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Preservation of quantum coherence after exciton-exciton interaction in quantum wells

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The dynamics of exciton-exciton interaction in quantum wells has been investigated by monitoring the time-resolved resonant secondary emission that follows excitation with linearly and circularly polarized light. Preservation of quantum beating in the cross-polarized emission demonstrates that spin relaxation can take place, for some scattering channels, without total quantum coherence loss. Interexciton electron exchange is the scattering mechanism that explains the persistence of the beating and, since it is sensitive to the fine structure of excitons, the shift by π in the phase of the beating observed in the experiment.

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Optical properties of low-dimensional semiconductor systems and devices depend strongly on the excitation density (n_X). For n_X below 3×10^{11} cm^{-2} and at low temperatures, excitons are stable and dominate the optical response of GaAs quantum wells (QW's) near the absorption edge. Within this regime of moderate excitation density ($n_X \geqslant 10^9$ cm^{-2}) exciton-exciton scattering plays a major role in the resonant response of QW's and, as the dominating phase-breaking mechanism, governs the dynamics of coherent transients. Since excitons confined in low-dimensional semiconductor systems have been suggested as possible candidates for quantum computation[1] it is important to understand the mechanisms involved in the loss of *optical* coherence (between the excitonic ensemble and the exciting light). The study of exciton dephasing has been most often attempted by means of nonlinear techniques.[2] Nevertheless, coherent features are also present in the linear response of QW's: the resonant secondary (nonspecular) emission (RSE) has a coherent component resulting from scattering with static disorder.[3] Thus time-resolved RSE can be used as a probe of the coherence of the excitonic ensemble providing information about scattering with dynamic disorder, such as exciton-exciton scattering, that leads to exciton dephasing.[4,5] Recent studies have shown that exciton-exciton interaction also contributes to the depolarization of RSE due to exciton spin relaxation.[6,7] The efficiency of such spin-relaxation channels increases with increasing excitation ellipticity. In the limiting case of linearly polarized excitation, the degree of polarization of RSE decays in a time scale comparable to typical dephasing times for GaAs QW's ($\sim$10 ps) and depends strongly on n_X. These experimental results are well explained in terms of interexciton exchange of carriers.[8,9] For circularly polarized excitation, on the other hand, spin relaxation is a much slower density-independent process.[10,11]

Periodic oscillations can appear in the time evolution of RSE due to the simultaneous resonant excitation of more than one excitonic transition. Beating between heavy-hole (hh) and light-hole (lh) excitons has been observed in the emission of wide QW's.[12,13] The visibility of the beating decays in a time scale of the order of 10 ps and depends on n_X. Determining the origin of the beating—whether it results from the quantum nature of the superposition of states or from classical interferences at the detector—is not trivial. Distinguishing between these two possibilities required performing a specific experiment in the case of beating observed in time-resolved four-wave mixing.[14] So far there are no conclusive studies about the nature of the beating present in the linear emission of QW's.

In this Rapid Communication we address two important issues regarding exciton coherence in QW's: the nature of hh-lh exciton beating in RSE and the effect of exciton-exciton interaction as a coherence-breaking mechanism. We studied the depolarization of time-resolved RSE from wide QW transitions after excitation with linearly polarized light in the density regime where exciton-exciton scattering is the dominant spin-relaxation mechanism. The improved time resolution of our experiment allowed the observation of several features indicating that spin relaxation can take place without coherence loss. We have identified the exciton-exciton scattering mechanism responsible for such effects.

The experimental results presented in this Rapid Communication correspond to a single 15-nm GaAs QW with $Al_{0.33}Ga_{0.67}As$ barriers (sample *A*) and a multiple-QW structure containing ten repetitions of 20-nm GaAs wells separated by GaAs/AlAs superlattice barriers (sample *B*). Both samples showed narrow emission linewidths at low temperatures, with full width at half maximum of 0.8 meV and 0.7 meV for samples *A* and *B*, respectively. We measured the time evolution of the RSE by means of two-color up-conversion spectroscopy (for more details about the experimental setup see Ref. 13). Excitation and gating pulses were 140-fs long (13-meV bandwidth), which kept the time resolution below 200 fs, shorter than in previous studies.[6,7] Excitation of the sample was done in the backscattering geometry with the laser pulse propagating parallel to the growth direction (z) of the sample. All measurements were done at 6 K.

