BIEXCITONS IN HgI_2 CRYSTALS

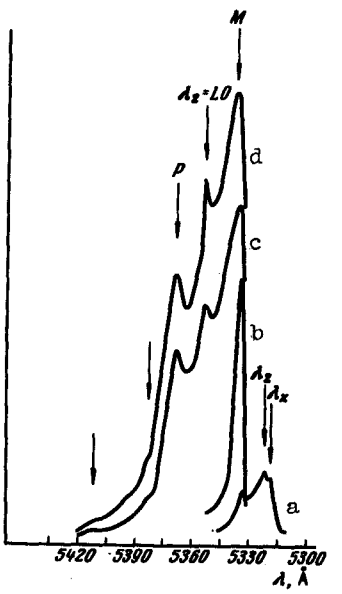
I.Kh. Akopyan, B.V. Novikov, M.M. Pimonenko, and B.S. Razbirin
Leningrad State University
Submitted 9 March 1973
ZhETF Pis. Red. 17, No. 7, 419 - 421 (20 April 1973)

The purpose of the study was to determine the properties of high-density excitons in HgI_2 crystals. Intensity excitation of the crystal revealed new emission bands. It is assumed that the most intense of these bands is due to biexcition annihilation.

HgI_2 is an exceedingly interesting object for the investigation of high-density excitons, for at 4.2°K the position of the exciton level n = 1 (λ = 5297 Å) in this crystal almost coincides with the wavelength of the second harmonic of the neodymium laser (λ = 5288 Å), so that it becomes possible to excite strongly the crystal at the frequency of the exciton transition. We have investigated the luminescence of HgI_2 crystals excited by such a laser. The pulse power range from 0.01 to 1.2 MW/cm^2; the pulse duration was approximately 50 nsec. According to approximate estimates, the maximum exciton concentration reached 10^{18} cm^{-3}. The luminescence spectra were photographed with an instrument having a dispersion 31 Å/mm. The samples, which were single-crystal plates with optical axes in the planes of the investigated surfaces, were cooled by direct immersion in liquid helium. Approximately 20 samples were studied.

At an excitation density less than 0.07 MW/cm^2, the luminescence spectrum of the HgI_2 crystal is similar in general outline to the spectrum obtained by excitation with a mercury lamp [1]. As a rule we have observed the two strongest emission lines of the bound excitons, λ_1 = 5317 Å and λ_2 = 5322 Å. At an excitation density W = 0.07 MW/cm^2 (see Fig. a), a new emission band M (λ = 5333 Å) of width 1 meV appears in the spectrum. With further increase of the excitation intensity, the band M becomes much stronger and predominates in the spectrum (Fig. b). The band is broadened and its maximum shifts towards the long-wave side of the spectrum. At a density W = 0.3 MW/cm^2, the maximum of the band is located at λ = 5337 Å, and the width amounts to 3 - 4 meV. At the same excitation there appear in the spectrum, in addition to the M band, also a new band P (λ = 5368 Å) 2 MeV wide and two weaker bands at 5383 Å and 5412 Å, of approximately the same width (Fig. c). A subsequent increase of the excitation to W = 1.2 MW/cm^2 causes further growth in the intensity of all the new bands. Starting with W = 0.1, all spectra contain the narrow line of the bound exciton λ^2 - LO (λ = 5345 Å).

Let us examine in greater detail the properties of the M band. The high intensity of the M band has made it possible to investigate the dependence of its intensity on that of the exciting light. We have found that up to W = 0.3 MW/cm^2 the M band is amplified superlinearly with increasing excitation, and then increases more slowly in the interval W = 0.3 - 1.2 MW/cm^2. A superlinear dependence of the luminescence intensity on the excitation is observed for interactions between excitons [2, 3]. The M band is shifted 15 meV towards the long-wave side of the exciton line n = 1 (λ = 5297 Å) [1]. It can



Luminescence spectra of HgI crystal at T = 4.2°K excited with a laser of wavelength 5288 Å with power (W) 0.07 (a), 0.2 (b), 0.3 (c), and 1.2 MW/cm^2 (d).

therefore be assumed that the M band is due not to Auger recombination of the free excitions, but more likely to radiative decay of the exciton molecule, as a result of which one exciton becomes annihilated and the second remains in the non-dissociated state. A similar process is observed, e.g., in the CuCl crystal [3]. In this model, the short-wave edge of the M band is separated from the free-exciton line by a distance equal to the binding energy E_i^M of the excitons in the molecule. The value of E_i^M determined in this case, 15 meV, is about half the exciton binding energy. Such a large value of E_i^M for a strongly anisotropic HgI_2 crystal agrees with the theoretical arguments of E.D. Gutlyanskii and V.E. Khartsiev, that the biexciton binding energy increases in crystals with anisotropy of the carrier effective masses. The high dissociation energy of the exciton molecule in HgI_2 is confirmed also by the weak temperature quenching of the M band, which was observed in the spectrum up to 50°K.

The M band has an asymmetrical Maxwellian-like contour with a sharp drop on the short-wave side of the spectrum. This is precisely the shape expected for the considered radiative biexcition decay, when the biexciton momentum is carried away by a non-dissociated exciton. The shift of the maximum of the M band towards the long-wave side can indicate an increase in the effective temperature of the biexcitons. This temperature estimated from the distance from the short-wave edge of the M band to its maximum, amounts to ∿30°K for strong excitations. At the same time, the lattice temperature does not exceed ∿10°K, as can be deduced from the absence of a displacement of the λ_2 - LO line [4].

The 5412 Å band is shifted by an amount E_i towards the long-wave side away from the M band and can correspond to Auger recombination of the biexciton, in which one exciton is annihilated radiatively and the other dissociates into a free electron and a hole. It is interesting to note that the weak band with λ = 5383 Å is located in that part of the spectrum where the next biexciton emission process can occur, namely annihilation of one exciton with simultaneous excitation of another one into the state n = 2.

The T band is separated from the M band by the energy of the LO phonon and can be the phonon replica of the M band. Another interpretation is also possible. The P band results from the interaction of excitons (Auger recombination) or from inelastic collision of the molecules [5].

The authors thank S.A. Moskalenko for a discussion of certain results.

[1] B.V. Novikov and M.M. Pimonenko, Fiz. Tekh. Poluprov. 4, 2077 (1970) [Sov. Phys.-Semicond. 4, 1785 (1971)].
[2] J.R. Hayns, Phys. Rev. Lett. 17, 860 (1966); C. Beniot a la Guillaume, J.M. Debever, and F. Salvan, Phys. Rev. 177, 567 (1969).
[3] A. Mysyrowicz, J.B. Grun, R. Levy, A. Bivas, and S. Nikitine, Phys. Rev. Lett. 26A, 615 (1968); H. Soama, T. Goto, T. Ohta, and M. Ueta, J. Phys. Soc. Japan 29, 697 (1970).
[4] B.V. Novikov and M.M. Pimonenko, Fiz. Tekh. Poluprov. 6, 771 (1972) [Sov. Phys.-Semicond. 6, 671 (1972)].
[5] H. Saito and S. Shionoya, Techn. Rep. ISSP A534 (1972).