**Bose-Einstein condensation of excitons in Cu$_2$O**

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We present a parameter-free model that estimates the density of excitons in Cu$_2$O, related to experiments that have tried to create an excitonic Bose-Einstein condensate. Our study demonstrates that the triplet-state excitons move along adiabats and obey classical statistics, while the singlet-state excitons are a possible candidate for forming a Bose-Einstein condensate. Finally we show that the results of this study do not change qualitatively in a two-dimensional exciton gas, which can be realized in a quantum well.

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**I. INTRODUCTION**

The field of Bose-Einstein condensation (BEC) has attracted much attention in recent years, especially in the context of trapped alkali-metal atoms (2). Much experimental effort has also been put in creating a Bose-Einstein condensate of excitons, both in three-dimensional crystals (3,4), and in quantum wells (10). Excitons are bound states of electrons and holes, which form in semiconductors after the electrons get excited from the valence to the conduction band with use of some external probe, a laser beam of light, for example. As the absorption spectrum of infrared electromagnetic radiation induces transitions from the 1s to the 2p level (12) under realistic conditions, and in Sec. V we investigate the behavior of a quasi-two-dimensional exciton gas. Finally we examine the two dominant mechanisms.

We also give suggestions and predictions for future experiments, which would put an end to the discrepancies described above. We also examine the conditions under which a Bose-Einstein condensate would form, since, as we show, this is possible for the paraexcitons. Finally, in connection with excitons in quantum wells, we demonstrate that our conclusions are unaffected by the density of the ortho and paraexcitons as function of their temperature, based on a parameter-free model. We also give suggestions and predictions for future experimental studies, which would put an end to the discrepancies described above. We also examine the conditions under which a Bose-Einstein condensate would form, since, as we show, this is possible for the paraexcitons. Finally, in connection with excitons in quantum wells, we demonstrate that our conclusions are unaffected by the two-dimensional exciton gas.

This paper is organized in the following way: we give in Sec. II a simple and analytical argument for the fact that the orthoexcitons move along adiabats. We estimate the density of the orthoexcitons and from that we extract the paraexciton density in Sec. III, discussing the long-time behavior of the paraexcitons. In Sec. IV we analyze the absorption spectrum of infrared electromagnetic radiation inducing transitions from the 1s to the 2p level under realistic conditions, and in Sec. V we investigate the behavior of a quasi-two-dimensional exciton gas.
nisms that determine the behavior of the orthoexcitons in Sec. VI, and summarize our results in Sec. VII.

II. ADIABATIC BEHAVIOR OF THE ORTHOEXCITONS

Let us first of all assume that the orthoexcitons and the paraexcitons occupy the same volume inside the crystal, and that they are determined by a common temperature. Our claim is that the orthoexcitons in this system move along adiabats as a result of the balance between two competing mechanisms: a heating mechanism that comes from an ortho-plus-ortho to para-plus-para conversion mechanism \[13\], and an acoustic-phonon cooling process \[14\]. A similar argument was used in Ref. \[14\], but the heating process was assumed to be an Auger mechanism \[13\]. However, in view of the more recent experimental measurements \[1\] and theoretical calculations \[13\], this Auger mechanism has a negligible rate. The model we present here is essentially free of adjustable parameters, which makes it very reliable \[13\].

Let us therefore give the general argument, and then examine each mechanism separately. We know from elementary thermodynamics that

\[
\frac{dU_o}{dt} = T dS_o - p_o dV + \mu_o dN_o, 
\]

where \(U_o\) is the internal energy, \(S_o\) is the entropy, \(p_o\) is the pressure, \(\mu_o\) is chemical potential, \(N_o\) is the number of species \(i\), and \(V\) is the volume of the exciton gas.

Solving Eq. (2) in terms of \(TdS_o\), and expanding the differential \(d(S_o/N_o)\) we get

\[
Td \left( \frac{S_o}{N_o} \right) = \frac{1}{N_o} \left[ dU_o - \left( \mu_o + \frac{T S_o}{N_o} \right) dN_o + p_o dV \right]. \tag{3}
\]

We show below that the orthoexcitons are in the classical regime, and for an ideal monatomic classical gas \(\mu_o + T S_o/N_o = 5k_BT/2\), and also \(p_o V = N_o k_BT\). Therefore, Eq. (3) can be written as

\[
Td \left( \frac{S_o}{N_o} \right) = \frac{1}{N_o} \left[ dU_o - \frac{5}{2} k_BT dN_o + n_o k_BT dV \right]. \tag{4}
\]

