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LETTER TO THE EDITOR

Quantum saturation of ortho-excitons near Bose–Einstein condensation

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Abstract. The quantum saturation of ortho-excitons relative to the Bose–Einstein condensation of para-excitons is explained in terms of indefiniteness of N_{ortho} in contrast to the conserved N_{para} as the exciting laser pulse declines in intensity.

Recently, Bose–Einstein condensation (BEC) has been observed in some dilute alkali atomic gases [1–4]. This discovery is strongly dependent on the technology of laser cooling and magnetic trapping of a small amount of atoms in a small region. Due to the heavy mass and the low density of these atoms, the transition temperature for these alkali atomic gases is extremely low (roughly 2 μK).

When we turn to condensed matter physics, examples of the effects of BEC abound. The condensation of Cooper pairs gives rise to superconductivity. That of liquid helium leads to superfluidity. Although interaction among particles is of crucial importance, a rough estimate of the scale of the transition temperature can be acquired via that of BEC in the noninteracting limit. As a result of the much lower electron mass in the Cooper pair and the much greater density in the liquid helium, the critical temperatures for these cases are much higher, roughly from $T_c \sim 4$ K for helium to tens of kelvin for superconductors. However, the role of interaction in the former and the overlapping of the Cooper pair wave functions in the latter render the role played by BEC less clearcut.

BEC has also been the subject of intense study in excitonic systems [5]. Wolfe and co-workers [6–8] have performed a pioneering set of experiments on the BEC of excitons in bulk semiconductors such as Cu_2O. The singlet para-excitons rather than the triplet ortho-excitons are favoured in the formation of a Bose–Einstein condensate since the electron–hole exchange interaction produces an ortho–para splitting of $\Delta\epsilon = 12$ meV, causing the singlet para-excitons to have a lower ground state. BEC has indeed been reported for singlet para-excitons in Cu_2O [7, 8].

Because the lifetimes of para-excitons in relatively pure, natural-growth crystals have been measured to be several microseconds [9], while the ortho-exciton lifetime at 2 K is typically 30 ns due to down conversion to para-excitons, and because the interconversion between ortho- and para-excitons is slower than their lifetimes, each species can be assumed to have a distinct and independent chemical potential, μ_i. On the other hand, the rapid collisions between the excitons lead to a single common internal gas temperature, T, within a few picoseconds, although this T may be different from the ambient lattice temperature. It is expected, therefore, that the triplet ortho-excitons would also undergo BEC independently at sufficiently low temperatures or high densities. However, no evidence for a condensed phase

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has been seen in the ortho-exciton spectra. Instead, the ortho-exciton gas displays what Wolfe *et al* termed quantum saturation [6]; i.e., $n/n_c = \text{constant} \lesssim 1$, where n_c is the critical particle density for BEC. The dimensionless chemical potential, $\alpha \equiv (\epsilon - \mu)/k_B T$, was observed [8] in stressed Cu_2O to saturate at $\alpha = 0.04$. Here ϵ marks the band bottom (zero kinetic energy level) of the ortho-exciton. We recall, of course, that the phase boundary for BEC is specified by $\alpha = 0$. Thus, the ortho-exciton gas never crosses the BEC phase boundary. In contrast, the para-exciton luminescence spectrum was observed to develop an extra component at low energy which is interpreted as a BEC of para-excitons [8], i.e., the BEC phase boundary is crossed! It is still something of a puzzle why the behaviours of the two species were so different.

In the following we shall attempt to give an explanation of the above puzzle, and to specifically answer why the ortho-exciton density $n(T)$ should obey the relation

$$n(T)/n_c(T) = \text{constant} \lesssim 1 \tag{1a}$$

or equivalently

$$\alpha \equiv \beta(\epsilon - \mu) = \text{constant} \gtrsim 0 \tag{1b}$$

so that

$$n(T) = CT^{3/2} \tag{1c}$$

as $n(T)$ itself varies over an order of magnitude. In other words, why should ortho-excitons exhibit quantum saturation?

