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Phase Diagram of Degenerate Exciton Systems

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Degenerate exciton systems have been produced in quasi–two-dimensional confined areas in semiconductor coupled quantum well structures. We observed contractions of clouds containing tens of thousands of excitons within areas as small as (10 μm)2 near 10 kelvin. The spatial and energy distributions of optically active excitons were determined by measuring photoluminescence as a function of temperature and laser excitation and were used as thermodynamic quantities to construct the phase diagram of the exciton system, which demonstrates the existence of distinct phases. Understanding the formation mechanisms of these degenerate exciton systems can open new opportunities for the realization of Bose–Einstein condensation in the solid state.

Over the last few years, the demonstration of Bose-Einstein condensation (BEC) in several atomic species confined in optical or magnetic traps has triggered intense interest (1, 2). A variety of quasi–particles with bosonic character are also found in condensed matter. In particular, semiconductors can sustain bound electron–hole (e-h) pairs, called excitons (Xs) that have small effective mass (on the order of the free electron mass) and behave as bosons in the hole (e-h) pairs, called excitons. Excitons (Xs) are a variety of quasi–particles with bosonic character of atomic species confined in optical or magnetic excitations (200). Theoretical work (3.4) suggests that Xs can undergo BEC at a critical temperature (Tc) of 1 K, a factor of 106 that for atoms.

The quasi–two-dimensional nature (quasi–2D) of CQWs raises important theoretical issues related to phase transitions in reduced dimensionality. BEC only occurs at T = 0 in a homogeneous 2D system of ideal bosons (14, 15) but can occur at finite T when they are confined in 2D traps (16, 17). Hence, producing local confined cold Xs systems may be a key step to realizing excitonic BEC. In that respect, applying the TC formulae for ideal bosons in a square 2D box (17) to the confined Xs observed in GaAs/AlGaAs CQWs gives TC ≈ 1 K for reachable experimental conditions.

We studied n~i~n GaAs/AlGaAs CQWs composed of two 8-nm GaAs wells separated by a 4-nm Al0.33Ga0.67As barrier (18). Collection of cold, dense indirect–X systems confined within localized areas of CQW samples was reported (19). Formation of photoluminescence (PL) ring patterns around localized excitation spots was also observed in CQW (20, 21). We present here a study of the phase diagram and clarify the formation mechanisms of X systems confined in localities, which, for brevity, we continue to refer to as “traps.” For the phase diagram, the thermodynamic quantities are the spatial and energy distributions (including emission energies and linewidths) of the optically active indirect Xs, measured with diffraction limit resolution by PL as a function of temperature (T) and laser excitation (P laser).

The phase diagram displays distinct regimes with one, in particular, that corresponds to the concentration of tens of thousands Xs in small areas (~10 μm)2 at low temperature and moderate excitation. Our investigations imply that the observed X degenerate systems are not caused by condensation of Xs preexisting in the CQW structure, but that indirect Xs are formed in a vacuum but in an extremely dense (~1023 particles per cm3) solid matrix. The X density, nX, can be widely varied with the photoexcitation, and when nX approaches the level where the interparticle spacing becomes comparable to aX, a crossover from Bose to Fermi statistics occurs. At sufficiently high densities, screening of the e-h coulomb attraction prevents the binding of e-h pairs, and the X phase transition occurs. In some semiconductors, a gas–liquid phase transition can also occur where the X gas condenses into an e-h liquid (3–5, 8, 9).

The crucial issue from a practical viewpoint is the realization of cold, statistically degenerate X systems. Because Xs are quasi–particles of excited semiconductors with a finite lifetime, this requires Xs to cool down by emitting phonons and to reach quasi–equilibrium in a time much shorter than their lifetimes. In semiconductor coupled quantum wells (CQWs) under a static electric field perpendicular to the QW plane (Z direction), the ground state is spatially an indirect X with the electron confined in one QW and the hole in the other (10, 11). This separation reduces the electron and hole wave function overlap, which results in an increased radiative lifetime that in our GaAs/AlGaAs CQWs samples is about two orders of magnitude that of the direct X (e-h pairs in the same QW) (11). The CQWs are embedded in a three-dimensional (3D) structure resulting in relaxation of the momentum conservation along Z; therefore, cooling to the lattice temperature by the emission of bulk longitudinal acoustic phonons is about 3 orders of magnitude faster for Xs in GaAs QWs than for that in the bulk (12). Finally, the e-h spatial separation gives rise to a repulsive dipolar X-X interaction that effectively screens the in-plane disorder potential (13).

