COMITATO NAZIONALE PER L'ENERGIA NUCLEARE
Laboratori Nazionali di Frascati

LNF-74/14(R)
1 Marzo 1974

G. Baldacchini: RADIATION EMITTED DURING EXCITON CONDENSATION.

Servizio Documentazione
dei Laboratori Nazionali di Frascati del CNEN
Casella Postale 70 - Frascati (Roma)

Laboratori Nazionali di Frascati del CNEN
Servizio Documentazione

LNF-74/14 (R)
1 Marzo 1974

G. Baldacchini: RADIATION EMITTED DURING EXCITON CONDENSATION. -

INTRODUCTION. -

In these last times a lot of interest was aroused by the discovery of new properties of excitons at high density in semiconductors[1].

It was foreseen long ago that excitons could originate molecular complexes[2] (biexcitons, etc.) and eventually undergo a Bose-Einstein condensation[3,4]. The former forecast was confirmed experimentally in a number of substances such as Si[5], Cu_2O[6], HgI_2[7], CdS and CdSe[8] etc. For the Bose condensation there is only one not completely convincing experimental evidence in CdSe[9] and the problem is still open.

A few years ago it was argued[10] that because of the strong analogy between the exciton and the Hydrogen atom, something similar to the Mott transition[11] could exist in a gas of free excitons (Wannier or extended type) at a given value of the density. Indeed because of the big radius of the excitons the observation of the Mott transition is much easier in a system of free excitons than in a gas of Hydrogen, for which a pressure of the order of $3 \cdot 10^6$ atmospheres[12] is needed. Without entering into details, for which I refer to a review article[13], I will be satisfied to say that such speculations brought to one of the major offspring of scientific results on the physics of semiconductors in the last years. Briefly it seems that at given values of density and temperature the gas of excitons condenses in drops[14], a few micron in radius, built-up of an electron-hole (e-h) plasma. Conversely some authors believe that the new effects are due to the formation of biexcitons[15] and eventually, at very high density, to drops of a normal biexciton liquid[16].

(+) - W. Czaya and C. F. Schwerdtfeger, at the RCA Labs in Zurich, observed what they believe to be a Bose-Einstein condensation of excitons in AgBr. Preprint to be published in Solid State Commun.

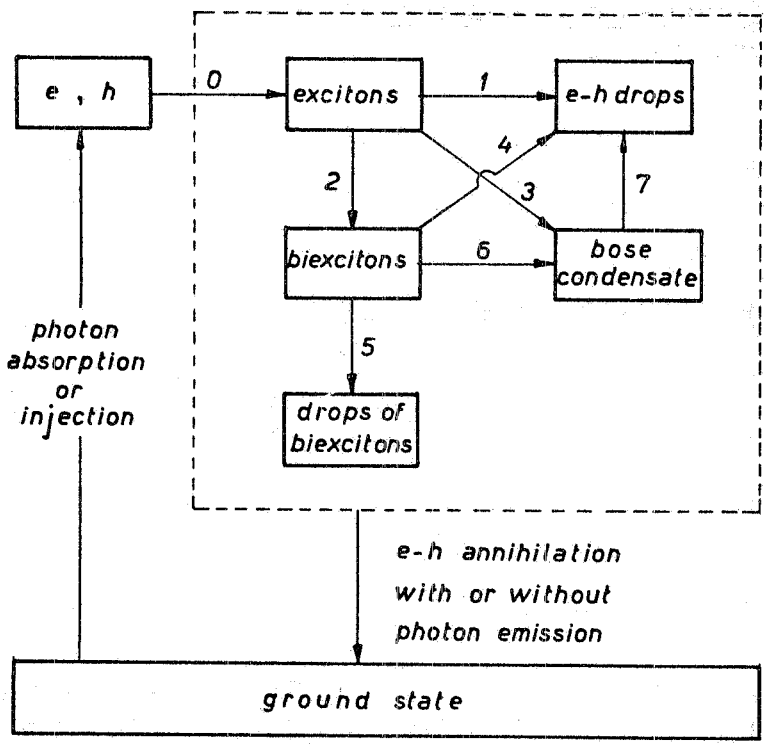

FIG. 1 - Block diagram of possible evolutions in time of a gas of excitons at high density in semiconductors.

