Spontaneous Emission of Excitons in GaAs Quantum Wells: The Role of Momentum Scattering

Hailin Wang, Jagdeep Shah, and T. C. Damen
AT&T Bell Laboratories, Holmdel, New Jersey 07733

L. N. Pfeiffer
AT&T Bell Laboratories, Murray Hill, New Jersey 07974
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Resonantly excited photoluminescence with femtosecond time resolution reveals the essential role of momentum scattering in emission from excitons in quasi-two-dimensional systems. The gradual rise of the luminescence provides a direct measure of exciton momentum scattering rates. An analysis of quantum beats in the luminescence determines exciton dephasing rates. The results show that the momentum scattering rate is considerably larger than the dephasing rate, indicating a breakdown of the impact approximation commonly assumed in coherent optical studies.

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Dynamics of excitons in quasi-two-dimensional semiconductors such as quantum wells (QW) has received considerable attentions in recent years. Radiative dynamics following resonant as well as nonresonant excitations has been extensively studied by using time-resolved photoluminescence (PL) [1–4]. The regime immediately following the resonant excitation, however, has yet to be investigated because of the limited time resolution in earlier measurements. This regime contains important information regarding the role of momentum scattering in optical interactions in semiconductors.

It is well known that conservation of momentum can lead to fundamental differences between optical emission processes of atomic vapors and of extended optical excitations in crystals. An atom recoils while emitting a photon leading to a Doppler shift of the transition [5]. In contrast, an exciton annihilates after the radiative recombination. In an ideal bulk crystal, an exciton with momentum $\mathbf{k}$ couples to only one particular photon mode because of the momentum conservation, leading to the formation of an exciton polariton that is stationary with respect to the electromagnetic vacuum fluctuation [6]. In an 2D system such as quantum wells (QW), an exciton with in-plane momentum $\mathbf{k}_{\|}$ smaller than the crossing of the exciton and photon dispersion lines can couple to a continuum of photon modes with wave vector $\mathbf{k} = (\mathbf{k}_{\|}, k_z)$, leading to irreversible radiative decay [7]. In the absence of momentum scattering, the energy-momentum conservation, however, still prevents emission of photons in directions other than the reflected or transmitted direction of the excitation beam. Photons detected from other directions, as in PL measurements must originate from excitons that have gone through momentum scattering. The very initial dynamics of the exciton PL should reflect the role of momentum scattering in exciton spontaneous emission.

The evolution of spontaneous emission immediately following an impulsive excitation also contains information on various coherences created between relevant electronic states. For example, spontaneous emission from an atom in a coherent superposition of two excited states can exhibit temporal oscillations (quantum beats) at a frequency corresponding to the energy splitting between the two excited states [8,9]. Decay of the oscillation is determined by the dephasing of the coherence between the two excited states. For excitons, dephasing of the optically induced coherence is strongly affected by the ubiquitous momentum scattering process.

In this paper we present time-resolved studies of resonantly excited PL with femtosecond resolution. The much-improved time resolution reveals the crucial role of momentum scattering in exciton spontaneous emission. The gradual rise of the PL provides a direct measure of exciton momentum scattering rates, information that is not available from nonlinear optical measurements such as four wave mixing (FWM). The presence of quantum beats in the PL further enables us to examine directly the relationship between momentum scattering and dephasing of the optically induced coherence. Measurement shows that the exciton momentum scattering rate is considerably larger than the dephasing rate, indicating a breakdown of the impact approximation commonly assumed in coherent optical studies.

We achieve, for the first time, the femtosecond time resolution for resonantly excited PL by upconverting the PL in a crystal ($LiIO_3$) with output at 1.5 μm from a synchronously pumped optical parametric oscillator. We used two high quality samples and obtained qualitatively the same results from both samples. Sample A (B) contains 15 periods of 130 Å (175 Å) GaAs quantum well and 150 Å $Al_{0.3}Ga_{0.7}As$ barrier. The PL linewidth for sample A (B) is 0.8 meV (0.2 meV) at 2 K and no Stokes shift was observed. For time-resolved measurements, the sample is excited with transform-limited pulses (14 meV spectral width) below the heavy hole (hh) resonance to minimize the excitation of free carriers. The PL is col-

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lected in a backscattering geometry and away from the reflected direction of the excitation beam (distinctions between resonant PL and resonant Rayleigh scattering due to the static disorders will be discussed later). All the measurements were performed at 10 K, and the range of exciton densities used is between 10^9 to 10^{10} cm^{-2}. Under these conditions excitons are nearly homogeneously broadened as shown by FWM measurements.