The differential \(dU_o\) is determined by the two mechanisms we mentioned. More specifically, as discussed in Sec. VI, the heating rate is equal to \(an_o\), \(a\) being a constant, i.e., it scales linearly with the orthoexciton density \(n_o\), whereas the acoustic-phonon cooling rate is equal to \(-bT^{3/2}\) (for \(T \gg T_l\), where \(T_l\) is the lattice temperature), \(b\) being a constant, i.e., it scales as \(T^{3/2}\). Therefore Eq. (3) implies that

\[
T \frac{d}{dt} \left( \frac{S_o}{N_o} \right) = \frac{an_o}{2} - \frac{bT^{3/2}}{2} \frac{k_BT dN_o}{N_o dt} + \frac{k_BT dV}{V dt}. \tag{5}
\]

Let us now examine the terms on the right side of Eq. (5). It turns out that the first two are the dominant ones.

More specifically as we show in Sec. VI the order of magnitude of these two terms (i.e., the typical energy-exchange rates that come from the phonon cooling and the ortho-to-para exchange heating) is \(\gtrsim 10\) meV/ns. On the other hand, concerning the last term that comes from the expansion of the orthoexcitons, a detailed study of this problem has given that the diffusion constant is \(D \sim 10^3\) cm\(^2\)/s \[20\]. Even at the initial stage of the expansion of the cloud when its radius is \(R \gtrsim 10\) \(\mu\), \(dV/(V dt) \sim D/R^2 \lesssim 1\) ns\(^{-1}\); thus this term is \(\lesssim 1\) meV/ns for \(T \approx 10\) K. Concerning the third term on the right side of Eq. (5) one has to distinguish between two different cases corresponding to two different experimental conditions: For long-pulse laser excitation, of order a few tens of nanoseconds, the population of orthoexcitons is essentially determined by the laser and it follows its profile (see, for example Fig. 4(a) of Ref. \[3\]). Therefore the characteristic timescale \(\tau_L\) that is related to this mechanism is a few tens of nanoseconds, and the corresponding term entering Eq. (5) is \(\sim k_BT/\tau_L\), or \(\lesssim 0.1\) meV/ns, and thus negligible. Under these conditions Eq. (5) takes the form

\[
T \frac{d}{dt} \left( \frac{S_o}{N_o} \right) \approx an_o - bT^{3/2}. \tag{6}
\]

Equation (6) is one of the basic results of this study. To see what this equation implies, it is instructive to give the two following formulas connecting the entropy, number, density, and temperature of the orthoexcitons (which are valid for any ideal Bose gas) \[21\].

![FIG. 1. The orthoexciton trajectories on the density-temperature plane. The solid line gives the root of the polynomial on the right side of Eq. (6), whereas the dashed line gives the root of the polynomial on the right side of Eq. (6).](image-url)
\[
\frac{S_o}{N_o k_B} = \frac{5}{2} \frac{g_3/2(z_o)}{2 g_3/2(z_o)} - \ln z_o; \quad (7)
\]

\[
\frac{n_o}{T^{3/2}} = \left(\frac{m k_B}{2 \pi h^2}\right)^{3/2} g_3/2(z_o), \quad (8)
\]

where \(z_o = e^{\mu_o/k_B T}\) and

\[
g_\alpha(z) \equiv \frac{1}{\Gamma(n)} \int_0^\infty x^{n-1} e^x - 1, \quad (9)
\]

with \(\Gamma(n)\) being the gamma function. From the above equations one sees that along lines of constant \(\mu_o/k_B T\), i.e., of constant \(z_o\), \(S_o/N_o\) and \(n_o/T^{3/2}\) are both constant. Therefore along adiabats, i.e., along lines with constant \(S_o/N_o, n_o \propto T^{3/2}\) and according to Eq. (4), the orthoexcitons approach an adiabat, and more specifically the one for which \(n_o = (a/b) T^{3/2}\). This is a line of “equilibrium” for the orthoexcitons, which comes as a balance between the heating and the cooling mechanisms. Using the values for the constants \(a\) and \(b\) given in Sec. VI, this adiabat is [see the solid line in Fig. 1]

\[
n_o \left(10^{16} \text{cm}^{-3}\right) \approx 10^{-2} \left(3 T^{3/2} \text{K}\right), \quad (10)
\]

where the notation \(n_o \left(10^{16} \text{cm}^{-3}\right)\) means that the density is to be measured in units of \(10^{16} \text{cm}^{-3}\), and correspondingly the temperature in Kelvin. The coefficient \(10^{-2}\) in Eq. (10) should be compared with \(\approx 3.3\) given by Eq. (6), which gives the phase boundary, and therefore this adiabat is in the regime where the excitons should not exhibit any sign of quantum degeneracy, since \(-\mu_o/k_B T \gg 1\). For a typical exciton temperature of order 20 K, the above equation implies that the orthoexciton density is of order \(10^{16} \text{cm}^{-3}\), which is consistent with that estimated experimentally in Refs. [5–8].