In Fig. 1 the various processes described above and others are represented schematically. Electron-hole pairs are created by photon absorption or by electrical injection through a junction[17]. They couple each other to form excitons and hence the various possibilities are clearly shown. It is worth-while to remind that such processes are possible only in times shorter than or comparables with the decaying time of the e-h annihilation. Eventually the electrons and holes recombine with the emission of photons or through non radiative processes. In Ge and Si (semiconductors with an indirect gap) the most studied materials until now, it is believed that the excitons go through 2 and 4 processes[1, 13]. In CdSe (semiconductor with a direct gap) the correct processes seem to be 2, 6[9] and 7[18] in order. And so on.

The way commonly used to distinguish a process from another was until now simply the study of the effects of the stationary states reached by the system. For instance the luminescence and its variations as a function of the pumping intensity, the magnetic field and the mechanical stress give the possibility to ascribe these effects to a Bose condensate, to a Biexciton gas or liquid or to an e-h drop. Unfortunately some times theoretical or phenomenological models for different states can be fitted equally well by experimental results. Even more the experimental results are often in contradiction each other[14, 19]. For all these reasons a clear recognition of the state in question is not always possible.

It is obvious that no doubts at all would remain if it would be possible to observe the fenomena relative to the transition from one state to another state. Indeed from the identification of the particular transition i(i = 0, 1, . . . , 7), Fig. 1, the initial and final states would be identified too. In the following I make a proposal of this kind.

PHASE TRANSITIONS AND PHOTON EMISSION. -

The passage of the exciton system from one state to another, see Fig. 1, can be interpreted as a phase transition[20, 21]. For instance processes like 1, 4 and 5 are interpreted as first order phase transition in the same way as vapor condenses into liquid. Process 3 represents clearly a second order phase transition analogous to the normal-superfluid transition of liquid Helium below T_λ .

It was suggested recently to measure the electromagnetic radiation emitted during a phase transition[22]. Indeed such processes are generally accompanied by motion of charges which are accelerated during short periods of time. Hence the spectral distribution of the radiation emitted contains in principle all the information relative to the transition on a microscopic scale. It is clear at this point that, if such radiation could be revealed, we would have on hand a magnificent tool to look directly at the transition itself.

A few years ago some calculations were made on the possibility to reveal the electromagnetic radiation emitted by free excitons during their abrupt deceleration by collision with the lattice[23]. A serious problem was to find out the right channel for energy dissipation. Ultimately photons or phonons emission? In the case just mentioned the conclusion was that, in spite of the production of phonons, electromagnetic radiation was emitted at a significant rate. Obviously this result cannot be generalized, but each case has to be considered separately because of the complexity of the phenomena involved.

Keeping well in mind the possibility of phonon generation it is worthwhile to estimate the main features of the electromagnetic radiation emitted by the processes shown in Fig. 1. Even if quantistic concept will be used through, the analysis of these phenomena is made essentially in a classical way.

As it is well known[24], the electric dipole moment, e. d. m. , of the exciton is given by :

$$P_{exc} = \int \psi_e^*(r)\, e\, r\, \psi_h(r)\, dr \qquad (1)$$

where ψ_e and ψ_h are the wavefunctions of the electrons and the holes, s-like and p-like in most semiconductors. Generally P_{exc} has a non ze

ro value ranging around 10^{-16} esu[25]. In any case even if $P_{exc} = 0$, it is always possible to create an e. d. m. by applying an external electric field smaller than its impact ionization value[26].

Now if we look at the processes of Fig. 1 we see that, each time the system undergoes a transformation i(i = 0, 1, . . . , 7), a very important role is played by the e. d. m. In 0 electrons and holes, which do not have an e. d. m., capture each other to form excitons with $P_{exc} \neq 0$. In 1 the exciton is destroyed and one has $P_{in} = P_{exc}$, $P_{fin} = 0$ ("in" stands for initial state, and "fin" for final state). In 2, $P_{in} = P_{ex}$, $P_{fin} = P_{biex}$. In 4, $P_{in} = P_{biex}$, $P_{fin} = 0$. In 7, $P_{in} = P_{exc}$ or P_{biex}, $P_{fin} = 0$. Only in transitions 3, 5 and 6 the e. d. m. does not change, because the excitons or biexcitons maintain their identity.

