Stimulated Scattering of Indirect Excitons in Coupled Quantum Wells: Signature of a Degenerate Bose-Gas of Excitons

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(Received 19 January 2001)

We observe and analyze strongly nonlinear photoluminescence kinetics of indirect excitons in GaAs/AlGaAs coupled quantum wells at low bath temperatures, ≥50 mK. The long recombination lifetime of indirect excitons promotes accumulation of these Bose particles in the lowest energy states and allows the photoexcited excitons to cool down to temperatures where the dilute 2D gas of indirect excitons becomes statistically degenerate. Our main result—a strong enhancement of the exciton scattering rate to the low-energy states with increasing concentration of the indirect excitons—reveals bosonic stimulation of exciton scattering, which is a signature of a degenerate Bose-gas of excitons.

DOI: 10.1103/PhysRevLett.86.5608 PACS numbers: 78.67.De, 63.20.Kr, 72.10.Di

Since the pioneering work of Keldysh and Kozlov [1], the composite boson nature of interacting Wannier-Mott excitons (which are composed of two fermions: electron and hole) has been analyzed in detail. The prediction of exciton condensation in momentum space as well as of a possible crossover from Bose-Einstein condensate (BEC) of weakly interacting excitons at low densities to the BCS-like condensate, called an excitonic insulator, at high electron-hole densities has initiated a large number of experimental efforts [2–5]. The basic property of BE quantum statistics is the stimulated scattering: The scattering rate of bosons to a state $\mathbf{p}$ is proportional to $(1 + N_{\mathbf{p}})$, where $N_{\mathbf{p}}$ is the occupation number of the state $\mathbf{p}$. At high $N_{\mathbf{p}} \gtrsim 1$ the scattering process is stimulated by the presence of other identical bosons in the final state. The stimulated scattering is a signature of quantum degeneracy and yields unambiguous evidence for the bosonic nature of excitons. Observation of stimulated scattering requires less restricted experimental conditions than those necessary for BEC: With reducing temperature and/or increasing density of excitons the occupation numbers of the low-energy states increase and the stimulated scattering arises as a precursor of the BEC or BEC-like phase transition. The latter refers to the collective condensed phases [6,7] which are expected for interacting quasi-two-dimensional (2D) excitons.

In order to obtain large, nonclassical population of the exciton states, one needs systems with a high exciton cooling rate and a long recombination lifetime (after an initial optical excitation an exciton gas should have enough time to decrease its effective temperature T toward the lattice temperature T_b) and with a small exciton translational mass M_x (the temperature T_0 of quantum degeneracy is inversely proportional to $M_x^{D/2}$, where D is the dimensionality of the exciton system). The cooling of hot photoexcited excitons down to the temperature of the cold lattice occurs via emission of bulk LA phonons and is much more efficient for 2D systems as compared to bulk semiconductors. This follows from the relaxation of momentum conservation in the z direction for 2D systems: the ground-state mode $E = 0$ is coupled to the continuum of the energy states $E \geq E_0$, rather than to the single energy state $E = E_0 = 2M_x v_s^2$ (v_s is the longitudinal sound velocity) as occurs in bulk materials (see the kinetic equation below). As a result, the characteristic thermalization time of excitons in GaAs quantum wells (QWs) is 2–3 orders of magnitude shorter than that in bulk GaAs.

In contrast to bulk semiconductors, artificially grown semiconductor nanostructures allow us to tune, to some extent, both the mass M_x and the radiative lifetime τ_{opt}. In our attempt to create and control a quantum degenerate gas of excitons we employ a new arrangement of the experiment: an extremely cold (the minimal cryostat temperature $T_b = 0.05$ K) optically pumped high-quality GaAs/AlGaAs coupled quantum well (CQW) in the presence of a static voltage V_g ($V_g \simeq 1$ V) and magnetic field B ($0 \leq B \leq 16$ T). The primary advantage of CQWs for observation of a quantum degenerate 2D exciton gas and, ultimately, a BEC-like phase transition is the long radiative lifetime τ_{opt} of indirect excitons. The long lifetimes originate from the spatial separation between the electron and hole layers. By changing V_g and B one can change the recombination rate and translational effective mass of long-lifetime 2D indirect excitons.

