solid), the full CR Hamiltonian without the $A^2$ term (black dashed), and the Jaynes-Cummings Hamiltonian (blue dotted). Only the full CR-cavity Hamiltonian reproduced the experimental data well, suggesting the non-negligible contribution of the $A^2$ term on the polariton frequencies, i.e., the system is close to the ultrastrong-coupling regime. (c) and (d) show the normalized fitting deviation as a function of $g/\omega_0$ ratio. The best fit achieved when, $g/\omega_0$ is 0.09 for Cavity 1 and 0.12 for Cavity 2, which are at the edge of the ultrastrong-coupling regime.
4.11 (a) Experimental transmission spectrum of Cavity 1 is reproduced by the transfer matrix method at 0 T. (b) Calculated cavity power transmittance for a CR active THz wave with realistic values of 2DEG density \(3 \times 10^{11} \text{ cm}^{-2}\) and mobility \(4 \times 10^6 \text{ cm}^2/\text{Vs}\) in magnetic fields up to 6 T. The calculation matches the experimental peak positions (yellow open circles) of the magneto-polaritons in Cavity 1.

4.12 (a) Calculated cavity power transmittance for linearly polarized THz probe pulse with a realistic 2DEG density for Cavity 2.

(b) Experimental transmittance of Cavity 2 from 0 T to 3.5 T with a color scale from 0 to 1 in transmittance. (c) Same experimental data with a color scale from 0 to 0.06 in transmittance. Magneto-polaritons and residual cavity mode are well resolved. They match the calculated spectrum in (a) well. The linewidths of the cavity mode and magneto-polaritons are instrument limited by the 80 ps time window.

5.1 Schematic phase diagram of an e-h pair system as a function of pair density and temperature at zero magnetic field.

5.2 Schematic phase diagram of an e-h system as a function of pair density and magnetic field. In the low density region, excitons become magnetoexcitons with their center of mass momentum coupled with internal energy. A correlated magnetoplasma is located in the high density region. Novel cooperative phenomena, e.g., superfluorescence [4], can appear in this region. In the extreme quantum limit of high magnetic fields, an insulating phase of noninteracting magnetoexcitons appears due to the e-h charge symmetry (‘hidden symmetry’). Adopted from Ref. [5], Figure 1.
5.3 Continuous-wave (a) absorption and (b) photoluminescence spectroscopy of undoped GaAs quantum wells from 0 T to 9 T at 5 K. The curves are vertically offset for clarity. .......................... 69

5.4 Temperature dependence of photoluminescence spectra at zero magnetic field. The bound exciton (BX) peak at 1.521 eV is not observed at a temperature higher than 20 K. The free exciton (FX) PL intensity increases with the temperature. .......................... 70

5.5 The peak positions of the observed interband transitions as a function of magnetic field. The red symbols are for absorption peaks and the blue symbols are for PL peaks. .......................... 70

5.6 (a) Schematic diagram of measuring exciton dynamics in undoped GaAs multiple QW with time-resolved THz magneto-spectroscopy. (b) Spectra of resonant optical pump pulse with the corresponding absorption spectra at 0 T and 9 T. The non-resonant excitation photon energy was kept at 1.6 eV in all measurements. .......................... 72

5.7 Photo-induced change in the real part of (a) conductivity and (b) permittivity under resonant excitation at 5 K and 0 T. The pump fluence was 150 nJ/cm$^2$. The 1s-2p transition is observed at 1.56 THz (or 6.4 meV). Probe delay time was 100 ps. .......................... 73

5.8 Dynamics of unbound e-h pairs and excitons after non-resonant excitation at 5 K and 0 T. Photoninduced changes in the real part of (a) conductivity and (b) permittivity are shown. The pump fluence was 150 nJ/cm$^2$. At 20 ps, $\Delta\epsilon(\omega)$ was negative, which is a characteristic metallic behavior. The 1s-2p exciton features was observed after 150 ps, and survived even after 1.8 ns. .......................... 74
5.9 Photo-induced conductivity spectra $\Delta \sigma(\omega)$ at various resonant pump fluences at 5 K and 0 T. The pump fluence from the bottom trace to the top were $1 \times 150$ nJ/cm$^2$ up to $40 \times 150$ nJ/cm$^2$. An excitonic Mott transition was observed at 5 K and 0 T. The $1s-2p$ transition totally vanished at the highest pump fluence, and the conductivity was dominated by an e-h plasma. The optical pump delay was 100 ps.

5.10 Calculated intraexciton transition energy and cyclotron frequencies of electrons and heavy holes as a function of magnetic field based on a 2D hydrogen model.

5.11 Photoinduced conductivity spectra as a function of magnetic field from 0 T to 10 T at (a) 15 ps and (b) 1 ns THz probe delay, after nonresonant excitation. At 15 ps, a hot e-h magnetoplasma shows two CR branches due to electrons and heavy holes. At 1 ns, both the $1s-2p_+$ and $1s-2p_-$ transitions were resolved.

5.12 Photoinduced conductivity spectra at various probe delay times and temperatures at 9 T. (a) Dynamics of magnetoexcitons after nonresonant excitation with 200 nJ/cm$^2$ fluence at 6 K. (b) Similar measurements to (a) but with resonant excitation of HH $1s$ excitons. The pump fluence was also 200 nJ/cm$^2$. (c) Temperature dependence of $\Delta \sigma(\omega)$ with resonant pumping, measured at 200 ps.

5.13 Photoninduced conductivity $\Delta \sigma(\omega)$ and permittivity $\Delta \epsilon(\omega)$ spectra at various resonant pump fluences at 5 K and 9 T. The pump fluence from the bottom trace to the top were $1 \times 200$ nJ/cm$^2$ up to $40 \times 200$ nJ/cm$^2$. The $1s-2p_-$ survived even at the highest fluence, which would be sufficiently large to drive the excitonic Mott transition at 0 T.
Chapter 1

Introduction

Condensed matter physics and quantum optics are two major branches of modern physics. Applying quantum optical concepts and methods to various collective and correlated excitations in solids has already resulted in a series of discoveries, including polariton condensation in semiconductor micro-cavities [6,7], quantum zero-point motions of mesoscopic optomechanical resonators [8,9], and collective excitations in spin ensembles for hybrid cavity quantum electrodynamics (QED) [10–13], just to name a few. From a condensed matter perspective, quantum optical models and techniques developed for many-atom systems provide valuable ideas and tools for understanding photo-induced non-equilibrium and coherent dynamics in interacting many-particle systems in solids, e.g., the discovery of superfluorescence in a solid state system [4].

From a quantum optics research and applications perspective, various solid state excitations offer novel properties, or even a new regime of light-matter interaction, i.e., ultrastrong coupling, which cannot be achieved with traditional atomic systems and may lead to novel quantum optical phenomena.

Among various solid state systems, quantum-confined semiconductor structures are ideal platforms on which to study non-equilibrium and coherent dynamics of interacting many particles in a highly controllable manner. In particular, two-dimensional (2D) semiconductor systems provide one of the cleanest condensed matter systems with ultralong coherence times, allowing us to excite and control macroscopic coherent phenomena. A perpendicular magnetic field quantizes the 2D system into
Landau levels and offers continuous tuning of Landau level spacing and their density of states. Electron-electron and electron-hole interactions are also modified correspondingly. Overall, a magnetic field offers a unique method for controlling semiconductor electronic structures, which is powerful for elucidating coherent dynamics of interacting particle states. Doping can also be accurately controlled in semiconductors, either electrically or optically, so that this system can accommodate quantum-degenerate fermions (electrons) and/or bosons (excitons), with repulsive and/or attractive Coulomb interactions, which lead to various phases of electrons and electron-hole (e-h) pairs.

The characteristic energies of many collective excitations, e.g., plasmons and phonons, are on the order of a milli-electron-volt (meV), which corresponds to the terahertz (THz) frequency range. In the case of quantum-confined semiconductor structures in magnetic fields, cyclotron resonance (CR), plasmon excitations, and intra-exciton transitions can be resonantly excited with THz radiation. Therefore, THz magneto-spectroscopy offers an unique probe of non-equilibrium dynamics and light-matter interactions with collective and correlated excitations in semiconductors with combined interests from both condensed matter physics and quantum optics.

In this dissertation, we focus on the coherent dynamics and light matter interactions of quantum-confined electrons and excitons in GaAs quantum wells in magnetic fields with time-domain THz magneto-spectroscopy. In high-mobility 2D electron gases (2DEG), we made the first observation of collective radiative decay, or superradiance, of CR. The decay rate of coherent CR oscillations increased linearly with the electron density in a wide range, which is a hallmark of superradiant damping. Our fully quantum mechanical theory provided a universal formula for the decay rate. We further achieved ultrastrong coupling of coherent CR with THz photons in
a high quality factor 1D photonic-crystal cavity. We directly observed time-domain vacuum Rabi oscillations, and the square root of $n_e$ dependence of collective Rabi splitting, where $n_e$ is the electron density. Superradiance decay of CR was significantly suppressed in the cavity, and an intrinsic CR linewidth as sharp as 5.6 GHz was resolved. In undoped GaAs quantum wells, we systematically investigated the non-equilibrium dynamics of e-h pairs using ultrafast optical-pump THz-probe spectroscopy. We simultaneously monitored the intra-excitonic $1s-2p$ transition, which splits into the $1s-2p_+$ and $1s-2p_-$ transitions in a magnetic field, and the CR of unbound carriers as a function of pair density, temperature, magnetic field, and probe delay time. We found that the $1s-2p_-$ feature is robust at high magnetic fields even under high excitation fluences, indicating magnetically enhanced stability of excitons. While mimicking some of the well-known phenomena in quantum optics of atomic and molecular gases, these results highlight some of the unique features of condensed matter systems due to strong many-body Coulomb interactions among carriers and open a door to the novel physics of THz many-body electrodynamics.

1.1 Scope of the thesis

In Chapter 2, we describe the main experimental setups used in this dissertation, including a time-domain THz magneto-spectroscopy setup and its time-resolved version. The GaAs quantum well samples and photonic-crystal cavities studied in this dissertation work are also shown in detail. In Chapter 3, we study the coherent dynamics of cyclotron resonance in high-mobility GaAs 2DEGs in free space, as a function of magnetic field, temperature, and electron density. We found that superradiance is the dominating decoherence process. Detailed quantum mechanical calculations for the superradiance damping rate are provided. In Chapter 4, we study the light-matter
interaction and coherent dynamics of CR in a high-Q 1D photonic-crystal cavity. Our experimental observations are compared with theoretical calculations based on both quantum mechanical and classical models. In Chapter 5, we present the dynamics of bound and unbound e-h pairs and excitonic Mott physics in photo-excited undoped GaAs quantum well at zero and finite magnetic fields. Finally, Chapter 6 summarizes the major results achieved in this dissertation work.
Chapter 2

Experimental Methods and Samples

In this chapter, we describe the main experimental setups used in this dissertation, including a time-domain THz magneto-spectroscopy setup and its time-resolved version. We also describe the GaAs quantum well samples studied in this dissertation work in detail, including the sample structures and selective etching of GaAs substrates as well as the construction of THz photonic crystal cavities.

2.1 Time-domain THz magneto-spectroscopy

The main experimental setup used in this dissertation work is a combination of THz time-domain spectroscopy (THz-TDS) and a superconducting magnet. It is a natural extension of conventional THz-TDS to include a high magnetic field, which provides access to the dynamics of various low-energy magnetic excitations in solid state materials.

Figure 2.1 is a schematic diagram of the setup. The 10 T superconducting magnet was manufactured by Oxford Instruments, Inc. (model Spectromag-10T), and equipped with a variable temperature insert with a usable temperature range of 1.6-300 K. Our laser source was a Ti:Sapphire regenerative amplifier (Clark MXR, Inc. CPA-2001) with 775 nm center wavelength, 1 kHz repetition rate, and 200 fs pulse width. The beam was split into two, one for THz generation, and the other for detection. THz generation was based on the optical rectification effect, in which the
The 775 nm fundamental beam undergoes difference frequency generation with itself and produces low-frequency (THz) radiation. The repetition rate of THz pulses was the same as that of the pump laser, i.e., 1 kHz. More importantly, all pulsed in THz pulse trains were phase-locked, thanks to the optical rectification effect, in which the phase of THz pulses was only determined by the intensity envelope of the 775 nm fundamental beam, not by its carrier envelope phase (CEP). Therefore, the CEP of THz pulses was stable. This property ensured that the THz waveforms were sampled and reconstructed from many different pulses. The nonlinear crystals used for both generation and detection were 1 mm thick ⟨110⟩-oriented zinc telluride (ZnTe).

THz detection was based on electro-optic (EO) sampling, which is the inverse process of optical rectification. The birefringence induced by the THz pulse in the ZnTe crystal is proportional to the THz electric field (the Pockels effect). Hence, the THz electric field information was encoded into the polarization state of the THz probe beam after going through the ZnTe crystal. The polarization of the probe beam was further analyzed by a combination of a Wallaston prism and a silicon balanced detector. To improve the signal-to-noise ratio (SNR), the THz generation beam was modulated by a mechanical chopper at a frequency of 500 Hz synchronized with the 1 kHz 775 nm pulse train. The electronic signal from the balanced detector was demodulated by a lock-in amplifier to extract the THz electric field at a specific time delay. By scanning the delay of the THz probe beam, we were able to reconstruct the entire THz waveform, which contained both amplitude and phase information.

