Spin Coherence and Electromagnetically Induced Transparency via Exciton Correlations

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We report experimental studies on exciton spin coherence induced via Coulomb correlations between excitons with opposite spins, including correlations associated with unbound as well as bound exciton pairs. Electromagnetically induced transparency resulting from the spin coherence is demonstrated in the transient optical response in GaAs quantum wells.

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Coherent nonlinear optical processes in semiconductors can provide an ideal laboratory for addressing the fundamental issue of how Coulomb correlations affect quantum coherences in a many-body system [1]. While recent experimental and theoretical studies have revealed important and ubiquitous roles of exciton-exciton correlations in these coherent processes [2–12], electromagnetically induced transparency (EIT) and related phenomena, in which destructive quantum interference plays an essential role, can provide a new and unique tool for probing the interplay between Coulomb correlations and quantum coherences in a highly nonlinear regime.

EIT reflects a simple and yet striking manifestation of quantum coherence and interference [13–15]. The essence of EIT is destructive interference arising from a nonradiative coherence. For the three-level system shown in Fig. 1(a), states $|1\rangle$ and $|2\rangle$ are coupled to state $|3\rangle$ via two resonant optical fields with Rabi frequency Ω_1 and Ω_2, respectively. The destructive quantum interference between the probability amplitudes associated with the $|1\rangle$ to $|3\rangle$ and the $|2\rangle$ to $|3\rangle$ transitions can lead to EIT and, in particular, a population trapped state $|\phi\rangle = \Omega^{-1}(\Omega_2|1\rangle - \Omega_1|2\rangle)$, where $\Omega = \sqrt{\Omega_1^2 + \Omega_2^2}$, that is decoupled from state $|3\rangle$. The basic physical process underlying EIT can also lead to phenomena such as lasing without inversion, slow light, adiabatic population transfer, and nonlinear optics at low light levels [13,14]. Successful observations of EIT and related phenomena in atomic systems have stimulated increasing efforts in extending these studies to semiconductors [16–19].

In this paper we report experimental studies on exciton spin coherence induced via Coulomb correlations between excitons with opposite spins. EIT resulting from the spin coherence is demonstrated in the transient optical response in GaAs quantum wells (QWs). The observation of EIT mediated via bound and especially unbound but still correlated exciton pairs demonstrates the importance of exciton-exciton correlations in a highly nonlinear regime and illustrates a unusual mechanism in which Coulomb correlations induce quantum coherences.

The band edge of a GaAs QW is characterized by the doubly degenerate conduction bands with $s = \pm 1/2$ and the doubly degenerate heavy hole (hh) and light hole (lh) valence bands with $j_z = \pm 3/2$ and $\pm 1/2$, respectively. Our studies were focused on the hh transitions that excite spin-up (σ^+ transition) and spin-down (σ^- transition) excitons, as shown in Fig. 1(b). Interactions between hh excitons with opposite spins take place through exciton-exciton correlations. These correlations can lead to the formation of bound and unbound exciton pairs (biexcitons). Effects of these correlations on optical processes can be described by microscopic theories based on dynamics controlled truncation schemes and also on the use of N-exciton many-body eigenstates [7–10].

As indicated schematically in Fig. 1(c), exciton spin coherence, i.e., coherent superposition of $|+\rangle$ and $|-\rangle$, can be induced by coupling $|-\rangle$ to $|+-\rangle_b$ or $|+-\rangle_u$ with a σ^+ polarized optical field and $|+\rangle$ to $|+-\rangle_b$ or $|+-\rangle_u$ with a σ^--polarized optical field, where $|+\rangle$, $|-\rangle$, $|+-\rangle_b$, $|+-\rangle_u$ denote the one-exciton (spin-up and spin-down) and the exciton pair (bound and unbound) states, respectively. The spin coherence arises from correlations between excitons with opposite spins. No spin coherence can be induced if only one-exciton states are involved [9,10]. The spin coherence also vanishes if exciton pair states can be factorized into a product state of single excitons. Note that using spin coherence for EIT differs

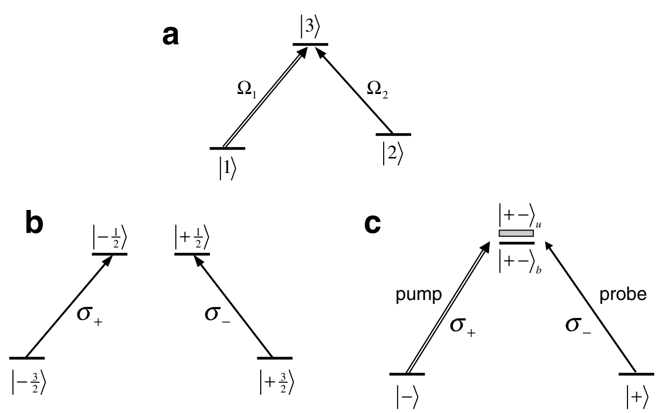

FIG. 1. (a) A Λ-type three-level system for EIT. (b) Level diagram for the hh exciton transition. (c) Transitions between the one-exciton and two-exciton states.

