

# Bose-Einstein Condensation of Excitons

Interest has been revived in high density excitons in semiconductors by some recent experiments that seem to indicate the existence of a new phase where excitons, or biexcitons undergo Bose-Einstein condensation.

If light of photon energy larger than the band gap is sent onto a semiconductor, free electron-hole pairs are formed. At low enough temperatures the electron-hole Coulomb attraction leads to a bound pair called exciton similar to a hydrogen atom. Excitement about high density excitons started in 1968, when V.M. Asnin and A.A. Rogachev<sup>1)</sup> from the Ioffe Physico-Technical Instit., Leningrad, showed that when germanium was hit by laser light of increasing intensity — thus creating excitons in the sample — a change of state of the exciton gas was taking place at some critical exciton density. This was very suggestive of a Mott-type insulator-metal transition of the exciton system which is expected when the exciton spacing is comparable with the exciton diameter. The idea that the lowest energy state of the exciton system was in fact "metallic" droplets was first suggested by L.V. Keldysh<sup>2)</sup> from the Lebedev Physical Institute in Moscow. Subsequent luminescence and light scattering experiments were soon carried out by Ya. Pokrovskii<sup>3)</sup> from the Institute of Radioengineering and Electronics of the Academy of Sciences in Moscow, and brought out that in the high density phase the excitons had dissolved into free electrons and holes, indeed forming "metallic" droplets, with typical radii of some microns, in thermal equilibrium with the exciton gas. Very good agreement was obtained by subsequent detailed work by Nozières and Combescot<sup>4)</sup> of Ecole Normale, Paris, and W.F. Brinkman and T.M. Rice<sup>5)</sup> of Bell Labs. Though this was quite satisfactory, it has also been clear that the free exciton gas and the electron-hole plasma droplet are not the only possible states for a collection of excitons. In the first place, excitons bear an extraordinary resemblance with hydrogen atoms, and can bind in pairs to form molecules, or "biexcitons", analogous to H<sub>2</sub>. Predicted very long ago, biexcitons have been observed in luminescence spectra of many crystals. The energetic competition between a biexciton gas and the droplet state has also been investigated by W.F. Brinkman and T.M. Rice<sup>6)</sup>

and the conclusion was that the details of the band structure play a crucial role.

Another possibility comes from Bose-Einstein statistics, which both excitons and biexcitons follow approximately, being made up with even numbers of fermions. The phenomenon of Bose-Einstein condensation (BEC), which occurs at a temperature  $T_c = h^2/12 k m (N/V)^{2/3}$  for N noninteracting perfect bosons of mass m in a volume V consists in a macroscopic population of particles in the lowest energy state. BEC might occur also for excitons (and biexcitons), as has been pointed out more than 10 years ago by Moskalenko<sup>6)</sup> from the Institute of Physics and Mathematics in Kishinev, Blatt and coworkers from New York University and Casella from IBM Watson Research Center, Yorktown Heights. Other systems, in which BEC is in discussion, are superfluids such as <sup>4</sup>He below the  $\lambda$ -point and Cooper pairs in a superconductor. The significance of experiments on exciton BEC lies in the new approach to this fundamental problem. One of the obstacles in the way of a possible condensation is the Pauli principle, which, by requiring antisymmetry of the total wave function to fermion exchange, prevents macroscopic occupancy of the exciton states, and thus removes that feature of Bose statistics which is crucial to BEC. It is, however, clear that this problem is not necessarily a serious one, e.g., as long as the exciton spacing is large enough to allow for the neglect of the exciton (or biexciton) size in comparison. The low temperature state in this case is only in principle distinct, but thermodynamically indistinguishable, from a Bose-Einstein condensate.

A second obstacle is interaction between the excitons, or biexcitons. Even in the large distance limit specified before, the net effect of the Pauli principle is equivalent to a repulsive exciton-exciton force. In addition to this, theorists like P.W. Anderson, S.T. Chui, and W.F. Brinkman<sup>7)</sup> of Bell Labs envisage at least another force, due to screening of the electron-ion potential, which seems to cancel the Pauli repulsion and leads to a net attractive exciton-exciton force. At the opposite limit of short distances, excitons, of course, attract each other, in the attempt to bind and form a biexciton. This overall attraction is