Figures 1(a) and 2 show the initial evolution of the PL excited resonantly with an excitation pulse centered 6 meV below the hh exciton energy. Circularly polarized fields were used to avoid effects of biexcitons [10]. The spike at zero delay is due to the scattering of the laser pulses from the sample surface, and the width of the spike shows the time resolution of the measurement. Unlike time-resolved resonant fluorescence in atomic vapors [9], the PL shown in Figs. 1(a) and 2 is characterized by a *gradual rise* after the excitation pulse. As we discussed earlier, excitons created by the excitation pulse cannot emit photons in directions other than the transmitted or reflected direction of the excitation pulse before the exciton momentum is randomized. The observed rise time for the exciton PL demonstrates the essential role of momentum scattering in exciton radiative recombination and provides quantitative information on momentum scattering of the excitons.

We examined the dependence of the PL rise time on detunings between the excitation pulse and the hh exciton. The spectrally broad excitation pulse (14 meV spectral width) used to achieve the necessary time resolution can excite hh excitons, light hole (lh) excitons, and free carriers. When the excitation pulse is 6 meV below the hh resonance, the estimated lh exciton (free carrier) population is less than 6% (3%) of the hh exciton population for the 130 Å QW. The rise time of the PL remains the same as we move the excitation pulse to lower energy while maintaining a constant exciton density. In this regime, effects of lh excitons and free carriers on the rise time are negligible compared to the hh excitons. Effects of lh excitons and free carriers, however, become important when the energy of the excitation pulse approaches the band gap. In particular, when the center of the pulse is a few meV above the hh exciton resonance, the PL rise time becomes resolution limited, reflecting strong interactions between the exciton and a large free carrier population.

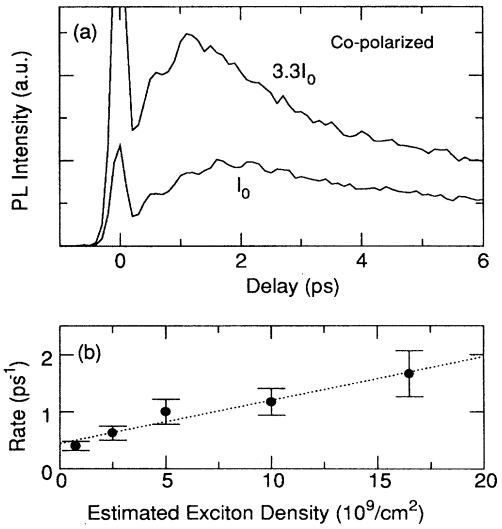

FIG. 1. (a) Initial time-resolved PL for sample A. I_0 corresponds to an exciton density of $5 \times 10^9/cm^2$. (b) Density dependence of the momentum scattering rate. The dashed line is a guide to the eye.

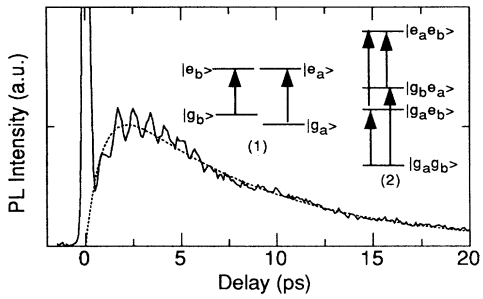

FIG. 2. Initial time-resolved PL for sample B. The exciton density used is $5 \times 10^9/cm^2$. The inset shows the energy structure responsible for the oscillation.

