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Intersubband exchange interaction induced by optically excited electron spins in GaAs/AlGaAs quantum wells

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Spin-dependent intersubband excitonic interactions have been investigated in GaAs/AlGaAs quantum wells by two-color pump and probe spectroscopy. We generated spin-polarized electrons in the lowest subband by resonant excitation of the heavy-hole exciton (E_1-HH_1) and observed polarization-dependent broadening of the second-subband exciton resonance (E_2-HH_2 and E_2-LH_1). The exchange interaction between the first and the second-subband excitons is found to play a crucial role in polarization-dependent spectral modulation as well as spin-independent Coulomb screening. © *2009 American Institute of Physics*. [DOI: 10.1063/1.3118584]

Polarization-dependent optical properties of semiconductors have been of great interest and extensively studied not only to manifest the physics of spin-related phenomena[1] but also to utilize the spin degree of freedom for application to novel spintronics devices.[2] In this context, optical nonlinearity and circular dichroism induced by spin-polarized electrons or excitons are important and essential. Recently, many experiments of time-resolved optical spectroscopy have explored exciton[3–7] and spin[8–11] dynamics in semiconductors and their quantum structures in depth. The mechanisms responsible for modulation of exciton resonance absorption at low excitation levels and low temperatures are phase space filling (PSF) and Coulomb screening (CS) which can be separated into short-ranged exchange interaction and long-ranged Coulomb correlation between exciton-exciton and free electron-hole pairs.[12,13] The PSF and the exchange interaction originate from the Pauli exclusion principle and are spin-dependent, while long-ranged CS is principally spin-independent. The PSF and exchange interaction cause bleaching and broadening of exciton absorption, and also reduce the exciton binding energy, which result in a blueshift in excitonic resonance.[4,5] As for the intersubband excitonic interaction, the most relevant effect is considered to be long-ranged CS rather than the exchange interaction[7] because of small overlap of the first and second-subband exciton wave functions.[7,14] It is quite intriguing to control the polarization dependence of excited excitonic states by optical or electrical spin injection. As far as we know, however, there is no report on the experimental study of polarization dependence of intersubband excitonic interaction in the presence of spin-polarized electrons or excitons. In this letter, we investigate the modulation of the second-subband exciton resonance via exchange interaction with photoexcited electron spins at the first subband in GaAs/AlGaAs quantum wells (QWs) by two-color pump-probe measurements.

The sample studied here was grown on (001) semi-insulating GaAs substrate by molecular beam epitaxy. It consists of 60 periods of 11-nm-thick undoped GaAs QWs separated by 10-nm-thick $Al_{0.3}Ga_{0.7}As$ barriers. For transmission and absorption measurements, the GaAs substrate was removed by selective chemical etching. Figure 1 shows the linear absorbance $\alpha L = -\ln(I_t/I_0)$ of the sample taken at 4.5 K. Here, I_t and I_0 are transmitted and incident light intensities, respectively. The first subband electron-heavy-hole (E_1-HH_1) and light hole (E_1-LH_1) exciton resonance peaks are observed at 1.543 and 1.548 eV, respectively. The absorption peaks at higher energies (1.639 and 1.652 eV) are the second-subband electron and the first subband light hole (E_2-LH_1), and the second-subband heavy-hole (E_2-HH_2) exciton resonance, respectively.

In two-color pump-probe measurements, we used synchronized two mode-locked $Ti:Al_2O_3$ lasers to generate pump and probe pulses (~110 fs) at 76 MHz. The pump beam was circularly polarized through a quarter-wave plate and the energy was tuned at the E_1-HH_1 resonance (1.543 eV). The intensity of the pump beam was 5 mW and the

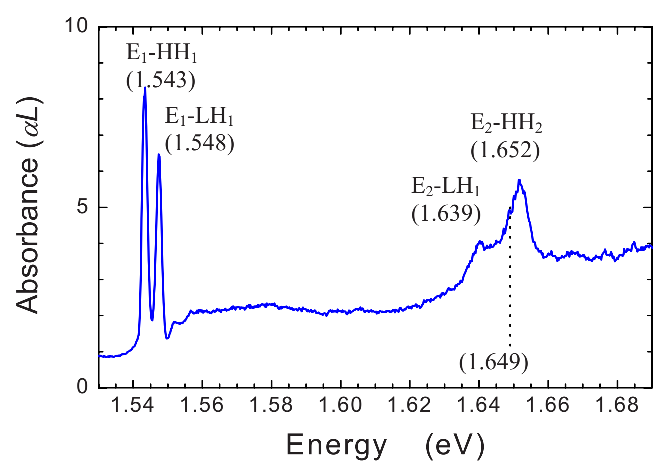

FIG. 1. (Color online) The absorption spectrum of the QWs taken at 4.5 K. Each absorption peak (with energy value) is labeled to the corresponding exciton resonance. The dotted line at 1.649 eV is an eye-guide [see also Fig. 2(c)].