To explain equations (1*a*), (1*b*) and (1*c*), we review the experimental data for clues. The main information can be gleaned from figures 4(a) and (b) and figure 1(c) in [7], and figures 2(a)–(d) and figure 3 in [6]. We shall focus on figures 4(a) and (b) from [7] here. Figure 4(a) shows that as the laser pulse intensity rises towards its peak value, n_{ortho} and n_{para} also rise similarly and closely follow each other. Correspondingly, the temperature T also rises over the same duration in such a way that $\beta(\epsilon - \mu) = \text{constant} \gtrsim 0$, or the curves of both n_{ortho} and n_{para} versus T closely parallel the BEC phase boundary, as shown in figure 4(b). Similar information is also shown in figure (2). While it is natural to expect the exciton densities to rise as the laser pulse gains in intensity, thereby exciting more and more electron–hole pairs, it is intriguing to see they would obey equation (1) separately according to figure 4(a), until we recall the origin of the chemical potential parameter μ. In the canonical ensemble, the parameter μ is invoked as a Lagrange multiplier to implement the condition of conservation of particles. For systems containing an indefinite number of particles, such as a photon gas or a phonon gas, there is no need for this parameter, resulting in $\mu = 0$ so that the free energy F is minimized. In the present case, the excitons of either species are created by the photons, whose increasing number is directly responsible for the change of the total N_{ortho} and N_{para}. So again, like the photons their creators, the indefinite total N's lead to $(\epsilon - \mu) \gtrsim 0$ which, in turn, yields

$$\begin{aligned} n &= CT^{3/2} \\ C &= 2.61\left(\frac{2\pi m k_B}{h^2}\right)^{3/2} \end{aligned} \tag{2}$$

for both species in the present system.

After the peak of the laser pulse, the light intensity declines. The short-lived ortho-excitons would not be able to sustain the value of n_{ortho} achieved at the peak of the pulse. So, N_{ortho} would also decline proportionally with the pulse, as shown in figure 4(a). The fact that there is almost no time lag as n_{ortho} starts to decrease immediately following the decline of the pulse implies an extremely short lifetime of the ortho-exciton in the scale of the plot [10]. This

indefiniteness of N_{ortho} leads again to the same relation between n_{orhto} and T as given by equation (2) during the declining stage of the laser pulse, as observed in figure 4(b). Thus the BEC boundary is never crossed by the ortho-excitons—quantum saturation! That the observed average $\epsilon_{ortho} - \mu_{ortho}$ derived from the spectra is slightly greater than zero may be caused by the spatial inhomogeneity of the gas density according to [6].

On the other hand, since the lifetime of the para-excitons is comparatively long [10], they will outlive the entire lifespan of the laser pulse. As the laser pulse declines from its peak value, n_{para}, not yet suffering from decay and still being pumped by the remnant of the pulse, can still increase for about 5 ns according to figure 4(a), before levelling off until way beyond the duration of the pulse. In the meantime, the gas temperature rises and falls roughly following the profile of the laser pulse according to figures 4(a) and (b), as expected on physical grounds. As n_{para} levels off, the total N_{para} is conserved due to its slow decay, unlike its behaviour during the rise of the pulse. The definiteness of the total N_{para} now requires, as usual, the existence of the chemical potential μ_{para}. As the temperature falls with the demise of the pulse to below T_c at $t \simeq 11$ ns from the start of the laser pulse according to figure 4(b) while N_{para} remains constant, BEC follows! Here T_c is given by equation related to equation 4,

$$T_c^{3/2} = (n_{para})/C. \tag{3}$$

This completes the resolution of the puzzle.

In conclusion, we have shown that the indefiniteness of N_{ortho}, which is related to the various relaxation processes of the excitons [10], is ultimately responsible for its quantum saturation near BEC.

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