In these last three cases the exciton complexes are abruptly decelerated from $K_{in} \simeq (3\ m\ KT/\hbar^2)^{1/2}$ to $K_{fin} = 0$ in 2 and 6, and to $K_{fin} < K_{in}$ in 5; K is the modulus of the wave vector of excitons or biexcitons and m their mass. We know from classical electrodynamic[27] that an e. d. m. does not radiate when it is decelerated as a whole. More exactly it generates quasistatic fields whose energy in the whole solid angle goes as $1/R^2$. Hence the only way for the e. d. m. to radiate in through a trasformation of the kinetic energy in rotovibrational energy. In this case the energy is quantized(x) and the emission of radiation is possible at given frequencies. Owing to the possibility for the kinetic energy to be dissipated through phonon generation, as previous reminded, I would have not mentioned the above possibility but for the unique features of the Bose-Einstein condensation. Indeed it is reasonable that all the boson-like particles, or a fraction, condense in phase each other. This means that the intensity of the emitted radiation, if any, could be proportional to N^2, as in stimulated emission, and not to N as in the random case; N is the number of particle which undergo the condensation. In conclusion, except for case 5, in 2 and 3 it is reasonable to expect emission of electromagnetic radiation, even if it is difficult to calculate the power level.

In processes 0, 1, 2, 4 and 7 the e. d. m. changes by an amount ΔP_i during the time t_i^*, characteristic of the particular transition (in the following I shall omit the subscript i). The energy radiated in the whole process by one particle is given by[27]:

$$E(\text{erg}) = \frac{2}{3c^3}\int_{-\infty}^{+\infty}\left[\frac{d^2 P(t)}{d t^2}\right]^2 dv \qquad (2)$$

(x) - This is a very delicate point which deserves more attention. Indeed many good quantum quantum numbers of free systems loose their meaning in a solid and I have to remember that excitons or biexcitons are "excitations" in a solid.

In order to integrate the above expression it is necessary to give an analitical form to P(t), which has to be reasonable from the physical point of view. I choose for P(t) a Fermi-Dirac distribution-like function:

$$P(t) = \frac{P}{e^{\frac{t-t_o}{t^*}} + 1} + P_{fin} \tag{3}$$

pratically the e. d. m. is always constant except around $t \simeq t_o$ for times of the order of t^*, which is the transition time. Inserting Eq. (3) in (2) and after tedious calculation, one obtains for E the following simple expression:

$$E(erg) = \frac{1}{45c^3} \frac{(\Delta P)^2}{t^{*3}} \tag{4}$$

This energy is emitted in a wide spectral range with an intensity distribution given by:

$$E(\omega) = \frac{2\pi}{3c^3} \frac{\omega^4 t^{*2}}{\sinh^2(\pi\omega t^*)} (\Delta P)^2 \tag{5}$$

in Fig. 2 the function $f(\omega t^*) = (3c^3/2\pi)(t^*/\Delta P)^2 \cdot E(\omega)$ is displayed versus ωt^*. As it can immediately be seen, pratically all the radiation is emitted at frequencies $\omega \lesssim (t^*)^{-1}$, as expected, with a very pronounced maximum for $\omega \simeq 0.61/t^*$.