The generated THz pulses were linearly polarized, and the polarization direction was fully determined by the polarization of the 775 nm laser beam during the optical rectification process. To further ensure the linear polarization, one pair of wire-grid polarizers was placed in the THz beam path before and after the sample. In most of
Figure 2.1: Schematic diagram of the time-domain THz magneto-spectroscopy system used in this work.
experiments in this dissertation, the electric field of THz pulses was vertical (defined to be $x$-polarized), which was perpendicular to the optical table. By rotating the detection ZnTe crystal and the polarizer after the sample by 90 degrees, the horizontal component ($y$-polarized) of the transmitted THz pulses was measured.

Figure 2.2 shows a typical time-domain THz waveform and frequency-domain spectrum obtained with a single scan. The average power of the 775 nm THz generation beam was about 200 mW. The time constant of the lock-in amplifier was 100 ms. The THz pulse duration was about 2 ps, with a bandwidth up to 2.6 THz, which was mainly limited by the phonon absorption in the 1 mm thick ZnTe crystal. The peak electric field strength is on the order of 100 V/cm at the sample with a beam size of about 3 mm in diameter at 1 THz. The pulse energy was about 10 pJ, and the average power was 10 nW.
2.2 Time-resolved THz magneto-spectroscopy

Different from conventional Fourier-transform infrared spectroscopy (FTIR) with a continuous-wave light source, ultrafast laser based THz-TDS provides a train of picosecond THz pulses, which allow one to perform time-resolved measurements with picosecond time resolution.

In a time-resolved pump-probe experiment, the sample is first excited by the pump pulse at time zero, and the subsequent dynamics of the system is probed by a second pulse after a certain time delay. Therefore, in addition to the THz generation and detection beams, we needed another beam for optical excitation with an independently controlled time delay. A schematic diagram of our time-resolved THz magneto-spectroscopy (TR-TMS) system is shown in Figure 2.3. Experimentally, two optical pump beams were added to the THz setup. The first optical pump beam was split from the THz generation beam by a second beam splitter, and had an average power of 100 mW at 775 nm (1.6 eV in photon energy). The second optical pump beam was from an optical parameter amplifier (OPA). By utilizing an amplified parametric down-conversion process, the OPA was capable of producing ultrashort pulses from the ultraviolet to the mid-infrared, if equipped with appropriate nonlinear crystals. In our particular experiment of probing exciton dynamics in GaAs, described in Chapter 5, a narrow-band pulse was required to resonantly excite the heavy-hole 1s exciton peak in a GaAs QW, which occurred at 814 nm at helium temperatures, with a line-width of 1 nm. We used the second harmonic (SH) of the idle beam from the OPA, and further applied a 4f pulse shaper to accurately select a very narrow frequency component. Wavelength tuning was achieved by moving the slit.

In the case of detecting photo-generated dilute exciton gases in GaAs quantum wells described in Chapter 5, the photo-induced THz change was extremely small
Figure 2.3: Schematic diagram of the time-resolved THz magneto-spectroscopy system (TR-TMS) used in this work.
(10^{-3}-10^{-4} \text{ times the original THz field}). In order to detect such a small change, we modulated the optical pump beam, and then demodulated the THz signal with a lock-in amplifier. In this method, the lock-in output voltage was directly proportional to the photo-induced THz electric field change $\Delta E(t)$.

With the modulation method, the smallest THz amplitude change the system was able to detect was $\sim 10^{-4}$ times the original THz field amplitude; in terms of power, it was nearly $10^{-8}$. The excellent sensitivity, combined with ultrafast time resolution, as well as low-temperature and high magnetic field capabilities, makes TR-TMS a very powerful tool for revealing the dynamics of low-energy excitations in various solid state systems.

2.3 GaAs quantum wells

The GaAs quantum well (QW) samples used in this study were grown by molecular beam epitaxy (MBE) at Sandia National Laboratory or Purdue University. A silicon $\delta$-doping technique was used in the growth of high-mobility 2DEG samples. A relatively high concentration of silicon dopants were accurately introduced to GaAs within 1-2 monolayers about 100 nm away from the GaAs well, where an electron gas was located. Hence, the major electron scattering mechanism, charged impurity scattering, was significantly reduced, and the electron mobility even reached $10^7 \text{ cm}^2/\text{Vs}$.

As shown in Figure 2.4, one of our high-mobility 2DEG samples contained a 30 nm thick single well, and the electron density and mobility were $3 \times 10^{11} \text{ cm}^{-2}$ and $4 \times 10^6 \text{ cm}^2/\text{Vs}$, respectively. We also used another lower electron density sample with a similar structure. Its density and mobility were $5 \times 10^{10} \text{ cm}^{-2}$ and $4.4 \times 10^6 \text{ cm}^2/\text{Vs}$, respectively.

The electron density of a 2DEG is sensitive to light illumination at helium tem-
Figure 2.4: Structure diagrams of a modulation doped high-mobility 2DEG (left) and an undoped GaAs multiple quantum well sample (right).
peratures. Electrons are able to jump from the Si $\delta$-doping layer into the QW by absorbing photons. Experimentally, we cooled down the sample under study to 4 K in the dark. Then, we open one of the side (Viogt) windows for room light illumination. By changing the illumination time (from seconds to 2 minutes), we were able to tune the QW electron density within a certain range, depending on the sample. The density was monitored by in-situ transport measurements. After illumination, it was important to put back window covers, to prevent any stray light induced density or mobility change on the sample. The two Faraday windows for THz measurements were covered with 1 mm thick silicon wafers to block near-infrared (NIR) and visible light but transmitted THz radiation. To erase the effect of illumination, samples were slowly warmed up to a temperature higher than 200 K.

As shown in Figure 2.4, the undoped GaAs multiple QW sample for exciton dynamics studies, contained 15 pairs a 20-nm-wide GaAs well and a 20-nm-wide Al$_{0.3}$Ga$_{0.7}$As barriers, which were sandwiched by two 500-nm-thick Al$_{0.3}$Ga$_{0.7}$As spacer layers. The total thickness of the multiple QW structure was 600 nm. Since we performed THz spectroscopy experiments in a transmission geometry, we needed to remove the GaAs substrate to avoid observing any photoinduced carrier effect in bulk GaAs. We utilized a mixture solution of citric acid (1 g C$_6$H$_8$O$_7$: 1 mg DI H$_2$O) with hydrogen peroxide (30% H$_2$O$_2$ and 70% H$_2$O by volume) to selectively etch away the GaAs substrate [14]. The optimized volume ratio between citric acid solution and hydrogen peroxide was 5:1. The etching speed ratio between GaAs and AlGaAs was about 100:1 at room temperature. More detailed procedures for preparing a 2DEG on a 50-$\mu$m-thick silicon and a thin undoped GaAs QW on sapphire are shown in Figure 2.5 and Figure 2.6, respectively.
Figure 2.5: Preparation procedures for a 2DEG sample on a thin Si wafer for inclusion in a THz photonic-crystal cavity.
Figure 2.6: Preparation procedures of undoped GaAs quantum well samples for investigating exciton dynamics.
2.4 THz 1D photonic-crystal cavities

Various methods have been used for constructing cavities in the THz range, including wave guide resonators [15], metamaterial resonators [16,17], and 1D [18] and 2D [19,20] photonic-crystal cavities (PCC). Among those methods, the simple 1D PCC design offers the highest quality factor, $Q$, and thus the narrowest linewidth. A linewidth as small as 30 MHz at 0.336 THz (corresponding $Q \sim 10^4$) [18], has been achieved by a three-layer silicon PCC with an air defect layer in the middle. As schematically shown in Figure 2.7, a perfect 1D photonic crystal contains a bandgap (or a stopband) in its transmission spectrum, due to resonant Bragg reflections. If the photonic crystal has a defect inside, a THz wave will be trapped by the defect, which leads to a transmission mode inside the bandgap. Different from its near-infrared and visible counterparts, a THz 1D PCC can utilize dielectric thin slabs and air as the high and low index materials, respectively. The air-Si combination provides a huge index contrast and thus can significantly reduce the number of layers needed on each side of the cavity. The 1D PCC is also a quite feasible design for our transmission THz-TDS system to study strong THz-matter coupling in thin film samples. It is convenient to place the thin film at the electric or magnetic field maximum for electronic or magnetic excitations, respectively.

In practice, we used commercial thin silicon wafers, with a refractive index of 3.4, as the high index material. A schematic diagram of the THz cavity used in our study is shown in Figure 2.8(a). The central Si piece is twice thicker than the others as the photonic defect. A thin 2DEG film (blue part) was transferred onto the center piece, where the electric field maximum is located. Figure 2.8(b) presents a photograph of the actual material we used for THz cavity construction, including a 50-µm-thick silicon wafer, and a 200-µm-thick copper film with a circular aperture in the middle.
Figure 2.7: Schematic diagram of (a) a 1D photonic-crystal and (b) a 1D photonic-crystal cavity.
as the air spacer. Figures 2.8(c) and (d) show side and front views of a constructed THz cavity with its holder. Non-magnetic screws and a top plastic piece were used to clamp the whole cavity to the aluminum sample holder, which was attached to the sample stick used for the magnet cryostat.

Figure 2.9(a) shows an experimental transmission spectrum measured for a cavity with two layers of Si on each side. Within the 2.5 THz range, the power transmission spectrum shows three transmission stop-bands, i.e., photonic band gaps. At the center of each stop-band is a sharp cavity mode. The full-width-at-half-maximum (FWHM) of the cavity mode was calculated to be 2.3 GHz, 2.8 GHz, and 2.6 GHz for the first, second, and third modes, respectively. The theoretically expected linewidth is around 1 GHz for a perfectly aligned cavity. The small discrepancy between experiment and theory is due to the unperfect parallelism of silicon wafers and copper thin films. The highest $Q$ achieved in this structure was 810 for the third cavity mode. Technically, in time-domain spectroscopy, a time window of $\Delta T$ provides a frequency resolution of $1/\Delta T$. We performed a 500 ps time window scan in the measurement shown above, which means that the linewidths of the cavity modes were not instrument-limited.
Figure 2.8: 1D photonic crystal cavity constructed by stacking thin silicon wafers and copper spacers. (a) Schematic structure diagram. The thin 2DEG film (blue) is transferred onto the center Si wafer. Air spacing is created by the thin copper films with holes in the middle. (b) Photographs of silicon wafer and copper spacer. (c) Side view and (d) front view of a THz cavity sample on the aluminum sample holder.
Figure 2.9: (a) Experimental transmission spectrum of a two-layer Si 1D PCC without a 2DEG. (b), (c) and (d) show, respectively, the first, second, and third cavity modes together with Lorentzian fits. The FWHM of the three modes were 2.3 GHz, 2.8 GHz and 2.6 GHz, respectively.
Chapter 3

Superradiant Decay of Electron Cyclotron Resonance in Two-Dimensional Electron Gases

3.1 Introduction

Understanding and controlling the dynamics of superposition states is of fundamental importance in diverse fields of quantum science and technology [21–23]. In particular, how an excited many-body system relaxes remains one of the fundamental questions in nonequilibrium statistical mechanics [24–26]. A Landau-quantized, high-mobility two-dimensional electron gas (2DEG) provides a uniquely clean and tunable solid-state system in which to explore coherent many-electron dynamics. A superposition of massively degenerate Landau levels (LLs) can be created by a coherent terahertz (THz) pulse through cyclotron resonance (CR) absorption [27], as shown in Figure 3.1. How rapidly the coherence of this many-body superposition state decays has not been well understood. Even though the CR frequency, $\omega_c$, is immune to many-body interactions due to Kohn’s theorem [28], the decoherence of CR can be affected by electron-electron interactions.

Theoretical studies predicted that the linewidth of CR should oscillate with the LL filling factor since the screening capability (i.e., the density of states at the Fermi energy) of a 2DEG oscillates with the filling factor [3, 29–33]. However, despite several decades of experimental studies of CR in 2DEGs using continuous-wave and incoherent methods [34–41], no clear evidence for the predicted CR linewidth os-
Figure 3.1: Coherent THz pulse creates a superposition of adjacent Landau levels with massive degeneracy. In the figure, the Landau level spacing equals $\hbar \omega_{\text{CR}}$, where $\omega_{\text{CR}}$ is the cyclotron frequency. The free induction decay of such a superposition state can be observed after the excitation pulse.

cancellations has been obtained for high-mobility, high-density samples, partly due to the ‘saturation effect’; i.e., when the conductivity is high, the 2DEG behaves as a metallic mirror, reflecting most of the incident light at the CR peak, resulting in an undesirable broadening of transmittance linewidths [42–44].

In this chapter, we describe results of our systematic study of CR decoherence in high-mobility 2DEGs by using time-domain THz magnetospectroscopy, measuring the CR decay time, $\tau_{\text{CR}}$, as a function of temperature ($T$), magnetic field ($B$), electron density ($n_e$), and mobility ($\mu_e$). As $T$ decreases, $\tau_{\text{CR}}$ increases due to reduced electron-phonon interaction, but $\tau_{\text{CR}}$ eventually saturates at low $T$. The low-$T$ saturation
value of $\tau_{CR}$ is uncorrelated with $\mu_e$; rather, the CR decay rate $\Gamma_{CR} \equiv \tau_{CR}^{-1}$ increases linearly with $n_e$. We developed a fully quantum mechanical theory for describing coherent CR, which clearly identifies superradiant (SR) damping [45, 46] to be the dominant decay mechanism. Namely, $\Gamma_{CR}$ is dominated by cooperative radiative decay at low $T$, which is much faster than any other phase-breaking scattering processes.

### 3.2 Experiments and results

We studied two samples of modulation-doped GaAs quantum wells grown by molecular beam epitaxy. Sample 1 had $n_e$ and $\mu_e$ of $1.9 \times 10^{11}$ cm$^{-2}$ and $2.2 \times 10^6$ cm$^2$/Vs, respectively, in the dark, while after illumination at 4 K they changed to $3.1 \times 10^{11}$ cm$^{-2}$ and $3.9 \times 10^6$ cm$^2$/Vs; intermediate $n_e$ values were achieved by careful control of illumination times. Sample 2 had $n_e = 5 \times 10^{10}$ cm$^{-2}$ and $\mu_e = 4.4 \times 10^6$ cm$^2$/Vs.

