Electromagnetically Induced Transparency in Semiconductors via Biexciton Coherence

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We report an experimental demonstration and theoretical analysis of electromagnetically induced transparency in a GaAs quantum well, in which the absorption of an exciton resonance is reduced by more than twentyfold. The destructive quantum interference in this scheme is set up by a control pulse that couples to a resonance of biexcitons. These studies illustrate that many-particle interactions, which are inherent in semiconductors and are often detrimental to quantum coherences, can also be harnessed to manipulate these coherences.

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Electromagnetically induced transparency (EIT) exploits destructive quantum interference to make an otherwise opaque medium almost transparent [1]. Coherent manipulation of quantum coherence and interference in the form of EIT in atomic systems has led to a variety of remarkable phenomena including lasing without population inversion [1], slow and stopped light [2,3], and stimulated Raman adiabatic passage [4]. Applications of EIT in diverse areas, such as all-optical switching and all-optical buffers [5], controlled generation of single photons [6], and quantum information processing [7], have also been actively pursued. The tremendous success of atomic EIT studies has stimulated considerable experimental and theoretical efforts in extending these studies to semiconductors. While numerous approaches for realizing EIT in semiconductors have been proposed and pursued, only a few observations of weak EIT signatures have been reported [8–10].

One difficulty in coherently manipulating optical excitations in semiconductors is the inherent and complex many-particle interactions between these excitations. It is now well established that Coulomb interactions between excitons can profoundly modify coherent excitonic nonlinear optical responses [11]. Earlier experimental demonstrations of EIT in interband optical transitions in semiconductors have required strong resonant optical excitations [10], leading to rapid decay of the underlying quantum coherences. The degree of quantum interference that can be achieved in these experiments is therefore severely limited.

In this Letter we report an experimental demonstration and theoretical analysis of a new EIT scheme, in which the absorption of an exciton resonance in a GaAs quantum well (QW) is reduced by more than twentyfold. The destructive quantum interference in this EIT scheme is set up by a control pulse that couples to a resonance of the exciton molecule (biexciton). The new EIT scheme avoids the excessive dephasing induced by strong resonant excitonic excitations that have hindered earlier EIT studies in semiconductors. The resulting EIT process, which resembles but differs from that in a cascaded three-level atomic system, can be qualitatively described by a microscopic many-particle theory. These studies also underscore that in order to induce strong EIT it is essential to compensate for effects of many-particle Coulomb interactions such as biexciton energy renormalization.

EIT can be realized in a three-level system [see Fig. 1(a)] where a control beam drives the $|2\rangle$–$|3\rangle$ transition and sets up a destructive interference for a weak probe beam coupling to the $|1\rangle$–$|2\rangle$ transition. For our effective three-level scheme, we focus on interband transitions in a GaAs QW between the conduction bands with

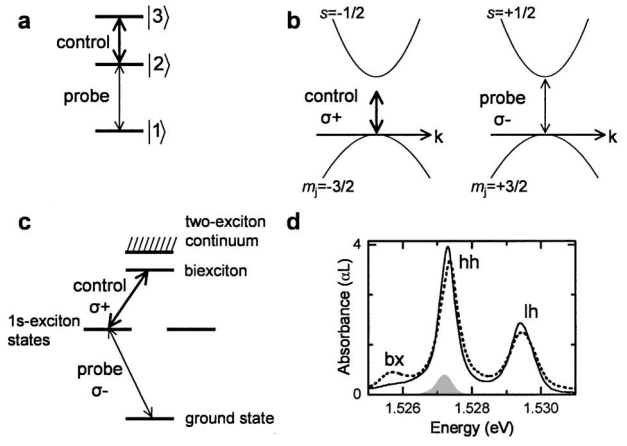

FIG. 1. (a) EIT scheme in a cascaded three-level system. (b) Band structure of a GaAs QW along with the optical selection rules. (c) Schematic of energy eigenstates for the ground, one-exciton, and two-exciton states. Included are only the 1*s*-exciton states and the corresponding lowest biexciton as well as the two-exciton continuum states. (d) Absorption spectra of a probe in the absence (solid lines) and presence (dashed lines) of a pump that was resonant with the hh exciton and had an energy flux of 40 nJ/cm^2. The probe had the opposite circular polarization of the pump and was delayed 10 ps with respect to the pump.

spin $s = \pm 1/2$ and the heavy-hole (hh) valence bands with $m_j = \pm 3/2$ [Fig. 1(b)]. Using circularly polarized light, we can excite σ^+ and σ^- excitons via σ^+ and σ^- transitions, respectively. While these two transitions share no common state, correlations (caused by the Coulomb interaction) between excitons with opposite spins can lead to the formation of bound two-exciton (biexciton) states as well as unbound two-exciton continuum [Fig. 1(c)]. EIT of the exciton resonance can in principle arise from destructive interference induced by a biexcitonic coherence, i.e., coherent superposition between the ground and biexciton states [12–14]. This is analogous but by no means equivalent to EIT in a cascaded three-level atomic system, since in our case the nonradiative coherence is induced via interactions between excitons. For the experimental realization, we used a circularly polarized control pulse to couple the transition between the exciton and biexciton states and a probe pulse with opposite circular polarization to measure exciton absorption, as indicated schematically in Fig. 1(c).

