Observation of Terahertz Gain in Two-Dimensional Magnetoexcitons

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We have observed photoinduced negative optical conductivity, or gain, in the terahertz frequency range in a GaAs multiple-quantum-well structure in a strong perpendicular magnetic field at low temperatures. The gain is narrow-band: it appears as a sharp peak (linewidth <0.45 meV) whose frequency shifts with applied magnetic field. The gain has a strict circular-polarization selection rule: it is observed only for hole-cyclotron-resonance-active polarization. Furthermore, the gain appears only when the exciton 1s state is populated, which rules out intraexcitonic transitions to be its origin. Based on these observations, we propose a possible process in which the stimulated emission of a terahertz photon occurs while two free excitons scatter into one biexciton in an energy and angular-momentum conserving manner.

An ensemble of correlated electron-hole (e-h) pairs, or excitons, in an insulating solid in an external magnetic field (B), or magnetoexcitons, provides a highly tunable nonequilibrium system in which to study quantum many-body phenomena [1]. The internal states of an exciton, which are analogous to the states of a hydrogen atom, can be probed by intraexcitonic transitions (i.e., the excitonic Lyman or Balmer series) using terahertz (THz) radiation (meV in photon energy) [2-22]. These transitions evolve as a function of B, exhibiting shifts and splittings [1, 12, 17, 20, 23], which provide insight into dark states that are not observable in interband optical experiments. It has been recently shown, by probing the 1s–2p transition [20], that two-dimensional (2D) magnetoexcitons in high B are extremely stable against an excitonic Mott transition [24,29], which would ordinarily transform the system from an insulating and bosonic exciton gas into a metallic and fermionic e-h plasma at high pair densities. Such an observation is consistent with the hidden symmetry of 2D magnetoexcitons [24-26], which prevents density-driven exciton dissociation. Further THz spectroscopy studies are needed to develop a microscopic understanding of many-body interactions within the 2D magnetoexciton system.

Here, we report an unexpected phenomenon observed in a 2D magnetoexciton system. We performed time-resolved and circular-polarization-resolved THz magneto-spectroscopy measurements on photoexcited GaAs quantum wells (QWs) in a B, and observed a spectroscopic feature that showed a negative value for the real part of the optical conductivity, indicating THz gain. The gain feature had a center frequency tunable by B and only appeared in the hole-cyclotron-resonance-active (hcRA) circularly polarized mode and at temperatures (T) lower than 10 K. We further found that the upper state of population inversion (USPI) was the 1s exciton state, which rules out any intraexcitonic transition as the gain transition. Based on these observations, we propose the following possible scenario to explain the THz gain: the lower state of population inversion (LSPI) is the biexciton ground state, implying that the scattering of two free 1s excitons into one biexciton caused THz stimulated emission. This process is complementary to the biexciton-exciton population inversion observed in GaAs quantum wires, which resulted in gain in the near-infrared (NIR) range [31].

Our experimental scheme is shown in Fig. 1(a). We performed optical-pump-THz-probe magneto-spectroscopy experiments on an undoped GaAs multiple-QW sample grown by molecular beam epitaxy. The QW sample contained 15 periods of alternating layers of 20-nm-wide GaAs wells and 20-nm-wide Al0.3Ga0.7As barriers. To ensure that photoinduced signals originated exclusively from the QW active region rather than the bulk GaAs buffer layer and substrate, we performed a substrate-removal procedure by wet chemical etching, and transferred the QW active region onto a transparent sapphire substrate.

Our laser system was a Ti:sapphire regenerative amplifier (1 kHz, 150 fs, 775 nm, Clark-MXR, inc.), which generated and detected THz probe pulses with ZnTe crystals and also fed an optical parametric amplifier system for NIR pump generation. The NIR pump photon energy \( E \) was tunable from 1.52 eV to 1.60 eV. The spectral width of the pump \( \Delta E \) was around 3 meV, after spectral filtering by a 4f pulse shaper, which was sharp enough...
Nonresonant continuum... THz quarter-wave plate [32, 33]. In and were circularly polarized by using an achromatic probe pulses covered a frequency range of 0.2–2.5 THz lay $\tau$ band continuum.

