BOSE-EINSTEIN CONDENSATION OF FREE EXCITONS IN AgBr

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ABSTRACT

Triplet excitons in AgBr are observed in photoluminescence at low temperatures with continuous near UV-excitation. The triplet exciton emission intensity varies strongly with temperature and excitation density. In particular this latter dependence supports the interpretation that these triplet excitons show Bose-Einstein condensation. The experiments also allow a determination of the critical temperature T_c. At low exciton densities ρ, T_c changes according to the ideal gas behaviour ($\sim\rho^{2/3}$) but differently at high exciton densities. By analysing the experimental data we arrive at some general conclusion about the observability of Bose-Einstein condensation of excitons.

I. INTRODUCTION

The AgBr samples were cut from very pure crystals originating from two different sources,1) and were kept in the dark until they were immersed in liquid helium. Care was taken in mounting the crystals in order to minimize strains. Further experimental details can be found elsewhere.2,3) Figure 1 shows the "TA_L"-phonon replicum of the exciton emission obtained with continuous near UV-excitation from an Ar^+-laser. The assignment of the phonon agrees with the recent inelastic neutron scattering data:4) It is the TO_L-phonon with the symmetry of the TA_L-mode. The broader emission in Fig. 1 has a line shape determined by a Boltzmann distribution with a temperature of about 10 K, independent of experimental conditions.2,5) This emission is due to the dipole allowed, phonon assisted, radiative recombination of singlet excitons. The sharp emission in Fig. 1 is attributed to a weakly dipole allowed, phonon assisted, radiative recombination of triplet excitons. The zero field singlet-triplet splitting is in agreement with magneto-absorption experiments[6)] and the weak dipole character of this transition is due to an admixture of Ag d-states to the valence-band states at the L-point.[3,7)] Thus triplet excitons are expected to have longer lifetimes than singlet excitons and consequently their temperature is lower.

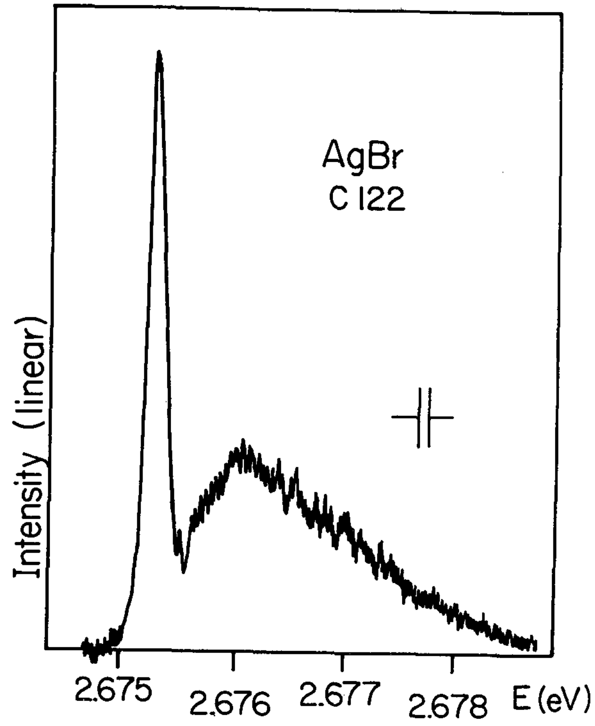


Fig.1. "TA_L"-phonon assisted exciton spectrum of AgBr obtained with laser excitation (3.40/3.52 eV) at 1.27 K.

II. PROPERTIES OF TRIPLET EXCITON LUMINESCENCE

The intensity of the triplet exciton luminescence depends strongly on temperature and on excitation density, compare Fig. 2. In these experiments[8)] the laser power has been kept constant but the beam has been focused differently. It is mainly the excitation density dependence which supports our interpretation: The rather sharp luminescence line of Fig. 1 is due to radiative recombination of triplet excitons which show Bose-Einstein condensation (BEC), *i.e.* which are in the total momentum K = 0 state. This model qualitatively explains our experiments.[3)] The intensity is proportional to N_o, the number of triplet excitons in the BEC-state which is given by