The EIT experiments were performed at 10 K in a GaAs QW with 15 periods of 17.5 nm GaAs wells and 15 nm $Al_{0.3}Ga_{0.7}As$ barriers, grown by molecular beam epitaxy. The control and probe pulses were derived from a mode-locked Ti:sapphire laser with 82 MHz repetition rate, and an external pulse shaper was also used to reduce the spectral width of the control pulse. The resulting nearly transform-limited control pulse had a duration of 6 ps and was focused to a 3×10^{-5} cm^2 spot size on the sample. The probe pulse had a duration of 150 fs and was focused to a smaller spot size of 4×10^{-6} cm^2 to probe a region of uniform excitation by the control pulse. The energy flux of the probe pulse was less than 1% of the control. Absorption spectra were obtained by spectrally resolving the probe pulse transmitted through the sample. To determine the spectral position of the biexciton, we excited the sample with a pump pulse that was resonant with the hh exciton and had a circular polarization opposite of the probe. The dashed line in Fig. 1(d) shows the induced biexciton resonance with a binding energy of 1.6 meV.

To demonstrate EIT of the exciton resonance, we applied a control pulse tuned to the vicinity of the exciton to biexciton transition. Figure 2(a) shows the absorption spectra measured by the probe with the control pulse at two different spectral positions. When the control pulse was at the biexciton resonance, a modest reduction in the absorption of the exciton resonance was observed and an absorption dip occurred at a frequency slightly higher than the hh exciton resonance. A much greater reduction in the exciton absorption, however, was obtained when the control pulse was between the exciton and biexciton resonances. In this case, the absorption dip occurred at the frequency of the hh exciton resonance and the absorption at the dip was reduced by more than twentyfold [the reduction in αL is 3.1, and the actual measured transmittance increases by a factor of $\exp(3.1) = 22$].

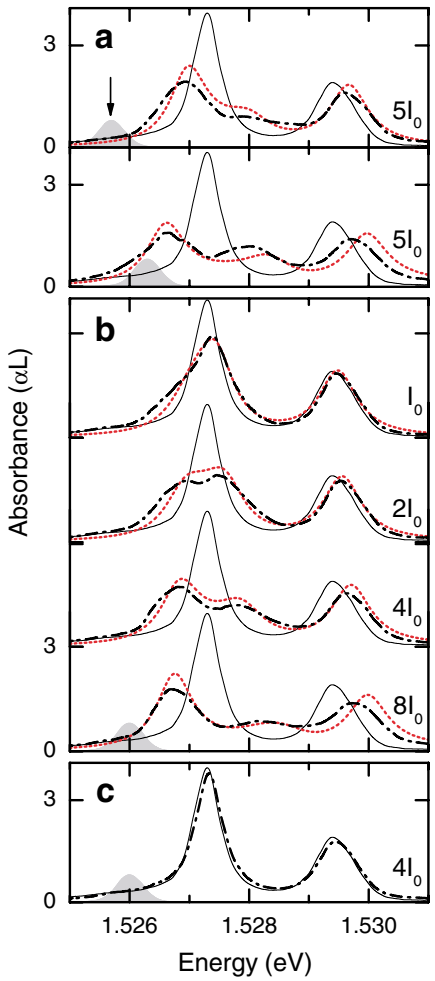

FIG. 2 (color online). Absorption spectra measured with the probe pulse in the absence (solid lines) and presence (dot-dashed lines) of the control pulse, with the probe and control having opposite circular polarization. The shaded area shows the spectrum of the control pulse. The arrow indicates the position of the biexciton resonance from Fig. 1(d). The energy flux of the control pulse is indicated in the figure, with $I_0 = 400$ nJ/cm^2 (corresponding to a peak intensity of order 100 kW/cm^2). For (a) and (b), the probe pulse arrives 1 ps before the peak of the control pulse. For (c), the probe pulse arrives 10 ps after the peak of the control pulse. Results of the full microscopic many-particle theory are shown as dotted lines.

For the above experiments, the absorption dip shifted to lower frequency when the control pulse was tuned to higher frequency, consistent with the two-photon resonance condition expected for EIT. The spectral positions of the dips, however, do not exactly correspond to EIT dips expected for an atomiclike system. As shown in Fig. 2(b), the absorption dip shifts to higher frequency as the intensity of the control pulse increases, a result unexpected for an atomiclike system.