Time-domain THz magnetospectroscopy experiments were performed with setup shown in Figure 2.1. The incident beam was linearly polarized by the first polarizer, and by rotating the second polarizer, the transmitted THz field was measured in both $x$- and $y$-directions [Fig. 3.2(a)]. Figure 3.2(b) shows transmitted THz waveforms in the time domain. Each blue dot represents the tip of the THz electric field, $E = (E_x, E_y)$, at a given time. The red traces are the projections of the waveforms onto the $E_x$-$t$ plane and $E_x$-$E_y$ plane. The top and middle traces show the transmitted THz waveforms at 0 T and 2.5 T, respectively. The 2.5 T trace contains long-lived oscillations with circular polarization. The bottom trace is the difference between the two, $E_{0T}(t) - E_{2.5T}(t)$, which is proportional to the THz-induced current at the CR frequency of the 2DEG [see Eq. (3.7)]. Hence, its decay time $\tau_{CR}$ can be directly and accurately determined through fitting with $A \exp(-t/\tau_{CR}) \cdot \sin(\omega_c t + \phi_0)$, where $A$
Figure 3.2: (a) A schematic of the polarization-resolved THz magneto-transmission experiment in the Faraday geometry. (b) Coherent cyclotron resonance oscillations in the time domain. Each blue dot represents the tip of the THz electric field at a given time. The red traces are the projections of the waveforms onto the $E_x$-$t$ and $E_x$-$E_y$ planes. The bottom trace is the difference between the top (0 T) and middle (2.5 T) traces.
Figure 3.3: (a) Magnetic field dependence of CR oscillations, showing peaks (blue) and valleys (red). (b) The frequency-domain version of (a). Black dashed line: linear fit with a cyclotron mass of $0.069m_e$. (c) Magnetic field dependence of $\tau_{\text{CR}}$ at 3 K. (d) Temperature dependence of $\tau_{\text{CR}}$ at 2.5 T. All the data are for Sample 1.

and $\phi_0$ are the CR amplitude and the initial phase, respectively.

Figure 3.3(a) shows CR oscillations at various $B$ for Sample 1 after illumination. The inter-LL spacing, or $\hbar\omega_c$, increases with $B$. Figure 3.3(b) shows the Fourier transform of the time-domain data in Fig. 3.3(a) into the frequency domain. A linear $B$ dependence of $\omega_c = eB/m^*$ provides electron cyclotron mass $m^* = 0.069m_e$, where $m_e = 9.11 \times 10^{-31}$ kg. As shown in Fig. 3.3(c), the variance of $\tau_{\text{CR}}$ with $B$ is small;
\( \tau_{\text{CR}} \) slightly decreases with increasing \( B \), but no oscillatory behavior is observed. Figure 3.3(d) shows that \( \tau_{\text{CR}} \) increases with decreasing \( T \) but saturates at \( \sim 9.5 \) ps when \( T \lesssim 10 \) K.

The values of \( \tau_{\text{CR}} \) at low \( T \) were much shorter than the DC scattering time, \( \tau_{\text{DC}} = m^* \mu_e/e \), of the same samples. Furthermore, there was no correlation between \( \tau_{\text{CR}} \) and \( \tau_{\text{DC}} \); in some cases, higher-mobility samples revealed shorter \( \tau_{\text{CR}} \) values. On the other hand, \( \tau_{\text{CR}} \) showed strong correlation with \( n_e \). As \( n_e \) was increased, \( \tau_{\text{CR}} \) was found to decrease in a clear and reproducible manner, as shown in Figs. 3.4(a) and 3.4(b). The low-density sample (Sample 2) exhibited the longest \( \tau_{\text{CR}} \) value of \( \sim 40 \) ps. Figure 3.4(c) shows that the decay rate, \( \Gamma_{\text{CR}} \), increases linearly with \( n_e \), which, as described below, is consistent with SR damping of CR.

A qualitative picture is as follows. A coherent incident THz pulse induces a polarization in the 2DEG, i.e., macroscopic coherence as a result of individual cyclotron dipoles oscillating in phase. The resulting free induction decay of polarization occurs in a SR manner, much faster than the dephasing of single oscillators. The SR decay rate, \( \Gamma_{\text{SR}} \), is roughly \( N \) times higher than the individual radiative decay rate, where \( N \sim n_e \lambda^2 \) is the number of electrons within the transverse coherence area of the incident THz wave with wavelength \( \lambda \). In an ultraclean 2DEG, \( \Gamma_{\text{SR}} \) is higher than the rates of all other phase-breaking scattering mechanisms. This scenario explains not only the \( n_e \) dependence of \( \tau_{\text{CR}} \) but also its weak \( B \) dependence as well as the saturation of \( \tau_{\text{CR}} \) at low \( T \).

Furthermore, the SR nature of CR emission not only dramatically speeds up the radiative decay but also makes CR radiation more directional and collinear with the excitation pulse. Thus, most of the CR radiation could be collected, allowing us to analyze the incident and radiated THz waves quantitatively. At the 2DEG, shown in
Figure 3.4: (a) Low-density sample (Sample 2) exhibiting the longest $\tau_{CR}$ of $\sim 40$ ps. (b) CR oscillations in Sample 1 with different densities by controlling the illumination time. (c) Decay rate as a function of density. Blue solid circle: Sample 2. Red solid circles: Sample 1. The blue dashed line represents Eq. (3.11) with no adjustable parameter, where $n_{GaAs} = 3.6$ and $m^* = 0.069m_e$ are used. (d) Incident $E_i$, reflected $E_r$ and transmitted $E_t$ THz pulses at the 2DEG. (e) The decay of the THz-pulse-excited energy in the 2DEG. The red dashed line is an exponential fit. About 80% of the energy relaxes through CR superradiance.
Fig. 3.4(d), the incident \((E_i)\), reflected \((E_r)\), and transmitted \((E_t)\) THz fields satisfy the boundary condition, \(E_i(t) + E_r(t) = E_t(t)\). With the full knowledge of \(E_t(t)\) at 0 T and 2.5 T as well as \(\sigma_{\text{cr}}(\omega)\), the optical conductivity of the 2DEG at 0 T, we obtained both \(E_i(t)\) and \(E_r(t)\) at 2.5 T. The THz-induced energy increase in the 2DEG, \(\Delta \varepsilon(t)\), is proportional to \(\int_0^t (n_{\text{GaAs}} |E_i(t')|^2 - n_{\text{GaAs}} |E_r(t')|^2 - |E_t(t')|^2) \, dt'\), shown in Fig. 3.4(e). If the energy is dissipated nonradiatively, i.e., via scattering, \(\Delta \varepsilon(t)\) would be a step function, as indicated by the black dotted line in Fig. 3.4(e). However, our data instead show that most of the absorbed energy goes back into the field, again supporting the SR picture. By fitting \(\Delta \varepsilon(t)\) with an exponential with a baseline, we found that the majority (~80%) of the energy decays radiatively; the other 20% could be due to imperfect collection and any residual scattering loss.

### 3.3 Theoretical analysis

Here, we present a quantum mechanical model for THz excitation and coherent CR emission of a 2DEG in a perpendicular \(B\), valid for an excitation pulse of an arbitrary duration with respect to \(\Gamma_{\text{cr}}^{-1}\) and \(\omega_c^{-1}\). We start from the master equation for the density operator in the coordinate representation, \(d\hat{\rho}/dt = -(i/\hbar)[\hat{H}, \hat{\rho}] + \hat{R}(\hat{\rho})\), where \(\hat{R}(\hat{\rho})\) is the relaxation operator. The Hamiltonian for an electron of mass \(m^*\) in a confining potential \(U(\mathbf{r})\) interacting with an optical and magnetic field described by the vector potential \(A = A_{\text{opt}} + A_B\) is

\[
\hat{H} = \frac{\hat{\mathbf{p}}^2}{2m^*} + U(\mathbf{r}) - \frac{e}{2m^*c} (\mathbf{A} \hat{\mathbf{p}} + \hat{\mathbf{p}} \mathbf{A}) + \frac{e^2}{2m^*} \mathbf{A}^2, \tag{3.1}
\]

where \(\mathbf{p} = -i\hbar \nabla\). In our case, the energy of the first-excited quantum-well subband is much higher than all energy scales relevant to the problem, and so we can assume that the electrons stay in the ground subband. Furthermore, in our case of relatively
modest $B$ and low-energy excitations, we can neglect any band nonparabolicity, and thus, the resulting LLs are equally spaced.

Care should be exercised in choosing the correct form of $\hat{R}(\hat{\rho})$. A standard empirical expression for the relaxation of the off-diagonal elements of the density matrix, $R_{mn} = -\gamma_{mn} \rho_{mn}$, can be used only within the rotating wave approximation (RWA) and under the assumption that the relaxation rate, $\gamma$, is much smaller than eigenfrequencies of the system. Neither of these approximations is valid in our case of an ultrashort excitation pulse and frequencies in the (sub)THz range. As was shown in Refs. [47] and [48], outside the RWA the standard relaxation term leads to spurious terms in the equations for quantum-mechanical averages of the dipole moment and populations, including violation of a standard relationship, $J = d$, between quantum mechanical averages of the current density ($J$) and dipole moment ($d$).

Following Ref. [47], we choose the relaxation operator in the coordinate representation and for $A = 0$ as

$$\hat{R} = -\gamma_\perp (\hat{\rho}_\perp - \hat{\rho}_\perp^*) - \gamma_\parallel (\hat{\rho}_\parallel - \hat{\rho}_\parallel,0), \quad (3.2)$$

where $\hat{\rho}_\perp, \parallel$ are the off-diagonal and diagonal components of the density operator, respectively, with corresponding transverse ($\gamma_\perp$) and longitudinal ($\gamma_\parallel$) relaxation rates, and $\hat{\rho}_\parallel,0$ is an equilibrium distribution of populations. For $A \neq 0$, Eq. (3.2) has to be transformed to preserve gauge invariance as specified in Ref. [47]. Using the master equation and Eq. (3.1), we can derive a set of equations for the quantum mechanical
averages of \( j, d \), and energy density \((W)\) of the system:

\[
\ddot{d} + 2\gamma_\perp \dot{d} + \frac{e}{m} \nabla U \bar{\rho} + \omega_c \mathbf{b} \times \dot{\mathbf{d}} = \frac{e^2}{4\pi\varepsilon_0 m} \bar{\rho} \mathbf{E}(t),
\]

(3.3)

\[
\dot{\mathbf{j}} = \mathbf{d},
\]

(3.4)

\[
\dot{W} + \gamma_\parallel (W - W_0) = \frac{\dot{\mathbf{d}} \mathbf{E}(t)}{4\pi\varepsilon_0},
\]

(3.5)

where \( \mathbf{d} = -e\mathbf{r}\bar{\rho}, \mathbf{j} = -(e/m)(\mathbf{p} + e\mathbf{A})\bar{\rho}, \mathbf{W} = \mathbf{H} \bar{\rho}, W_0 \) is an equilibrium energy density, \( \mathbf{b} \) is a unit vector along \( \mathbf{B} \parallel \mathbf{z} \), and \( \mathbf{E} \) is the THz electric field. The overbar means taking the trace with the density matrix, i.e., \( \Bar{g}\rho = \int g(\mathbf{r})\rho(\mathbf{r}, \mathbf{r}')\delta(\mathbf{r} - \mathbf{r}')\,d^3r\,d^3r' \) [49].

Since all electrons are in the ground subband, i.e., effectively 2D with no confinement potential transverse to \( \mathbf{B} \), we can drop the term containing \( \nabla U \). We also assume that the excitation pulse is too weak to perturb populations. Then from Eqs. (3)-(5) we can obtain the following equation for the circularly polarized current \( j_+ = j_x - ij_y \):

\[
\frac{dj_+}{dt} + (i\omega_c + 2\gamma_\perp)j_+ = \alpha E_+(t),
\]

(3.6)

where \( E_+ = E_x - iE_y \), \( \alpha = \omega_p^2/4\pi \), and \( \omega_p = \sqrt{e^2\bar{\rho}/\varepsilon_0 m^*} \) is the plasma frequency.

The electric field acting on the current in Eq. (3.6) consists of the excitation pulse \( \mathbf{E}_0 = (E_{0x}(t), 0, 0) \) and the field radiated by the current, \( \mathbf{e} = (e_x, e_y, 0) \). Note that \( \bar{\rho} = n_e/L \) is the electron volume density, where \( L \) is the thickness of the 2DEG layer. There is an interesting characteristic feature of an electron system with a parabolic band, i.e., equally spaced LLs: the current and its radiation are determined by the total \( n_e \) and independent of how the electrons are distributed among the LLs. Therefore, our results remain valid even at room temperature and low \( B \).