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focused spot size on the sample was around 100 μm in diameter. Roughly estimating, about a few 10^{10} cm^{-2} excitons were generated in each QW by a single pump pulse. Because the hole spin relaxation time is sufficiently short, the polarization-dependent optical response is due to the electron spins. In time-resolved Faraday-rotation (TRFR) measurements,[10] the probe beam was linearly polarized and its Faraday-rotation angle θ_F caused by spin-dependent dichroism in the QWs was detected by a balanced detector. In polarization-dependent transmission measurements, on the other hand, the probe beam was right-(σ^+) or left-(σ^-) circularly polarized through another quarter-wavelength wave plate, and the transmission $[I_t(\sigma^{\pm})]$ or differential transmission $[I_{dt}(\sigma^{\pm})]$ were detected by a lock-in technique. The time delay Δt between pump and probe pulses were controlled by a mechanical delay line. Both pump and probe beams were fed into a cryostat with superconducting magnet, in which the sample was set in Voigt geometry.

First, we studied the spin relaxation time T_1 and the spin decoherence time T_2^* in the lowest subband (E_1) by TRFR measurements. We tuned the photon energies of both pump and probe beams at E_1-HH$_1$ resonance, and measured θ_F as a function of Δt. Figure 2(a) shows a semilog plot of $\theta_F(\Delta t)$. When $\Delta t < 50$ ps, a fast decay component (time constant $\sim$25 ps) is observed, which may be attributed to the hole spin relaxation. The remaining electron spins relax with $T_1 \sim 110$ ps, which is much shorter than the recombination lifetime of electron-hole pairs ($\sim$2.7 ns). By applying an in-plane magnetic field B, we monitored the Larmor precession of electron spins. Figure 2(b) shows $\theta_F(\Delta t)$ measured at $B=4$ T when the sample was excited by σ^+ (squares) and σ^- (circles) polarized pump beams. The exponentially decaying oscillations of $\theta_F(\Delta t)$ reveal the absolute value of the electron g-factor $|g|=0.236$, and $T_2^* \sim 100$ ps.

Next, we carried out two-color TRFR measurement at $B=4$ T by setting the probe energy at 1.649 eV, at which θ_F is maximum in the energy range between E_2-LH$_1$ and E_2-HH$_2$ resonance (indicated by a dotted line in Fig. 1). In Fig. 2(c), the data of $\theta_F(\Delta t)$ under excitation with σ^+ and σ^- polarized pump beams are shown in the same manner as Fig. 2(b). We observed clear exponentially decaying oscillations similar to those when the E_1-HH$_1$ state is probed. When the probe energy is set away from the exciton absorption peaks, on the other hand, almost no θ_F is observed. This clearly indicates that the imbalance of the spin population in the E_1 level results in the polarization-dependent second-subband (E_2) exciton resonance. It should be noted that in Figs. 2(b) and 2(c) the phase of TRFR oscillations with σ^+ and σ^- excitation are altered, in spite of the fact that the probe energy [1.649 eV in Fig. 2(c)] is closer to E_2-HH$_2$ than E_2-LH$_1$ exciton resonance. The sign of the Faraday rotation depends on circular birefringence, i.e., difference in the refraction indices for σ^+ (n^+) and σ^- polarized lights (n^-), $n^+ - n^-$. The sign change in the signals shown in Figs. 2(b) and 2(c) is a result of the sign of $n^+ - n^-$ at 1.543 and 1.649 eV. This can be seen from the sign of $d\Delta\alpha/dE$ (approximately proportional to $n^+ - n^-$ in the present case), where $\Delta\alpha$ is the absorption coefficient difference between the two polarizations, at E_1-HH$_1$ peak at 1.543 eV (Fig. 3) and at 1.649 eV [Fig. 4(b)].