Recently stimulated scattering has been observed in the system of microcavity (MC) polaritons, which are coupled modes of excitons and photons [8,9], as well as in a degenerate gas of Bose atoms [10]. Note that contrary to MC polaritons, the coupling of indirect excitons with photons is negligible and so the degenerate exciton gas is physically analogous to the degenerate gas of Bose atoms. However,

 0031-9007/01/86(24)/5608(4)$15.00

because M_x is 5–6 orders of magnitude smaller than typical atomic masses, the degeneracy temperature T_0 for excitons is much larger than that for Bose atoms and reaches 1 K at experimentally accessible densities of indirect excitons in CQWs.

In this Letter, we report an observation of a strong enhancement of the exciton scattering rate to the low-energy exciton states as the concentration ρ_{2D} of the indirect excitons is increased. This finding reveals the stimulated exciton scattering which, in turn, directly points out quantum degeneracy of the exciton gas.

An $n^+ - i - n^+$ GaAs/AlGaAs CQW structure was grown by molecular-beam epitaxy. The i region consists of a single pair of 8 nm GaAs QWs separated by a 4 nm $Al_{0.33}Ga_{0.67}As$ barrier and surrounded by 200 nm $Al_{0.33}Ga_{0.67}As$ barrier layers. The n^+ layers are Si-doped GaAs with $N_{Si} = 5 \times 10^{17}$ cm^{-3}. The electric field in the z direction is monitored by the external gate voltage V_g applied between n^+ layers ($V_g = 1$ V throughout the paper). The small in-plane disorder in the CQW is indicated by the photoluminescence (PL) linewidth $\sim$1 meV. The 50 ns laser excitation pulse has a rectangular shape with the edge sharpness $\sim$0.5 ns ($\hbar\omega = 1.85$ eV and the repetition frequency is 1 MHz). The experiments were performed in a He^3/He^4 dilution refrigerator or He^4 cryostat. Excitation and PL collection were performed normal to the CQW plane by means of a 100 μm optical fiber positioned $\sim$300 μm above 350 $\times$ 350 μm mesa (the total sample area was $\sim$4 mm^2).

The ground state of an optically pumped CQW at $V_g = 1$ V refers to indirect excitons constructed from electrons and holes in different layers (Fig. 1a). Unlike the direct exciton, the energy of the indirect exciton increases with ρ_{2D}, due to the net repulsive interaction between indirect excitons dipole oriented in the z direction. The energy shift allows us to estimate the concentration ρ_{2D} of the indirect excitons using the formula $\delta E = 4\pi\rho_{2D}e^2d/\varepsilon_b$ (d is the effective separation between the electron and hole layers): The strongest optical excitations $W = 10$ W/cm^2 (during the excitation pulse) used in our experiments yield the maximum $\rho_{2D}^{max} \simeq 2 \times 10^{10}$ cm^{-2}.

The PL kinetics of indirect excitons are shown in Fig. 1b. At low optical excitations, after a rectangular excitation pulse is switched off, the PL intensity of indirect excitons decays nearly monoexponentially (Fig. 1b). In contrast, at high excitations, right after the excitation pulse is switched off, the PL intensity first jumps up and starts to decay only in a few nanoseconds (Fig. 1b). The rate, τ_{rise}^{-1}, of the PL intensity enhancement after the excitation pulse is switched off increases strongly with increasing exciton density (Figs. 1d and 1e), lowering temperature (Fig. 1f), and reducing magnetic field (Fig. 1g).