From the boundary conditions on both sides of the 2DEG, i.e., the continuity of the electric field and the jump in the magnetic field \((b_x, b_y, 0)\) radiated by the current, \( b_+(z = +0) - b_+(z = -0) = \mu_0 j_+ L \), together with Maxwell’s equations relating \( \mathbf{e} \) and
b fields in the outgoing radiation, we can obtain the radiation field on the surface of the 2DEG,
\[ e_+ (1 + n_{GaAs}) = -\mu_0 j_+ L, \] (3.7)
where \( e_+ = e_x - ie_y \), \( n_{GaAs} = 3.6 \) is the refractive index of GaAs substrate. This gives the final equation for the current:
\[ \frac{dj_+}{dt} + (i\omega_c + \Gamma_{CR}) j_+ = \alpha E_{0x}(t), \] (3.8)
where the CR decay rate, \( \Gamma_{CR} \), now includes the collective radiative contribution proportional to \( n_e \):
\[ \Gamma_{CR} = \Gamma_{scatt} + \Gamma_{SR}, \] (3.9)
where
\[ \Gamma_{scatt} = 2\gamma_\perp, \] (3.10)
\[ \Gamma_{SR} = \frac{\omega_p^2 L}{(1 + n_{GaAs})c} = \frac{e^2 n_e}{m^*(1 + n_{GaAs})\epsilon_0 c}. \] (3.11)

As shown by the dashed line in Fig. 3.4(c), Eq. (3.11) reproduces the observed linear \( n_e \) dependence of \( \Gamma_{CR} \) without any adjustable parameter, strongly supporting the notion that SR damping dominates the CR decay process in these high-\( \mu_e \) samples.

For low-\( n_e \) and low-\( \mu_e \) samples, \( \Gamma_{scatt} \) is not negligible compared to \( \Gamma_{SR} \), and thus, the values of \( \Gamma_{CR} \) are expected to deviate from \( \Gamma_{SR} \), as seen in Fig. 3.4(c). For rough estimation, one can assume that \( \Gamma_{scatt} \approx \Gamma_{DC} = \tau_{DC}^{-1} = e/m^*\mu_e \) in Eq. (3.9). For example, the value of \( \tau_{CR} \) (\( = \Gamma_{CR}^{-1} \)) estimated in this manner for Sample 2 is \( \sim 44 \) ps, which agrees well with the measured value (40±10 ps). Figure 3.5(a) plots \( \Gamma_{CR} - \Gamma_{DC} \) versus \( \Gamma_{SR} \) for four representative data points in the present study as well as data previously reported for 2DEGs with different values of \( n_e \) and \( \mu_e \) [1, 2]. The linear
Figure 3.5: (a) The measured values of $\Gamma_{\text{CR}} - \Gamma_{\text{DC}}$ versus $\Gamma_{\text{SR}}$ given by Eq. (3.11) for four representative data points in the present study and values from Refs. [1,2]. The solid line has a slope of 1. (b) $\Gamma_{\text{scatt.}} \equiv \Gamma_{\text{CR}} - \Gamma_{\text{SR}}$ as a function of $\sqrt{B}$. Red solid line: linear fit. Blue dashed line: prediction based on short-range scattering [3].

relationship with a slope of 1 seen in this plot proves the validity of the following convenient formula

$$\tau_{\text{CR}} \simeq \frac{m^*}{e} \left[ \frac{e n_e}{(1 + n_{\text{GaAs}})\epsilon_0 c} + \frac{1}{\mu_e} \right]^{-1} \quad (3.12)$$

for estimating $\tau_{\text{CR}}$ from the knowledge of $n_e$ and $\mu_e$.

Finally, Eq. (3.9) allows us to determine $\Gamma_{\text{scatt.}}$ as $\Gamma_{\text{CR}} - \Gamma_{\text{SR}}$. In particular, we interpret the small but non-negligible $B$-dependence of $\Gamma_{\text{CR}}$ shown in Fig. 3.2(c) to be the $B$-dependence of $\Gamma_{\text{scatt.}}$. Figure 3.5(b) shows $\Gamma_{\text{CR}} - \Gamma_{\text{SR}}$ versus $\sqrt{B}$, which exhibits an approximately linear relationship, consistent with theoretical predictions based on short-range scattering [3,29].
3.4 Conclusion

We studied the decay dynamics of Landau-quantized 2DEGs coherently and resonantly excited by THz pulses. We found that the decay rate of coherent cyclotron oscillations increases linearly with electron density, which we interpret as evidence of superradiance. Our detailed quantum mechanical calculations confirmed this interpretation, reproducing our experimental observation quantitatively without any adjustable parameter. Overall, this study demonstrates the cooperative nature of decay dynamics of a quantum-degenerate, interacting electron system, even though its resonant frequency is independent of many-body interactions.
Chapter 4

Strong Coupling of Collective Electron Cyclotron Resonance and THz Cavity Photons

4.1 Introduction

Light-matter interaction is one of the central topics in the modern condensed matter physics research, with great importance in both fundamental science and device applications. Matter and photons inside a small cavity can behave completely differently from those in free space and reflect the quantum nature of light and even the vacuum. The study of interactions between discrete matter transitions and cavity photons has come to be known as cavity quantum electrodynamics (QED) [50].

Figure 4.1 schematically shows a two-level matter system coupled with a cavity, where $g$ represents the coupling strength, while $\kappa$ and $\gamma$ are the cavity and matter decay rates, respectively.

![Figure 4.1: Schematic diagram for a two-level matter system coupled with a cavity, where $g$ represents the coupling strength between the two-level system and the cavity field, while $\kappa$ and $\gamma$ are the cavity and matter decay rates, respectively.]

- Weak coupling: $2g < \kappa, \gamma$
- Strong coupling: $2g > \kappa, \gamma$
- Ultrastrong coupling: $g \geq \omega_0$
decay rates, respectively. Depending on the magnitude of the coupling strength $g$, system losses ($\kappa$ and $\gamma$), and the resonance frequency $\omega_0$, light-matter interaction in a cavity can be divided into three regimes in general. They are the weak-coupling regime ($2g < \kappa, \gamma$), the strong-coupling regime ($2g > \kappa, \gamma$), and the ultrastrong-coupling regime ($g \gtrsim \omega_0$). In the weak-coupling regime, light-matter interaction can be treated as a perturbation, but the spontaneous emission rate can still be modified by the presence of the cavity (the Purcell effect) [51]; absorption or emission of light is irreversible between the matter and the cavity in this regime. The strong-coupling regime exhibits much more interesting features, where the Rabi splitting can be spectrally resolved. In the time domain, it ensures that at least one cycle of Rabi oscillations can happen before either matter excitations lose their coherence or photons leak out of the cavity. There is repetitive energy transfer between the matter and cavity, which leads to reversible absorption and emission. Nonperturbative treatments of light-matter interaction are required in order to describe the dressed states. When the Rabi splitting is comparable with the resonance frequency, the system is known to be in the ultrastrong coupling regime. In this regime, the rotating wave approximation breaks down, namely, the anti-resonance terms in the Rabi model cannot be neglected anymore, which results in a measurable shift (known as the Bloch-Siegert shift [52]) in polariton frequencies, as well as a possible quantum phase transition [53–55].

Light-matter interaction can be dramatically enhanced by coherent coupling of $N$ atoms with the same light field, as pointed out in Dicke’s celebrated paper on superradiance in 1954 [45]. Since then, many-oscillators systems have attracted an enormous mount of interest. In such systems, single photon excitation is shared by $N$ atoms, and the Rabi splitting is enhanced by $\sqrt{N}$ times the value of an individual atom. It is
known as the collective Rabi splitting. In addition to enhanced light-matter coupling, even more fascinating physics can emerge when the collective Rabi splitting exceeds the resonance frequency. Theory predicts that an increasing coupling strength can eventually drive a quantum phase transition, known as the Dicke superradiant phase transition (SPT) [53–55]. During the SPT, the ground state becomes a light-dressed polariton state (or a superradiant state), instead of the normal vacuum-matter ground state, since it is lower in energy, which leads to such intriguing notion as ‘unstable vacuum’ and ‘spontaneous photon generation’. However, if the Hamiltonian of the light-matter system includes a quadratic term of field vector potential (the so-called $A^2$ term, or the diamagnetic term), which is the case in most of atomic and solid state systems, SPT will be suppressed [56, 57]. Light and matter resonances will be effectively decoupled even with very strong coupling [58]. Although under debate [59,60], graphene, on the other hand, may be free from the $A^2$ term, thanks to its unique linear energy dispersion, and is a candidate for realizing a genuine SPT in a solid.

In practise, on the cavity side, it is easier to achieve larger $g/\omega_0$ ratios by reducing the cavity frequency. Furthermore, a longer wavelength provides a greater coherent area, which also helps to establish collective behaviors involving more individual oscillators. In this sense, strong light-matter interaction are more readily studied with THz and microwave cavities than with their visible and near-infrared (NIR) counterparts. THz cavity-QED is particularly interesting because the characteristic energies of many collective excitations in condensed matter systems are on the order of 1 meV, which is located in the THz frequency range. Interactions between correlated excitations with THz cavity photons remain largely unexplored, not to mention their dynamics.

On the matter side, collective Rabi splitting has been demonstrated in atomic
systems, as well as in superconducting qubits, and, most recently in ferromagnetic systems in the microwave range [11, 12]. Achieving collective Rabi splitting in semiconductors have also attracted tremendous interests, since semiconductor quantum structures have superior purity and excellent tunability among all solid states systems. In quantum well embedded micro-cavities, vacuum Rabi splitting [61], and exciton-polariton condensation [7] have been observed. However, the vacuum Rabi splitting observed in such systems is not a fully collective effect, and no $\sqrt{N}$ dependence has been reported. Intersubband transitions in GaAs quantum wells have also been studied for ultrastrong coupling physics [62–66], and an ultrafast switch-on of strong coupling has been demonstrated [64]. However, an intersubband transition requires an out-of-plane light field due to the selection rule, which leads to inconvenient sample structure and experimental geometries. In addition, the fast intersubband relaxation limits the lifetime of the coupled modes. An experimental breakthrough came from strongly coupled electron cyclotron resonance (CR) with a near-field metamaterial resonator array in a GaAs 2DEG. Owing to the small mode volume and large dipole moment, a record high ratio of $g/\omega_0 = 0.87$ has been reported [67,68], which suggests that CR in high-mobility 2DEG is a promising system for achieve ultrastrong light-matter coupling in the THz range. However, due to the lossy metamaterial resonator and the superradiant decay of CR, the lifetime of magneto-polaritons was quite short, which limits further studies and device applications. Alternatively, photonic crystal cavities (PCCs) are able to provide ultralong cavity lifetimes [18], which are much longer than a state-of-the-art superconducting metamaterial resonator [69]. With a reasonable mode volume, PCCs are promising platforms for THz cavity QED studies. Another motivation for coupling CR with a high-$Q$ THz cavity is to suppress the superradiance decay, and achieve a long CR decoherence time.
In this chapter, we describe our observation of strong coupling between coherent cyclotron resonance and photons in 1D THz PCCs, with a cooperativity (defined as \(4g^2/\kappa\gamma\)) up to 360. We directly observed vacuum Rabi oscillations in the time domain. We further observed a \(\sqrt{N}\) dependence of vacuum Rabi splitting with respect to the 2DEG carrier density, which is a signature of collective Rabi splitting. A ratio of \(g/\omega_0 = 0.12\) was achieved with just a single quantum well and moderate electron density. Furthermore, superradiance decay of CR in high-mobility 2DEGs was significantly suppressed by the presence of a high-\(Q\) THz cavity. As a result, we observed ultra-narrow magneto-polariton resonances and an intrinsic CR linewidth as small as 5.6 GHz. Our observations can be well described by both quantum mechanical and classical theories. This work sets a landmark towards promising quantum optics and cavity-QED applications with CR in semiconductors, including the possible realization of a superradiance phase transition with coherent CR in graphene. Our method also applies to various strongly correlated systems with collective many-body excitations in the THz range. It opens a door to the fascinating physics of THz many-body cavity QED.

4.2 Experiments and results

Two GaAs quantum well samples containing high mobility 2DEGs were used in this study. Sample A has a density of \(3 \times 10^{11}\) cm\(^{-2}\) and a mobility of \(4 \times 10^6\) cm\(^2\)/Vs. Sample B had a lower density (\(\sim 5 \times 10^{10}\) cm\(^{-2}\) at 4 K), and a mobility of \(4 \times 10^6\) cm\(^2\)/Vs. As schematically illustrated in Figures 4.2(a) and (b), our 1D THz PCC was constructed by stacking thin silicon wafers (50 \(\mu m\) for Bragg mirrors, and 100 \(\mu m\) for the central defect layer) and copper spacers (200 \(\mu m\)) designed for a fundamental cavity mode around 0.4 THz. The 2DEG sample, which was 4.5 \(\mu m\) thick after
Figure 4.2: 1D THz photonic crystal cavity (PCC) with a high-mobility 2DEG (Cavity 1). (a) Schematic diagram for CR involving two adjacent Landau levels coupled with a THz cavity field. (b) 1D THz PCC structure. Two silicon layers are placed on each side of the center defect layer. The blue part is the transferred 2DEG thin film. (c), (d), and (e), calculated electric field amplitude distribution inside the cavity for the 1st, 2nd, and 3rd cavity modes, respectively. 2DEG is located at the field maximum for all three cavity modes. (f) Experimental power transmittance for Cavity 1. Three sharp cavity modes are clearly resolved in the middle of each stopband. Zoom-in spectra for the three modes are shown in (g), (h), and (i), together with Lorentzian fitting. The FWHM is 2.6 GHz, 5 GHz, and 3.8 GHz for the 1st, 2nd, and 3rd mode, respectively.
substrate removal, was placed on the central silicon piece, where the electric field
had its maximum, with two layers of Si on each side as a Bragg mirror. Thanks to
the large contrast of refractive index between silicon and vacuum, only a few layers
of silicon were required to achieve sufficient cavity confinement of THz wave with
high $Q$ values, in contrast to Bragg cavities for near-infrared waves, which would
require a large number of layers to achieve similar $Q$ values. The calculated electrical
field distribution inside the cavity is shown in Figures 4.2(c), (d), and (e), for the
fundamental, second, and third modes, respectively. The spatial overlap of the 2DEG
and electrical field maximum ensured the strongest light-matter coupling. Two types
of cavities were employed in this study. In Cavity 1, we used a 100-$\mu$m-thick silicon
wafer as the central defect layer, while, in Cavity 2, a 100 $\mu$m thick sapphire wafer
was used. Most of the experimental data shown in this chapter are from Cavity 1.