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in the immediate vicinity of the traps through the distinct photoassisted capture and injection of electrons and holes.

The PL images of the indirect-X cloud for three temperatures $T = 1.7$ K, 8 K, and 20 K (Fig. 1A) at a low and uniform laser excitation power, $P_{\text{laser}} = 50$ $\mu$W, show that contraction of the X cloud is evident with decreasing bath temperature. The cloud radius decreases more than an order of magnitude to $\sim 10$ $\mu$m, whereas the peak intensity increases by a factor of 50. The 2D PL images at $T = 1.7$ K (Fig. 1B) for three $P_{\text{laser}}$ values shows that with increasing $P_{\text{laser}}$, the X cloud first contracts and then expands, exhibiting an annular shape with a darker central region that also shrinks and then expands with $P_{\text{laser}}$. The color bar scale on the figure shows the local density of Xs estimated from the PL intensity by the methods described below. From these images, we deduce the integrated PL intensity ($I_{\text{PL}}$), the root-mean-square radius ($\sigma_{\text{PL}}$), the skewness (asymmetricity), and kurtosis ($K_{\text{PL}}$—concentration of high peaks as compared with a Gaussian distribution). These parameters are used as thermodynamic quantities to characterize the X fluid.

The phase diagram of $\sigma_{\text{PL}}(T, P_{\text{laser}})$ is shown in Fig. 2A. At either high temperature or low excitation intensity the X cloud is extended, whereas both low temperature and significant excitation are required for concentration of Xs in a small area. Indeed, $\sigma_{\text{PL}}$ is at its minimum ($\sim 10$ $\mu$m) for $T < 4$ K and moderate excitation ($P_{\text{laser}} = 35$ $\mu$W to 500 $\mu$W). In that range, the kurtosis phase diagram $K_{\text{PL}}(T, P_{\text{laser}})$ (Fig. S1) demonstrates that the X distribution is more concentrated than a Gaussian, despite the central intensity dip. It is noteworthy that $\sigma_{\text{PL}}$ exhibits steepest variations in a narrow domain—nearly vertical for $P_{\text{laser}} \approx 20$ $\mu$W and diagonal for $P_{\text{laser}} \geq 20$ $\mu$W around $\sigma_{\text{PL}} = 30$ $\mu$m, which reveals the existence of distinct phases. At fixed $T \leq 10$ K, as $P_{\text{laser}}$ is increased, the X cloud first contracts when $P_{\text{laser}}$ crosses $\sim 20$ $\mu$W, plateaus over a small range of moderate intensities, then expands. At fixed $P_{\text{laser}}$ as $T$ decreases, the X cloud contracts and experiences a sharp reduction in size near $T = 10$ K. This behavior is seen more clearly in $T$-dependent cuts of $\sigma_{\text{PL}}$ for five $P_{\text{laser}}$ values between 25 $\mu$W and 6 mW (Fig. 2B). For $P_{\text{laser}} = 25$ $\mu$W, $\sigma_{\text{PL}}(T)$ exhibits a clear kink near $T = 10$ K that confirms the sudden cloud contraction. However, that kink becomes less pronounced at higher $P_{\text{laser}}$ as $\sigma_{\text{PL}}$ becomes progressively $T$ independent. This transition is further supported by the corresponding normalized $I_{\text{PL}}(T)$ curve (Fig. 2C), which, at low excitation, shows a strong $T$ dependence with an almost steplike increase between $T = 15$ K and 10 K. As the excitation density increases, the magnitude of $I_{\text{PL}}(T)$ increases by a factor of 100 but its $T$ dependence becomes progressively flatter.