The time-integrated PL intensity remains almost constant with V_g variation, while the decay time changes by several orders of magnitude [11]. Hence, radiative recombination is the dominant mechanism of the decay of

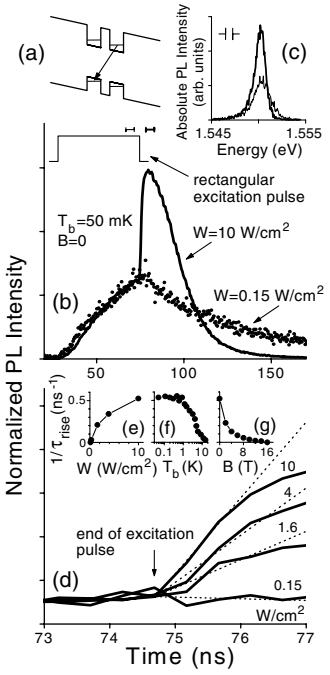

FIG. 1. (a) Energy band diagram of CQWs. (b) PL kinetics of indirect excitons at high and low excitations. The laser excitation pulse and the time intervals for spectrum integration are shown schematically above. (c) The PL spectra of indirect excitons at $W = 10$ W/cm^2 measured in 5 ns time intervals just before and after the end of the excitation pulse shown in (b). The spectral resolution is shown on the left. (d) PL kinetics of indirect excitons near the end of the excitation pulse at different excitations. The rate $1/\tau_{rise}$ of the PL rise right after the end of the excitation pulse is presented in (e), (f), and (g) vs W at $T_b = 50$ mK and $B = 0$, T_b at $W = 10$ W/cm^2 and $B = 0$, and B at $W = 10$ W/cm^2 and $T_b = 50$ mK, respectively. The fast enhancement of the PL intensity is observable only for high-quality CQWs and only at high excitations, low magnetic fields, and low temperatures.

indirect excitons in the high-quality samples studied. For delocalized, in-plane free, 2D excitons only the states with small center-of-mass momenta $|\mathbf{p}_\parallel| \leq p_0 \simeq E_g\sqrt{\varepsilon_b}/c$ (E_g is the band gap and ε_b is the background dielectric constant) can decay radiatively by resonant emission of bulk photons [12] (see Fig. 2a). Thus the PL dynamics is determined by the occupation kinetics of the optically active low-energy states $E \leq E_{p_0}$ ($E_{p_0}/k_B = p_0^2/2M_xk_B \simeq$ 1.2 K at $B = 0$). The LA-phonon-assisted relaxation of hot photoexcited indirect excitons into the optically active low-energy states results in a rise of the PL signal, while optical recombination of the low-energy indirect excitons results in a decay of the PL intensity. The end of the excitation pulse is accompanied by a sharp drop in the exciton temperature as caused by switching off the generation of hot indirect excitons and, as a result, the PL intensity and the occupation numbers $N_{E\leq E_{p_0}}$ abruptly rise within a few nanoseconds right after the trailing edge of the excitation pulse. The changes of the rate τ_{rise}^{-1} of the PL intensity enhancement with varying exciton density,

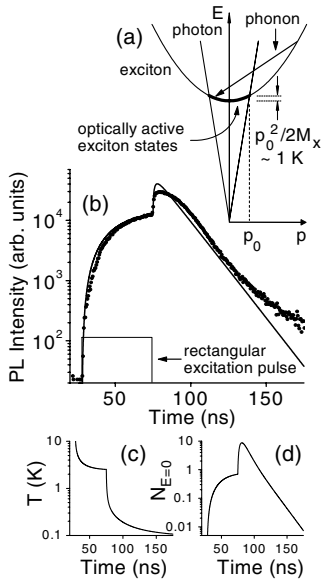

FIG. 2. (a) Energy diagram for the LA-phonon-assisted relaxation and optical decay of indirect excitons. The bold sector of the exciton dispersion indicates the radiative zone. The calculated dynamics of (b) the PL intensity of indirect excitons, (c) the effective exciton temperature, and (d) the ground-state occupation number (lines). The parameters used in the simulations refer to the experimental PL kinetics at $W = 10$ W/cm^2, $T_b = 50$ mK, and $B = 0$ shown in (b) by points ($M_x = 0.3m_0$, $2\tau_R \simeq 13.5$ ns, and $D_x = 6.5$ eV). A quantum degenerate gas of indirect excitons with $N_{E=0} \gg 1$ builds up in a few ns time domain after the optical excitation.

temperature, and magnetic field (Figs. 1d–1g) correspond to changes of the LA-phonon-assisted exciton scattering rate to the optically active exciton states $E \leq E_{p_0}$ (the influence of the recombination is negligibly small for this time domain).

