Vanishing and Emerging of Absorption Quantum Beats from Electron Spin Coherence in GaAs Quantum Wells

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We report experimental studies of absorption quantum beats induced by electron spin coherence in GaAs quantum wells. Absorption quantum beats occur for strongly localized excitons, but nearly vanish for mobile excitons in the third order nonlinear optical response. Pronounced quantum beats for mobile excitons emerge in an unusual fifth order process. These results, along with a qualitative analysis based on the use of N-exciton eigenstates, elucidate how the manifestation of electron spin coherence in the excitonic nonlinear optical response can differ fundamentally from that in an atomic system.

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Recent optical studies on electron spin coherence in semiconductors have shown that spin coherence can be preserved over remarkably long time and length scales [1]. This is in marked contrast to most other forms of optically induced quantum coherences in semiconductors, which are extremely fragile. The robust electron spin coherence provides a promising platform for pursuing optical manipulation and control of quantum coherences in semiconductors and for developing coherent semiconductor quantum devices [2]. Of special importance to these efforts is the understanding of how electron spin coherence manifests in nonlinear optical processes.

Coherent nonlinear optical processes in a semiconductor are strongly influenced by inherent many-body Coulomb interactions and differ greatly from those in a dilute atomic system, as shown by extensive studies using excitonic coherences in quantum wells (QWs) [3,4]. These studies, however, are limited by the extremely short lifetime of the excitonic coherence. In this regard, the robust electron spin coherence also provides us with a special model system to further explore and deepen our understanding of coherent optical interactions in semiconductors.

Electron spin coherence as a nonradiative coherent superposition can lead to coherent oscillations or quantum beats (QBs) in a variety of transient optical processes. QBs occurring in time-resolved photoluminescence and especially time-resolved Faraday rotation have been used with remarkable success to probe the dynamics of the spin coherence [5,6]. In comparison, QBs in transient differential absorption (DA), or pump probe, are highly sensitive to details of the underlying nonlinear optical interactions and are thus a natural probe of these interactions [7–10].

In this Letter, we report experimental studies of absorption QBs induced by electron spin coherence in GaAs QWs. In these QWs, interface fluctuations can lead to strong localization of excitons in the low energy tail of the exciton absorption. Excitons near the line center, however, remain mobile with a localization length much greater than the exciton Bohr radius [11–13]. We show that, while strongly localized excitons behave like an atomic system, mobile excitons feature behaviors that are fundamentally different from those of an atomic system. For these excitons, the QBs nearly *vanish* in the lowest order ($\chi^{(3)}$ or third order) nonlinear optical response, but emerge in a fifth order, or $\chi^{(5)}$, response with an unusual dependence on the probe intensity. A qualitative analysis of the experimental results using the approach of N-exciton eigenstates [14] further illustrates how exciton-exciton (x-x) interactions, especially the absence of these interactions, profoundly affect the manifestation of electron spin coherence in coherent nonlinear optical processes.

For a GaAs QW subject to a relatively weak magnetic field in the plane of the QW, J_z for the heavy-hole (hh) valence band remains an approximate good quantum number near the band edge. The electron spin in the s-like conduction band, however, lines up with the external magnetic field. Coupling the two electron spin states to the same hh valence band via two dipole-optical transitions can induce an electron spin coherence, as shown schematically in Fig. 1(a) where we take the optical fields to be σ-polarized and thus include only the $J_z = 3/2$ *hh* band.

The electron spin coherence can be probed with transient DA that measures the change in the absorption of a

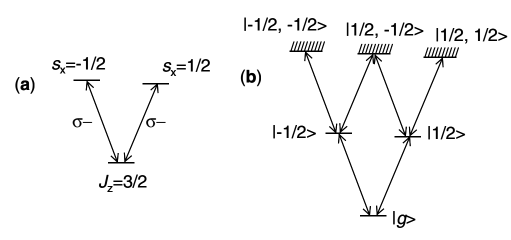

FIG. 1. (a) Dipole selection rules for transitions between the conduction and the $J_z = 3/2$ valence bands in a weak magnetic field along the x axis. (b) Schematic for the energy eigenstates of the ground, one-exciton, and two-exciton states. The states are labeled only by the electron spins.