The PL intensity is only sensitive to the radiative recombination of optically active, bound or unbound e-h pairs with center of mass momentum $K \sim 0$ (22, 23). It may not reflect the X density, which, for CQW Xs, is more directly related to the PL peak energy ($E_x$) blue shift due to the indirect-X interaction (24). Therefore, to better characterize the energy distribution of the indirect-X systems, we used the PL scans that provide local spectroscopic information with a $\Delta \nu \sim 1.7$ $\mu$m spatial resolution (Fig. 3A). In Fig. 3B, the black curve gives the indirect-X PL intensity profile, $I_{\text{PL}}(y)$ (spectrally integrated over the indirect-X peak), for $P_{\text{laser}} = 200$ $\mu$W and $T = 1.7$ K. Consistent with the annular structure of the Fig. 1B images, $I_{\text{PL}}(y)$ exhibits a central dip surrounded by two intense maxima. The $E_x$ profile (magenta curve) shows that the blue shift increases monotonically from the cloud edges to a maximum at the center. Thus, the indirect Xs at the cloud center are dense but hot ($K > 0$), not as optically active as those cold ($K \sim 0$) ones in the bright annulus. This is consistent with the variations of the PL linewidth $\Delta \nu I_{\text{PL}}(y)$ (red curve), which narrows sharply where $I_{\text{PL}}(y)$ is maximal and broadens a little at the center. This behavior is observed in the region where the indirect-X cloud is concentrated. But for lower $P_{\text{laser}}$ (less than about 100 $\mu$W), the $E_x$ profile reported previously (19) first appears, and then, near the center, $E_x$ gradually flattens.
The \( I_{\text{pl}(s)}(y) \) mean square radius, \( \sigma_{\text{pl}(s)} \), and the PL linewidths at the two minima, \( \Delta E_{\text{pl}(s)} \), are plotted versus \( P_{\text{laser}} \) (Fig. 3C) for \( T = 1.7 \) K. The \( \sigma_{\text{pl}(s)}(P_{\text{laser}}) \) curve agrees with the behavior seen in Fig. 2A, i.e., the X cloud contracts to a small area, \( \sigma_{\text{pl}(s)} \approx 7 \, \mu m \), at low \( T \) for moderate \( P_{\text{laser}} \). Interestingly, the linewidth \( \Delta E_{\text{pl}(s)} \) (\( P_{\text{laser}} \)) curves show a parallel variation versus \( P_{\text{laser}} \). At low \( T \), where phonons are scarce, the low-density PL linewidth in a QW is dominated by the in-plane potential fluctuations, an effect particularly pronounced in the statistically deformed wells. In the case of indirect Xs, however, the repulsive dipole-dipole interaction screens the fluid forms in regime II where the environmental fluctuations are smoothed out and the effects of scattering reduced or even suppressed.

To better understand the X collection mechanisms, we first characterized the sample under zero applied electric fields (Fig. 4A), where the PL of direct Xs dominates and the localized indirect-X cloud disappears. Molecular beam epitaxially grown QW structures have typical thickness fluctuations on the order of one monolayer, corresponding to about a 0.5-meV energy variation for our 8-nm QWs. The direct-X PL peak has a constant energy across the trap within our 0.1 meV resolution; therefore, QW thickness fluctuations are unlikely to be the main cause of collection. A weak photocurrent is also observed, and at the trap center, a broad (\( \Delta E \approx 10 \) meV) low-intensity emission \( \approx 1.565 \) eV is seen. This signals the presence of local inhomogeneous fields and/or current flows. Using the PL from the heavily doped \( n^+ \)-GaAs layer, we determined that the Fermi level lowered by \(-1 \) meV at the trap center, which indicated a lighter local electron concentration that may be related to variation in doping and/or vacancies or a photogenerated carrier drain.