One can also study the dynamics of orthoexcitons. To do this, it is convenient to introduce the dimensionless quantity \(\alpha = -\mu_o/k_B T\) and rewrite Eq. (4) in the form [14]

\[
\frac{d\alpha}{dt} = -\frac{\alpha - \alpha_s}{\tau_s}, \quad (11)
\]

where \(\alpha_s\) is the value of \(\alpha\) along the specific adiabat with \(n_o = (a/b) T^{3/2}\). Also

\[
\tau_s = \frac{T}{b} \left[\frac{\partial (S_o/N_o)}{\partial n_o}\right]_T \approx \frac{5\zeta(5/2) k_B}{2\zeta(3/2) aT^{1/2}}. \quad (12)
\]

Equation (11) has to be solved along with the rate equations describing the change in the exciton density of each species,

\[
\frac{dn_o}{dt} = G_o(t) - \frac{n_o}{\tau_{o,p}} + \frac{n_p}{\tau_{p,o}} - \frac{n_o}{\tau_o}, \quad (13)
\]

\[
\frac{dn_p}{dt} = G_p(t) - \frac{n_p}{\tau_{p,o}} + \frac{n_o}{\tau_{o,p}} - \frac{n_p}{\tau_p}, \quad (14)
\]

where \(G(t)\) is the laser production rate of excitons, \(\tau_i\) are the intrinsic radiative lifetimes, \(\tau_{o,p}\) is the rate for the ortho-to-para conversion process, and \(\tau_{p,o}\) is the rate for the (reverse) para-to-ortho conversion process. As shown in Ref. [3], \(\tau_{p,o}^{-1} \approx cn_o\) (\(c\) is a constant that is given in Sec. VI), whereas as explained in Sec. VI the reverse process is thermally supressed, \(\tau_{p,o} \ll \tau_{o,p}\). In addition the radiative lifetimes of excitons are very long (longer than microseconds), and thus Eqs. (13) and (14) take the simple form

\[
\frac{dn_o}{dt} \approx \frac{n_o}{\tau_{o,p}}, \quad (15)
\]

\[
\frac{dn_p}{dt} \approx \frac{n_p}{\tau_{p,o}}. \quad (16)
\]

Equations (13) and (14), viewed as densities being functions of time, do not depend on the exciton temperature and can be integrated. Therefore Eqs. (13), (15), and (16) can be integrated to give \(n_o(t), n_p(t),\) and \(T(t)\) [14]. However we are interested in \(n_o(T)\), i.e., in the dependence of \(n_o\) on the exciton temperature \(T\), and that is given directly by Eq. (11), which implies that the orthoexcitons approach the adiabat with \(\alpha = \alpha_s\) on a timescale of order \(\tau_s\), which is of order of a few tens of picoseconds under typical exciton temperatures.

In another class of experiments short laser pulses (on the order of 100 ps) have been used in order to excite the crystal. In this case one must include the third term on the right side of Eq. (3) which, however, does not change appreciably our basic results. More precisely Eq. (3) takes the following form in this case

\[
\frac{T}{dt} \left(\frac{S_o}{N_o}\right) \approx an_o - \frac{b}{T} - \frac{5c}{2} k_B T n_o. \quad (17)
\]

The dashed line in Fig. 1 shows the root of the polynomial \(an_o - \frac{b}{T} - \frac{5c}{2} k_B T n_o = 0\). Although the deviation from the adiabatic behavior is not substantial, it is in agreement with the experimental observations, as one can see in Fig. 2 of Ref. [7], for example. The open circles in this graph correspond to short laser pulse excitation (\(\approx 100\) ps) and they show some small deviation from the adiabats, as opposed to the solid circles (corresponding to long pulses, \(\sim 10\) ns) that follow very closely an adiabat.