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from the earlier proposal based on the use of Raman coherence between two valence bands [19].

The samples studied were (001) GaAs/AlGaAs QWs grown by molecular beam epitaxy. Three different structures with well width of 10, 13, and 17.5 nm were used and qualitatively the same results were obtained. For brevity, we present here results obtained from the sample that contains 10 periods of 10 nm GaAs well and 10 nm $Al_{0.3}Ga_{0.7}As$ barrier. For transient optical studies, a pump-probe configuration was used. The pump and probe beams were derived from a mode-locked Ti:sapphire laser with a repetition rate of 82 MHz. External pulse shapers were used for the generation of nearly transform-limited pulses with the desired spectral bandwidth and duration. For the data presented here, the durations of the pump and probe pulses were 6 ps and 150 fs, respectively. The spot sizes of the pump and probe were 3×10^{-5} and 4×10^{-6} cm^2, respectively, and the probe energy flux was less than 1/100 of the pump. All measurements were performed at 10 K.

Figure 2(a) shows experimental results where we measured the absorption of a weak probe beam with σ^- circular polarization by spectrally resolving the probe after its propagation through the sample. The sample was excited with a pump beam resonant with the hh excitonic transition. The absorption spectrum with both the pump and probe having the same circular polarization is shown as the dashed curve in Fig. 2(a). The magnitude of the optical Stark splitting observed is in agreement with earlier spectral and time domain studies [20,21] and with the Rabi oscillation period obtained in separate time domain measurements (not shown). The absorption spectrum when the pump and probe have the opposite circular polarization but with other conditions unchanged is shown as the solid curve in Fig. 2(a). In addition to the biexcitonic resonance below the hh exciton resonance, a pronounced absorption dip occurs in the σ^- exciton resonance. Figures 2(b) and 2(c) further show that the wavelength of the absorption dip follows that of the pump and thus satisfies the characteristic two-photon resonance condition for the exciton spin coherence. The wavelength dependence, along with the polarization configuration used, indicates that the absorption dip results from the exciton spin coherence.

The coherent nature of the absorption dip can be demonstrated directly by its dependence on the delay between the pump and probe. As shown in Figs. 2(a) and 2(d)–2(f), the absorption dip as well as the optical Stark splitting is most pronounced when the probe arrives slightly before the peak of the pump and nearly vanishes when the probe arrives 6 ps after the peak of the pump. This distinctive time dependence is characteristic of coherent optical processes when the probe pulse is much shorter than both the pump pulse and the exciton dephasing time (a few ps). In these experiments, coherent nonlinear responses such as optical Stark splitting and EIT depend on the pump field after the arrival of the probe and within a duration on the order of the relevant dephasing time. Note that the strongly asymmetric response in both optical Stark splitting and EIT near the zero delay is due to the blueshift of the overall exciton resonance induced by excitons excited by the leading part of the pump pulse [22].