The THz probe pulse arrived at the sample at time delay $\tau$ after the NIR pump pulse excited the sample. THz probe pulses covered a frequency range of 0.2–2.5 THz and were circularly polarized by using an achromatic THz quarter-wave plate [32, 33]. In $B$, due to the opposite signs of Lorentz forces, electron cyclotron resonance (CR) and hole CR respond to circularly polarized THz light with opposite helicities. We denote the two circular polarizations as the electron-CR-active ($e$CRA) mode and the $h$CRA mode. THz conductivities for the $e$CRA and $h$CRA modes, denoted as $\sigma_+$ and $\sigma_-$, respectively, were measured.

Figure (b) displays an energy-level diagram showing the pump and probe transitions used in both nonresonant and resonant pumping experiments. Figure (c) shows an experimental THz magnetoconductivity map obtained in the nonresonant pumping case. Here, the pump fluence ($F$) was 400 nJ/cm$^2$, and other experimental conditions were $T = 2$ K, $E = 1.60$ eV, and $\tau = 900$ ps. The pump photon energy was 75 meV above the heavy-hole exciton 1s energy at 0 T; note that we will focus on heavy-hole excitons in this article. Therefore, the pump created a hot $e$-$h$ plasma in the continuum when it arrived at the sample (orange arrow). Carrier cooling and exciton formation ensued in the next 900 ps (black dashed arrow), so that by the time the THz probe arrived at the sample, part of the $e$-$h$ pairs were occupying the 1s exciton state. The 2p state split into two ($2p_+$ and $2p_-$) in $B$ due to the nonzero angular momentum. The THz probe light with $e$CRA ($h$CRA) circular polarization induced the 1s–2p$_+$ (1s–2p$_-$) intraexcitonic transition, as shown by the red solid arrows.

The upper (lower) panel of Fig. (c) maps $\text{Re}(\sigma_+)$ for $e$CRA polarization ($\text{Re}(\sigma_-)$ for $h$CRA polarization) as a function of frequency and magnetic field. Two absorption lines with $\text{Re}(\sigma_+ > 0$ can be observed in the upper panel. The line that starts from 1.6 THz at $B = 0$ T and increases in frequency with $B$ is the 1s–2p$_+$ transition. The line that starts from zero frequency at 0 T and increases linearly with $B$ is due to the CR of free electrons ($e$-CR), which are nonresonantly created and have not relaxed to the 1s exciton state.

In the lower panel of Fig. (c) showing $\text{Re}(\sigma_-)$, the $e$-CR and 1s–2p$_+$ transitions are not observed, because their polarization selection rules are not satisfied. In contrast, the 1s–2p$^-$ transition, which is excitable by $h$CRA-polarized radiation, appears; its transition frequency first decreases and then increases with $B$ due to the combined effect of Zeeman splitting and diamagnetic shift [1]. Most notably, there is a narrow line exhibiting $\text{Re}(\sigma_- < 0$ on the low frequency side of the colormap, indicating gain. The gain peak frequency increases with $B$ and only appears in the $h$CRA polarization mode. The gain feature does not appear in measurements on bulk GaAs single crystals under the same experimental condition [34].

Figure (d) shows a $\text{Re}(\sigma_-)$ spectrum (i.e., for $h$CRA polarization) taken under the following conditions: $E = 1.527$ eV, $T = 4$ K, $B = 7$ T, $F = 250$ nJ/cm$^2$, and $\tau = 300$ ps.

FIG. 1. (a) Schematic diagram of the experimental geometry. (b) Energy-level diagram showing the transitions used for the optical pump and circularly polarized THz probe for both resonant and nonresonant pumping experiments. (c) Experimental THz magnetoconductivity maps for nonresonant pumping with $T = 2$ K, $E = 1.60$ eV, $F = 400$ nJ/cm$^2$, and $\tau = 900$ ps. The upper and lower panels in (c) map $\text{Re}(\sigma_+)$ and $\text{Re}(\sigma_-)$, respectively. Black dashed lines are guides to the eye. (d) $\text{Re}(\sigma_-)$ spectrum obtained in a resonant pumping experiment with $E = 1.527$ eV, $T = 4$ K, $B = 7$ T, $F = 250$ nJ/cm$^2$, and $\tau = 300$ ps.
ergies while we kept $T = 4$ K, $B = 7$ T, $F = 250$ nJ/cm$^2$, and $\tau = 15$ ps. A linear absorbance spectrum becomes bright (the absorption feature marked as “$h$–CR” in the colormap). The frequencies of the gain feature and the $h$–CR absorption are close, as shown in the two THz spectra on the right, corresponding to the two vertical cuts at $E = 1.527$ eV and $E = 1.570$ eV, corresponding to the two vertical cuts marked by the black dashed lines in the map.