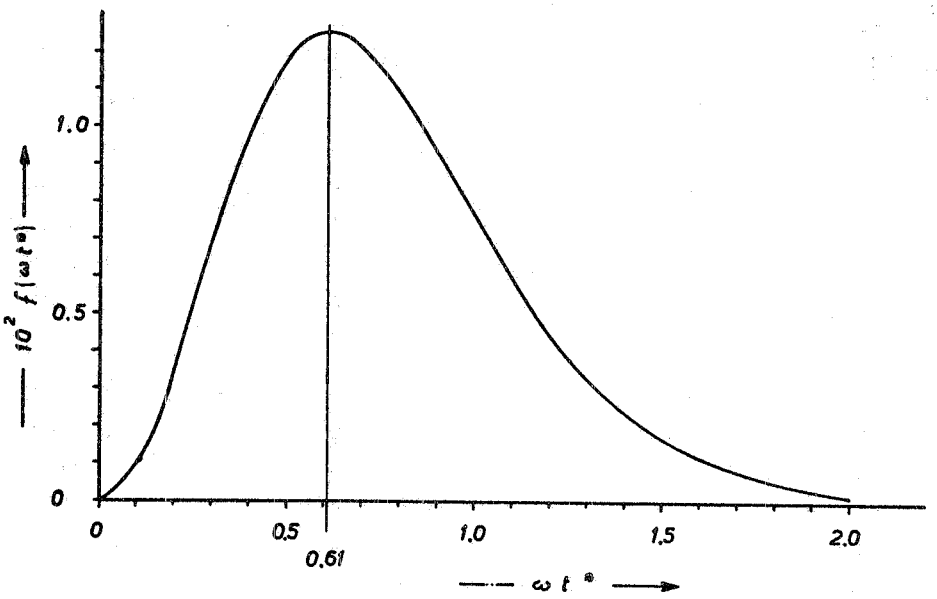

FIG. 2 - Frequency distribution of the electromagnetic radiation emitted by the disappearence of an electric dipole moment in a time t^*.

GENERAL REMARKS AND CONCLUSIONS. -

In the previous paragraphs I have analyzed in some detail the kinetic of an exciton gas at high density and I have outlined in a few cases the features of the electromagnetic radiation that could be emitted during the transitions from one state to another. By measuring the total energy emitted and its frequency distribution it is possible to go back to such parameters as the transition time, t^*, and the e.d.m. variation, ΔP, which are characteristic of some transitions. Hence it would be possible to recognize through the transition the initial and the final state.

The order of magnitude of the energy emitted during such transitions can be guessed by using experimental and theoretical data actually on hand. For instance in process 1, see Fig. 1, the e.d.m. goes to zero, $\Delta P = P_{in} = P_{exc}$, in a time $t^* \simeq d/v$. Here d[28] is the thickness of the surface layer of the drop where the exciton density varies from the gas density value, n, to the liquid density value, n_o, ($n_o > n$ at equilibrium) and v is the exciton velocity. For $d \simeq 50$ A and $v = 10^5$ cm/sec one obtains $t^* \simeq 10^{-11}$ sec. Using this value for t^* and $\Delta P \simeq 10^{-16}$ e.s.u.[25], eq. (4) gives for the energy emitted during the whole process $E \simeq 10^{-32}$ erg. With a very intense short pulse, $\sim 10^{-12}$ sec[9], it is possible to create $\sim 10^{18}$ excitons/cm^3 which condense in e-h drops in a time $(\Delta t) \simeq (n^{1/3} v)^{-1} \simeq 10^{-11}$ sec[29]. Hence the power delivered by the transition of the whole system is $p = (nE/\Delta t) \simeq \simeq 10^{-3}$ erg/sec. This energy is emitted in the microwave region of the electromagnetic spectrum and centered around $\nu_{max} \simeq 0.6/2\pi t^* \simeq \simeq 10^{10}$ sec^{-1} (i.e. $\sim$ 10 GHz)(x). The absorption of microwave by the sample and drops does not pose a serious problem at least on some materials[30, 31] and hence this radiation could be detected.

As shown above, the power emitted by the drop formation is $\sim 10^{-10}$ watts or less in most cases, a very low value not easy to be measured. However some progresses in microwave detection have been made in these last times using Josephson devices[32]. In particular with a Dayen bridge[33] some authors obtained at 10 GHz a NEP (Noise Equivalent Power) = 210^{-14} watt with one cycle of bandwithd!

At this point it is obvious that the detection and the analysis of the radiation emitted, if any, during a phase transition , pose technical problem very difficult to solve. But what a reward! The knowledge of the microscopic development in space and in time of a phase transition. In the specific case of the exciton condensation it would be possible to reach a clear understanding of all the complex phenomena involved.

(x) - For comparison the Power emitted by one cm^2 of Black Body surface at 1°K is about 6×10^{-5} erg/sec and centered at $\sim$ 50 GHz.

I am indebted with Prof. C. Pellegrini, whose advices in the early stage made this work possible, and with Dr. G. Paternò for some information. Thanks are due to Prof. G. Bassani for his usefull criticism on fondamental problems which I greatly appreciated.