Figure 4.2(f) shows a broadband THz transmission spectrum of the coupled cavity-
2DEG structure at 4 K. Three photonic bandgaps can be found as transmission stop-
bands. At the center of each stop-band, a sharp cavity modes is seen. As shown
in Figures 4.2(g), (h), and (i), the full-width-at-half-maximum (FWHM) of the first,
second, and third cavity modes was found to be 2.7 GHz, 5 GHz, and 3.8 GHz, at
0.407 THz, 1.218 THz, and 2.020 THz, respectively. The linewidth would have been
1 GHz for each mode in the case of a perfectly aligned cavity. The quality factor,
$Q$, for these three modes was 150, 243, and 532, respectively. These values are much
higher than what has been reported for metamaterial resonators [67,68]. A 500-ps-
long time window used in the experiments ensured that the measured linewidths in
Cavity 1 were not instrument limited.

By varying the magnetic field, we were able to continuously change the detunings
between CR and the cavity modes. A anticrossing behavior expected for strong
light-matter coupling was clearly resolved, as shown in Figure 4.3. Two ultranarrow magneto-polariton branches were formed through the hybridization of CR and the cavity mode. For the fundamental mode, zero detuning happened when the magnetic field strength was at 1.00 T, while the FWHM for both the upper and lower polariton modes is 4.2 GHz, and the cavity mode was 2.7 GHz. The vacuum Rabi splitting \((2g)\) was \(74 \pm 1\) GHz, yielding \(2g/(\kappa/2 + \gamma/2) = 17\), which corresponds to 360 in cooperativity, defined as \(4g^2/\kappa\gamma\). It unambiguously shows strong coupling between electronic CR and THz cavity photons. Unlike the two magneto-polariton modes, a cavity mode remains almost unchanged with magnetic field, since it originates from the transmission of the CR inactive circular polarization component in the linearly polarized THz probe.

Strong coupling between CR and the second and third cavity modes is shown in Figures 4.4(a) and (b). The vacuum Rabi splittings of the second and third mode were \(66 \pm 1\) GHz and \(60 \pm 1\) GHz, respectively. These values are slightly smaller than the vacuum Rabi splitting observed for the fundamental mode, while the polariton linewidth is larger, especially for the 3rd mode, likely due to unperfect surface contact between the 2DEG and the central silicon wafer. The higher-order modes are much more susceptible to imperfections due to their shorter wavelengths; hence, we believe that the polariton modes are inhomogeneously broadened in the second and third cavity modes. The peak positions for magneto-polariton modes are presented in Figure 4.4(c), which will be discussed in more detail using quantum mechanical and classical models in Section 3.3 and 3.4, respectively. Cavity 2 shows a larger vacuum Rabi splitting of 90 GHz for both the fundamental and second cavity modes, due to a better 2DEG-sapphire interface. However, the linewidths of the cavity modes and polaritons in Cavity 2 data were instrument-limited because of a 80 ps time window.
Figure 4.3: Anticrossing of CR and the first cavity mode in Cavity 1. The residual cavity mode results from the CR inactive component of the linearly polarized THz beam. Transmission spectra at different magnetic fields are vertically offset for clarity. The magnetic field is increased from 0.4 T (bottom) to 1.4 T (top).
Figure 4.4: Anticrossing of CR with (a) the second cavity mode and (b) the third cavity mode in Cavity 1. In (a), the magnetic field is increased from 2.4 T (bottom) to 3.4 T (top). In (b), the magnetic field is increased from 4.6 T (bottom) to 5.2 T (top). (c) Summary of measured peak positions of the magneto-polariton branches for the three cavity modes, the value $\nu$ indicate the filling factor at the resonances.
Figure 4.5: Anticrossing of CR with (a) the fundamental cavity mode and (b) the second cavity mode in Cavity 2. All traces are vertically offset for clarity. In (a), the magnetic field is increased from 0.5 T (bottom) to 1.35 T (top) with a step size of 0.05 T. In (b), the magnetic field is increased from 2.1 T (bottom) to 3.2 T (top) with a step size of 0.05 T. The linewidth of both cavity modes and polaritons is limited by the 80 ps time window. (c) Peak positions for the magneto-polariton branches for the first (blue) and second (red) cavity modes.

In analogy to the physics of many-atom light-matter interactions, one crucial question to be answered is whether the strong coupling observed here is a fully coherent behavior among a large number of individual electrons in the 2DEG. Figure 4.6 shows three cases of magneto-polariton manifestation with different electron densities, when CR is in resonance with the fundamental cavity mode. At zero detuning, the vacuum Rabi splitting \(2g\) equals the splitting between the upper and lower polariton modes.
Figure 4.6: (a) Vacuum Rabi splittings observed for 2DEGs with three different electron densities in Cavity 1. CR is resonant with the fundamental mode. (b) Square root of $n_e$ dependence evidencing the collective nature of vacuum Rabi splitting.

which exhibits a square root dependence on the electron density $n_e$, which is strong evidence for collective Rabi splitting. This observation proves that the billions of electrons interact with the same cavity field in a fully coherent manner. By extrapolation, the vacuum Rabi splitting for CR of a single electron is 0.14 MHz. By coupling with multiple layers of a 2DEG with a higher density, the vacuum Rabi splitting can be further increased.

In the strong-coupling regime, the most pronounced feature of light-matter interaction is the coherent repetitive energy transfer between the matter resonance and the cavity photons as observed in atomic [70], semiconductor exciton-polariton [71, 72], and spin ensemble systems [11]. Because THz-TMS is a time-domain technique, we are naturally able to see vacuum Rabi oscillations directly in the time domain. Experimentally, to minimize the background and only probe the circularly-polarized
magneto-polaritons, we measured the \( y \)-polarization component, \( E_y \), of the transmitted THz wave, while the incident THz wave was fully \( x \)-polarized. We measured \( E_y \) in positive (+\( B \)) and negative (−\( B \)) fields, and took the difference \( \Delta E_y = E_y(+B) - E_y(-B) \), to further eliminate the background. With this method, \( \Delta E_y \) only comes from the circularly-polarized magneto-polaritons and the CR inactive mode with the counter-circular polarization. As such, the measured \( \Delta E_y \) signal shows strong beating between the two polariton modes and CR inactive modes, as shown in Figure 4.7 top trace. By numerically filtering out the center cavity mode, clear beating between the two magneto-polaritons alone can be resolved, as shown in the bottom trace in Figure 4.7. As indicated by the arrows, at each beating node energy is stored in the 2DEG CR. The average time separation between two adjacent beating nodes was about 13-15 ps, matching the \( 2g \) splitting in the frequency domain, as shown in Figure 4.7(b). The beating lives up to dozens of picoseconds, indicating a long intrinsic CR decoherence time.

The decay of CR in free space is dominated by collective radiative decay, or superradiance, in high-mobility 2DEGs, as discussed in Chapter 3. However, in a strongly coupled cavity-2DEG system, the emitted CR radiation cannot escape from the high-
\( Q \) cavity and thus re-excites coherent CR. Hence, the reversible emission and absorption in a strongly coupled cavity-2DEG system strongly suppresses the superradiant decay, revealing the intrinsic CR decoherence time. Figure 4.8(a) presents the temperature dependence of transmission spectra of the ultranarrow magneto-polariton peaks at zero detuning for the first cavity mode in Cavity 1. The linewidth of magneto-
polariton modes significantly increases above 20 K. Polaritons totally vanish at 80 K. The CR inactive cavity mode, on the other hand, remains almost unchanged as the temperature increases, serving as an excellent linewidth reference. At zero detun-
Figure 4.7: (a) Vacuum Rabi oscillations in the time domain. Cyclotron resonance is resonantly coupled with the second cavity mode at 2.975 T in Cavity 1. $\Delta E_y = E_y(+2.975 \ T) - E_y(-2.975 \ T)$ is the measured difference between the transmitted THz waveforms taken at +2.975 T and -2.975 T in the $y$-polarization direction, as shown by the top black trace. More clear vacuum Rabi oscillations can be observed by removing the residual cavity mode due to the CR inactive THz component with a numerical notch filter (bottom blue trace). The beating nodes of two magneto-polaritons are indicated by arrows. (b) Frequency-domain spectrums for $\Delta E_y$ before and after the removal of residual cavity mode.
ing, the linewidths of the lower (LMP) and upper (UMP) magneto-polaritons are the same, \( \gamma_{\text{LMP}} = \gamma_{\text{UMP}} = (\kappa + \gamma_{\text{CR}})/2 \), where \( \gamma_{\text{CR}} \) is the CR decay rate. With Lorentzian fitting, \( \gamma_{\text{CR}} \) can be obtained through \( \gamma_{\text{CR}} = \gamma_{\text{LMP}} + \gamma_{\text{UMP}} - \kappa \), which is the average between LMP and UMP. As shown in Figure 4.8(b), the CR decay time \( \tau_{\text{CR}} \left(1/\gamma_{\text{CR}}\right) \) in the cavity is \( 57 \pm 4 \text{ ps} \) at 2 K, which is much longer than the superradiance-limited decay time \( \tau_{\text{CR}} \) with a value of 10 ps in free space. The dramatic enhancement of CR decay time is totally opposite to the Purcell effect in a high-\( Q \) cavity expected in the weak-coupling regime, and can only be understood within the framework of strong coupling, where superradiant decay is suppressed by the reversible absorption and emission. Therefore, the \( \tau_{\text{CR}} \) measured in our high-\( Q \) is the intrinsic CR decay time due to scattering. Above 20 K, \( \tau_{\text{CR}} \) is approaching \( \tau_{\text{DC}} \), where piezoelectric scattering and polar optical phonon scattering dominate. At 2 K, the intrinsic CR decay time is still lower than the DC momentum scattering time \( \tau_{\text{DC}} \). The decoherence mechanism at this temperature range is still a topic under investigation [73].

### 4.3 Quantum Mechanical Picture

The Jaynes-Cummings (JC) model is widely used in describing light-matter interactions in two-level systems. The JC Hamiltonian reads

\[
H_{\text{JC}} = H_{\text{atom}} + H_{\text{cavity}} + H_{\text{int}}
\]

\[
H_{\text{atom}} = \frac{1}{2} \hbar \omega_{\text{atom}} \sigma_z
\]

\[
H_{\text{cavity}} = \hbar \omega_0 a^\dagger a
\]

\[
H_{\text{int}} = \hbar g (a^\dagger \sigma_- + a \sigma_+)
\]

where \( \sigma_x, \sigma_y, \) and \( \sigma_z \) are the Pauli matrices of the two-level system, \( \sigma_\pm = \sigma_x \pm i\sigma_y \), and operators \( a, a^\dagger \) are the annihilation and creation operators for cavity photons. \( H_{\text{int}} \)
Figure 4.8: (a) Temperature dependence of magneto-polaritons in Cavity 1 at zero detuning with the fundamental cavity mode from 2 K to 80 K with Lorentzian fitting (black dashed lines). Traces are vertically offset for clarity. (b) Temperature dependence of CR decay time $\tau_{CR}$ in free space (red solid circles), in cavity (blue solid circles) and DC momentum scattering time $\tau_{DC}$ (black open circles).
is the light-matter interaction term, where the coupling strength is represented by $g$. The operator $\sigma_\pm$ creates/annihilates one atomic excitation. In the JC model, the Rabi splitting $\Omega$ equals $\sqrt{\Delta^2 + 4g^2(n + 1)}$, where $\Delta = \omega_{\text{atom}} - \omega_0$ is the detuning and $n$ is the averaged number of photons inside the cavity. At zero detuning ($\Delta = 0$) and zero photon case ($n = 0$), the atom can still be coupled with the cavity vacuum state through quantum fluctuations. In this case, the Rabi splitting $\Omega = 2g$ is known as vacuum Rabi splitting. One assumption made in the JC model is the rotating wave approximation (RWA), which neglects the other two terms ($a\sigma_-$ and $a^\dagger\sigma_+$) in $H_{\text{int}}$, which is valid when $g \ll \omega_0$. This condition is generally satisfied if the system is far from the ultrastrong coupling regime.

In our case, however, a Landau-quantized 2DEG is not a two-level system in general; rather, it is an ensemble of many harmonic oscillators. In addition, the large dipole moment of CR provides strong light-matter coupling. It has been shown that $g$ can go beyond $0.1\omega_0$ [67,68], where the RWA is not well satisfied. Furthermore, the diamagnetic term needs to be included to describe the diamagnetic response of free electrons. Therefore, the JC model is not expected to be able to describe our CR-cavity system well. Instead, we used a more accurate Hamiltonian, which is written in the following way [74]:

$$H_{\text{tot}} = H_{\text{CR}} + H_{\text{cavity}} + H_{\text{int}} + H_{\text{dia}} \quad (4.5)$$

$$H_{\text{CR}} = \hbar\omega_{\text{CR}}b^\dagger b \quad (4.6)$$

$$H_{\text{cavity}} = \hbar\omega_0 a^\dagger a \quad (4.7)$$

$$H_{\text{int}} = \hbar ga(b^\dagger - b) + \hbar ga^\dagger(b^\dagger - b) \quad (4.8)$$

$$H_{\text{dia}} = \frac{\hbar g^2}{\omega_{\text{CR}}}(a^\dagger a^\dagger + a^\dagger a + aa^\dagger + aa) \quad (4.9)$$

where $H_{\text{CR}}$ describe the energy of the 2DEG in a magnetic field with frequency $\omega_{\text{CR}}$. 
and $H_{\text{cavity}}$ is for the cavity mode at $\omega_0$. Operators $b, b^\dagger$ are the annihilation and creation operator for collective CR excitations, respectively. The coupling strength between CR and a cavity photon is $g$. The last term is the diamagnetic term; mathematically, it is the quadratic term of the vector potential $A$, i.e., the so-called $A^2$ terms. The pre-factor $\frac{g^2}{\omega_{\text{CR}}}$ means that the $A^2$ term is small in the weak coupling regime but may have measurable effects or can even become dominant in the ultra-strong coupling regime where $g/\omega_{\text{CR}} \sim 1$.