The measured PL kinetics is strongly *nonlinear* (Figs. 1d and 1e): the observed increase of $\tau_{\rm rise}^{-1}$ with increasing exciton concentration ρ_{2D} (or W) proves that the scattering is stimulated by the final exciton state occupancy which, in turn, is high, $N_{\mathbf{p}\leq\mathbf{p}_0} \gtrsim 1$. The observed decrease of $\tau_{\rm rise}^{-1}$ with increasing temperature (Fig. 1f) results from a thermal reduction of the occupation of the low-energy exciton states $N_{\mathbf{p}\leq\mathbf{p}_0}$. The observed decrease of $\tau_{\rm rise}^{-1}$ in the presence of a magnetic field (Fig. 1g) is mainly caused by an increase of the magnetoexciton mass $M_x = M_x(B)$ (in our CQW M_x increases by a factor of 9.9 at $B = 16$ T, according to the theory [13]). Because of exponentially decreasing occupation numbers of the excited excitonic states which are coupled to the ground state ($N_{E\geq E_0} \propto e^{-E_0/T} = e^{-2M_x v_s^2/T}$), the increase of M_x relaxes the sharp reduction of the effective exciton temperature T right after the excitation pulse. As a result, at $t \geq \tau_{\rm pulse}$ the stimulated population of the low-energy states weakens, $N_{E=0}^{\rm max}$ decreases, and $\tau_{\rm rise}^{-1}$ smoothly drops with increasing B.

The theoretical calculations presented below are in good agreement with the experimentally observed PL dynamics of indirect excitons (Fig. 2b) and magnetoexcitons [14]. Thermalization of indirect excitons, initially distributed below the threshold for optical phonon emission, can be understood from the LA-phonon-assisted relaxation kinetics into the ground-state in-plane mode $\mathbf{p}_\parallel = 0$. The kinetic equation is given by [15] $\partial N_{E=0}/\partial t = \frac{2\pi}{\tau_{\rm sc}} \int_1^\infty d\varepsilon \times \varepsilon\sqrt{\varepsilon/(\varepsilon-1)}\,|F_z[a\sqrt{\varepsilon(\varepsilon-1)}]|^2 \mathcal{L}(N_{E=0}, n_\varepsilon^{\rm ph}, N_\varepsilon)$, where the collision integrand is $\mathcal{L}(N_{E=0}, n_\varepsilon^{\rm ph}, N_\varepsilon) = [N_\varepsilon(1 + n_\varepsilon^{\rm ph}) + N_{E=0}(N_\varepsilon - n_\varepsilon^{\rm ph})]$, $\varepsilon = E/E_0 = E/(2M_x v_s^2)$ is the dimensionless energy, $n_\varepsilon^{\rm ph} = 1/[\exp(\varepsilon E_0/k_B T_b) - 1]$, and N_ε are the distribution functions of bulk LA phonons and indirect excitons. The characteristic scattering time is given by $\tau_{\rm sc} = (\pi^2\hbar^4\rho)/(D_x^2 M_x^3 v_s)$, where ρ is the crystal density and D_x is the deformation potential. The form factor $F_z(\chi) = [\sin(\chi)/\chi][e^{i\chi}/(1 - \chi^2/\pi^2)]$ refers to an infinite CQW confinement potential. This function describes the relaxation of the momentum conservation in the z direction and characterizes a spectral band of LA phonons, which effectively interact with indirect excitons. The dimensionless parameter $a \sim 1$ is given by $a = (L_z M_x v_s)/\hbar$, where L_z is a QW thickness.