- 8) GULLY, W.J., OSHEROFF, D.D., LAWSON, D.T., RICHARDSON, R.C., and LEE, D.M., *Phys. Rev. A* **8** (1973) 1633.
- 9) OSHEROFF, D.D., GULLY, W.J., RICHARDSON, R.C., and LEE, D.M., *Phys. Rev. Lett.* **29** (1972) 920.
- 10) PAULSON, D.N., KOJIMA, H., and WHEATLEY, J.C., *Phys. Rev. Lett.* **32** (1974) 1098.
- 11) LEGGETT, A.J., *Phys. Rev. Lett.* **31** (1973) 352, and *Ann. Phys. (N.Y.)* **85** (1974) 11.
- 12) OSHEROFF, D.D., and BRINKMAN, W.F., *Phys. Rev. Lett.* **32** (1974) 584.
- 13) BOZLER, H.M., BERNIER, M.E., GULLY, W.J., RICHARDSON, R.C., and LEE, D.M., *Phys. Rev. Lett.* **32** (1974) 875.
- 14) WEBB, R.A., KLEINBERG, R.L., and WHEATLEY, J.C., *Phys. Rev. Lett.* **33** (1974) 145.
- 15) AHONEN, A.I., HAIKALA, M.T., KRUSIUS, M., and LOUNASMAA, O.V., (1974, to be published).
- 16) PAULSON, D.N., KOJIMA, H., and WHEATLEY, J.C., *Phys. Lett.* **47 A** (1974) 457.
- 17) WEBB, R.A., GREYTAK, T.J., JOHNSON, R.T., and WHEATLEY, J.C., *Phys. Rev. Lett.* **30** (1973) 210.
- 18) KOJIMA, H., PAULSON, D.N., and WHEATLEY, J.C., *Phys. Rev. Lett.* **32** (1974) 141.
- 19) YANOF, A.W., and REPPY, J.D., *Phys. Rev. Lett.* **33** (1974) 631.
- 20) ALVESALO, T.A., ANUFRIYEV, Yu. D., COLLAN, H.K., LOUNASMAA, O.V., and WENNERSTRÖM, P., *Phys. Rev. Lett.* **30** (1973) 962.
- 21) ALVESALO, T.A., COLLAN, H.K., LOPONEN, M.T., and VEURO, M.C., *Phys. Rev. Lett.* **32** (1974) 981, and *J. Low Temp. Phys.* (1975, to be published).
- 22) ANDERSON, P.W., and MOREL, P., *Phys. Rev.* **123** (1961) 1911.
- 23) BALIAN, R., and WERTHAMER, N.R., *Phys. Rev.* **131** (1963) 1553.
- 24) ANDERSON, P.W., and BRINKMAN, W.F., *Phys. Rev. Lett.* **30** (1973) 1108.
- 25) MERMIN, N.D., and AMBEGAOKAR, V., *Nobel Symposium 24* (Academic Press, London), p. 97.
- 26) SHUMEIKO, V.S., *Zh. Eksp. Teor. Fiz.* **63** (1972) 621; translation: *Soviet. Phys. JETP* **36** (1973) 330.
- 27) PAULSON, D.N., JOHNSON, R.T., and WHEATLEY, J.C., *Phys. Rev. Lett.* **30** (1973) 829.
- 28) LAWSON, D.T., GULLY, W.J., GOLDSTEIN, S., RICHARDSON, R.C., and LEE, D.M., *J. Low Temp. Phys.* **15** (1974) 169.
- 29) WÖLFLE, P., *Phys. Rev. Lett.* **30** (1973) 1169.

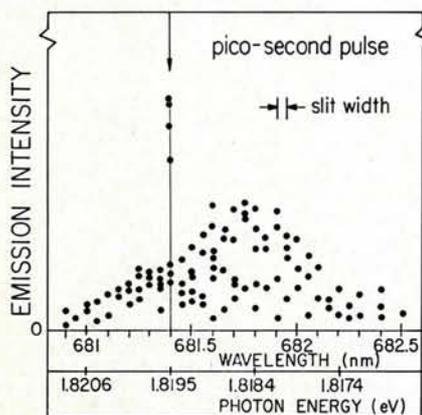


Fig. 1. Luminescence spectrum of a CdSe crystal at 1.8 K under the pico-second light pulse excitation (slit width: 0.05 nm); from ref. 9.

generally believed to make BEC very unlikely, and is held responsible for the failure of observing it so far. If one has biexcitons however, this force is probably reduced, and at short distance is missing altogether, because fermions are already paired. It is thus somewhat more natural to expect biexcitons to condense, at least in some restricted density range. Many of the properties to be expected from such a condensate have been investigated a few years ago by E. Hanamura of Tokyo's Institute for Solid State Physics<sup>8)</sup>.

First reports of what could be an observation of biexciton BEC came in spring 1973 also from this Institute, by S. Shionoya<sup>9)</sup> and his group. By sending ultrashort light pulses as intense as 50 MW/cm<sup>2</sup> onto a crystal of CdSe, they could observe radical changes in the luminescence spectra. Above some critical light intensity a new sharp line appears that has been associated with biexcitons condensed in the  $K = 0$  state as shown in Fig. 1. The problem with these experiments is that biexcitons in CdSe decay as fast as  $10^{-10}$  sec after having formed. This makes a high density of biexcitons, and the possible BEC, a transient phenomenon in time, a very difficult system to deal with if other experiments have to be made to test Shionoya's conclusions. CuCl crystals, which suffer less from this problem because biexcitons have a longer lifetime, are now being tried in Shionoya's group, and the reported preliminary observations seem to be once again in favour of biexciton BEC.

A more consistent step seems to be the very recent observation by W. Czaja and C.F. Schwerdtfeger of RCA Labs, Zürich<sup>10)</sup> who report a new sharp emission line for not too strongly excited AgBr.