Figure 2(c) shows the absorption spectrum obtained with the probe pulse arriving 10 ps after the control pulse. The spectrum is nearly identical to the linear absorption spectrum obtained in the absence of the control pulse,

indicating minimal real absorption of the control pulse. This nearly complete recovery of the absorption spectrum shows that the reduction in the absorption of the exciton resonance in the above experiments is primarily a coherent process with only minimal contributions from incoherent bleaching. This also rules out contributions from incoherent two-photon absorption processes that can induce biexcitonic absorption resonance in probe absorption spectra.

We have applied a microscopic many-particle theory to understand the physical processes underlying the above EIT experiment. Since that analysis is rather involved, we will only briefly summarize the theory and present the numerical results at the end of this Letter. A comprehensive account of the theory will be published elsewhere. To make direct connections with the well-known concepts from atomic physics, here we will also discuss a phenomenological model that illustrates the essential physical mechanism of the observed EIT process.

The pair of optical transitions indicated in Fig. 1(c), in which the control couples to the exciton-to-biexciton transition and the probe couples to the exciton transition, resembles the well-established cascaded three-level EIT system. These transitions feature simultaneous one- and two-photon resonances and thus dominate the nonlinearity in the vicinity of the exciton. In comparison, the pair of transitions not shown in Fig. 1(c), in which the control couples to the exciton transition and the probe couples to the exciton-to-biexciton transition, are not doubly resonant and therefore not expected to have a large contribution to the observed nonlinearity. Consequently, our phenomenological model is based on the cascaded three-level system. To first order in the probe field and all orders of the control field, and within the rotating wave approximation, the density matrix equations in a rotating frame for the three-level system are given by [1]

$$\dot{\rho}_{xg} = [i(\omega_p - \omega_x) - \gamma]\rho_{xg} + i\Omega_p/2 + i\Omega_c\rho_{bg}/2,$$
$$\dot{\rho}_{bg} = [i(\omega_p + \omega_c - \omega_b) - \gamma_b]\rho_{bg} + i\Omega_c\rho_{xg}/2, \qquad (1)$$

where ρ_{xg} and ρ_{bg} are the exciton and biexciton coherence, respectively, ω_x (ω_b) and γ (γ_b) are the resonance frequency and decay rate for the exciton (biexciton) coherence, and ω_c (ω_p) and Ω_c (Ω_p) are the optical frequency and Rabi frequency for the control (probe). We take the biexciton binding energy $\hbar(2\omega_x - \omega_b)$ to be $5\hbar\gamma$. Figure 3 shows the calculated probe absorption spectra. A pronounced absorption dip occurs for small γ_b but vanishes for large γ_b [see Fig. 3(a)], verifying that the EIT dip results from destructive interference induced by the biexciton coherence. The degree of the induced transparency is determined by both γ_b and Ω_c. Note that, although the EIT process discussed here resembles that of an atomic system, the biexciton coherence itself is a direct result of many-particle Coulomb correlations and can thus lead to behaviors qualitatively different from those of an atomic system.

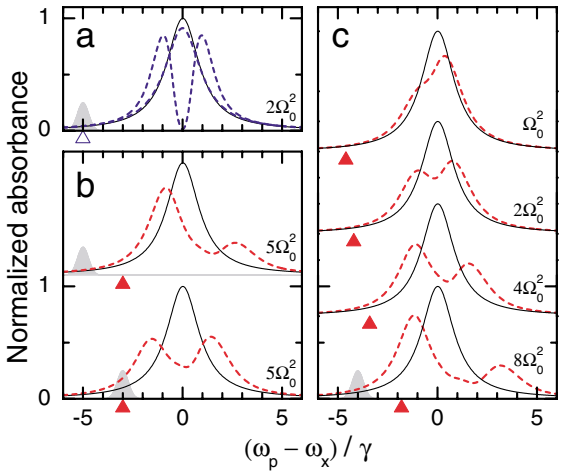

FIG. 3 (color online). Probe absorption spectra in the absence (solid line) and presence (dashed line) of a control obtained from the cascaded three-level model. The control and probe durations are $10\gamma^{-1}$ and $0.1\gamma^{-1}$, respectively, with the probe arriving $1\gamma^{-1}$ before the control. The peak control intensity is indicated in the figure, with $\Omega_0^2 = 2\gamma^2$. The shaded area shows the control spectrum. The open and solid triangles mark the position of the nonrenormalized and renormalized (at the peak of the control pulse) resonance frequency for the exciton-to-biexciton transition, respectively. (a) Without biexciton energy renormalization, for $\gamma_b = 0.1\gamma$ (deep EIT dip) and 10γ (no dip). (b),(c) With biexciton energy renormalization, with $\gamma_b = \gamma$ and $\beta = 0.2\gamma^{-1}$, and for various control spectra and intensities.