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probe induced by a pump. In this experiment, the pump excites an electron spin coherence. The nonradiative coherence forms in the second order of the pump field and oscillates with a frequency determined by the Zeeman splitting of the two electron spin states. The probe interacts with the spin coherence, inducing a nonlinear polarization propagating along the same direction. The optical field generated by this polarization is homodyne detected with the transmitted probe field. The DA response versus the delay between the pump and probe can thus measure directly the temporal evolution of the spin coherence.

The experimental studies were carried out at 10 K on four different undoped (001) GaAs QW samples grown by molecular beam epitaxy. Qualitatively the same results have been obtained in all four samples. Here, we present the results from two samples. Sample A (B) contains 15 (1) periods of 13 (17.5) nm GaAs wells and 15 nm $Al_{0.3}Ga_{0.7}As$ barriers. The samples, etched and glued onto a sapphire disk, were mounted in Voigt geometry in a superconducting magnet. To separate contributions from the mobile and the strongly localized excitons, we used an external pulse shaper to narrow the bandwidth of the optical pulse from a mode-locked Ti:sapphire laser (82 MHz repetition rate). The resulting nearly transform limited pulse has a bandwidth of 0.35 nm and a duration of 4 ps. Both the pump and probe had the same circular polarization. Note that we have carried out additional studies to confirm the well-known behaviors of exciton localization [11–13]. The mobile excitons near the *hh* exciton absorption line center feature a diffusion coefficient and a dephasing rate much greater than those of the strongly localized excitons at the low energy absorption tail.

Figure 2 shows transient DA responses obtained from sample A. The inset also shows the linear transmission spectrum and the spectra of the laser pulse. As shown in Fig. 2(a), pronounced long-lived oscillations were observed for hh excitons at the low energy absorption tail. The oscillation frequency scales linearly with the magnetic field with the electron g factor, $|g_e| = 0.26$. These oscillations are absorption QBs induced by the electron spin coherence. In contrast, for DA studies near the line center, the QBs vanished under otherwise similar conditions, as shown in Fig. 2(b). The vanishing of the QBs is unexpected based on atomiclike considerations of the three-level system shown in Fig. 1(a).

The long-lived component in the DA response for mobile excitons is due to a quasithermal exciton population formed via exciton-phonon scattering, *x*-*x* scattering, and spin relaxation of holes. The lifetime of this quasithermal population at 10 K can range from a few hundred ps to ns, depending on the well width [15]. Electron spin coherence, however, is expected to persist in spite of these relaxation processes.

The intensity dependence of the DA response, especially the dependence on the probe intensity, further reveals the unusual behavior of absorption QBs induced

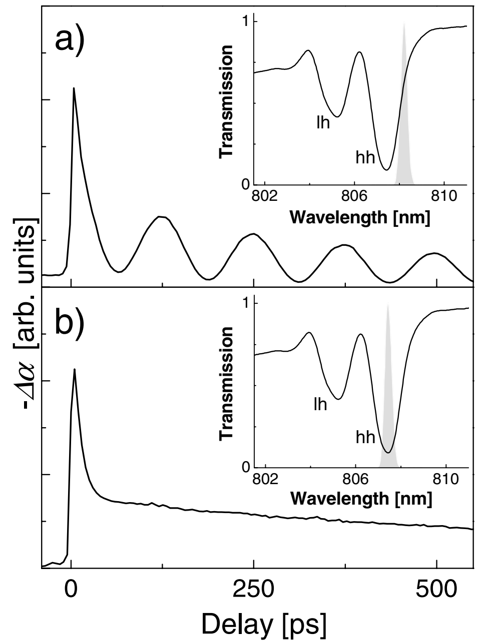

FIG. 2. DA response from sample A at $B = 2$ T for (a) strongly localized and (b) mobile excitons, with an average intensity of $I_{pump} = 10I_{probe} = 8$ W/cm^2. The inset shows the transmission spectrum and the laser spectra used for the DA study, where hh and lh denote the heavy-hole and light-hole exciton resonance, respectively.

by the electron spin coherence. Note that in the $\chi^{(3)}$ limit, $\Delta\alpha$ scales linearly with the pump intensity but is *independent* of the probe intensity. The corresponding nonlinear polarization scales quadratically with the pump field and linearly with the probe field.