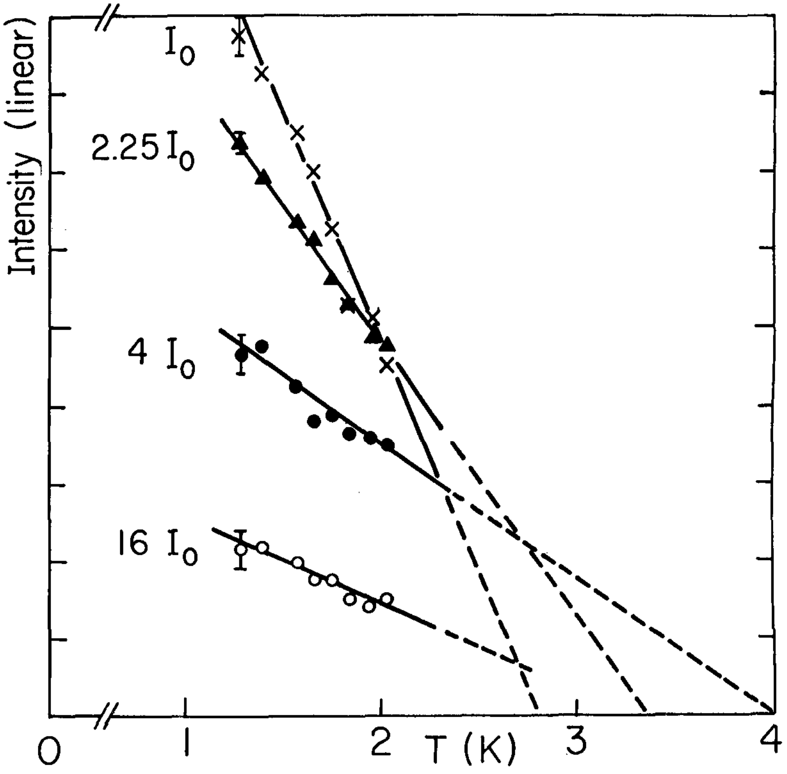
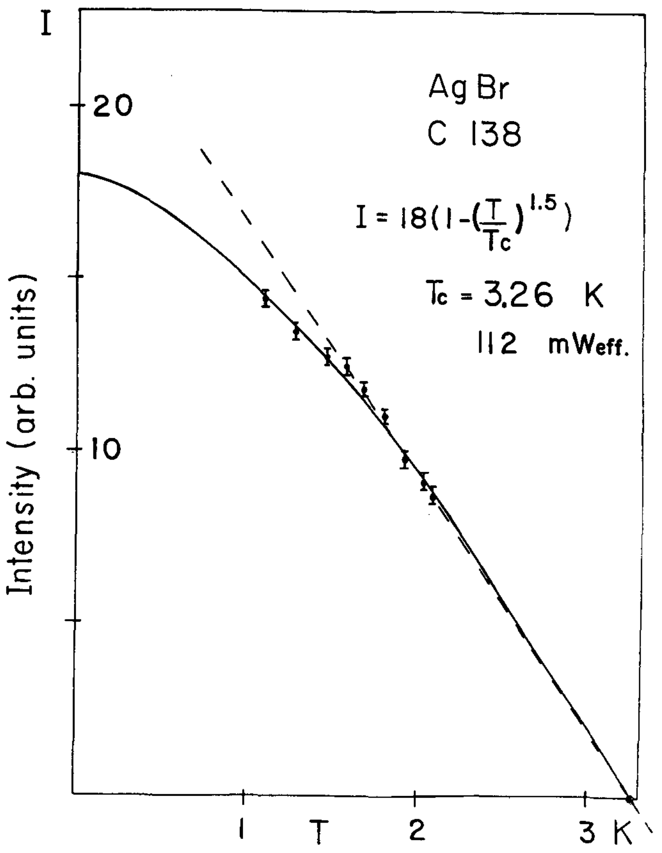


Fig.2. Intensity as a function of temperature and excitation density (unnormalized). The numbers are relative excitation densities.

$$N_o = N(1 - (\frac{T}{T_c})^{3/2}) , \qquad (1)$$

for an ideal gas. N is the total number of excitons and T_c is the critical temperature as obtained by a linear extrapolation of the luminescence intensity as a function of temperature to the value zero. The only free parameter is N. As exemplified in Fig. 3 our most accurate data can be fitted quite well to a dependence according to eq. (1). Also the variation with excitation density displayed in Fig. 2 is in qualitative agreement with our model: By increasing the excitation density one increases the total exciton density ρ and consequently T_c will increase. For an ideal gas, for instance, $T_c(\rho)$ is given by

Fig.3. Experimental intensities obtained for $i_{eff} = 112$ mW fitted to an ideal Bose gas temperature dependence.



$$T_c = \frac{2\pi\hbar^2}{Mk} (\rho)^{2/3} \frac{1}{[\zeta(3/2)]^{2/3}} , \quad \zeta(3/2) = 2.612\ldots \qquad (2)$$