The intensity dependence of the rise time further confirms that momentum scattering determines the rise time of the PL. Figure 1(b) shows that the inverse of the rise time (Γ in later discussions) increases with increasing exciton densities. The data are obtained when the center of the excitation pulse is at or more than 6 meV below the hh exciton. Both exciton-exciton scattering and scattering from interface disorder contribute to momentum relaxation at low temperature. The exciton-exciton scattering rate increases with increasing exciton densities and should eventually dominate the scattering process [11]. For the data shown in Figs. 1(a) and 2 exciton-exciton scattering is the leading contribution to momentum scattering.

We further studied the dependence of the PL rise time on the direction of the emission. PL along the reflected direction requires no momentum scattering and is expected to rise instantaneously. The overwhelming scattering of the laser pulse from the sample surface, however, has prevented us from measuring the emission rise time near the reflected direction. The rise time obtained at relatively large but different angles does not differ beyond the experimental uncertainty (the external angle between the emission and reflection is varied from 20° to 70°). The dependence of the rise time on the incident angle is expected to be weak since the inverse of the difference of the exciton wave vectors involved (of order 1000 Å is large compared with both the scale of interface disorder and the exciton Bohr radius that can

be considered as a characteristic scale for exciton-exciton scattering.

The much-improved time resolution also allows us to access the regime of spontaneous emission from a coherent superposition state of Wannier excitons. The PL in Figs. 1(a) and 2 features an initial temporal oscillation. The period of the oscillation corresponds to the hh and lh exciton splitting of the sample. As expected, decay of the oscillation becomes faster with increasing exciton densities, and the depth of the oscillation diminishes as we move the excitation pulse to lower energies. The oscillations are more pronounced for sample B, as shown in Fig. 2, because the ratio of the lh exciton to hh exciton population is larger for sample B than for sample A due to the smaller splitting (5 meV) between the hh and lh excitons for sample B.

Assuming the exciton momentum scattering time is much shorter than the initial decay time of the exciton PL, a simple rate equation analysis ignoring the initial oscillation shows that the rise and initial decay of the exciton PL can be described by $[1 - \exp(-\Gamma t)]\exp(-\kappa t)$, where Γ and κ are the momentum scattering rate and the initial PL decay rate, respectively. A numerical fit to the time evolution, shown in Fig. 2, gives $\Gamma^{-1} = 1$ ps and $\kappa^{-1} = 8$ ps. The decay time is in good agreement with earlier time-resolved studies that used excitation pulses with 4 meV spectral width [4]. Decay of the thermalized exciton distribution at much longer time scales (not shown in 2) also agrees with the earlier measurement [4]. A separate numerical fit to the oscillation gives a decay time of 2.5 ps for the oscillation.

We now discuss the physical origin of the quantum beats in the PL measurement. Quantum beats in spontaneous emission of atoms have been well understood and can be used as a guide for understanding quantum beats in exciton PL [8,9]. Quantum beats in FWM and Rayleigh scattering of semiconductors have been studied in detail in recent years [12,13]. Since we used circularly polarized excitation fields, the hh and lh transitions involved can be characterized by two independent transitions similar to that shown in energy structure (1) of Fig. 2. For two atoms (or excitons), energy structure (1) is equivalent to energy structure (2) where $|g_a g_b\rangle$ is the ground state, $|g_a e_b\rangle$ and $|e_a g_b\rangle$ are two excited states. Spontaneous emission from a coherent superposition of two excited states can lead to temporal oscillations, as is discussed in detail for atomic systems [8,9]. The frequency of the oscillation corresponds to the energy splitting between the two excited states. The amplitude of the oscillation is proportional to [8]

$$\mu_{eg}^{(a)}\mu_{ge}^{(b)}\langle e_b|\rho|g_b\rangle\langle g_a|\rho|e_a\rangle + \text{c.c.}, \quad (1)$$

where μ is the dipole moment operator, and ρ is the density matrix operator. As indicated by Eq. (1), decay of the oscillation in this case is determined by $\gamma_a + \gamma_b$, where γ_a and γ_b are the dephasing rates for the two transitions. More specifically, γ_a and γ_b are dephasing rates for off-diagonal matrix element $\langle g_a|\rho|e_a\rangle$ and $\langle e_b|\rho|g_b\rangle$, respectively. In analogy, the decay rate of the quantum beats in Figs. 1(a) and 2 should be given by $\gamma_{\text{hh}} + \gamma_{\text{lh}}$, the sum of the hh and lh exciton dephasing rates.