In our experiments with $T_b = 50$ mK one has that $k_B T_b \ll E_0$ and, therefore, $n_{\varepsilon\geq 1}^{\rm ph} \ll 1$ ($E_0/k_B \simeq 540$ mK at $B = 0$). As a result, LA-phonon vacuum is realized for the LA-phonon-assisted relaxation of indirect excitons [16]. For a degenerate exciton gas with $N_{E=0} > 1$ the stimulated kinetics proportional to $N_{E=0}$ is dominant, and the kinetic equation reduces to $\partial N_{E=0}/\partial t = \Lambda N_{E=0}$. In the presence of a continuous generation of hot indirect excitons the increment Λ proportional to $N_{\varepsilon\geq 1} - n_{\varepsilon\geq 1}^{\rm ph}$ is positive (this is *Fröhlich's inversion condition* [17] and is the counterpart of the population inversion condition in optical lasers [18]), and high occupation numbers of the low-energy states effectively develop with time.

Because of exciton-exciton interaction, at concentrations $\rho_{2D} \geq 10^9$ cm^{-2} the thermalization occurs through the quasiequilibrium thermodynamic states characterized by the BE distribution function $N_E^{\rm eq}(\rho_{2D}, T)$ and the effective temperature T of indirect excitons [19]. Here, $N_E^{\rm eq} = (1 - e^{-T_0/T})/(e^{E/k_B T} + e^{-T_0/T} - 1)$ and the degeneracy temperature is given by $T_0 = (\pi\hbar^2\rho_{2D})/(2M_x k_B)$. Within the relaxational thermodynamics the occupation number of the ground-state mode is given by $N_{E=0} = e^{T_0/T} - 1$, where T and $T_0 \propto \rho_{2D}$ are time dependent.

The PL dynamics of indirect excitons is modeled numerically by three coupled differential equations for the exciton temperature $T(t)$, the effective optical lifetime $\tau_{\rm opt}(T, \rho_{2D})$, and the concentration $\rho_{2D}(t)$, respectively. The generation and heating of indirect excitons, due to the excitation pulse, is included in the PL dynamics. Namely, according to the design of our experiments, we assume that at $0 \leq t \leq \tau_{\rm pulse} \approx 50$ ns the pump pulse creates hot indirect excitons at the energy $E^{(0)} \simeq 20$ meV above the bottom of the exciton band. The latter value refers to the generation process by resonant injection of indirect excitons from the optically populated states of direct excitons

(Fig. 1a). Figure 2b shows that the numerical simulations reproduce the observed PL dynamics. According to our calculations plotted in Fig. 2c, the end of the excitation pulse is indeed accompanied by a sharp drop of the effective temperature. According to Fig. 2d, the PL jump after the pump pulse is accompanied by $N_{E=0} \gg 1$, i.e., a strongly degenerate Bose-gas of indirect excitons builds up. In this case, the stimulated relaxation of excitons effectively populates the ground-state mode $\mathbf{p}_\parallel = 0$.

At the first few nanoseconds after the pump pulse, the thermalization kinetics is approximated by $k_B T(\tau) = E_0/\ln(\tau)$ and $N_{E=0}(\tau) \sim \tau^{k_B T_0/E_0}$, where the dimensionless time is given by $\tau = (2\pi^{3/2} E_0 t)/(k_B T_0 \tau_{\rm sc})$. Thus the power-law occupation dynamics of the ground-state mode accelerates at $\tau \geq 1$ with increasing $\rho_{\rm 2D}$.

Finally we discuss briefly an issue of in-plane disorder on the exciton PL kinetics. Stimulated scattering can occur to localized states as well as delocalized states. The existence of a broad PL line is no bar in principle to bosonic statistics of the individual states. If the disorder is strong enough to localize excitons on a scale approaching their Bohr radius, then confinement of multiple excitons in one single-particle state will induce strong interactions between the excitons and suppress the coherent occupation. So stimulated scattering will survive certainly if $l_{\rm loc} < \lambda$ but will likely require that $l_{\rm loc} \gg a_{\rm Bohr}$; the second criterion is equivalent (in the worst case, if disorder is on short length scales) to a PL linewidth much less than the exciton binding energy (well satisfied in our samples), and at best (if the disorder is on long length scales) to a much weaker constraint [20].

We appreciate valuable discussions with D. S. Chemla, N. A. Gippius, L. V. Keldysh, and Y. Yamamoto. Support of this work by the RFBR, QUEST, EPSRC, and INTAS is gratefully acknowledged.

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