Furthermore, one would argue that with increased density a possibly existing exciton-exciton interaction becomes increasingly effective and therefore the low temperature intensities will be decreased as is expected from the theory of a weakly interacting Bose gas, compare for instance.[3)]

III. EXCITATION DENSITY DEPENDENCE

By observing the luminescence spectrum Fig. 1 as a function of excitation density we have checked at several points that the result is independent whether the laser power is varied at constant illuminated area or whether the focus has been changed at constant laser power, provided the data are normalized with respect to the same illuminated area. We therefore believe that drastic heating effects are not present.

The exciton spectrum in Fig. 1 is inherently broadened by the width of the emitted phonon (~0.14 meV $\simeq$ 1.68 K$^{2)}$) due to the indirect band gap in AgBr. Thus the observed intensity $I(\omega)$ is proportional to the density of states convoluted with this effective "slit width" ω_o.

$$I(\omega) \sim \frac{1}{\omega_o} \int_{\omega-\frac{1}{2}\omega_o}^{\omega+\frac{1}{2}\omega_o} d\omega' \sqrt{\omega'} \; n(\omega') \quad , \tag{3}$$

where $n(\omega')$ is the triplet exciton distribution. It is therefore impossible to perform a detailed line-shape analysis since ω_o is of the order of the bath temperature and since the high energy tail of the spectrum is masked by the much broader singlet exciton emission. However, some information about the exciton statistics can be obtained from an analysis of the observed dependence of the luminescence intensity on excitation density. To simplify the analysis we consider in Fig. 4 only the difference, ΔI, of the intensities of the sharp line of Fig. 1 observed at two different temperatures as a function of excitation power i_{eff}.$^{9)}$ At very weak excitation the difference varies linear with i_{eff} and therefore also linear with ρ. This is expected if the exciton follows Boltzmann statistics. At some higher value of the excitation a drastic deviation occurs until ΔI is practically independent of the excitation density and therefore also of the total exciton density. This behaviour is expected if BEC occurs in an ideal gas, since all particles added to the system go into the condensate, whereas the thermal occupation depends only on temperature and not on density. In forming the intensity difference ΔI the density dependent terms cancel. The full horizontal line in Fig. 4 corresponds practi-

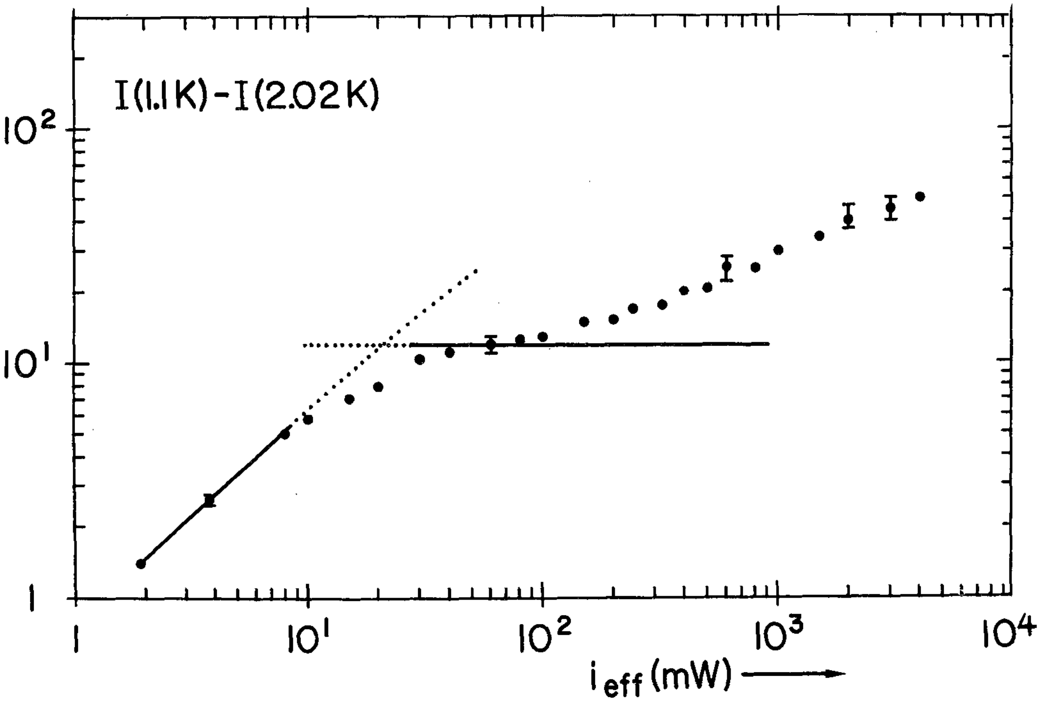


Fig. 4. Intensity difference ΔI = I(1.1 K) - I(2.02 K) as a function of laser power.

