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Coexistence of Bose-Einstein paraexcitons with Maxwell-Boltzmann orthoexcitons in Cu_2O

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Time-resolved photoluminescence studies in Cu_2O at high excitation levels and intermediate lattice temperatures ($T_L \simeq 30$ K) exhibit a nearly classical orthoexciton distribution with gas temperature somewhat higher than T_L during the excitation pulse. Simultaneously, a paraexciton spectrum with width less than half that of the classical distribution is observed during the pulse, indicating the quantum-statistical nature of this high-density component.

Few excitonic systems have been observed to display Bose-Einstein statistics. Biexcitons in CuCl, and excitons in Ge and Cu_2O have exhibited such effects.[1-4] Time-resolved photoluminescence studies of highly excited Cu_2O at low temperature have demonstrated a quantum-degenerate orthoexciton gas which is near Bose-Einstein condensation (BEC).[4] Further experiments[5-7] have investigated the two-component nature of this system by examining both the (triply degenerate) orthoexcitons and the (singly degenerate) paraexcitons which are split by an exchange energy of 12 meV. The most intriguing results are that the lower-lying paraexcitons, produced at densities above the critical density for condensation, exhibit an anomalous spectrum and a rapid spatial expansion, suggestive of BEC and superfluidity of excitons.

To gain further insights into this unusual quantum system, we now investigate the exciton gas in Cu_2O under somewhat different experimental conditions. The previous experiments[3-7] were conducted at a lattice temperature of about 4 K. The present work is conducted at higher lattice temperatures, in an effort to increase the fraction of paraexcitons in the gas due to an increased ortho-to-para conversion rate at higher temperatures.[8,9] Indeed, we find that, in contrast to the quantum distributions observed at low crystal temperatures, the orthoexciton spectrum displays a nearly classical (Maxwell-Boltzmann) energy distribution indicative of a low-density gas. Simultaneously, the paraexcitons exhibit a spectrum which is narrower than the k_BT width associated with the lattice temperature of about 30 K, clearly demonstrating the quantum-statistical nature of a high-density gas.

For these studies a natural-growth crystal of Cu_2O is cooled by a cold ^{4}He gas in a Janis varitemp cryostate. The crystal is excited by a cavity-dumped Ar^+ laser ($\lambda = 514$ nm, pulse width = 10 ns) which produces a localized gas of excitons near the crystal surface (laser absorption length $\simeq 1.6\ \mu$m). At the high pulse-repetition rates (10^5–10^6 Hz), the crystal lattice reaches a steady-state temperature determined by the average laser power and the helium cooling rate. The lattice temperature is determined by the orthoexciton energy distribution measured long after the pulse. A Maxwell-Boltzmann distribution is observed, which has been shown to agree well with the lattice temperature in low power continuous-excitation experiments.[4] The luminescence spectra obtained by time-resolved photon counting[4-6] are shown in Fig. 1. The solid lines in Figs. 1(a) and 1(b) are the paraexciton spectra at two sampling times. The strong off-scale signal at the right-hand side is due to an orthoexciton replica. The dashed lines in Figs. 1(a) and 1(b) are the simultaneous orthoexciton spectra, shifted in energy and normalized in