Time-resolved RSE spectra of sample *A* for identical n_X but corresponding to different excitation and detection conditions are plotted in Fig. 1. The excitation pulse was either circularly polarized (a) or linearly polarized along the x direction (b) and the detected emission was copolarized (solid lines) and cross-polarized (dashed lines) with respect to the excitation. Copolarized emission has an intense spike at t

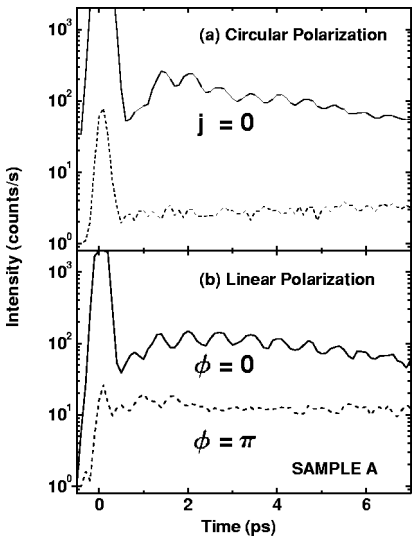

FIG. 1. Time evolution of the RSE from the 15-nm single-QW sample after excitation with circularly (a) and linearly (b) polarized light. Solid lines correspond to copolarized emission whereas dashed lines denote cross-polarized emission. Estimated excitation density is 3×10^9 cm^{-2}. φ is the phase of the beat modulation with respect to the origin at zero time.

$=0$ corresponding to the excitation pulse scattered at the sample's surface. The presence of beating with the periodicity of the hh-lh exciton energy splitting (6 meV) in the copolarized emission is a common feature for circularly and linearly polarized excitation. The first maximum of the beats coincides with the arrival of the excitation pulse so the phase of the modulation $\varphi=0$. The cross-polarized emission is severely suppressed for circular polarization at early times, as seen in Fig. 1(a). On the other hand, we detect intense cross-polarized emission in the first 2 ps after excitation with linearly polarized light, see Fig. 1(b). Furthermore, the cross-polarized emission still shows beating with the same period but it has its phase shifted by π with respect to the beating of the copolarized component ($\varphi=\pi$). To the best of our knowledge this feature has never been reported before. In the current experiment the system evolves freely with time after the arrival of the excitation pulse in contrast to phase-shifted hh-lh exciton beats observed in four-wave mixing[15] and coherent control[16] experiments, in which a second excitation pulse interacts with the excitonic polarization, altering the phase of the beats.

In order to obtain a deeper insight into the persistence of the beats, we have investigated the effects of changing the excitation density. Time-resolved RSE from sample *B* after excitation with linearly polarized light is displayed in Fig. 2. The lower energy splitting between hh and lh excitons in sample *B* increases the beating amplitude and improves the visibility of the beating at early times. Again in this case, the cross-polarized emission is intense at early times after excitation and shows π-shifted hh-lh exciton beating. These effects disappear when exciting with circularly polarized light, as for sample *A*, and when decreasing n_X. The latter case is shown in Fig. 2(c). Exchange interaction between the exciton electron and hole governs spin relaxation at low n_X.[10,17] The