REFERENCES. -

(1) - G. Baldacchini, Frascati Report LNF-73/52 (1973) (In italian).
(2) - M. A. Lampert, Phys. Rev. Letters 1, 450 (1958).
(3) - S. A. Moskalenko, Sov. Phys. Solid State 4, 199 (1962).
(4) - J. M. Blatt, K. W. Boerand and W. Brandt, Phys. Rev. 126, 1691 (1962).
(5) - J. R. Haynes, Phys. Rev. Letters 17, 860 (1966) (The new emission ascribed to the biexiton here was later explained by other authors(13) as coming from exciton droplets).
(6) - E. F. Gross and F. I. Kreingold, JETP Letters 12, 68 (1970) (The existence of the biexitons in CU_2O was later questioned by Y. Petroff et al. in Phys. Rev. Letters 29, 1558 (1972)).
(7) - I. Kh. Akopyan, B. V. Novikov, M. M. Pimonenko and B. S. Razbirin, JETP Letters 17, 299 (1973).
(8) - S. Shionoya, H. Saito, E. Hanamura and O. Akimoto, Solid State Commun. 12, 223 (1973).
(9) - H. Kuroda, S. Shionoya, H. Saito and E. Hanamura, Solid State Commun. 12, 533 (1973).
(10) - V. M. Asnin and A. A. Rogachev, JETP Letters 7, 360 (1968).
(11) - N. F. Mott, Phil. Mag. 6, 287 (1961).
(12) - G. F. Chapline Jr., Phys. Rev. B6, 2067 (1972).
(13) - Ya. Pokrovskii, Phys. Stat. Solid (a) 11, 385 (1972).
(14) - Ya. Pokrovskii and K. I. Svistunova, Sov. Phys. Semicond. 4, 409 (1970).
(15) - V. M. Asnin, Yu. N. Lomasov and A. A. Rogachev, JETP Letters 18, 144 (1973).
(16) - V. M. Asnin, A. A. Rogachev and N. I. Sablina, Sov. Phys. Solid State 14, 332 (1972).
(17) - V. Marrello, T. F. Lee, R. N. Silver, T. C. McGill and J. W. Mayer, Phys. Rev. Letters 31, 593 (1973).
(18) - J. Bille, T. Fischer, H. Liebing and W. Ruppel, Proc. 11 Internat. Conf. on the Phys. of Semicond. Warszawa-Poland (1972), pag. 183.
(19) - V. M. Asnin, A. A. Rogachev and N. I. Sablina, Sov. Phys. Semicond. 5, 1607 (1972).

(20) - L. V. Keldysh, Sov. Phys. Uspekhi 13, 292 (1970).
(21) - Z. A. Insepov and G. E. Norman, Sov. Phys. JETP 35, 1198 (1972).
(22) - V. N. Bogomolov, E. K. Kudinov and Yu. A. Firsov, Sov. Phys. Solid State 14, 1788 (1973).
(23) - S. G. Ryzhanov, Sov. Phys. J. 11, 125 (1968).
(24) - R. S. Knox, Theory of Excitons (Academic Press, 1963).
(25) - S. G. Ryzhanov, Sov. Phys. J. 10, 77 (1967).
(26) - T. Yao, K. Inagaki and S. Mackawa, Solid State Commun. 13, 533 (1973).
(27) - L. Landau and E. Lifshitz, The Classical Theory of Fields (Addison-Wesley, 1951), Chap. 9.
(28) - L. M. Sander, H. B. Shore and L. J. Sham, Phys. Rev. Letters 31, 533 (1973).
(29) - V. F. Elesin and Yu. V. Kopaev, Sov. Phys. JETP 36, 767 (1973).
(30) - B. M. Ashkinadze and V. V. Rozhdestvenskii, JETP Letters 15, 261 (1972).
(31) - J. C. Hensel and T. G. Phillips, Proc. 11 Internat. Conf. on the Phys. of Semicond., Warszawa-Poland (1972), pag. 671.
(32) - B. N. Taylor, J. Appl. Phys. 39, 2490 (1968).
(33) - M. T. Levinsen, Rev. Phys. Appl. 9, 135 (1974).