cally to the excitation-density range shown in Fig. 2. It is important to note that in agreement with the above explanation the temperature dependence of the intensity Fig. 3 only applies to the nearly horizontal range. Equation (1) cannot be fitted to the temperature dependences at very low excitation. At very high excitations we expect to leave the region for BEC again, as will be explained in the next section. In the whole range of Fig. 4 the observed line shape does not change since it is broadened by ω_o as explained above. This is also the reason why no excitation or temperature thresholds have been observed at the onset of BEC.

In the intermediate range between the linear and the almost horizontal part in Fig. 4 we have 1.1 K $\leq T_c \leq$ 2.02 K and the upper and lower limit allow a simple correlation between T_c and ρ. To explain the finer details of Fig. 4 a more refined model has to be created which among other effects has to include the exciton-exciton interaction, for which direct experimental evidence exists.$^{9)}$

IV. PHASE DIAGRAM FOR BEC

In order to obtain more quantitative information about $T_c(\rho)$ we have studied the following model:$^{10)}$ A Bose system whose particles interact only *via* two-body forces is subjected to the Bogoliubov treatment for weak interactions (the operators a_o and a_o^+ are replace by $\sqrt{N_o}$; only those interaction terms are retained in the Hamiltonian which are linear in N_o). Furthermore it is assumed that the interaction can be described by specifying a scattering length, a. Using the identity

$$< N_o > = N - \sum_k{}' < a_k^+ a_k > \quad , \tag{4}$$

where the sum represents the thermally excited bosons, the average number $< N_o >$ of particles in the condensate can be calculated as a function of T. The critical temperature is specified by the condition $< N_o > = 0$, whose solution is obtained numerically as a universal relation between t_c and ρa^3. The dimensionless temperature t is defined in Fig. 5. The region below the full curve is the two phase region for BEC (condensate and thermally excited bosons), above this curve only thermally excited bosons (normal gas) exist. In the low density region the theory is expected to be of reasonable accuracy. At high densities the results are only qualitatively correct. Also shown in Fig. 5 are experimentally determined T_c for excitons from two different AgBr samples. A scattering length $a = 2a_{ex}$ has been chosen, *i.e.* excitons are treated as hard spheres with a radius a_{ex} ($a_{ex} \simeq 50$Å exciton Bohr radius). One experimental value (●) is fitted to the theoretical curve. However, the corresponding density agrees reasonably well with various other estimates.$^{3)}$ The points (▲) are the values determined