From the discussion we have presented up to now there is a contradiction: on the one hand spectroscopically the orthoexcitons show a high degree of quantum degeneracy \([1, 2]\). On the other hand, we argued that the orthoexcitons should not show any sign of quantum degeneracy (under equilibrium conditions), in agreement with the experimental data presented in Refs. [5–8]. We will not address this issue here, which is an open question for future studies.
III. QUANTUM DEGENERACY OF PARAEXCITONS

Let us now turn to the paraexcitons. Their radiative lifetime has been determined to be on the order of milliseconds \(^{[8,22]}\), which allows them to form a cold gas, with a temperature very close to \(T_1\) on these timescales. Since on such timescales and for such low temperatures, all the excitons will have converted to paraexcitons, they are also expected to establish a relatively high density, which we estimate now, given the orthoexciton density we got earlier. Statistically due to the multiplicity of the orthoexcitons, one expects that \(N_o/N_p = 3\). Therefore the paraexciton density (assuming that all the orthoexcitons get converted into paraexcitons) is four times the one we found earlier. However, as we argued the temperature is expected to get very close to that of the lattice for late times, i.e., for times much larger than the exciton - acoustic phonon scattering time (on these timescales the ortho-to-par conversion process will have converted all the orthoexcitons to paraexcitons and there will be no heating due to this mechanism). Therefore for an initial exciton temperature \(T_i = 40\) K, the paraexciton density can get up to \(10^{17}T_i^{3/2}(40 \text{ K}) \text{ cm}^{-3}\), as Eq. (10) implies, where \(T_i\) is to be measured in units of 40 K. Even for a density of \(10^{17} \text{ cm}^{-3}\) the critical temperature for Bose-Einstein condensation of the paraexcitons is approximately 2 K, i.e., liquid-helium temperatures. One important conclusion of this analysis is, therefore, that the paraexcitons in the experiments that have been performed \(^{[3,8]}\) should be very close to the phase boundary, or they might have even crossed it already. In the experiment of Ref. \(^{[8]}\) the paraexcitons were reported to have crossed the phase boundary for Bose-Einstein condensation. The whole analysis was based on spectroscopically analyzing the (only) phonon-assisted recombination line of paraexcitons, which is very weak, and is close to other, much stronger lines. An alternative way of probing experimentally the degree of quantum degeneracy of paraexcitons is presented in the following section.

IV. ABSORPTION SPECTRUM OF RADIATION INDUCING INTERNAL TRANSITIONS: PROBING THE QUANTUM DEGENERACY

In a possible experiment \(^{[22]}\) that has been proposed and has been studied theoretically in Ref. \(^{[12]}\), it was shown that the absorption spectrum of infrared radiation inducing internal transitions of the excitons from the 1s to the 2p level is very sensitive to the degree of quantum degeneracy of the gas. Therefore, in such an experiment one should be able to observe the contribution of the orthoexcitons and the paraexcitons to the absorption separately, with an energy separation of order \(\Delta E\) (assuming that the width of each distribution is of order \(k_B T \ll \Delta E\)). Here \(\Delta E\) is the energy splitting between the orthoexcitons (lying higher than) the paraexcitons due to the exchange interaction \(^{[11]}\). At the zone center \(\Delta E \approx 12 \text{ meV in Cu}_2\text{O}\), which corresponds to approximately 150 K. In a temporal study of this experiment, following a short laser-pulse excitation (like the one already used of a few hundred picoseconds) the contribution of the orthoexcitons to the absorption of infrared radiation would vanish within a timescale of order \(\tau_{o,p}\), i.e., of order nanoseconds under typical conditions. On the other hand the contribution of the paraexcitons to the absorption would last much longer, on a timescale of order of their radiative lifetime, i.e., milliseconds \(^{[9,22]}\). As shown in Ref. \(^{[12]}\) the appearance of two distinct peaks in the absorption spectrum of paraexcitons would signal the presence of a Bose-Einstein condensate, since in the condensed phase one deals with a two-component system and the two peaks would indicate the two different collective modes of it. But even if the paraexcitons have not crossed the phase boundary, but they are highly degenerate, that would still show up clearly in the absorption spectrum \(^{[2]}\). An additional advantage of this method is that it does not depend on the strength of the radiative recombination lines (which is very weak for the paraexcitons, as we mentioned in the preceding section).

It is interesting that if the lattice temperature increases and becomes of order the orthoexciton-paraexciton splitting, that would result in more or less equal rates for the orto-to-para and the para-to-ortho conversion processes, and would decrease the paraexciton density substantially \(^{[23]}\). In such a case the orthoexciton lifetime would be determined by the radiative lifetime, i.e., microseconds \(^{[8,22]}\). Therefore keeping \(T_i\) as low as possible is very crucial, since it (i) enhances the paraexciton density, and (ii) determines the temperature of the paraexcitons for late times.