The magnitude of the vacuum Rabi splitting, $2g$, is determined by the CR dipole moment of an individual electron ($d$), cavity field strength ($E_{\text{Vac}}$), and the number of electrons ($N$). $2g$ for a single quantum well can be written as [74]

$$2g = 2d \cdot E_{\text{Vac}} \cdot \sqrt{N} = \sqrt{\frac{2e^2\omega_{\text{CR}}n_e\omega_0}{\epsilon m^* L_z}}$$  \hspace{1cm} (4.10)$$

where $n_e$ is the 2DEG electron density, $m^*$ is the electron effective mass, and $L_z$ is the effective cavity length. In our THz PCC cavity, the $E_{\text{vac}}$ is on the order of $10^{-3}$ V/cm. In our experiments, the actual electric field of the THz probe at the cavity frequency was between 0.005 to 0.1 V/cm. The corresponding photon number was between $10^1$ to $10^4$. The reason we still observed vacuum Rabi splitting is because a GaAs 2DEG in a magnetic field in an electromagnetic field is a linear system. Different from two-level atoms, the coupling strength does not depend on the driving field strength. Hence, the Rabi splitting in a cavity-2DEG system does not have a square root dependence on the cavity photon number. This is confirmed by the THz field strength dependence of polariton positions shown in Figure 4.9, in which the THz probe field was attenuated to the $10^{-3}$ V/cm level, approaching the vacuum field value, and yet, the polariton positions remained unchanged.

The quadratic form of $H_{\text{tot}}$ makes exact diagonalization possible, through the
Figure 4.9: The probe field strength dependence of the polariton splitting. (a) THz probe field. The peak electric field from bottom to top is 5, 10, 20, 30, and 45 V/cm, respectively. The corresponding THz field at the cavity frequency is \( \sim 10^{-3} \) times smaller than the peak field. (b) The corresponding polariton splitting for each case. The splitting remain unchanged from the case of \( 10^3 \) averaged cavity photons to the few photons quantum limit.
generalized Hopfield method [75]. It is equivalent to a Bogoliubov transformation on bare CR and cavity photon operators. The operators of new normal modes, magneto-polaritons, are a linear combination of \( a, a^\dagger, b, \) and \( b^\dagger \). The Hopfield transformation matrix is the following [74,75]:

\[
M = \begin{pmatrix}
\omega_0 + 2 \frac{g^2}{\omega_{CR}} & ig & -2 \frac{g^2}{\omega_{CR}} & ig \\
-ig & \omega_{CR} & ig & 0 \\
2 \frac{g^2}{\omega_{CR}} & ig & -\omega_0 - 2 \frac{g^2}{\omega_{CR}} & ig \\
ig & 0 & -ig & -\omega_{CR}
\end{pmatrix}
\] (4.11)

By diagonalizing \( M \), one is able to obtain the frequencies of magneto-polaritons from the eigenvalues. With the experimental cavity frequency and effective mass values, we varied \( g \) to fit the experimental data. The best fit is determined by minimizing the standard deviation between fitting curves and the measured points. Figures 4.10(a) and (b) present the best fits with three different Hamiltonians for both cavities, including the full CR-cavity Hamiltonian \( H_{\text{tot}} \), \( H_{\text{tot}} \) without the diamagnetic term (\( A^2 \) term), and the JC Hamiltonian \( H_{\text{JC}} \). Figures 4.10(c) and (d) show the normalized standard deviations of fitting results as a function of \( g/\omega_0 \) for both Cavities 1 and 2. At the minimum deviation (<1%), \( g/\omega_0 \) is 0.09 for Cavity 1 and 0.12 for Cavity 2. The full CR-cavity Hamiltonian provides the best fit, while the other two fail to show the non-negligible blue shift of the polariton modes. With \( g/\omega_0 \) around 0.1, the change induced by the \( A^2 \) term is on the order of 0.01\( \omega_0 \). Thanks to the ultranarrow linewidth of magneto-polaritons, we observed this 1% contribution from the \( A^2 \) term, indicating that our system is reaching the ultrastrong coupling regime.
Figure 4.10: (a) and (b) present the best fits to our experimental data of the two cavities using three different Hamiltonians—the full CR-cavity Hamiltonian (black solid), the full CR Hamiltonian without the $A^2$ term (black dashed), and the Jaynes-Cummings Hamiltonian (blue dotted). Only the full CR-cavity Hamiltonian reproduced the experimental data well, suggesting the non-negligible contribution of the $A^2$ term on the polariton frequencies, i.e., the system is close to the ultrastrong-coupling regime. (c) and (d) show the normalized fitting deviation as a function of $g/\omega_0$ ratio. The best fit achieved when, $g/\omega_0$ is 0.09 for Cavity 1 and 0.12 for Cavity 2, which are at the edge of the ultrastrong-coupling regime.
4.4 Classical Electrodynamic Picture

The strong coupling of CR and THz cavity fields can be described by classical electrodynamics as well. Rabi splitting itself is not a purely quantum mechanical effect. In the classical picture, the vacuum Rabi splitting can be explained as normal mode coupling between CR and cavity modes. That is, the upper and lower magneto-polaritons can be viewed as normal modes of the coupled system. To reproduce all the experimental features with classical electromagnetism, we utilized the transfer matrix method, which is a powerful tool for describing optical properties of multilayered structures, to calculate the transmission spectra of our 2DEG-THz cavity structure.

The transfer matrix $M = \begin{pmatrix} m_{11} & m_{12} \\ m_{21} & m_{22} \end{pmatrix}$ of a given layer connects electric and magnetic fields at the two surfaces of the layer. Assuming that an electromagnetic wave is propagating from the left to the right through a layer with thickness $d$ and index $n$, we have the following relation between the $(E_0, H_0)$ and $(E_1, H_1)$, which are the electric and magnetic fields at the left and right surfaces of the layer, respectively:

$$\begin{pmatrix} E_0 \\ Z_0 H_0 \end{pmatrix} = \begin{pmatrix} m_{11} & m_{12} \\ m_{21} & m_{22} \end{pmatrix} \begin{pmatrix} E_1 \\ Z_0 H_1 \end{pmatrix} = \begin{pmatrix} \cos \delta & i \sin \delta/n \\ i \sin \delta & \cos \delta \end{pmatrix} \begin{pmatrix} E_1 \\ Z_0 H_1 \end{pmatrix}$$ (4.12)

where $\delta = nd\omega/c$ is the phase shift, and $Z_0$ is vacuum impedance. The field transmissivity, $t$, can then be written as

$$t = \frac{E_1}{E_0} = \frac{2n_0}{n_0m_{11} + n_0n_1m_{12} + m_{21} + n_0m_{22}},$$ (4.13)

where $n_0$ and $n_1$ are the refractive indices of the left and right sides of the layer, respectively. For an $N$-layer structure, the total transfer matrix is given by the product of individual transfer matrices, $M_{\text{total}} = M_1 M_2 \cdots M_N$, and the overall $t$ can be obtained solely from the matrix elements of $M_{\text{total}}$. 
Figure 4.11: (a) Experimental transmission spectrum of Cavity 1 is reproduced by the transfer matrix method at 0 T. (b) Calculated cavity power transmittance for a CR active THz wave with realistic values of 2DEG density \(3 \times 10^{11} \text{ cm}^{-2}\) and mobility \(4 \times 10^6 \text{ cm}^2/\text{Vs}\) in magnetic fields up to 6 T. The calculation matches the experimental peak positions (yellow open circles) of the magneto-polaritons in Cavity 1.
In our 1D THz PCC, the refractive index of silicon and vacuum are 3.4 and 1, respectively, with no frequency and magnetic field dependence. The complex refractive index of a 2DEG thin film can be obtained through the Drude model. The elements of the Drude conductivity tensor of a 2DEG in a magnetic field applied in the $z$ direction are given by

$$
\sigma_{xx} = \frac{\sigma_0 \omega \tau}{(1 - i \omega \tau)^2 + (\omega_{CR} \tau)^2}, \quad \sigma_{xy} = \frac{\sigma_0 \omega_{CR} \tau}{(1 - i \omega \tau)^2 + (\omega_{CR} \tau)^2},
$$

$$
\sigma_{CRA} = \sigma_{xx} + i \sigma_{xy}, \quad \sigma_{CRI} = \sigma_{xx} - i \sigma_{xy},
$$

(4.14)

where $\sigma_0$ is the DC conductivity, $\sigma_{xx}$ and $\sigma_{xy}$ are the longitudinal and Hall conductivities, respectively, and $\sigma_{CRA}$ and $\sigma_{CRI}$ are the conductivities for the CR active and CR inactive modes, respectively.

Figure 4.11 shows good agreement between experimentally measured (red solid curve) and theoretically calculated (blue dashed curve) transmission spectra for the CR active mode where the transfer matrix method was used for the calculation. The actual density and mobility of the 2DEG were used in the calculation. The calculated normal mode splitting is 86 GHz for all three cavity modes. This value is close to the experimental value of 74 GHz for the first mode. For Cavity 2, excellent agreement between calculation and experiments was achieved, as shown in Figure 4.12. The peak positions of the magneto-polaritons and the residual cavity mode are well reproduced by our classical model. In contrast to the ultrasharp magneto-polaritons observed within the photonic stop-bands, a much broader CR linewidth is observed outside the photonic stop-bands due to the unsuppressed superradiance decay, as shown in Figure 4.12(b); interestingly, this feature is also reproduced by the classical model, as shown in Figure 4.12(a), indicating that superradiance is not a purely quantum mechanical concept.
Figure 4.12: (a) Calculated cavity power transmittance for linearly polarized THz probe pulse with a realistic 2DEG density for Cavity 2. (b) Experimental transmittance of Cavity 2 from 0 T to 3.5 T with a color scale from 0 to 1 in transmittance. (c) Same experimental data with a color scale from 0 to 0.06 in transmittance. Magneto-polaritons and residual cavity mode are well resolved. They match the calculated spectrum in (a) well. The linewidths of the cavity mode and magneto-polaritons are instrument limited by the 80 ps time window.
4.5 Conclusion and prospects

We demonstrated strong light-matter coupling between the cyclotron resonance of a two-dimensional electron gas and THz cavity photons in a high-$Q$ 1D photonic-crystal cavity, with an unprecedentedly high cooperativity of up to 360. We observed vacuum Rabi oscillations directly in the time domain, and the observed Rabi splitting exhibited a $\sqrt{n_e}$ dependence on the electron density ($n_e$), which is the signature of collective Rabi splitting [12, 45, 70, 76]. The observed anticrossing behaviors of magneto-polaritons can be well described by a full quantum mechanical treatment. A small but definite blue shift was observed for the polariton frequencies, which can be attributed to the $A^2$ term in the light-matter interaction Hamiltonian. Classical electromagnetic theory using the transfer matrix method also reproduced the observed peak positions of the polaritons well. Furthermore, since absorption and emission are reversible in the strong coupling regime, the superradiance decay of cyclotron resonance in high-mobility 2DEGs (see Chapter 3) was significantly suppressed by the presence of the high-$Q$ THz cavity. As a result, we observed ultra-narrow magneto-polariton resonances, with corresponding intrinsic CR linewidths of 5.6 GHz (or a CR decoherence time of 57 ps) at 2 K.

Our demonstration of strong light-matter coupling in high-$Q$ THz cavities opens up various new possibilities for quantum optical studies using CR in semiconductors. Nonlinearities of a cavity-QED system are key to successful generation of non-classical states, such as squeezed light and entangled photons. Although CR in GaAs 2DEGs is not suited for this purpose because of the small conduction-band nonparabolicity in GaAs, narrow-gap semiconductors such as InSb and InAs, with inherently strong nonparabolicity, are promising materials for exploring THz quantum optics. With state-of-the-art high-mobility ($\sim 10^5$ cm$^2$/Vs) InSb 2DEGs [77], it should be possible
to isolate two adjacent Landau levels from the non-equally spaced Landau ladder with a proper density and magnetic field. Such a high-mobility, massively-degenerate quasi-two-level solid-state system guarantees both strong light-matter interaction and long-lived coherence and thus can be expected to exhibit a variety of exotic nonlinear THz quantum optical phenomena.

The cyclotron resonance of massless Dirac fermions in graphene is expected to provide an even more nonlinear system. Although this is currently a subject under debate [59, 60], light-matter interaction in graphene, being different from InSb, may be free from the $A^2$ term due to the linear energy-momentum relationship, which is ideal for the realization of the Dicke superradiance phase transition [59]. Regardless of the theoretical expectations and debates, it will be very important to establish experimental facts concerning strong light-matter interactions in massless Dirac fermion systems, which are largely nonexistent. Our THz cavities can be readily applied to graphene, but the current technical obstacle is the rarity of high-mobility large-area graphene samples suitable for THz spectroscopy.

Another important and intriguing research direction to be enabled by InSb- or graphene-cavity systems is to investigate the role of electron-electron interactions in the strong light-matter coupling regime. Due to the breakdown of Kohn’s theorem in strongly nonparabolic systems such as InSb and graphene, the energies of magneto-polaritons may be modified by electron-electron interactions. Some many-body features have been reported for interband magneto-optical absorption in a near-infrared cavity [78]. It will be very interesting to see how such a system behaves with much stronger light-matter interaction in the THz range, especially in the fractional quantum Hall regime.