For strongly localized excitons and at low pump and probe intensities, the amplitude of the QBs scales linearly with the pump intensity and is nearly independent of the probe intensity, as shown in Figs. 3(a) and 3(b). This indicates that the QBs result from a $\chi^{(3)}$ process. At higher pump and probe intensities, the DA response saturates. The amplitude of the QBs becomes sublinear with the pump intensity and decreases with increasing probe intensity.

In contrast, for mobile excitons the QBs, which at low probe intensities nearly vanish, emerge with increasing probe intensity, as shown in Fig. 3(c). The amplitude of the QBs rises linearly with the probe intensity and becomes sublinear at higher probe intensities. A linear dependence of the QB amplitude on the probe intensity means that the corresponding nonlinear polarization scales with the third order of the probe field.

In spite of the striking difference in the probe intensity dependence, the pump intensity dependence for mobile excitons is qualitatively the same as that for strongly localized excitons, as shown in Fig. 3(d). The observed linear pump intensity dependence confirms that in both cases electron spin coherence forms in the second order of the pump field.

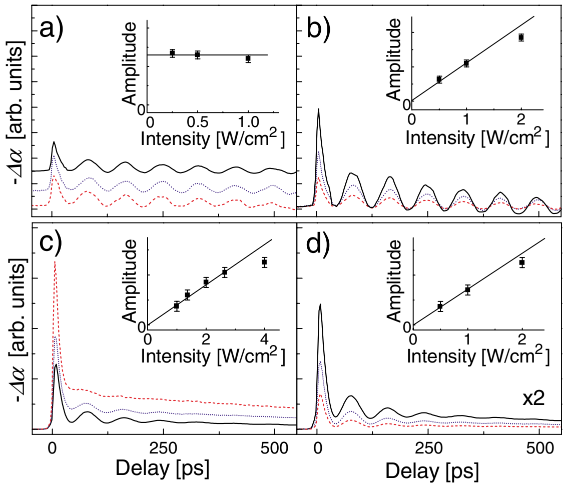

FIG. 3 (color online). The probe and pump intensity dependence of the DA response from sample A for strongly localized (a),(b) and mobile (c),(d) excitons at $B = 3$ T. (a) $I_{pump} = 0.5$ W/cm^2 and $I_{probe} = 0.25$, 0.5, 1 W/cm^2 (the baselines are shifted for display clarity). (b) $I_{probe} = 1$ W/cm^2 and $I_{pump} = 0.5$, 1, 2 W/cm^2. (c) $I_{pump} = 2$ W/cm^2 and $I_{probe} = 1$, 2, 4 W/cm^2. (d) $I_{probe} = 4$ W/cm^2 and $I_{pump} = 0.5$, 1, 2 W/cm^2. Dashed, dotted, and solid curves correspond to increasing intensities for either probe or pump. The insets show the corresponding probe and pump intensity dependence of the QB amplitude (in arbitrary units) obtained from a numerical fit, with the solid line as a guide to the eye.

The probe intensity dependence shown in Fig. 3(c) is not due to effects of propagation or strong absorption at the exciton line center [9]. Qualitatively the same results have been observed in QWs with different thickness and especially in single QWs. Sample B, which is a single QW, exhibits a probe intensity dependence similar to that in Fig. 3(c), as shown in the inset of Fig. 4. Figure 4 also shows that at high probe intensities, absorption QBs can dominate the DA response ($|g_e| = 0.31$ was obtained for sample B).

The observation that the QB amplitude increases with probe intensity as shown in Figs. 3(c) and 4 is highly unusual. DA responses arise from excitations induced by the pump but not the probe. For most nonlinear optical processes, increasing the probe intensity saturates the DA response. As shown in Fig. 3(c), while the amplitude of the QBs rises with the probe intensity, the rest of the DA response saturates strongly and decreases with increasing probe intensities.