In the following, we consider the mechanisms responsible for the polarization dependence of exciton resonance

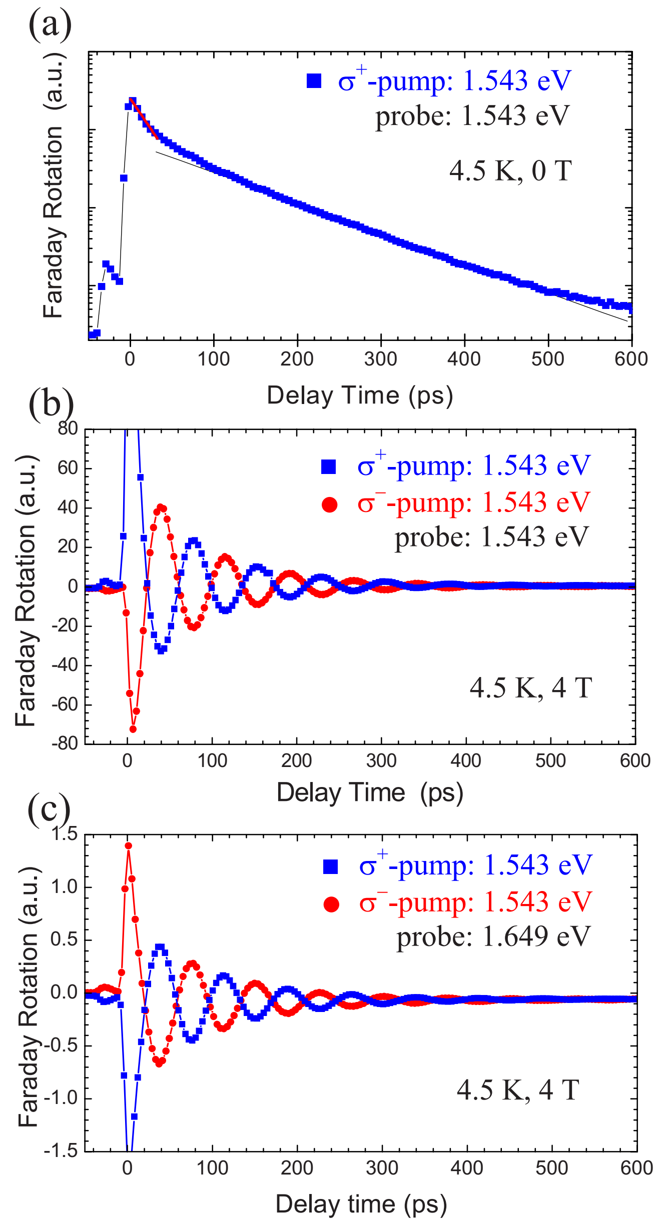

FIG. 2. (Color online) Results of TRFR measurements. (a) $\theta_F(\Delta t)$ measured at $B=0$ T with pump and probe energies set at 1.543 eV, in which a response seen at $-50 < \Delta t < 0$ ps is most likely an interference effect, and (b) $B=4$ T. (c) $\theta_F(\Delta t)$ measured at $B=4$ T with the probe energy set at 1.649 eV (indicated by dotted line in Fig. 1).

and the difference between occupied (E_1) and unoccupied (E_2) levels. First, we investigate the exciton absorption peaks of E_1-HH$_1$ and E_1-LH$_1$ after resonant excitation of E_1-HH$_1$ exciton by σ^+-pump beam. We measured the probe-energy dependence of transmission $I_t(\sigma^{\pm})$ at $\Delta t=40$ ps. Figure 3 shows the absorbance $-\ln[I_t(\sigma^{\pm})/I_0(\sigma^{\pm})]$ in log-scale to see closely the difference between $I_t(\sigma^+)$ (squares) and $I_t(\sigma^-)$ (triangles). The inset shows the vertically expanded figure around the E_1-LH$_1$ resonance. As a reference, we also show the absorbance before excitation of E_1-HH$_1$ exciton by a pump pulse ($\Delta t=-20$ ps), which was taken by measuring the transmitted probe intensity with both pump and probe beams linearly polarized. One can see the E_1-HH$_1$ and E_1-LH$_1$ resonance peaks become smaller and blueshifted at $\Delta t=40$ ps, while the change in the line-widths of their peaks is not clearly observed. In addition, we observed energy dif-

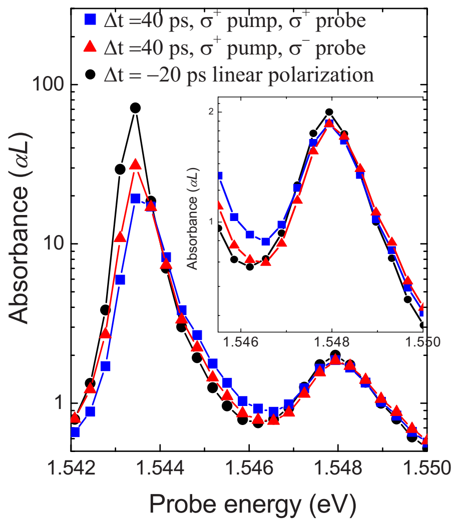

FIG. 3. (Color online) (a) Log-scale plots of the absorbance at Δt=40 ps for σ^+ (squares) and σ^- (triangles) polarized probe beams in the range of E_1-HH_1 to E_1-LH_1 exciton resonance. The inset is an extension around E_1-LH_1. The absorbance taken before the excitation (Δt=−20 ps) with both pump and probe linear-polarized is shown (circles) as reference.