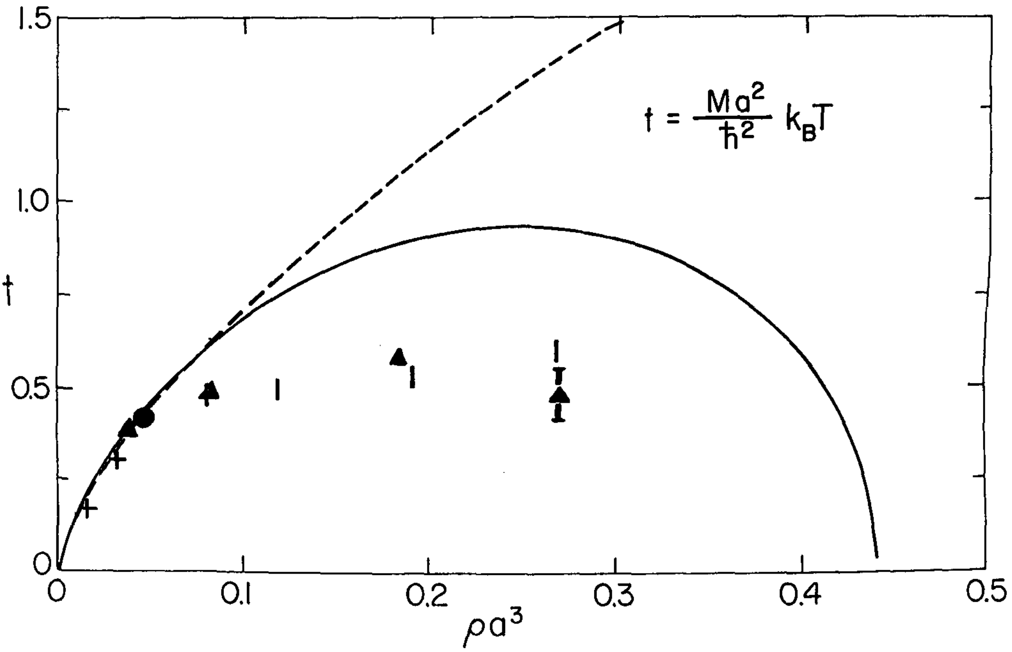


Fig.5. Temperature *vs.* density (normalized units). Dashed curve: Ideal Bose gas. Solid line: Weakly interacting Bose gas. Experiments: See text.

from the measurements shown in Fig. 3. For a second sample the points (+) are determined as described in the section above, the bars (I) have been obtained by extrapolation (compare Fig. 3). The agreement is reasonable considering the as yet unsatisfactory experimental determination of ρ and T_c and considering the simplifications of the theoretical model.

V. CONCLUSIONS

The model calculation and the analysis of our experiments suggest the following more general statements:

1) Excitons form an interacting Bose gas which implies the existence of a maximum critical temperature (Fig. 5),

$$T_{c\ max} \simeq \frac{\hbar^2}{Mka^2} \qquad (\sim 7\ \text{K for AgBr}) \qquad (5)$$

At temperatures above $T_{c\ max}$ BEC cannot exist at all. Therefore thermalization of the excitons is important which requires long exciton lifetimes.

2) As a further consequence of the interaction there exists a maximum density above which no BEC will occur even at $T = 0$. Therefore, in order to observe BEC of excitons, the excitation should not be too high, independent of heating effects.

Since these considerations are general they will apply to the case of BEC of biexcitons as well. The following statements, however, are more related to BEC of excitons in indirect materials:

3) Due to the finite phonon width no sharp thresholds due to the onset of BEC will be observed in the emission and absorption spectra.

4) Our analysis demonstrates the importance of experimental information in addition to bare luminescence spectra, as for instance their dependences on temperature and excitation density.

5) The importance of some material parameters has been discussed in ref. 3). Another important property is the exciton-exciton interaction. An estimate11) of the influence of the electron-phonon interaction in the rather strongly polar AgBr seems to indicate that the attractive interaction between excitons is considerably reduced.12) However, a more detailed investigation is necessary.

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REFERENCES

1) P. Junod, CIBA-GEIGY Photochemie, Fribourg, Switzerland; J. Malinovski, Institute of Photoprocesses, Bulgarian Academy of Sciences, Sofia, Bulgaria.

2) W. Czaja and C. F. Schwerdtfeger: Solid State Comm. **15** (1974) 87.

3) D. Baeriswyl and W. Czaja: RCA Review **36** (1975) 5.

4) H. Kanzaki, S. Sakuragi, S. Hoshino, G. Shirane and Y. Fujii: Solid

State Comm. 15 (1974) 1547;

W. von der Osten and B. Dorner: Solid State Comm. 16 (1975) 43.

5) W. von der Osten and J. Weber: Solid State Comm. 14 (1974) 1133.

6) M. Matsushita: J. Phys. Soc. Japan 35 (1973) 1688.

7) G. M. Mason: Phys. Rev. B11 (1975) 5094;

J. Tejeda, N. J. Shevchik, W. Braun, A. Goldmann and M. Cardona: Phys. Rev. B (1975) to be published.

8) W. Czaja and C. F. Schwerdtfeger: *Proc. XII Int. Conf. Physics of Semicond., Stuttgart*, 1974, ed. M. H. Pilkuhn (Teubner, Stuttgart, 1974) p.142.

9) W. Czaja: *Proc. XII Congrès Européen de Spectroscopie Moléculaire, Strasbourg, France* 1975, ed. M. Grossmann, S. G. Elkomoss, and J. Ringeissen (Elsevier, Amsterdam 1976) p.155.

10) W. Czaja and D. Baeriswyl: *Proc. 14th Int. Conf. Low Temp. Physics, Helsinki* 1975 ed. M. Krusius and M. Vuorio, Vol.4 (North Holland Pub. Co., Amsterdam, 1976) p.429.

11) E. Tosatti: Phys. Rev. Letters 33 (1974) 1092.

12) Personal communication E. Tosatti: 1975. Although the mechanisms considered in ref. 11) are basically correct, the influence of the repulsive part has been overestimated.