We have further investigated the electric field–dependent PL and the photocurrent characteristics of the samples under different excitation conditions: (i) various excitation energy \( h \nu \), (ii) different excitation spot sizes—uniform or localized (a spot of \( \approx 5 \) to \( \approx 10- \mu m \) diameter), and (iii) continuous wave (CW) or pulsed excitation. In the experiments presented thus far, e-h pairs are photogenerated uniformly over a large area by a HeNe laser at \( h \nu = 1.96 \) eV, \( \approx 20 \) meV above the \( Al_{0.33}Ga_{0.67}As \) barrier’s bandgap \( E_{\text{bg}} \). The effects reported above are readily observed for excitation energy \( h \nu \) between \( E_{\text{bg}} \) and \( E_{\text{bg}} \) of \( \approx 2.34 \) eV. Note that they disappear sharply when \( h \nu \) is tuned below \( E_{\text{bg}} \) or above \( E_{\text{bg}} \), even though the excitation is adjusted to create about the same density of e-h pairs in the QW.

Last, we performed two other sets of experiments: a comparison of \( I_{\text{pl}} \) from the cloud...
under uniform CW excitation and remote (∼100 μm away) localized excitation, and transient measurements under remote, localized 10- to 20-ns pulsed excitation at hν = 1.95 eV. In the first case, shown in Fig. 4B, under uniform excitation, IP, varies approximately linearly with P laser, whereas under remote localized excitation (see Fig. 4B, inset, X-cloud PL images), IP, first grows linearly and then exhibits a kink followed by a superlinear growth at P laser ≈ 200 μW. In the second case, the transient photocurrent response consists of a brief and intense pulse followed by a weak, steady long-lived (nearly 10-μs) component. Correspondingly, the PL dynamics (Fig. 4C) shows an extremely long decay time of the indirect Xs. These results imply that the indirect-X clouds are formed not only by the collection of photogenerated carriers, but also by a continuous formation of indirect Xs through photoinduced carrier transport and injection across the n’-i-n” structure.

Putting all these observations together, we propose the following model. The localized dark spot at the center of the X cloud is associated with a pinhole that funnels a current filament through the n’-i-n” structure. This source of current is smaller than or on the order of our spatial resolution of ∼2 μm. In the absence of photoexcitation, it contributes only to the leakage current, but provides a localized source of majority carriers of one type. For low excitation density, corresponding to regime I, carriers of the opposite type flow toward the current source by the carrier imbalance mechanism proposed and modeled in (29, 30). Indirect Xs are formed where the two types of carriers merge, which results in an extended indirect-X system. As the photoexcitation increases, at some point, both types of carriers are injected into CQWs, which results in an X density strongly peaked at center (Fig. 3B) and a concentrated cloud as shown in the central image of Fig. 1B. This corresponds to regime II. As the excitation density increases further to reach regime III, effects of the underlying Fermi statistics become apparent. The picture that has emerged is quite different from that of a condensation of Xs preceding in the structure. Nevertheless, we have found conditions of temperature and photoexcitation that result in the formation of highly statistically degenerate cold Xs systems where quantum mechanical collective effects can be observed. The insights gained in the formation mechanisms of the degenerate indirect X systems can lead to new strategies for producing confined cold X systems artificially in a more controllable way.

Fig. 4. (A) Spatially resolved spectra at the center of and 40 μm from the trap under zero applied electric fields. (B) Integrated PL (IP) with a uniform CW (black squares) and remote (∼100 μm away) localized (red circles) HeNe excitations. The blue dashed line is a guidance of linear dependence. The inset PL images are taken with P laser = 15, 290, and 1100 μW. The color bar scale represents the estimated optically active X density for images scaled by ×1, ×5, and ×10, respectively. (C) Dynamics of the PL from the indirect-X cloud measured with an ∼20-ns pulsed localized (−10-μm diameter spot) excitation of hν = 1.95 eV 100 μm away. The laser has an average P laser = 26 μW and 100 kHz repetition rate. The PL exhibits a long decay that can be divided approximately into three stages with decay time τ ∼ 180 ns, 1 μs, and 5 μs. This signals a continuous formation of indirect Xs, because the lifetimes of indirect Xs are typically on the order of 100 ns only.

References and Notes
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Materials and Methods
Fig. S1
Movie S1
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