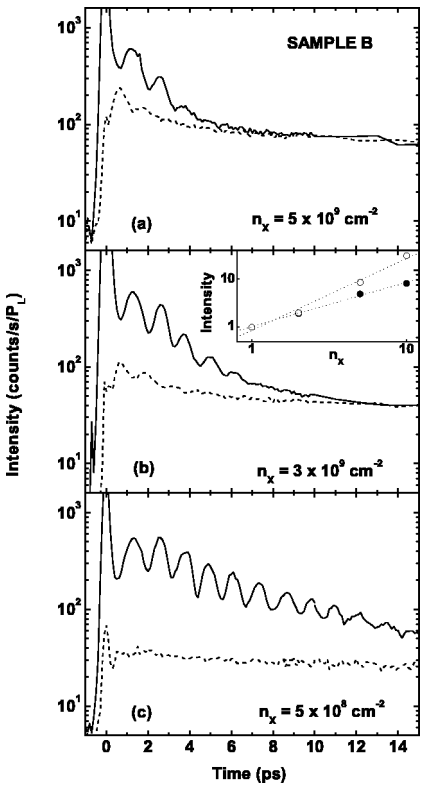

FIG. 2. Time evolution of the copolarized (solid line) and cross polarized (dashed line) RSE from the 20-nm multiple-QW sample following excitation with linearly polarized light. The emission intensity is normalized to the laser power (P_L). The inset shows the intensity of the copolarized (●) and cross polarized (○) emission integrated for the first 10 ps as a function of P_L. All data are normalized to those corresponding to $n_X=5\times10^8$ cm^{-2}.

dynamics of the emission depends strongly on n_X at early times after excitation (range shown in Fig. 2). We observe that the rise and decay rates of both components of the emission increase for increasing n_X, as expected from scattering into nonradiative states.[6,7] Also the depolarization rate of the emission becomes much faster: full depolarization takes place in less than 10 ps for $n_X=5\times10^9$ cm^{-2} [see Fig. 2(a)]. But the most noticeable effect is the strong superlinearity found in the intensity of the cross polarized emission against the laser power (P_L), while the copolarized emission scales linearly with P_L. This effect is also shown in the inset of Fig. 2(b) where the intensity of the emission integrated for the first 10 ps after excitation is plotted against P_L (the $t=0$ spike has been avoided). The solid circles correspond to the intensity of the copolarized emission and the open circles are the integrated intensities of the cross-polarized emission. We have fitted the experimental data by $I\propto P_L^y$ finding $y=0.93\pm0.06$ and 1.46 ± 0.12 for the co-polarized and cross-polarized emission, respectively.

Superlinear intensity dependence on n_X can be a signature of biexciton generation. However, low-temperature cw photoluminescence spectra of the transitions investigated with different laser power do not show any feature that could be interpreted as biexcitons in the excitation density regime of our experiment. Moreover, we do not observe the fast non-

exponential decay at early times attributed to biexcitons.[18] The persistence of the beating and its phase shift is not related to strain effects either since we do not observe any splitting in the excitonic transitions produced by strain.[19,20] We also disregard wave-function anisotropy effects due to exciton confinement on the plane of the well as we do not observe cross polarized emission at early times after σ excitation [Fig. 1(a)]. Such an effect, observed in narrow QW transitions, modifies the absorption selection rules and gives rise to depolarization without coherence loss.[21,22] The remaining mechanism responsible for the RSE depolarization and the nonlinearities described above is exciton-exciton scattering.

The stochastic character of exciton-exciton scattering implies that it destroys the optical coherence of the excitonic ensemble. Consequently, cross polarized emission should be fully incoherent and the presence of beating can only be understood as a quantum effect: i.e., the excitonic states created by the short excitation pulse reaching the sample at $t=0$ are quantum superpositions of the form

$$|\mathbf{Q},S\rangle = a|\mathbf{Q},S\rangle_{\mathrm{hh}} + b|\mathbf{Q},S\rangle_{\mathrm{lh}}, \tag{1}$$

where $\mathbf{Q}$ is the center-of-mass wave vector and S accounts for the spin state. The coefficients a and b are given by the oscillator strength and the intensity of the laser pulse at the energies of the transitions. Valence-band mixing is negligible in the proximity of the zone center and hh and lh excitons have well-defined energies. Thus the beating observed in RSE is a direct consequence of the time evolution of the exciton wave function.