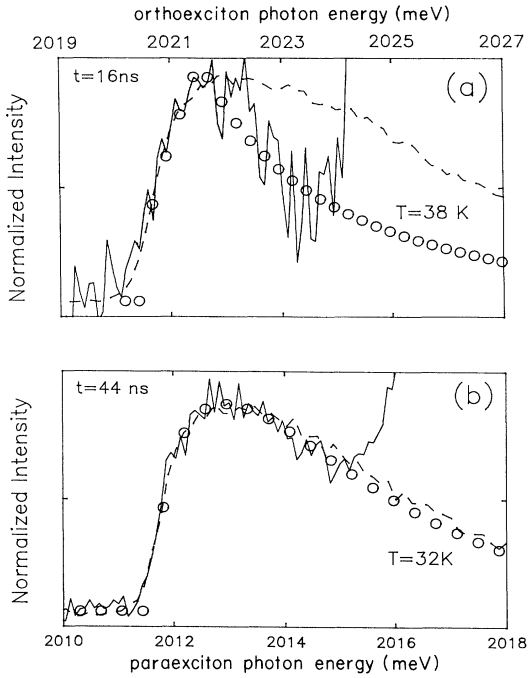

FIG. 1. (a) The orthoexciton (dashed line) and paraexciton (solid line) distributions during laser-pulse excitation. The laser has a peak incident power of about 16 W and is focused to approximately 30 μm. The two spectra are normalized to the same height and shifted in energy for comparison. The respective photon energy scales are shown at the top and the bottom of the figure. The orthoexciton distribution fits a Maxwell-Boltzmann (MB) distribution with $T = 38$ K. The paraexciton distribution is obscured by an orthoexciton line at energies above 2015 meV. The open circles represent a 1.3-meV-broadened Bose-Einstein distribution with $\alpha = 0.07$ and $T = 38$ K. (b) The distributions well after the laser pulse. Both fit a MB distribution with $T = 32$ K (open circles).

intensity to compare with the paraexciton component.

During the pulse [e.g., $t=16$ ns in Fig. 1(a)], the orthoexciton energy distribution fits quite well to a Maxwell-Boltzmann distribution with temperature $T_{ex}=38$ K. This is slightly higher than the temperature observed at later times. The time evolution of the spectral full width at half maximum (FWHM), which equals $1.8k_BT_{ex}$ for a classical distribution, is plotted as the open circles in Fig. 2(a). At late times, the temperature approaches a constant value, $T_{ex}\simeq 32$ K, which we interpret as the steady-state lattice temperature T_L.

The most striking result is the behavior of the paraexciton distribution. During the pulse, the paraexciton spectrum is considerably narrower than that of the orthoexcitons, as seen in Fig. 1(a). The full width at half maximum of this spectrum is less than $0.9k_BT_L$ where $1.8k_BT_L$ is the spectral width of a classical gas at the lattice temperature [Fig. 1(b)]. At a later sampling time, $t=44$ ns, the paraexciton spectrum broadens to match the orthoexciton distribution. The time evolution of the spectral width for the paraexcitons is shown as the solid circles in Fig. 2(a).

The narrowing of this spectrum at high gas density must be due to Bose-Einstein statistics. To get an estimate of the chemical potential of the paraexciton gas, we have fit the spectrum in Fig. 1(a) to a Bose-Einstein distribution,

$$I(E)=I_oE^{1/2}/[\exp(\varepsilon+\alpha)-1]\,, \qquad (1)$$

where $\varepsilon=E/k_BT$ and $\alpha=-\mu/k_BT$ are the kinetic energy and chemical potential, normalized to k_BT. Assuming $T=38$ K, determined from the orthoexciton spectrum, the open circles in Fig. 1(a) correspond to $\alpha=0.07$. To obtain a good fit to the low-energy edge of the data, a broadening of 1.3 meV is required. This is a factor of 2 larger than the spectral resolution of 0.7 meV. The source of this broadening is unknown, but similar effects were observed in the low-temperature experiments and ascribed to interactions in the gas (e.g., collisional broadening or interaction with a condensate).[5,6] The extra broadening observed in the degenerate paraexciton spectrum leads to some uncertainty in the value of α (0.05–0.15) determined from the fit.

These spectral measurements indicate that the density of the paraexcitons is much greater than that of the orthoexcitons. This conclusion can be tested by comparing the relative luminescence signals for the two species. Previous work[10] has shown that the ratio of radiative efficiency for these two luminescence replicas is 500. Figure 2(b) compares the orthoexciton luminescence intensity ($2.018\text{ eV}<E<2.028$ eV) to that of the paraexcitons ($2.011\text{ eV}<E<2.015$ eV), multiplied by 500 to show the relative densities of the two species. Indeed the paraexciton gas is about 5 times more dense than the orthoexciton gas, which is consistent with the respective spectral distributions.