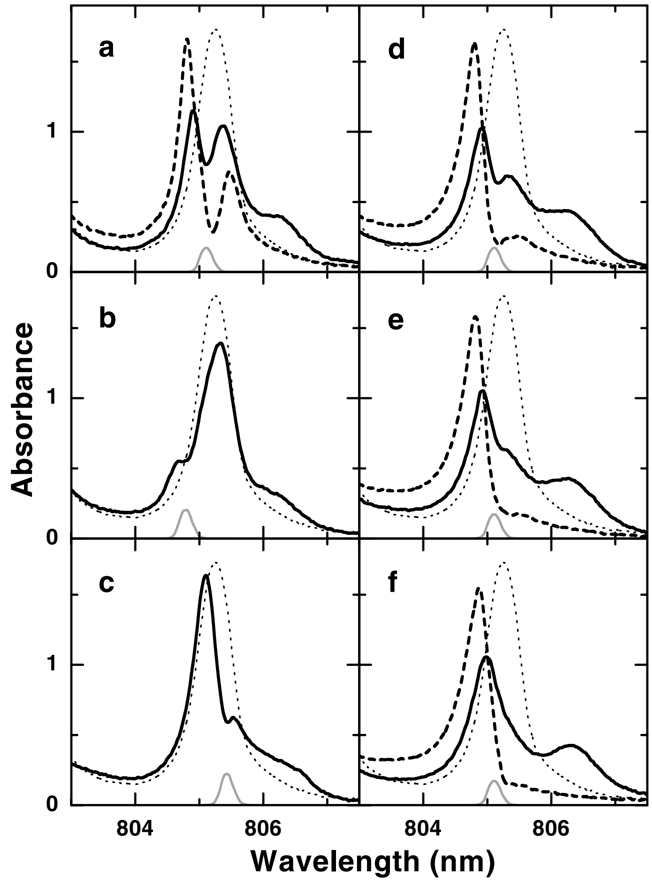

FIG. 2. Absorption of a σ^- probe in the presence of a σ^- (dashed) and a σ^+ (solid) pump, and in the absence of the pump (dotted). The pump spectrum is shown at the bottom of each figure. The probe arrives 3 ps before the pump for (a)–(c) and 0, 2, 6 ps after the pump for (d), (e), and (f), respectively. The pump pulse energy flux is 400 nJ/cm^2.

The absorption dip in Fig. 2 is not due to spectral hole burning since hole burning is an incoherent process and also since the pump beam cannot generate a spectral hole in the inhomogeneous distribution when the optical Stark splitting exceeds the inhomogeneous linewidth [23]. Note that the use of pump pulses with relatively long durations avoids the complication of spectral oscillations encountered in earlier pump-probe experiments where pump pulses with shorter durations were used [21,24].

It is interesting to point out that for the solid curve in Fig. 2(a), there is no absorption dip in the biexciton resonance in spite of the optical Stark splitting in the σ^+ exciton resonance induced by the pump. In this configuration, a biexcitonic two-photon coherence, i.e., coherent superposition of $|+-\rangle_b$ and the ground state, is required for an absorption dip to occur in the biexciton resonance. The absence of this absorption dip reflects that the biexcitonic two-photon coherence is more fragile against dephasing processes such as exciton-exciton scattering than the exciton spin coherence.

To single out a Λ-type three-level system for analogy with atomic studies, we have devised an experiment using bound instead of unbound exciton pairs because the one-exciton to unbound two-exciton transitions cannot be separated in energy from other dipole transitions. Since both $|+\rangle$ and $|-\rangle$ are initially unoccupied, we prepared the system with an incoherent population of spin-up excitons by applying a σ^+ polarized prepulse with a duration of 3 ps and an energy flux of 100 nJ/cm^2. The delay between the pump and the prepulse is 10 ps, long compared with the exciton dephasing time but still short compared with the exciton spin-flip time (~ 50 ps). In this case, $|+\rangle$, $|-\rangle$, and $|+-\rangle_b$ realize a Λ-type three-level system with only one lower state ($|+\rangle$) initially populated.

The experimental conditions for Fig. 3 are similar to those for Fig. 2 except that now the pump pulse is resonant with the exciton to bound biexciton transition and a prepulse injects incoherent spin-up excitons as discussed above. The dotted curve in Fig. 3(a) shows the biexcitonic

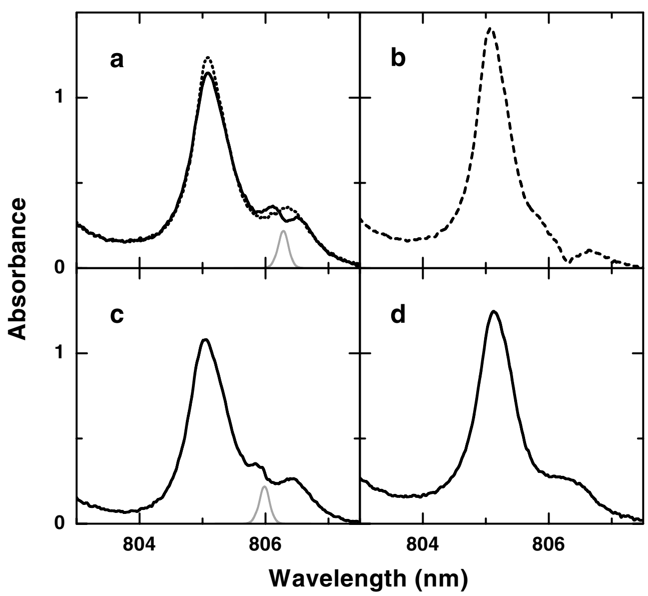

FIG. 3. Absorption of a σ^- probe in the presence of a σ^- (dashed) and σ^+ (solid) pump, and in the absence of the pump (dotted), when the sample is prepared with incoherent spin-up excitons. The pump spectrum is shown at the bottom of (a) and (c) and is the same for (a), (b), and (d). The probe arrives 3 ps before the pump except for (d) where the probe arrives 6 ps after the pump. The pump pulse energy flux is 800 nJ/cm^2.