Let us analyze the role of the spin in a dense exciton gas from a theoretical point of view. We have followed the approach of Ciuti *et al.*[8] to this problem extending it to the case of a quantum superposition as Eq. (1). *A* Coulomb scattering process of the form

$$(1s,\mathbf{Q},S)+(1s,\mathbf{Q}',S')\rightarrow(1s,\mathbf{Q}+\mathbf{q},S_f)+(1s,\mathbf{Q}'-\mathbf{q},S_f'), \tag{2}$$

is considered. The spin part of the wave function S is determined by the circularly polarized optical selection rules in zinc-blende QW's (D_{2d} symmetry). Thus circularly polarized light propagating along the z direction creates excitons with $|J_z=\pm 1\rangle$ for $\sigma^{\pm}$ helicity and linearly polarized photons create linearly polarized excitons: $|x\rangle=(|+1\rangle+|-1\rangle)/\sqrt{2}$ and $|y\rangle=(-|+1\rangle+|-1\rangle)/\sqrt{2}$. The *s*-conduction band has two spin states $s_z=\pm\frac{1}{2}$ and the *p*-valence band has four spin states split into the hh band $j_z=\pm\frac{3}{2}$ and the lh band $j_z=\pm\frac{1}{2}$. There are four optically active excitonic states with z component of the total angular momentum $J_z=j_z+s_z$: the hh excitons, $|\pm 1\rangle_{\mathrm{hh}}=|\pm\frac{3}{2}\mp\frac{1}{2}\rangle$, and lh exciton states $|\pm 1\rangle_{\mathrm{lh}}=|\pm\frac{1}{2}\pm\frac{1}{2}\rangle$. Notice the different fine structures of hh and lh excitons. In order to match the experimental conditions, we considered scattering processes involving excitons in the vicinity of the zone center and therefore the exciton center-of-mass wave vectors of the initial and final states are close to zero. The antisymmetry of the two-exciton wave function under the exchange of identical particles gives four different contributions to the scattering amplitude of the process described in Eq. (2): the direct term corresponding to classical Coulomb interaction between the two excitons; the exciton-exciton exchange interaction term (simultaneous exchange of electrons and holes); and two terms corresponding to the interexciton exchange of holes and electrons, respectively. Due to symmetry considerations,[8] the first two contributions tend to zero for scattering processes with $\mathbf{q}\rightarrow 0$. Within our experimental conditions, i.e., $\mathbf{Q}\approx\mathbf{Q}'\approx\mathbf{q}\approx 0$, the dominant mechanisms of interaction correspond to hole-hole and electron-electron exchange. Moreover, as demonstrated by Ciuti *et al.*,[8] in the region of small $\mathbf{q}$ the scattering amplitude is independent of the electron and hole masses and can be written as

TABLE I. Scattering amplitudes of the allowed spin scattering channels. Other inelastic scattering channels, such as scattering into nonradiative states, are not included in this table.

$\lvert S\rangle$	$\lvert S'\rangle$	$\lvert S_f\rangle$	$\lvert S_f'\rangle$	$S^e(S,S',S_f,S_f')$	$S^h(S,S',S_f,S_f')$
$\lvert x\rangle_{\mathrm{hh}}$	$\lvert x\rangle_{\mathrm{hh}}$	$\lvert x\rangle_{\mathrm{hh}}$	$\lvert x\rangle_{\mathrm{hh}}$	1/2	1/2
$\lvert x\rangle_{\mathrm{hh}}$	$\lvert x\rangle_{\mathrm{hh}}$	$\lvert y\rangle_{\mathrm{hh}}$	$\lvert y\rangle_{\mathrm{hh}}$	1/2	1/2
$\lvert x\rangle_{\mathrm{lh}}$	$\lvert x\rangle_{\mathrm{lh}}$	$\lvert x\rangle_{\mathrm{lh}}$	$\lvert x\rangle_{\mathrm{lh}}$	1/2	1/2
$\lvert x\rangle_{\mathrm{lh}}$	$\lvert x\rangle_{\mathrm{lh}}$	$\lvert y\rangle_{\mathrm{lh}}$	$\lvert y\rangle_{\mathrm{lh}}$	1/2	1/2
$\lvert x\rangle_{\mathrm{hh}}$	$\lvert x\rangle_{\mathrm{lh}}$	$\lvert x\rangle_{\mathrm{hh}}$	$\lvert x\rangle_{\mathrm{lh}}$	1/2	
$\lvert x\rangle_{\mathrm{hh}}$	$\lvert x\rangle_{\mathrm{lh}}$	$\lvert y\rangle_{\mathrm{hh}}$	$\lvert y\rangle_{\mathrm{lh}}$	$-1/2$	