The present experiments demonstrate that, even at lattice temperature of 32 K, it is possible to produce a gas of paraexcitons exhibiting quantum statistics. This is the first time that we have observed spectra which are narrower than those predicted for a Maxwell-Boltzmann distribution at the lattice temperature. In the previous work,[3–7] both orthoexciton and paraexciton components displayed temperatures well above the lattice temperature, due in part to Auger heating of the gas.[11] In the present case, the increased lattice temperature causes a more rapid thermalization of the gas, in particular, a more efficient conversion of orthoexcitons to paraexcitons and probably shorter exciton lifetimes. While the spatial distribution of excitons has not been measured in this case, it is expected that the gas occupies a smaller volume than that at lower temperature because of the increased scattering with phonons at higher lattice temperatures. At low temperatures, it was shown that the orthoexciton and paraexciton components occupy the same effective volume. Assuming this to also be true in the present case, our experiments illustrate the interesting coexistence of a quantum-statistical paraexciton gas with a classical orthoexciton gas.

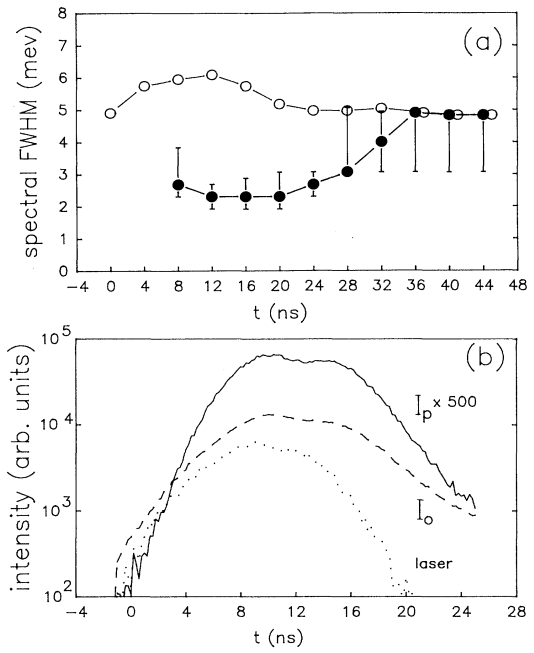

FIG. 2. (a) The spectral FWHM of each energy distribution as a function of time (open circles: orthoexciton; solid circles: paraexciton). The laser pulse is 10 ns wide, centered at $t=10$ ns. (b) The spectral integrated luminescence intensity of orthoexciton (dashed line) and paraexciton (solid line) as a function of the time. The laser pulse is the dotted line. The paraexciton line is multiplied by 500 so that the intensities represent the relative densities of the two species. The spectral sampling ranges are proportional to the widths of the two lines.

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[10]The paraexciton density may be estimated either from fitting the Bose-Einstein spectral distribution or by comparing the relative luminescence intensities of orthoexcitons and paraexcitons. The latter method relies on a measurement of the relative absorption strengths (equal to 50) of the Γ_5 phonon replicas of the two species made by P.D. Bloch and C. Schwab, Phys. Rev. Lett. **41**, 514 (1978). Due to the orthoexciton degeneracy of 3, this measurement implies that the orthoexciton optical matrix element is a factor of $\frac{50}{3}=17$ greater than that of the paraexciton. The degeneracy factor was mistakenly omitted in Refs. 5 (text) and 6 (Figs. 10 and 11), leading to an overestimate of the paraexciton density by a factor of 3. Thus the relative radiative efficiency of the Γ_3 orthoexciton and the Γ_5 paraexciton replicas is 500, not 1500, as assumed in those references. The main conclusions of Refs. 5 and 6 remain unchanged, and the factor of 3 correction brings the density of paraexcitons estimated by their luminescence intensity into closer agreement with the density derived from their kinetic-energy distribution.

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