The gradual rise and presence of pronounced oscillations in the time-resolved PL, shown in Fig. 2, probe directly the relationship between exciton dephasing and momentum scattering. Figure 2 shows that the oscillation persists after the rise of PL. Similar behaviors are also observed in the range of exciton densities we used (10^9 to 10^{10} cm^{-2}). The rise time (1 ps) in Fig. 2 represents the momentum scattering time for hh excitons since PL comes primarily from the hh exciton. The decay time (2.5 ps) for the oscillation corresponds to a hh exciton dephasing time of 5 ps, assuming γ_{lh} is nearly the same as γ_{hh}, as shown by separate FWM measurements and also by earlier studies [14]. We note that separate dephasing measurements using FWM under similar experimental conditions are in general agreement with the result of PL measurements. Although it is desirable to perform FWM and time-resolved PL under identical conditions, the upconversion setup has prevented us from performing both measurements simultaneously.

The above analysis leads to an unexpected conclusion that the hh exciton dephasing time (5 ps) is much longer than the momentum scattering time (1 ps). Although there exists no microscopic description for exciton dephasing, the concept of dephasing is well established. For an excitonic system, quasielastic momentum scattering induces a phase shift in the probability amplitude of an individual state (or exciton). This induced phase shift leads to a change in the optically induced coherence given by $\delta\rho_{eg} = [\exp(i\theta) - 1]\rho_{eg}$, where θ is the relative phase shift between the excited and ground state, resulting in decay of the *macroscopic average* of the optically induced coherence (dephasing). In the limit of the impact approximation where scattering occurs instantaneously with regard to all other time scales except the inverse of the optical transition frequency, the scattering process can be considered Markovian and there exists no correlation between scattering events. For dilute atomic vapors where the impact approximation applies, one expects the dephasing rate to be greater than or at least equal to one half of the scattering rate [15]. The result that the exciton momentum scattering rate is 5 times the dephasing rate indicates the inadequacy of the impact approximation for exciton scattering and suggests the presence of correlation between scattering events [16]. The breakdown of the impact limit is likely due to the fact that the ratio of the exciton Bohr radius to the interexciton distance is much larger than the corresponding ratio for dilute atomic vapors. When the collision duration becomes comparable to the time separation between scattering events, successive scattering events can no longer be considered independent. The inadequacy of the impact limit is also hinted at in a recent study where a sublinear relationship between the exciton dephasing rate and exciton

density is suggested through the polarization dependence of the excitonic FWM response [17].

Finally, we discuss briefly the distinction between spontaneous emission and resonant Rayleigh scattering. Under resonant excitations it becomes very difficult to distinguish PL from resonant Rayleigh scattering in the initial regime [13]. Physically, the incoherent exciton PL is due to the coupling of the exciton with the vacuum and can be considered as coming from a fluctuating dipole induced by the vacuum field [18]. In contrast, the coherent Rayleigh scattering results from radiation of an induced macroscopic polarization, and decay of the Rayleigh scattering is determined by the dephasing of the induced polarization [18]. Decay of the PL shown in Fig. 2 is much slower than that expected for the resonant Rayleigh scattering, indicating that contributions from Rayleigh scattering are small. We also note that recent theoretical studies of the initial time evolution of resonant Rayleigh scattering suggest that the presence of static disorder can lead to a finite rise time in Rayleigh scattering [19]. The rise time observed in our studies, however, is strongly density dependent, showing that the rise is primarily due to exciton-exciton scattering as discussed earlier.

In conclusion, resonantly excited PL with femtosecond time resolution allows us to probe the very initial stage of exciton spontaneous emission, a regime that was not accessible previously. Our measurements demonstrate the crucial role of momentum scattering in the radiative process of extended optical excitations in semiconductors. The gradual rise and the presence of quantum beats in the PL probe directly the relationship between exciton dephasing and momentum scattering. The result reveals the failure of the impact limit and provides important new insight into the fundamental problem of dephasing processes in semiconductors. These studies should help to stimulate further efforts to formulate a microscopic description for exciton dephasing.

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