Beyond semiconductors, there are even more opportunities for unique THz cavity-
QED studies with 1D PCCs. Unlike the near-field coupling of metamaterial resonators, our THz cavity scheme applies to both 2D and bulk materials, which make it feasible for various strongly correlated systems with collective many-body excitations in the THz range, including long-lived THz antiferromagnetic resonances in bulk materials [79–81] and collective superconducting amplitude mode (Higgs mode) in conventional superconductors [82]. Our demonstration of strong light-matter interaction in high-$Q$ THz cavities opens a door to a plethora of new possibilities to combine the traditional disciplines of many-body condensed matter physics and quantum optics of cavity QED.

Finally, the ultrafast nature of THz-TDS is expected to lead to the creation of another exciting research topic for THz cavity-QED research, i.e., ultrafast dynamics of strong light-matter interaction, including quantum quench. Nonequilibrium dynamics of many-body systems is one of the least understood topics in condensed matter physics and currently actively investigated in the community of ultracold atoms [26]. With the addition of strong light-matter coupling to nonequilibrium systems, exotic quantum mechanical phenomena may appear, such as the dynamic Casimir effect [83], through, e.g., an ultrafast nonadiabatic change of light-matter interaction [64].
Chapter 5

Ultrafast Dynamics of Excitons and Magneto-excitons in GaAs Quantum Wells

5.1 Introduction

An electron-hole (e-h) pair system is a simple two-component system, but it has various possible phases brought about by long-range Coulomb interactions, including an e-h plasma, an exciton gas, e-h droplets [84], and Bose-Einstein condensations (BEC) [85]. Each of these phases has its origin in the coexistence and interplay of the repulsive and attractive interactions of similar magnitudes, as well as the capability of exhibiting either Bose-Einstein or Fermi-Dirac statistics for individual electrons/holes or excitons, respectively.

A schematic phase diagram for e-h pairs as a function of temperature and pair density is shown in Figure 5.1. The Coulomb attraction between an electron and a hole creates a bound e-h pair, i.e., an exciton. At low temperatures, excitons behave as an insulating Bose gas in the dilute limit, where the exciton Bohr radius, $a_B$, is much smaller than the interexciton distance ($d$). The e-h pair system can be driven through an excitonic Mott transition from the insulating excitonic gas into a metallic e-h plasma, where $a_B > d$. This is an insulator-to-metal transition induced by density-dependent Coulomb interactions; the resulting metallic state corresponds to a half-filled Hubbard band in the regular picture of Mott physics [86]. Within a certain temperature and pair density range, a coexisting phase of excitons, biex-
Figure 5.1: Schematic phase diagram of an e-h pair system as a function of pair density and temperature at zero magnetic field.
citons, and e-h droplets may appear. Droplets are a classical condensation of the e-h pairs and have been observed mainly in indirect bandgap semiconductors such as silicon [87] and germanium [88]. In direct bandgap semiconductors, however, droplet formation is much more difficult, due to the relatively short exciton lifetimes (∼1 ns), which are not sufficiently long for excitons to cool and reach thermal equilibrium. However, an interesting observation of quantum droplets, or ‘dropletons’, has been reported for GaAs quantum wells, which is a direct bandgap system [89]. At very low temperatures, quantum condensation phenomena are expected to happen in e-h systems. On the bosonic side, the possibility of Bose-Einstein condensation (BEC) of excitons has attracted enormous amounts of attention and efforts over the last several decades [85]. Thanks to the small reduced masses and large permittivities, exciton condensation phenomena are expected to happen at much high temperatures than their atomic counterparts [90]. With giant exciton binding energies, 2D semiconductors of transition metal dichalcogenides may be able to achieve BEC even at room temperature. On the fermionic side, the Bardeen-Cooper-Schrieffer (BCS) like pairing of electrons and holes in $k$-space is expected to happen, at least transiently. The BCS pairing of electrons and holes is expected to open an energy gap at the Fermi surface, and thus, a quantum-degenerate electron-hole plasma is turned into an exciton insulator phase [91].

The e-h pair phase diagram can be modified by an external magnetic field in a profound manner. As schematically shown in Figure 5.2, in the low density regime, the major change of excitons induced by a magnetic field is the coupling between the exciton center-of-mass (COM) momentum and their internal energy [92–94], which are obviously decoupled at zero field. This new type of excitons are called magnetoexcitons. In the high magnetic field and high density regime is the correlated
Figure 5.2: Schematic phase diagram of an e-h system as a function of pair density and magnetic field. In the low density region, excitons become magnetoexcitons with their center of mass momentum coupled with internal energy. A correlated magnetoplasma is located in the high density region. Novel cooperative phenomena, e.g., superfluorescence [4], can appear in this region. In the extreme quantum limit of high magnetic fields, an insulating phase of noninteracting magnetoexcitons appears due to the e-h charge symmetry (‘hidden symmetry’). Adopted from Ref. [5], Figure 1.
magnetoplasma phase of e-h pairs. Strong Landau quantization and Fermi degeneracy increase the density of states of electrons and holes and reduce the phase space for possible decoherence scattering processes, which leads to a longer phase coherence time. Because of that, novel cooperative phenomena may appear in this dense correlated plasma, e.g., superfluorescence [4] with Fermi-edge enhancement [95]. The excitonic Mott transition is also expected to move toward higher densities due to shrinking of exciton wavefunctions in a high magnetic field.

The most interesting theoretical prediction for 2D excitons in a strong perpendicular magnetic field is that exciton-exciton interactions should completely vanish due to an e-h charge symmetry, the so-called hidden symmetry, which results in an exact cancellation of Coulomb interactions between excitons, creating ultrastable magnetoexcitons [96–98]. An intuitive picture for the hidden symmetry theory is that the exciton wavefunction is so strongly shrunk by the extremely high magnetic field that the average electron-hole distance becomes negligible compared with the interexciton distance [96]; hence, excitons behave as point-like charge neutral particles without long-range interactions. The existence of the hidden symmetry phase is totaly due to the e-h charge symmetry and should be distinct from the excitonic Mott transition. A previous experimental study suggested that a hidden symmetry phase appears abruptly at a filling factor ($\nu_e, \nu_h$) of exactly two in a 2D e-h pair system [99], namely, when the all carriers are accommodated by the lowest Landau level.

Experimental studies of exciton dynamics in high magnetic fields are very limited [92–94, 100, 101]. Probing excitons and e-h plasmas in semiconductors usually employs interband photoluminescence (PL) and absorption spectroscopy. However, there are two inherent limitations in utilizing interband optical transitions for monitoring exciton dynamics. First, interband spectroscopy is based on the detection of
photons associated with the creation (i.e., absorption) or annihilation (i.e., PL) of excitons in semiconductors. Due to the momentum conservation requirement, combined with the negligibly small momentum of photons, the COM momentum of such a created/annihilated exciton is almost zero. Namely, excitons with nonzero COM momenta are dark (or inaccessible) for interband techniques. Second, it is difficult to clearly identify the spectral features for bound and unbound carriers at the same time. Many nonexcitonic effects affect interband transitions, including bandgap renormalization, which can also interfere with the determination of exciton binding energy. These two limitations are overcome in THz spectroscopy, which is a technique based on intraband excitations. Different from the interband case, intraband transitions maintain the total number of excitons/e-h pairs. In addition, it can access to those interband ‘dark excitons’ with finite COM momenta. The latter advantage is crucial especially for magnetoexcitons, which have nontrivial internal energy dispersions with respect to the COM momentum [102]. The excitonic features that are probed in THz spectroscopy are intraexciton transitions, especially the 1s-2p transition. These bound carrier resonances are distinct from the Drude-like response of unbound carriers; for example, the exciton internal transitions are essentially uninfluenced by bandgap renormalization. One is able to monitor the dynamics of bound and unbound e-h pairs at the same time. Experimentally, THz time-domain spectroscopy has been proven to be a powerful tool for investigating excitonic dynamics and Mott physics in semiconductors [87,103–108]. With continuously tunable pair density, temperature, and probe delay time, one can systematically examine excitons in different phases on an ultrafast time scale, and in a high magnetic field.

In this chapter, we present a systematic experimental study of e-h pairs in photoexcited undoped GaAs quantum wells in perpendicular magnetic fields up to 10 T,
using optical-pump THz-probe spectroscopy. We simultaneously monitored the intraexcitonic 1s-2p transition (which splits into the 1s-2p+ and 1s-2p− transitions in a magnetic field) and the cyclotron resonance of unbound electrons and holes as a function of magnetic field, temperature, e-h pair density, and probe time delay. We found that the 1s-2p− feature is robust at high magnetic fields even under high excitation fluences, indicating magnetically enhanced stability of excitons.

5.2 Continuous Wave Spectroscopy

One undoped GaAs multiple QW samples was used in this study. Each sample contained 15 pairs of a 20-nm GaAs well and a 20-nm Al0.3Ga0.7As barrier, sandwiched by two 500-nm-thick Al0.3Ga0.7As spacer layers. The total thickness of the multiple QW layer was 600 nm. The GaAs substrate was removed to avoid observing any photoinduced carrier effects in bulk GaAs (see Chapter 2 for more details).

Absorption and photoluminescence (PL) spectroscopy with a continuous-wave light source can characterize the linear interband optical properties of the sample under weak excitation. As shown in Figure 5.3(a), at zero magnetic field, a pronounced absorption peak is located at 1.5233 ± 0.0005 eV, which is well separated from all other absorption features. It is the heavy-hole (HH) 1s exciton resonance, which has a linewidth of 1 meV indicating the good quality of the quantum wells. The HH 2s state and the light-hole (LH) 1s state are overlapping each other. Their peak positions are at 1.5297 ± 0.0005 eV and 1.5302 ± 0.0005 eV, respectively. Therefore, the energy separation between the 1s and 2s state of HH excitons is 6.4 ± 0.5 meV, which is also the photon energy required for the 1s-2p transition, since the 2s and 2p states are degenerate at zero magnetic field. When a magnetic field is applied, the HH 1s peak slightly shifts toward the high frequency side, which is the diamagnetic
shift of excitons. With increasing magnetic field, LH 1s and HH 2s states further separate. The linewidth of the HH 1s peak increases with increasing magnetic field due to spin and orbital angular momentum splitting. Figure 5.3(b) shows the PL spectra from 0 T to 9 T. The low-energy PL peak is due to bound excitons, while the higher-energy peak is due to free excitons. The separation between the bound exciton and the free exciton PL is 1.4 meV at any magnetic field, which is 16 K in temperature, consistent with the temperature dependence shown in Figure 5.4. The linewidth of the free exciton PL feature is 1 meV at zero field, slightly decreases with increasing magnetic field, and reaches 0.6 meV at 9 T. The peak positions of the exciton absorption and PL features are shown in Figure 5.5. The Stokes shift is 0.6 meV at 0 T.
Figure 5.4: Temperature dependence of photoluminescence spectra at zero magnetic field. The bound exciton (BX) peak at 1.521 eV is not observed at a temperature higher than 20 K. The free exciton (FX) PL intensity increases with the temperature.

Figure 5.5: The peak positions of the observed interband transitions as a function of magnetic field. The red symbols are for absorption peaks and the blue symbols are for PL peaks.
5.3 Exciton Dynamics at Zero Magnetic Field

We studied exciton dynamics in the undoped GaAs multiple QW sample by time-resolved THz magneto-spectroscopy. As schematically shown in Figure 5.6 (a), at time zero, an optical excitation pulse created HH 1s excitons (free e-h pairs) in the quantum wells in the case of resonant (nonresonant) excitation. For resonant excitation, a 4f pulse shaper was used as a tunable ultranarrow bandpass filter to select the frequency component that is resonant with the HH 1s state, as shown in Figure 5.6(b). In the case of nonresonant excitation, the sample was excited by a 1.6 eV pulse, which is ~70 meV higher than the HH 1s state energy and thus created a hot e-h plasma in the continuum at time zero. The THz probe pulse transmitted through the QW at a certain time delay, ∆t, measured the instantaneous response of the exciton/e-h pair system. By varying ∆t, we were able to obtain snapshots of the dynamics of the system.

More specifically, what we measured was the optical-pump-induced change of the transmitted THz field, ΔE(t). Combining it with a separated reference measurement of THz waveform without optical excitation, ΔE_{ref}(t), we were able to obtain the photoinduced change in the complex dielectric function, Δε(ω), and the complex optical conductivity, Δσ(ω), through the following equations [106]:

\[
\Delta \varepsilon(\omega) = \frac{\Delta E(\omega)}{\Delta E(\omega) + E(\omega)} \left( \frac{1 + n_S + B}{A} - \varepsilon(\omega) \right)
\]

(5.1)

\[
A = i\omega \left( \frac{1}{c} - i\omega d_L (1 + n_S) \right) D_{GaAs}
\]

(5.2)

\[
B = -i\omega \left( n_S D_{tot} + D_{AlGaAs} n_{AlGaAs}^2 \right)
\]

(5.3)

where ΔE(ω) and E(ω) are the complex Fourier transform of ΔE(t) and E_{ref}(t), respectively, n_S is the refractive index of the sapphire substrate, n_{AlGaAs} is the refractive index of the Al_{0.3}Ga_{0.7}As spacers, D_{GaAs} (= 300 nm) is the total thickness
Figure 5.6: (a) Schematic diagram of measuring exciton dynamics in undoped GaAs multiple QW with time-resolved THz magneto-spectroscopy. (b) Spectra of resonant optical pump pulse with the corresponding absorption spectra at 0 T and 9 T. The non-resonant excitation photon energy was kept at 1.6 eV in all measurements.
Figure 5.7: Photo-induced change in the real part of (a) conductivity and (b) permittivity under resonant excitation at 5 K and 0 T. The pump fluence was 150 nJ/cm². The 1s-2p transition is observed at 1.56 THz (or 6.4 meV). Probe delay time was 100 ps.

of GaAs layers, \( D_{\text{AlGaAs}} (= 1.3 \, \mu m) \) is the total thickness of the \( \text{Al}_{0.3}\text{Ga}_{0.7}\text{As} \) layers, \( D_{\text{tot}}=D_{\text{GaAs}}+D_{\text{AlGaAs}} \), and \( d_L (= 500 \, \text{nm}) \) is the thickness of the AlGaAs spacers.