V. A TWO-DIMENSIONAL EXCITON GAS

As a final remark we comment on the possibility of confining the excitons and creating a quasi-two-dimensional gas, like in quantum wells \(^{[10]}\). As we show in Sec. VI in two dimensions the acoustic-phonon cooling rate scales linearly with the temperature for \(T \gg T_1\). On the other hand the ortho-to-para conversion mechanism is unaffected by the dimensionality of the system. Thus the argument we gave earlier in Eq. (2) now takes the form

\[
T \frac{d}{dt} \left( \frac{S_o}{N_o} \right) \approx a \sigma_o - bT, \tag{18}
\]

where \(\sigma_o\) is the orthoexciton surface density. For \(S_o/N_o\) to be constant in two dimensions \(\sigma_o \propto T\), and therefore according to Eq. (13) the orthoexcitons in \(\text{Cu}_2\text{O}\) would still move along an adiabat \(\sigma_o = (b/a)T\). In addition a two-dimensional gas is expected to undergo a Kosterlitz-Thouless transition \(^{[24]}\) to a superfluid state along lines on which \(\sigma_o \propto T\) and thus the orthoexcitons would
also move along the phase boundary for the Kosterlitz-Thouless transition without crossing it, as opposed to the paraexcitons that would have the chance of undergoing this phase transition.

VI. ORTHO TO PARA CONVERSION AND ACOUSTIC-PHONON COOLING

Let us now examine in detail the two processes we mentioned earlier, starting with the spin-exchange mechanism, where two orthoexcitons collide, exchange their electrons or their holes, resulting into two paraexcitons in the final states. This process has been studied in detail in Ref. [13] and the decay time $\tau_{o,p}$ has been calculated to be in very good agreement with experiment [4], with essentially no adjustable parameter. The decay rate was shown to be given by

$$\tau_{o,p}^{-1} \approx 5n_o(10^{16}\text{ cm}^{-3})\text{ ns}^{-1}. \quad (19)$$

The reverse process is thermally suppressed for temperatures much lower than $\Delta E/k_B$. The ortho-to-para conversion is also a heating mechanism, since there is a gain of energy $\Delta E$ per particle. Therefore the heating rate due to this process is given by

$$\frac{1}{N_o} \left( \frac{\partial U_o}{\partial T} \right)_{o,p} = \frac{\Delta E}{\tau_{o,p}} \approx 60n_o(10^{16}\text{ cm}^{-3})\text{ meV/ns}. \quad (20)$$

We conclude with the cooling process of excitons due to their scattering with phonons. The excitons interact with the lattice vibrations, and since the lattice temperature $T_l$ is kept low (i.e., a few K), the excitons, which in general have a higher temperature, are cooled down. At the low energies we consider here the process is dominated by collisions with acoustic phonons. The corresponding cooling rate per particle is given by

$$\frac{1}{N_o} \left( \frac{\partial U_o}{\partial T} \right)_{\text{ph}} \approx -0.56T^{3/2}(K)\left(1 - \frac{T_l}{T}\right)\text{ meV/ns}. \quad (21)$$

The above cooling rate scales as $T^{3/2}$ for $T \gg T_l$, with the matrix element due to deformation potential theory, the acoustic-phonon energy, and the density of states in three dimensions each contributing a factor of $T^{1/2}$ (they scale linearly with the momentum exchange). For a two-dimensional gas the density of states is constant, and the cooling rate is proportional to $T$ is this case.

VII. SUMMARY

In conclusion, based on a parameter-free model, we have given estimates for the density of the excitons in Cu$_2$O as a function of temperature. The triplet state excitons are expected to move along adiabats, and to obey classical statistics. We have argued that this adiabatic behavior is a result of the competition between a spin-exchange heating mechanism, and an acoustic-phonon cooling process. An open question that needs to be investigated is the chemical potential of orthoexcitons, which, although they obey classical statistics, is very close to zero.

In addition, according to our model, the singlet-state excitons, are a possible candidate for forming a Bose-Einstein condensate. Finally, we have shown that in a two-dimensional exciton gas the basic conclusions of our study remain the same.

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It has been argued that a phonon-assisted mechanism also converts orthoexcitons to paraexcitons \[18,19\]. We ignore this mechanism here, but since its rate scales as \( T^{3/2} \), even if it is non-negligible, it does not change the results of the present study, apart from pushing the orthoexcitons to an adiabat which is even further away from the phase boundary, and enhancing the paraexciton density.

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