To understand the striking difference in the absorption QBs between the strongly localized and the mobile excitons, we present a qualitative analysis based on the use of N-exciton eigenstates [14]. As shown in Fig. 1(b), we label the N-exciton energy eigenstates using only the spins of the electrons since the hole state is the same for the excitons involved: $|g\rangle$ is the ground state, $|1/2\rangle$ and $|-1/2\rangle$ are one-exciton states with electron spin $s_x = 1/2$ and $-1/2$, respectively, $|1/2, 1/2\rangle$, $|1/2, -1/2\rangle$, and $|-1/2, -1/2\rangle$ are the relevant two-exciton states. In this notation, the electron spin coherence corresponds to a coherent superposition of $|1/2\rangle$ and $|-1/2\rangle$. To the first order in the probe field, the probe can interact with the spin coherence via $|g\rangle$ and $|1/2, -1/2\rangle$, states that couple to both $|1/2\rangle$ and $|-1/2\rangle$ through dipole-optical transitions as shown in Fig. 1(b).

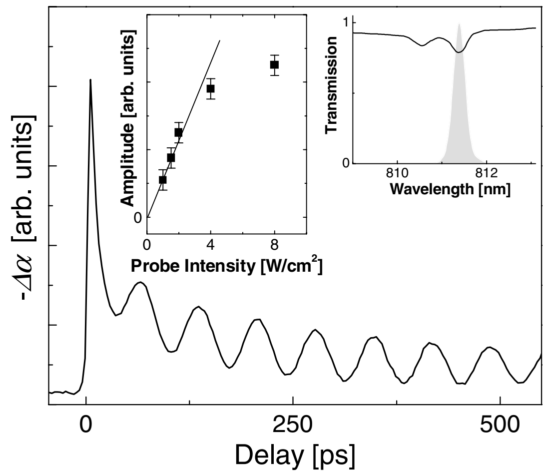

FIG. 4. DA response from sample B at $B = 3$ T and with $I_{probe} = 15 I_{pump} = 12$ W/cm^2. The insets show the transmission spectrum, the laser spectrum, and the probe intensity dependence of the QBs amplitude at $I_{pump} = 0.8$ W/ cm^2 with the solid line as a guide to the eye.

Absorption QBs induced by the electron spin coherence depend critically on the interaction between the two excitons in $|1/2, -1/2\rangle$. For strongly localized excitons, the strong *on-site* repulsion between the holes prevents the formation of $|1/2, -1/2\rangle$. The absorption QBs thus behave like those in a three-level system, as evidenced in Figs. 3(a) and 3(b). In contrast, for mobile excitons, the localization length is large compared with the exciton Bohr radius and two-exciton states thus become accessible. Interactions between mobile excitons underlying these two-exciton states depend on the relative spin orientation of the electrons and holes. If the electrons have the same spin and the holes also have the same spin, the exchange interaction between the electrons and that between the holes can lead to an overall repulsive *x-x* interaction. If the electrons have opposite spin and the holes also have opposite spin, the attractive Coulomb correlation between excitons can lead to the formation of a bound two-exciton state (biexciton). For $|1/2, -1/2\rangle$, the electrons have opposite spin, but the holes have the same spin. In this case, the attractive Coulomb correlation can to a large extent be *canceled* by the exchange repulsion between the holes.

The absence of a strong *x-x* interaction between the two excitons in $|1/2, -1/2\rangle$ can profoundly change the manifestation of the electron spin coherence in the DA response. Most notably, the nonlinear polarization induced by coupling the probe field to the spin coherence via $|g\rangle$ can destructively interfere with that induced via

$|1/2, -1/2\rangle$. Complete destructive interference occurs in the limit that the transitions between $|g\rangle$ and the one-exciton states are identical to the respective transitions between the one-exciton states and $|1/2, -1/2\rangle$. In this ideal noninteracting boson limit, the spin coherence can still be excited by the pump. The nonlinear response from the spin coherence, however, vanishes. A similar type of destructive interference has also been discussed in the context of absorption QBs from intervalence band coherence, although in that case strong *x-x* interactions are still present [8].