ference between σ^+ and σ^- absorption peaks for E_1-HH_1 (~100 μeV) and E_1-LH_1 (~50 μeV), respectively. The decrease and blueshift in E_1-HH_1 exciton resonance peaks can be attributed to the PSF and exchange interaction between excitons. The polarization-dependent energy splitting of E_1-HH_1 and E_1-LH_1 can also be explained by the exchange interaction.[4,5,9]

Next, we investigate the intersubband excitonic interaction. Compared to the case of the first subband excitons, the polarization-dependent modulation for the second-subband exciton is considerably smaller. Thus we measured the differential transmission $I_{dt}(\sigma^{\pm})$ by measuring the change in the transmitted probe beam caused by the σ^+-excitation of E_1-HH_1 excitons. Figure 4(a) shows $I_{dt}(\sigma^{\pm})(\Delta t)$ at E_2-HH_2 exciton resonance (1.652 eV). At $\Delta t \sim 0$ ps, both $I_{dt}(\sigma^{\pm})$ increases due to the spin-independent CS.[11] The difference between $I_{dt}(\sigma^+)$ and $I_{dt}(\sigma^-)$ is seen up to $\Delta t \sim 350$ ps. In Fig. 4(b), $I_{dt}(\sigma^{\pm})$ at Δt=40 ps are plotted as a function of probe energy by squares $[I_{dt}(\sigma^+)]$ and triangles $[I_{dt}(\sigma^-)]$. The oscillatory features common to both $I_{dt}(\sigma^+)$ and $I_{dt}(\sigma^-)$ around E_2-LH_2 and E_2-HH_2 resonance peaks originate from the broadening of the exciton absorption by long-ranged CS.[7] In addition, the difference in the amplitudes of $I_{dt}(\sigma^{\pm})$ at E_2-HH_2 and E_2-LH_1 exciton resonance is clearly observed. This shows that the spin-dependent intersubband exchange modulates the second-subband excitonic states. Spin-dependent energy shift is, however, not resolved for E_2-LH_1 and E_2-HH_2 resonance peaks. This can be understood by the fact that in the present experiment the density of the photoexcited carriers is of the order of 10^{10} cm^{-2}, while PSF and intersubband exchange renormalization become significant when the carrier density is as high as 10^{12} cm^{-2}.[4,14]

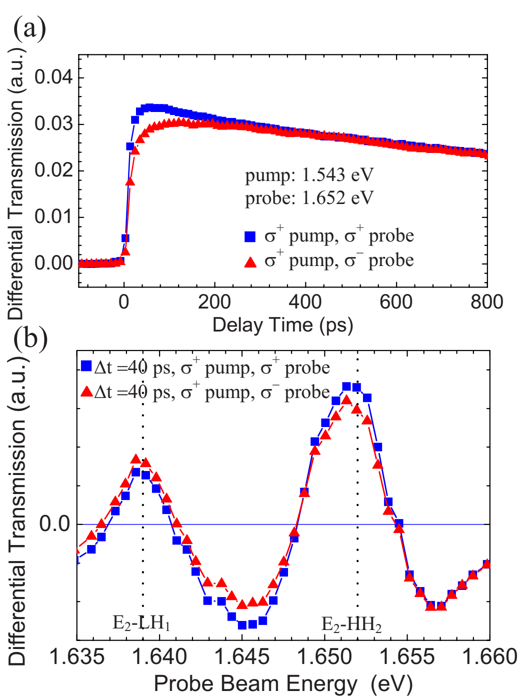

FIG. 4. (Color online) (a) Differential transmission $I_{dt}(\sigma^{\pm})$ at probe energy=1.652 eV with σ^+ pump (1.543 eV). (b) $I_{dt}(\sigma^{\pm})$ at Δt=40 ps are shown in the range of E_2-LH_2 and E_2-HH_2 exciton resonance.

In conclusion, we investigated spin-dependent intersubband excitonic interactions in GaAs/AlGaAs (100) QWs by two-color pump and probe spectroscopy. We found polarization-dependent broadening but no energy shift in the second-subband exciton resonance, which indicate that the short-range exchange interaction plays crucial role in intersubband excitonic interactions.

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