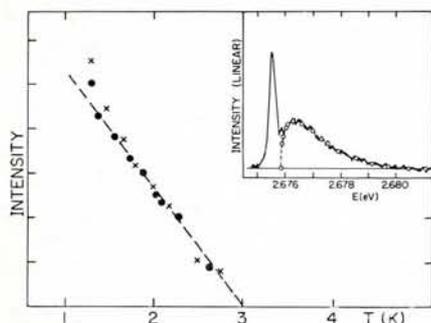


Fig. 2. AgBr: Luminescence intensity as a function of temperature for the sharp emission line (see insert) occurring at the low energy side of the free exciton  $TA_1$ -assisted emission.

As shown in Fig. 2, the peak appears at about 3 K, and grows rapidly on cooling. The temperature, at which the peak appears, increases with light

intensity but the magnitude of the peak is reported first to increase and then to decrease for increasing light intensity. These circumstances, along with the absence of ordinary biexciton luminescence, led the RCA group to conclude that single exciton BEC is taking place in their samples. If  $T_c$  is taken to be 3 K and the indirect exciton mass for AgBr as about one free electron mass one arrives at a critical exciton concentration of  $3 \times 10^{16}$  cm<sup>-3</sup>. On his side, Hanamura wonders whether biexcitons do not play any role, as the biexciton binding energy should not be too low in AgBr (about 10 K). Shionoya, on the other hand, has strong doubts whether any condensation can be observed with intensities as low as RCA's — about 200 W continuous wave. Nothing definitive can be said until the actual densities can be arrived at unambiguously.

Theoretically, one can at least argue that polar semiconductors — of which AgBr is an example — ought to be the best bet for single exciton BEC for not too high densities. This is so because in this case strong exciton-lattice coupling leads to lattice-induced exciton-exciton forces, whose net sum amounts, for large spacings, to a repulsion which has the right

magnitude to give, jointly with the Pauli repulsion, a cancellation of the otherwise attractive force. In that regime, excitons would be essentially free, in which case BEC follows necessarily. This could, however, only occur as a transient phenomenon, since the biexciton binding is anyway not suppressed by these lattice forces.

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#### REFERENCES

- 1) ASNIN, V.M., and ROGACHEV, A.A., *Sov. Phys. JETP Letters* **7** (1968) 360; ROGACHEV, A.A., *Proc. IX. Internat. Conf. Semiconductors, Moscow 1968*, p. 407
- 2) KELDYSH, L., *Proc. IX. Internat. Conf. Semiconductors, Moscow 1968*, p. 1307; "Excitons in Semiconductors", *Izd. Nauka, Moscow 1971* e.g. review paper by POKROVSKII, Ya., *Phys. Stat. Sol. (A)* **11** (1972) 385
- 3) COMBESCOT, M., and NOZIERES, P., *J. Phys. C* **5**, (1973) 2369
- 4) BRINKMAN, W.F., and RICE, T.M., *Phys. Rev. B* **7**, (1973) 1508
- 5) MOSKALENKO, S.A., *Sov. Phys. Solid State* **4** (1962) 199; BLATT, J.M., BOER, K.W., and BRANDT, W., *Phys. Rev.* **126** (1962) 1691; CASSELLA, R.C., *J. Phys. & Chem. Solids* **24** (1963) 19
- 6) ANDERSON, P.W., CHUI, S.T., and BRINKMAN, W.F., *J. Phys. C* **5** (1972) L119
- 7) HANAMURA, E., *Proc. XI. Internat. Conf. Semiconductors, Warsaw 1972* p. 711; *Solid State Commun.* **11** (1972) 485
- 8) KURODA, H., SHIONOYA, S., SAITO, H., and HANAMURA, E., *Solid State Commun.* **12** (1973) 533; *J. Phys. Soc. Japan* **35** (1973) 534
- 9) CZAJA, W., and SCHWERDTFEGER, C.F., *Solid State Commun.* **15** (1974) 87, and *Proc. XII. Internat. Conf. on Semiconductor Phys., Stuttgart 1974*, in press.

On August 30, 1974, we learned of the unexpected and sudden death of

### Josef-Maria Jauch

Professor at the Department of Theoretical Physics, the University of Geneva, Switzerland.

The very same day the book

## Physical Reality and Mathematical Description

edited by Charles P. Enz and Jagdish Mehra, was published as a tribute to Josef-Maria Jauch on the occasion of his sixtieth birthday, September 20, 1974.

Through his scientific work Jauch has justly earned an honored name in the community of theoretical physicists. Through his teaching and a long line of distinguished collaborators he has put an imprint on modern mathematical physics.

A number of Jauch's scientific collaborators, friends and admirers have contributed to this collection of 34 essays, which reflect to some extent Jauch's own wide interests in the vast domain of theoretical physics. The book includes a personal introduction by the editors and a list of principal publications of J. M. Jauch. (xxiii + 552 pp., Dfl. 125,- / US \$ 39.50).



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