resonance, corresponding to the $|+\rangle$ to $|+-\rangle_b$ transition, obtained in the absence of the pump. This resonance is induced by the incoherent pumping from the prepulse and vanishes without the prepulse or when the probe and prepulse have the same circular polarization. A pronounced absorption dip occurs in the biexcitonic resonance when the pump and probe have the opposite circular polarization, as shown in the solid curve in Fig. 3(a). Optical Stark splitting of the biexcitonic resonance when the pump and probe have the same circular polarization but with other conditions unchanged can also be seen in Fig. 3(b). Similar to the behaviors of the absorption dip in Fig. 2, the dip in Fig. 3 follows the wavelength of the pump [Fig. 3(c)] and vanishes when the probe pulse arrives 6 ps after the pump pulse while the biexcitonic resonance still persists [Fig. 3(d)].

The absorption dip in both Figs. 2 and 3 can be understood in terms of EIT. In these experiments, the σ^- polarized probe and σ^+ polarized pump induce a spin coherence. The induced spin coherence can lead to destructive interference, resulting in a dip in the absorption spectrum measured by the probe (EIT). The absorption dip occurs when the probe and pump have the same wavelength (the two-photon resonance condition for the spin coherence) and when $\Omega_{\rm pump}$ exceeds $\gamma_{\rm spin}$ and $\sqrt{\gamma \cdot \gamma_{\rm spin}}$, where $\Omega_{\rm pump}$ is the Rabi frequency of the pump and $\gamma_{\rm spin}$ and γ are dephasing rates of the spin coherence and the relevant dipole coherence, respectively [13].

For our measurements, the probe duration is short compared with the pump duration, $1/\gamma_{\rm spin}$, $1/\gamma$, and $1/\Omega_{\rm pump}$. To illustrate how spin coherence evolves and leads to destructive interference under these transient conditions, we have used a phenomenological Λ-type three-level model. The density matrix equations for the three-level system are solved numerically to the first order in the probe field and to all orders in the pump field [14]. Results obtained with resonant pump and probe beams and with $\Omega_{\rm pump} = 2\gamma$ at the peak of the pump pulse are shown in Fig. 4. Figure 4(b) shows the rise and decay of the spin coherence ρ_{+-} after the probe pulse, and further indicates that ρ_{+-} is phased for destructive interference (Re$\rho_{+-} < 0$). Figure 4(c) shows that the absorption dip resulting from the destructive interference depends sensitively on $\gamma_{\rm spin}$ and the magnitude of the induced spin coherence.

For the absorption dip in Fig. 2, in which the pump is resonant with the exciton transition instead of the exciton to bound biexciton transition, the pump plays two very different roles. The pump excites a polarization of spin-up excitons and also couples to the $|-\rangle$ to $|+-\rangle_u$ transition. This latter role, which can be counterintuitive because $|-\rangle$ is initially unexcited, is essential for EIT. The coupling of the pump to the $|-\rangle$ to $|+-\rangle_u$ transition, along with the coupling of the probe to the $|+\rangle$ to $|+-\rangle_u$ transition as shown schematically in Fig. 1(c), induces the exciton spin coherence and leads to EIT.

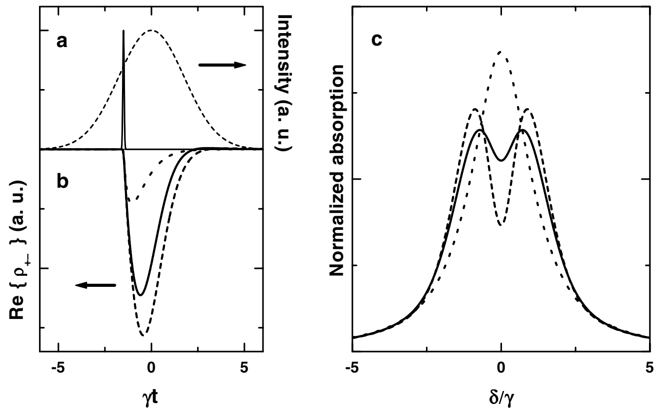

FIG. 4. Theoretical results based on the three-level model discussed in the text. (a) The temporal profile of the pump (dashed) and probe (solid) pulses. (b) Evolution of the spin coherence ρ_{+-}. (c) Absorption of a σ^- probe in the presence of a σ^+ pump with the probe pulse spectrally resolved after the sample (δ is the detuning with respect to the pump). For (b) and (c), dotted, solid, and dashed curves are obtained with the same γ and with $\gamma_{\text{spin}} = 5\gamma$, γ, and 0.5γ, respectively.