Pumping with higher $E$ photons creates excitons in excited states, such as $2s$ and $3s$; a Mott transition occurs for these excited excitons, and the resulting free heavy-hole CR absorption becomes bright (the absorption feature marked as “$h$–CR” in the colormap). The frequencies of the gain feature and the $h$–CR absorption are close, as shown in the two THz spectra on the right, corresponding to the two vertical cuts at $E = 1.527$ eV and $E = 1.570$ eV in the color map. However, when the gain feature appears in the resonant pumping case, the $h$–CR absorption is clearly absent, reflecting the absence of a Mott transition for $1s$ excitons.

Figure 2(a) shows Re($\sigma_-$) spectra at different temperatures. We kept the other experimental parameters at $E = 1.527$ eV (resonant pumping), $B = 7$ T, $F = 250$ nJ/cm$^2$, and $\tau = 15$ ps. There is a clear trend that the amplitude of the Re($\sigma_-$) < 0 dip due to the gain feature decreases with increasing $T$. At around 10 K, the gain feature disappears.

Furthermore, we performed pump fluence dependent measurements for quantifying the maximally achievable THz gain under NIR pumping with increasing intensity. Figure 3(b) shows Re($\sigma_-$) spectra at various pump fluences from $F = 25$ nJ/cm$^2$ to 750 nJ/cm$^2$ while we kept $E = 1.527$ eV (resonant pumping), $T = 2$ K, $B = 7$ T, and $\tau = 15$ ps. We fit these spectra with a model in which Re($\sigma_-$) consists of a Lorentzian dip at 0.38 THz (the gain feature), a Lorentzian peak at 1.75 THz (the $1s$–$2p_-$ transition), and a broadband background [34]. Integrated weights and linewidths of the gain and the $1s$–$2p_-$ transition peaks versus pump fluence. (d) Gain weight is proportional to the $1s$–$2p_-$ transition weight.

The gain feature appears at the smallest $F$ and grows with increasing $F$ until 250 nJ/cm$^2$. Above 250 nJ/cm$^2$, the gain feature saturates and eventually merges with the strong Re($\sigma_-$) > 0 background; the region where Re($\sigma_-)$ < 0 decreases and eventually disappears at 750 nJ/cm$^2$. The maximum gain coefficient achieved is 0.5 cm$^{-1}$, which appears at the center of the narrow gain band at 250 nJ/cm$^2$ (the photoexcited e-h pair density is 7.95 x 10$^9$ cm$^{-2}$ per well). The weight of the $1s$–$2p_-$ transition also shows saturation. When we plot the weight of the gain versus that of the $1s$–$2p_-$ transition, proportionality is seen; see Fig. 3(d). This relation suggests that the exciton $1s$ state is the USPI, assuming that the gain coefficient should be proportional to the inverted population difference. This conclusion agrees with the pump photon energy dependence study in Fig. 2. The observation also rules out impurity-related transitions to be the cause of the gain because impurities should saturate.

**FIG. 2.** Re($\sigma_-$) map versus THz frequency and pump photon energy at $T = 4$ K, $B = 7$ T, $F = 250$ nJ/cm$^2$, and $\tau = 15$ ps. The top panel shows a linear absorbance spectrum for the QW at the same $T$ and $B$. The right panel shows two THz spectra at $E = 1.527$ eV and $E = 1.570$ eV, corresponding to the two vertical cuts marked by the black dashed lines in the map.

**FIG. 3.** (a) Temperature dependence of Re($\sigma_-$) spectra obtained with $E = 1.527$ eV (resonant pumping), $B = 7$ T, $F = 250$ nJ/cm$^2$, and $\tau = 15$ ps. (b) Pump fluence dependence of Re($\sigma_-$) spectra obtained with $E = 1.527$ eV (resonant pumping), $T = 2$ K, $B = 7$ T, and $\tau = 15$ ps. (c) Integrated peak weights and linewidths of the gain and the $1s$–$2p_-$ transition peaks versus pump fluence. (d) Gain weight is proportional to the $1s$–$2p_-$ transition weight.
We succinctly summarize the essential experimental facts here. The gain feature is narrow-band, is tunable with \( B \), only appears for hCRA circular polarized radiation and at \( T < 10 \) K, and requires finite \( 1s \) state population. The last point shows that the \( 1s \) exciton state is the USPI, which narrows down the choices for the LSPI because the \( 1s \) state is the ground state of excitons. Any state that is energetically lower and has an excitonic origin must be from a more complicated \( e-h \) complex where an exciton binds to an additional object so that the total energy of the bound \( e-h \) complex is lower than its constituents in the state where they are free to move about. Following this logic, we discuss below a possible physical origin of the LSPI.