$$H_{SS'}^{S_fS_f'} = e[S^h(S,S',S_f,S_f')+S^e(S,S',S_f,S_f')], \tag{3}$$

where e is a real factor (see Appendix B in Ref. 8) and

$$S^h(S,S',S_f,S_f') = \sum_{s_e,j_h,s_{e'},j_{h'}} \langle s_e,j_h|S\rangle\langle s_{e'},j_{h'}|S'\rangle \times\langle S_f|s_e,j_{h'}\rangle\langle S_f'|s_{e'},j_h\rangle \tag{4}$$

and

$$S^e(S,S',S_f,S_f') = \sum_{s_e,j_h,s_{e'},j_{h'}} \langle s_e,j_h|S\rangle\langle s_{e'},j_{h'}|S'\rangle \times\langle S_f|s_{e'},j_h\rangle\langle S_f'|s_e,j_{h'}\rangle. \tag{5}$$

This theoretical model predicts that exciton-exciton scattering does not rotate the spin polarization of two-exciton states with equal circular polarization, in good agreement with our experimental observations. For linearly polarized excitons, on the other hand, there are a number of allowed spin-scattering channels. Table I summarizes the scattering amplitudes of some of the allowed scattering channels for an initial two-exciton state of the form $|x\rangle|x\rangle$. A quantitative analysis of all possible channels has been performed in Refs. 6 and 7. Here we only study the case in which either the exciton spin polarization is preserved or is rotated by 90°. We distinguish between hh-hh exciton, lh-lh exciton, and hh-lh exciton scattering. While there are two contributions to the scattering amplitude due to hh-hh exciton interaction and equally for lh-lh exciton interaction, only electron exchange accounts for the hh-lh exciton scattering amplitude since heavy and light holes are distinguishable particles. The hh-hh

and lh-lh exciton scattering amplitudes are equal in these two cases. On the contrary, for the mixed hh-lh exciton term, the scattering into the cross-polarized state introduces an additional phase of π with respect to the channel that preserves the spin polarization (notice the minus sign of the scattering amplitude). This additional phase proves that this scattering mechanism is sensitive to the fine structure of excitons.

Now we can calculate straightforwardly the possible spin-relaxation channels for a quantum superposition as Eq. (1) by separating all the possible contributions to the scattering amplitude, i.e., hh-hh, lh-lh, and hh-lh, that are already listed in Table I. Starting from an initial two-exciton state of the form $(a|x\rangle_{\mathrm{hh}}+b|x\rangle_{\mathrm{lh}})(a|x\rangle_{\mathrm{hh}}+b|x\rangle_{\mathrm{lh}})$, most scattering channels destroy the quantum superposition. However, electron-electron exchange interaction can scatter such an initial state into two possible active final states of the form

$$(a'|x\rangle_{\mathrm{hh}}+b'|x\rangle_{\mathrm{lh}})(a'|x\rangle_{\mathrm{hh}}+b'|x\rangle_{\mathrm{lh}}) \tag{6}$$

and

$$(a'|y\rangle_{\mathrm{hh}}-b'|y\rangle_{\mathrm{lh}})(a'|y\rangle_{\mathrm{hh}}-b'|y\rangle_{\mathrm{lh}}), \tag{7}$$

that preserve quantum coherence ($a'=a\times e$ and $b'=b\times e$ and e is real) of the initial state. These two final states evolve with time in the same way as Eq. (1) and therefore we should still expect beating in the emission. Moreover, the negative sign in Eq. (7) implies a π shift in the phase of the beating in perfect agreement with our experimental results.

In conclusion, we have studied the dynamics of the spin polarization of resonantly photogenerated excitons with improved time resolution in a regime where exciton-exciton scattering dominates exciton spin relaxation. We report on the persistence and change in phase of hh-lh exciton beating in the spin-relaxed component of the emission after excitation with linearly polarized light. This has two important implications: it demonstrates the quantum nature of the RSE beating; and more importantly, it proves that exciton-exciton scattering can preserve quantum coherence. We have identified that interexciton electron exchange is the scattering mechanism responsible for such effects. Moreover, the different fine structure of hh and lh excitons manifests itself in the π shift observed in the phase of the beating.

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