The dependence of the spectral position of the EIT dip on the intensity of the control shown in Fig. 2(b) highlights an important difference between EIT induced by the biexciton coherence and EIT in an atomic system. For a fixed ω_c, the spectral position of the EIT dip is set by the resonance frequency of the biexciton coherence. Since a biexciton is a bound state of two excitons, the biexciton energy is determined by the sum of the σ^+ and σ^- exciton energies minus the biexciton binding energy, all of which are affected by exciton-exciton interactions, leading to renormalization in both exciton and biexciton energies. At relatively low intensity for the σ^+ polarized control, we simulate effects of the biexciton energy renormalization by replacing ω_b with $\omega_b + \beta\Omega_c^2$ in Eq. (1), where β is a phenomenological constant, and ignore energy renormalization for the σ^- exciton.

Figure 3(b) illustrates that renormalization in the biexciton energy leads to a corresponding shift in the spectral position of the EIT dip, and qualitatively accounts for the experimental results shown in Fig. 2(a). Figure 3(c) shows the calculated probe absorption spectra at various Ω_c with other parameters including ω_c fixed. With increasing control intensity, the EIT dip shifts gradually to higher energy due to the increasing blueshift of the biexciton energy, as found experimentally in Fig. 2(b).

The exciton-biexciton system has been investigated extensively for optical Stark effects. Reports of exciton

redshifts and Coulomb memory effects due to biexcitons, which can be viewed as a precursor for the EIT discussed here, can be found in [15]. Autler-Townes splitting of biexcitons has also been observed in two-photon absorption spectra of biexcitons [16]. In spite of these extensive efforts, EIT occurring at the exciton resonance via biexciton coherence has not been reported previously. As we have discussed above, to observe the strong EIT process at the exciton resonance, it is essential that the biexciton energy renormalization be properly compensated. In addition, it is also important that the control duration is long or at least comparable to the biexciton decoherence time.

As noted above, we have also formulated and applied a microscopic theory that can provide a more detailed and satisfactory understanding of the experimental results and can have a predictive capability [13]. The theory is based on a picture of interacting excitons and on the dynamics-controlled truncation (DCT) formalism [17]; details of our formulation are given in [18,19]. Within this formalism, a coherent biexciton correlation function plays the same pivotal role in EIT as that played by ρ_{bg} in the phenomenological model. The analog of the decay rate γ_b in the phenomenological model is the decay rate (Γ_b) of the biexciton correlation function. In comparison to the phenomenological three-level model that contains only effects and excitation pathways deemed most important to the observed EIT process, the DCT approach systematically treats nonlinear optical processes from both bound and continuum two-exciton states as well as from transitions that are not doubly resonant. The DCT approach has often been applied to calculate nonlinear responses up to third order in the light field amplitude, e.g., [16]. For the present application, we have generalized the theory in Refs. [18,19] to include the effects of interactions between the coherent biexciton with other control-beam produced excitons, which yields the renormalization of the biexciton energy. At low intensities, the theory gives the biexciton energy shift proportional to the control intensity and approximately given as twice the average of the correlation-induced level shifts of the constituent exciton states [which include both optically active and inactive excitons, e.g., that formed by $s = -1/2$ and $m_j = +3/2$ in Fig. 1(b)]. With the resulting equation, we have evaluated the optical response to the first order in the probe field but to all orders in the control field [20].

The absorption spectra calculated from the microscopic many-particle theory are shown as dotted lines in Fig. 2, where $\Gamma_b = 0.9\ \mathrm{ps}^{-1}$ (twice the exciton dephasing rate) and the Rabi frequency corresponding to I_0 indicated in Fig. 2 is $0.2\ \mathrm{ps}^{-1}$. The theory is in very good qualitative agreement with the experiment. In particular, the nonlinear dependence of the overall magnitude of the absorption reduction on the control intensity is well described by the theory. The small difference between the theoretical and experimental results in Fig. 2 is likely due to inhomogeneous broadening and higher-order correlations and nonlinear biexciton energy shifts that have not been included. The microscopic theory confirms the physical mechanisms illustrated by the phenomenological model and establishes rigorously the link between EIT in an atomic system and a similar EIT-like process induced via many-particle Coulomb correlations in a semiconductor.

In summary, we have demonstrated strong EIT in a GaAs QW by coupling a control pulse to a biexciton resonance. The EIT is induced by a biexciton coherence and exhibits behaviors distinctively different from those of atomic systems. This ability to induce strong EIT via many-particle Coulomb correlations also points to an effective avenue for controlling and utilizing quantum coherences in an interacting many-particle system.

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