We propose that the LSPI is the biexciton ground state, and therefore, the THz gain transition is from the two-exciton state (denoted as \( 2X \)), describing two free \( 1s \) excitons, to the biexciton ground state (denoted as \( X_2 \)). As shown in the schematic diagram in Fig. 4(a), when the NIR pump resonantly creates excitons (\( X \)), the \( 2X \) state is automatically populated. At \( B \neq 0 \), the \( 2X \) state shifts, and splits into a multiplet due to several choices of the spin alignment of the two excitons. The NIR pump populates the multiplet evenly, but the exciton spin rapidly decays within \( \tau \) so that the lowest state of the multiplet, \( |↓↓⟩ \), is most populated when the THz probe arrives. The gain transition is from \( |↓↓⟩ \) to \( X_2 \). Whether or not such a transition is able to cause stimulated emission, or even radiatively allowed, has not been discussed or investigated in previous studies. Note that this process has some similarity to the \( X_2 - X \) population inversion observed in quantum wires [31], which, however, required the \( X_2 \) density to be higher than the \( X \) density and the gain appeared in the NIR. Below, we provide additional supporting evidence that our proposed scenario can explain our experimental observations, assuming that this radiative transition is allowed.

First, based on this scenario, the energy of the THz gain should be the biexciton binding energy \( E_B \), which can be measured independently by photoluminescence (PL) spectroscopy. Figure 4(b) shows PL spectra at \( T = 4 \) K in \( B \) up to 6 T. There are peaks due to biexcitons (brown arrows) and bound excitons (green arrows) on the lower energy side of the free exciton \( 1s \) peak (black arrows) at \( T < 10 \) K, which is the \( T \) range in which the THz gain was observed. See Ref. [31] for details of PL peak assignments. \( E_B \) measured from Fig. 4(b) is plotted against \( B \) in Fig. 4(c) together with the center frequency of the observed THz gain feature. The trend that the gain frequency increases with \( B \) matches the behavior of \( E_B \) determined from PL measurements, but \( E_B \) is slightly larger in frequency overall. We explain this discrepancy by noting that the \( E_B \) in PL measurements is estimated by the energy difference between the biexciton peak and the center of the free exciton \( 1s \) peak, which is also the center of the \( 2X \) multiplet, while the gain is originating from \( |↓↓⟩ \), which is the lower edge of the multiplet. This explains a slight overestimation of \( E_B \).

Second, we examine how our proposed scheme can explain the robust polarization selection rule of the THz gain. It is known that the LSPI, namely, the \( X_2 \) state, is a spin singlet state with zero total angular momentum. Therefore, the polarization selection rule of the gain transition is determined by the spin angular momentum carried by the \( |↓↓⟩ \) state in Fig. 4(a). Both the sign of the exciton \( g \) factor [37] and hole-angular-momentum mixing [38] due to valence band hybridization are important. We took both factors into consideration and confirmed that the wavefunction of the \( |↓↓⟩ \) state possesses a component that only supports THz gain for the hCRA polarization, while no gain is expected for the eCRA polarization [34].

In summary, by performing optical-pump-THz-probe experiments on undoped GaAs quantum wells at low temperatures in a strong perpendicular magnetic field,
we observed a narrow-band, circularly polarized THz gain feature whose frequency shifts with the magnetic field. From systematic experiments as a function of magnetic field, temperature, pump fluence, pump photon energy, and probe polarization, we confirmed that the upper state of population inversion is the 1s exciton state. We described a possible scenario of the biexciton ground state being the lower state of population inversion and proposed a specific transition scheme for the gain to appear. Our observations not only provide new insight into the physics of many-exciton states and radiative transitions in a complex 2D e-h system in a strong magnetic field but also open up possibilities for developing tunable THz lasers based on 2D magnetoeexcitons.

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