Bose-Einstein Condensation of Excitons

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This note discusses the question as to whether quasi-particles, such as excitons, i.e., nonlocalized excited states of solids, can fulfill necessary conditions for a Bose-Einstein condensation, and whether such condensation can be observed. Although uncertainties of present data on excitons preclude precise numerical predictions, it is concluded that under certain experimentally attainable circumstances excitons fulfill the necessary conditions, i.e., condensation is possible. Ways of detecting the condensation are considered, and a specific experiment is proposed.

SINCE the Bose-Einstein condensation has been important in elucidating the anomalies of liquid helium, this concept has been successful recently also in the interpretation of superconductivity in terms of a condensation of electron pairs.[1–3] If the condensation of such composite statistical entities is a valid concept, all such quasi-particles of integral spin can exhibit Bose-Einstein condensation, provided they fulfill at least the following two necessary conditions: (I) the chemical potential of these particles is a valid thermodynamic variable, and (II) their density can be increased beyond a critical value n_{cr}, given by the condition that the mean interparticle distance is comparable to the de Broglie wavelength, $\lambda\!\!\!^{-}$, of these particles, of kinetic energy $\sim kT$ and effective mass m_{eff}.

$$n_{\mathrm{cr}}^{-\frac{1}{3}}\sim \lambda\!\!\!^{-}_{\mathrm{cr}}\simeq\hbar(m_{\mathrm{eff}}kT_{\mathrm{cr}})^{-\frac{1}{2}}. \quad (1)$$

It is important to find new systems which can give further and direct evidence of a Bose-Einstein condensation of quasi-particles.

Excitons have been invoked for the interpretation of certain optical properties of solids.[4–5] They are nonlocalized excited states which can be regarded as hydrogenlike states of electron-hole pairs bound by their mutual effective Coulomb interaction in the crystal.[6] This suggests that excitons can behave as quasi-particles which obey Bose-Einstein statistics. They fulfill condition (I). This assumes that they cannot associate into polyexcitons, which is unlikely to occur for excitons of effective mass comparable to or less than the electron mass. Condition (II), Eq. (1), defines a critical exciton density which must be produced by the absorption of light quanta. This circumstance sets excitons apart from the other above-mentioned systems. There is no reason to expect, however, that this will affect basically the statistical behavior of the excitons.

For given incident flux of light quanta $I_0(\omega)$ of frequency ω, the maximum average density of excitons with extinction coefficient $\kappa(\omega)$ is produced in a crystal of thickness $\sim\kappa^{-1}$. In such a crystal, the mean exciton density is given by

$$n_{\mathrm{exc}}\sim q\tau\kappa I_0, \quad (2)$$

where q is the quantum yield for exciton formation and τ the exciton life time. By Eqs. (1) and (2), condensation can occur below a critical temperature T_{cr} given by

$$T_{\mathrm{cr}}\simeq(\hbar^2/m_{\mathrm{exc}}k)(q\tau\kappa I_0)^{\frac{2}{3}}. \quad (3)$$

Many experiments have been reported which permit some inferences about the constants in Eq. (3). Rewriting Eq. (3) in the form

$$\tau=CT_{\mathrm{cr}}^{\frac{3}{2}}, \quad (4)$$

and choosing $q\simeq1$, $\kappa\sim3\times10^5$ cm^{-1}, as has been reported for many substances, and a light intensity, accessible by conventional methods, within the wavelength range of the exciton absorption $I_0\sim10^{17}$ cm^{-2} sec^{-1} we estimate $C\lesssim10^{-6}$. The upper limit corresponds to the (for our discussion) unfavorably high value $m_{\mathrm{exc}}\simeq m_e$, where m_e is the electron rest mass; in many substances, m_{exc} is considerably smaller than m_e. Hence, condition (II) is fulfilled at liquid helium temperatures, for example, for lifetimes as have been measured via energy transport phenomena.[7] More recently, some arguments have been advanced against the interpretation of the energy transport phenomena in photoconductors in terms of exciton migration. Theoretical estimates lead to shorter life-

[1] M. R. Schafroth, Phys. Rev. **96**, 1442 (1954); *Solid-State Physics*, edited by F. Seitz and D. Turnbull (Academic Press Inc., New York, 1960), Vol. 10, p. 295 ff.
[2] J. M. Blatt, Progr. Theoret. Phys. (Kyoto) **23**, 447 (1960); **24**, 851 (1960); Phys. Rev. Letters **7**, 82 (1961).
[3] L. Onsager, Phys. Rev. Letters **7**, 50 (1961).
[4] I. Frenkel, Phys. Rev. **37**, 17, 1276 (1931); J. Exptl. Theoret. Phys. U.S.S.R. **6**, 647 (1936).
[5] H. Haken, Fortschr. Physik **6**, 271 (1958).
[6] G. H. Wannier, Phys. Rev. **52**, 91 (1937).
[7] Cf., e.g., G. Diemer, G. J. Van Grup, and W. Hoogenstaaten, *Advances in Semiconductor Science* (Pergamon Press, New York, 1958), p. 182; M. Balkanski, J. Phys. Chem. Solids **6**, 401 (1958); **8**, 179, 193 (1959); J. O. Kessler and A. R. Moore, *Proceedings of the International Conference on Semiconductor Physics, Prague, 1960* (Czechoslovakian Academy of Sciences, Prague, 1961), p. 804.

times than those quoted above.[8] Apparently, the question of lifetimes is too unsettled at present for dependable estimates of the condensation temperature. Still, in view of the development of new monochromatic light sources, it is to be expected that condition (II) can be fulfilled for a number of substances already at liquid helium temperature.

Given that conditions (I) and (II) are fulfilled, the question remains how one can establish experimentally whether, in fact, condensation occurs.

Since excitons are electrically neutral, we cannot expect to detect such condensation by spectacular electrical effects. However, properties of excitons which normally are influenced by the lattice dynamics should change abruptly on condensation. For example, the absorption or emission lines of excitons, which are normally broadened predominantly by inelastic scattering and by fluctuations in the local field, should approach the natural linewidth in the condensed state. Energy transport phenomena can be affected by the decoupling of excitons from the lattice dynamics on condensation. Ortho-excitons (of spin 1) should show a change in their paramagnetic resonance on condensation owing to the accompanying change in the spin-lattice coupling. A more exciting manifestation of the Bose-Einstein condensation of ortho-excitons would be their spontaneous magnetization ("ferromagnetism") which will occur when the Weiss field due to the condensed excitons exceeds the internal field due to paramagnetic impurities.

Under realistic assumptions, the last effect is unlikely to occur, and the next to last barely within the limits of present experimental detectability. The effect on the linewidth, however, seems best suited for experimental verification. For example, at a given sufficiently low temperature, the exciton line will be affected by the light intensity as follows: As the light intensity exceeds a critical value, the intensity of the normal line is reduced while the narrow line of the condensed state builds up correspondingly.

The outcome of such an experiment will shed new light on the problem of the statistics of quasi-particles and on the dynamical properties of excitons. Experiments along these lines are in progress.

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[8] J. W. Allen, *Proceedings of the International Conference on Semiconductor Physics, Prague, 1960* (Czechoslovakian Academy of Sciences, Prague, 1961), p. 435.