The qualitative descriptions presented here are aimed at illustrating the role of exciton-exciton correlations underlying the coherent optical phenomena shown in Fig. 2 and 3. Detailed microscopic descriptions of exciton-exciton correlations developed recently have been very successful in describing the effects of these correlations in nonlinear optical processes to the third and fifth order of the applied field [3–5,8–10]. A satisfactory microscopic description of correlations and quantum coherences involved in EIT requires a solution to all orders of the pump field, which currently represents a considerable theoretical challenge. The basic understanding gained from these recent developments along with appropriate analogies with atomic systems forms the basis of our current qualitative understanding. It should be noted that we have limited our discussions to correlations and coherences up to the level of two-exciton states. In principle, quantum coherences of N-exciton states with $N \geq 3$ can also contribute. However, there has been no evidence of bound triexcitons or three-exciton coherences in GaAs QWs.

In summary, we have demonstrated EIT resulting from exciton spin coherence induced via exciton-exciton correlations. In addition to its potential technological implications, EIT in semiconductors opens up a new avenue for exploring the interplay between Coulomb correlations and quantum coherences in a many-body system.

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[1] D. S. Chema and J. Shah, Proc. Natl. Acad. Sci. U.S.A. **97**, 2437 (2000).
[2] H. Wang *et al.*, Phys. Rev. Lett. **71**, 1261 (1993).
[3] P. Kner *et al.*, Phys. Rev. Lett. **81**, 5386 (1998); S. R. Bolton *et al.*, Phys. Rev. Lett. **85**, 2002 (2000).
[4] G. Bartels *et al.*, Phys. Rev. B **51**, 11 217 (1995); G. Bartels *et al.*, Phys. Rev. Lett. **81**, 5880 (1998).
[5] C. Sieh *et al.*, Phys. Rev. Lett. **82**, 3112 (1999).
[6] G. Chen *et al.*, Science **289**, 1906 (2000).
[7] M. Combescot and R. Combescot, Phys. Rev. Lett. **61**, 117 (1988).
[8] Th. Östreich, K. Schönhammer, and L. J. Sham, Phys. Rev. Lett. **74**, 4698 (1995).
[9] V. M. Axt and Stahl, Z. Phys. B **93**, 195 (1994); V. M. Axt and S. Mukamel, Rev. Mod. Phys. **70**, 145 (1998).
[10] C. Sieh *et al.*, Eur. Phys. J. B **11**, 407 (1999).
[11] F. Quochi *et al.*, Phys. Rev. Lett. **80**, 4733 (1998).
[12] M. Saba *et al.*, Phys. Rev. Lett. **85**, 385 (2000).
[13] S. E. Harris, Phys. Today **50**, No. 7, 36 (1997).
[14] M. O. Scully and M. S. Zubairy, *Quantum Optics* (Cambridge University Press, Cambridge, United Kingdom, 1997).
[15] E. Arimondo, in *Progress in Optics*, edited by E. Wolf (Elsevier Science, Amsterdam, 1996), p. 274.
[16] A. Imamoglu and R. J. Ram, Opt. Lett. **19**, 1744 (1994).
[17] G. S. Agarwal, Phys. Rev. A **51**, R2711 (1995).
[18] G. B. Serapiglia *et al.*, Phys. Rev. Lett. **84**, 1019 (2000).
[19] M. Lindberg and R. Binder, Phys. Rev. Lett. **75**, 1403 (1995).
[20] A. Schülzgen *et al.*, Phys. Rev. Lett. **82**, 2346 (1999).
[21] M. Saba *et al.*, Phys. Rev. B **62**, 16 322 (2000).
[22] N. Peyghambarian *et al.*, Phys. Rev. Lett. **53**, 2433 (1984).
[23] P. Meystre and M. Sargent III, *Elements of Quantum Optics* (Springer-Verlag, Berlin, 1990).
[24] M. Lindberg and S. W. Koch, Phys. Rev. B **38**, 7607 (1988).