Details of the above nonlinear optical process can be illustrated by using phenomenological density matrix equations (for brevity, we use $|+\rangle$, $|-\rangle$, and $|a\rangle$ to denote $|1/2\rangle$, $|-1/2\rangle$, and $|1/2, -1/2\rangle$, respectively). In the $\chi^{(3)}$ limit, the interaction of the probe with the electron spin coherence, $\rho^{(2)}_{-+}$, induced by the pump, leads to two pairs of density matrix elements: $\rho^{(3)}_{-g}$, $\rho^{(3)}_{a+}$, and $\rho^{(3)}_{+g}$, $\rho^{(3)}_{a-}$. Including only processes related to the electron spin coherence, we can write the phenomenological equations of motion for $\rho^{(3)}_{-g}$ and $\rho^{(3)}_{a+}$ as

$$\dot{\rho}^{(3)}_{-g} = -(i\omega_1 + \gamma_1)\rho^{(3)}_{-g} - i\mu_{+g}E_{pr}\rho^{(2)}_{-+}/\hbar, \tag{1a}$$

$$\dot{\rho}^{(3)}_{a+} = -(i\omega_2 + \gamma_2)\rho^{(3)}_{a+} + i\mu_{a-}E_{pr}\rho^{(2)}_{-+}/\hbar, \tag{1b}$$

where μ_{+g}, ω_1, γ_1, and μ_{a-}, ω_2, γ_2 are the dipole matrix element, resonance frequency, and dephasing rate for the respective one-exciton and two-exciton transitions. As shown in Eq. (1), the term that couples the probe field, E_{pr}, to $\rho^{(2)}_{-+}$ has the opposite sign for $\rho^{(3)}_{-g}$ and $\rho^{(3)}_{a+}$, leading to the destructive interference between the corresponding nonlinear polarizations. Similar destructive interference also occurs between nonlinear polarizations induced by $\rho^{(3)}_{+g}$ and $\rho^{(3)}_{a-}$.

Note that complete destructive interference is not expected because of residual *x-x* interactions underlying $|1/2, -1/2\rangle$ and also because of band filling arising from the fermionic nature of electrons and holes. Nevertheless, Figs. 3(c) and 4 indicate that in the $\chi^{(3)}$ limit, the absorption QBs are negligible compared with the overall DA response, reflecting the fact that band filling plays only a minor role in excitonic nonlinear optical response as shown in extensive earlier studies [4].

In the limit of a vanishing $\chi^{(3)}$ response from the electron spin coherence, higher order nonlinear optical processes become important. In an order-by-order perturbation analysis, the probe can further couple to $\rho^{(3)}_{a+}$, $\rho^{(3)}_{a-}$, $\rho^{(3)}_{+g}$, $\rho^{(3)}_{-g}$ and then to the resulting fourth order density matrix elements through respective transitions including those that involve $|1/2, 1/2\rangle$ and $|-1/2, -1/2\rangle$, generating a nonlinear polarization to the third order of the probe field and a $\chi^{(5)}$ response. Strong destructive interference due to the weak *x-x* interaction underlying $|1/2, -1/2\rangle$ still persists in the higher order process. The $\chi^{(5)}$ response, however, can now arise from *strong x-x* interactions such as those inherent in $|1/2, 1/2\rangle$ and $|-1/2, -1/2\rangle$. The amplitude of the QBs in this $\chi^{(5)}$ response increases linearly with the probe intensity. The unusual intensity dependence shown in Figs. 3(c) and 4 thus provides us with the crucial information on how electron spin coherence manifests in the coherent nonlinear optical response from mobile excitons.

In summary, absorption QBs induced by electron spin coherence in a QW have shown that the manifestation of the spin coherence in coherent nonlinear optical processes in a semiconductor can differ fundamentally from that in an atomic system. While strongly localized excitons behave like an atomic system, absorption QBs from mobile excitons reveal behaviors that are unique to weakly interacting extended optical excitations. Previous nonlinear optical studies of excitonic coherences have emphasized the effects of strong many-body Coulomb interactions. For electron spin coherence, however, it is the *absence* of a strong underlying *x-x* interaction that leads to remarkable coherent nonlinear optical processes, including the vanishing of the QBs in the $\chi^{(3)}$ limit and the emerging of QBs in an unusual $\chi^{(5)}$ process. In this regard, electron spin coherence with its robustness and weak underlying *x-x* interactions opens up a new domain for exploring and understanding the interplay between quantum coherences and many-body interactions in an interacting many-particle system.

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