Figure 5.7 shows the observed HH exciton 1s-2p transition under resonant excitation at 0 T and 5 K. The fluence of the near-infrared pump was 150 nJ/cm², and the THz probe delay time was 100 ps. The 1s-2p transition was located at 1.55 THz, which is 6.4 meV in energy, matching well the 6.4 meV energy separation between the HH 1s and 2s states measured by CW absorption spectroscopy. The corresponding exciton binding energy, \( E_b \), is 7.2 meV for a 2D hydrogen-like exciton. No clear Drude-like component was found in the low frequency region, indicating very few unbound e-h pairs; that is, the e-h pairs system was in the excitonic gas phase.

In the case of nonresonance excitation (Figure 5.8), hot e-h pairs were created by the 1.6 eV pump pulse with 150 nJ/cm² in fluence at 5 K and 0 T. \( \Delta \epsilon(\omega) \) was negative, no exciton feature was observed, and the system was in the metallic plasma phase. As the THz probe delay increased, the 1s-2p transition emerged at 150 ps, and
Figure 5.8: Dynamics of unbound e-h pairs and excitons after non-resonant excitation at 5 K and 0 T. Photoninduced changes in the real part of (a) conductivity and (b) permittivity are shown. The pump fluence was 150 nJ/cm$^2$. At 20 ps, $\Delta \epsilon(\omega)$ was negative, which is a characteristic metallic behavior. The 1s-2p exciton features was observed after 150 ps, and survived even after 1.8 ns.
Figure 5.9: Photo-induced conductivity spectra $\Delta \sigma(\omega)$ at various resonant pump fluences at 5 K and 0 T. The pump fluence from the bottom trace to the top were $1 \times 150$ nJ/cm$^2$ up to $40 \times 150$ nJ/cm$^2$. An excitonic Mott transition was observed at 5 K and 0 T. The $1s-2p$ transition totally vanished at the highest pump fluence, and the conductivity was dominated by an e-h plasma. The optical pump delay was 100 ps.

became much clearer at 400 ps, then dominated the conductivity at later times, which showed the dynamics of e-h pair cooling and exciton formation. Excitons survived even after 1.8 ns of THz probe delay.

Figure 5.9 presents the pump fluence dependence of $\Delta \sigma(\omega)$ for resonant excitation. At low fluences, $\Delta \sigma(\omega)$ shows a pronounced exciton feature. As the density of HH 1s excitons increases with increasing pump fluence, the exciton internal transition broadens very quickly, while the Drude-like response also emerges in the low frequency region. Eventually, at the highest pump fluence ($40 \times 150$ nJ/cm$^2$), the $1s-2p$ transition totally vanished, and the system was turned into a correlated e-h
plasma. The carrier scattering time and e-h pair density were extracted by Drude fitting of the highest fluence trace. The obtained scattering time was 0.1 ps, and the sheet density was $2.86 \times 10^{12} \text{cm}^{-2}$, which corresponds to $1.9 \times 10^{11} \text{cm}^{-2}$ per quantum well. Therefore, in our system, the Mott density is on the order of $10^{11} \text{cm}^{-2}$ for a single 20-nm-wide quantum well, which is consistent with Ref. [104]. However, different from Ref. [104], no clear red-shift of the exciton 1$s$-$2p$ feature was found. It was difficult to trace the 1$s$-$2p$ peak position, since the exciton internal transition broadened quickly, and peaks were not well defined. Interestingly, in bulk indirect bandgap semiconductors, the 1$s$-$2p$ transition of 3D excitons remained at almost the same frequency until the excitonic Mott transition occurred [107,108], which is similar to what we observed.

### 5.4 Exciton Dynamics at Finite Magnetic Fields

A magnetic field lifts the degeneracy of the 2$p$ states, which have nonzero orbital angular momenta. Hence, the 1$s$-$2p$ transition splits into 1$s$-$2p_+$ and 1$s$-$2p_-$ in a magnetic field. In the Landau level (LL) picture, the 1$s$-$2p_+$ (1$s$-$2p_-$) transition can be viewed as a transition where the electron (hole) is excited from the lowest to the first LL while the hole (electron) stays in the lowest LL. Figure 5.10 shows calculated intraexciton transition energies as a function of magnetic field based on a 2D hydrogenic model within the effective-mass approximation. The electron and heavy hole effective masses used in this calculation were $0.067m_e$ and $0.51m_e$, respectively.

We measured photoinduced THz conductivity spectra at various magnetic fields from 0 T to 10 T under 1.6 eV nonresonant excitation at 5 K. As shown in Figure 5.11, two distinct CR features due to electrons and heavy holes were observed for the hot e-h magnetoplasma at 15 ps. The measured effective masses of electrons and heavy
holes were $0.069m_e$ and $0.53m_e$, respectively. At 1 ns, magnetoexcitons were formed. We observed both the $1s-2p_+$ and $1s-2p_-$ transitions, as well as the CR of unbound electrons. They match the calculation in general. It is worth noting that, at the intersection between e-CR and the $1s-2p_-$ transition, no anticrossing behavior was observed. This can be understood as a consequence of the small interaction between these two modes due to the opposite helicities.

Exciton dynamics at high magnetic fields are the main focus of this study. Figure 5.12 presents the dynamics and temperature dependence of magnetoexcitons at 9 T. For the case of nonresonant excitation, the formation process of magnetoexcitons is shown in Figure 5.12(a). Heavy-hole CR and the tail of electron CR were observed at 5 ps, and the $1s-2p_-$ feature appeared after 100 ps. For resonant excitation, the $1s-2p_-$ transition appeared right after pumping and showed a weak dependence on
Figure 5.11: Photoinduced conductivity spectra as a function of magnetic field from 0 T to 10 T at (a) 15 ps and (b) 1 ns THz probe delay, after nonresonant excitation. At 15 ps, a hot e-h magnetoplasma shows two CR branches due to electrons and heavy holes. At 1 ns, both the $1s-2p_+$ and $1s-2p_-$ transitions were resolved.
Figure 5.12: Photoinduced conductivity spectra at various probe delay times and temperatures at 9 T. (a) Dynamics of magnetoeexcitons after nonresonant excitation with 200 nJ/cm$^2$ fluence at 6 K. (b) Similar measurements to (a) but with resonant excitation of HH 1s excitons. The pump fluence was also 200 nJ/cm$^2$. (c) Temperature dependence of $\Delta\sigma(\omega)$ with resonant pumping, measured at 200 ps.
Figure 5.13: Photon-induced conductivity $\Delta \sigma(\omega)$ and permittivity $\Delta \epsilon(\omega)$ spectra at various resonant pump fluences at 5 K and 9 T. The pump fluence from the bottom trace to the top were $1 \times 200 \text{ nJ/cm}^2$ up to $40 \times 200 \text{ nJ/cm}^2$. The $1s-2p_-$ survived even at the highest fluence, which would be sufficiently large to drive the excitonic Mott transition at 0 T.

The probe time delay within 900 ps, indicating a long exciton lifetime. Thermal ionization of magnetoexcitons after resonant pumping is presented in Figure 5.12(c). The oscillator strength was transferred from the $1s-2p_-$ transition to the heavy hole CR ($\sim 0.5 \text{ THz}$) and electron CR, which was outside our THz bandwidth. At 60 K, the exciton feature vanished. The thermal energy at 60 K is 5.2 meV, which is close to the exciton binding energy.

Figure 5.13 illustrates the pump fluence dependence of $\Delta \sigma(\omega)$ and $\Delta \epsilon(\omega)$ at 9 T. The $1s-2p_-$ peak survived at the highest pump fluence ($40 \times 200 \text{ nJ/cm}^2$), which would already be sufficient to drive the excitonic Mott transition at 0 T. This suggests that the $1s-2p_-$ feature was robust at high magnetic fields even under high excitation fluences, indicating magnetically enhanced stability of excitons. The position of the
$1s-2p_-$ peak also remained the same. The e-h pair density was on the order of $10^{11}$ cm$^{-2}$ per quantum well. The corresponding filling factor at 9 T was less than two, and thus the system was in the magnetic quantum limit. Therefore, hidden symmetry may be responsible for the robustness of magnetoexcitons we observed here. At high pump fluences, a conductivity feature emerged around 1 THz, which could be due to the roton-like minimum in the COM dispersion of the $2p_-$ magnetoexciton state [102].

5.5 Conclusion

We presented a systematic experimental study of e-h pairs in photoexcited undoped GaAs quantum wells in perpendicular magnetic fields up to 10 T, using optical-pump/THz-probe spectroscopy. We simultaneously monitored the intraexcitonic $1s-2p$ transition and the cyclotron resonance of unbound electrons and holes as a function of temperature, e-h pair density, magnetic field, and optical pump photon energy. The $1s-2p$ energy was 6.4 meV, which corresponds to 7.2 meV in binding energy. We observed the exciton formation process with nonresonant excitation. With resonant pumping, we observed an excitonic Mott transition at zero magnetic field. The Mott density was on the order of $10^{11}$ cm$^{-2}$ for a single 20-nm quantum well. In magnetic fields, we observed both the $1s-2p_+$ and $1s-2p_-$ transitions. We further resolved the formation dynamics of magnetoexcitons, whose time scale was on the order of 100 ps. Thermal ionization of magnetoexcitons was also demonstrated at 60 K. Most importantly, we found that the $1s-2p_-$ feature at 9 T was more robust even under high excitation fluences than the $1s-2p$ feature at 0 T, indicating magnetically enhanced stability of excitons.
Chapter 6

Conclusions

In this thesis, we investigated the coherent dynamics and light-matter interactions of quantum-confined electrons and excitons in GaAs quantum wells in magnetic fields using time-domain THz magneto-spectroscopy. First, we investigated the free-space decay dynamics of cyclotron resonance (CR), which were coherently and resonantly excited by THz pulses. We found that the decay rate of CR oscillations increases linearly with the electron density, which we interpreted as evidence of collective radiative decay, or superradiance. Our detailed quantum mechanical calculations confirmed this interpretation, reproducing our experimental observations quantitatively without any adjustable parameter.

Next, we investigated the light-matter interaction and coherent dynamics of the same Landau-quantized 2DEG system when it is strongly coupled with a high-$Q$ 1D photonic-crystal cavity. We simultaneously achieved ultrastrong coupling between coherent CR and THz photons, as well as an ultralong coherence time of magneto-polaritons. The corresponding cooperativity was up to 360, and the $\Omega/\omega_0$ ratio was 0.12. We observed vacuum Rabi oscillations directly in the time domain, and the observed Rabi splitting exhibited a $\sqrt{n_e}$ dependence on the electron density ($n_e$), which is the hallmark of collective Rabi splitting. The observed anticrossing behaviors of magneto-polaritons can be well described by a full quantum mechanical treatment. A small but definite blue shift was observed for the polariton frequencies, which can be attributed to the $A^2$ term in the light-matter interaction Hamiltonian, indicating
that the system reached the ultrastrong coupling regime. Furthermore, due to the reversible absorption and emission in the strong coupling regime, the superradiance decay of CR in high-mobility 2DEGs was significantly suppressed by the presence of the high-$Q$ THz cavity. As a result, we observed ultra-narrow magneto-polariton resonances, with corresponding intrinsic CR linewidths of 5.6 GHz (or a CR decoherence time of 57 ps) at 2 K.

Finally, we presented a systematic experimental study of e-h pairs in photo-excited undoped GaAs quantum wells in perpendicular magnetic fields up to 10 T. We simultaneously monitored the intraexcitonic $1s$-$2p$ transition and the cyclotron resonance of unbound electrons and holes as a function of temperature, e-h pair density, magnetic field, and the optical pump photon energy. The $1s$-$2p$ energy was 6.4 meV, which corresponds to 7.2 meV in binding energy. We observed the exciton formation process with nonresonant excitation. With resonant pumping, we observed an excitonic Mott transition at zero magnetic field. The Mott density was on the order of $10^{11}$ cm$^{-2}$ for a single 20-nm quantum well. In magnetic fields, we observed both the $1s$-$2p_+$ and $1s$-$2p_-$ transitions. We further time-resolved the formation dynamics of magneto-excitons, which occurred on the time scale of $\sim$100 ps. Thermal ionization of magneto-excitons was also demonstrated at 60 K. Most importantly, we found that the $1s$-$2p_-$ feature was extremely robust at high magnetic fields even under high excitation fluences, compared to the 0 T case, indicating magnetically enhanced stability of excitons.

In summary, we revealed the collective nature of decay dynamics and light-matter interaction of a quantum-degenerate, interacting electron system. In the correlated e-h pair system, we observed exciton formation, an excitonic Mott transition, and magnetically enhanced stability of excitons. All of these results were enabled by
coherent time-domain THz magneto-spectroscopy, with ultrahigh sensitivities and picosecond time resolution. Our demonstrations of nonequilibrium dynamics and strong light-matter interaction of quantum-degenerate fermionic and bosonic gases in semiconductors with coherent THz magneto-optics provide good examples of combining the traditional disciplines of many-body condensed matter physics and quantum optics.
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