Screening of the inverse hydrogen-like series in BiI_3 with two-photon carrier generation

V. F. Agekyan, N. N. Vasil'ev, V. P. Gladkikh, and R. I. Shekhmamet'ev
A. A. Zhdanov Scientific-Research Institute of Physics, Leningrad State University

(Submitted 23 January 1984)
Pis'ma Zh. Eksp. Teor. Fiz. **39**, No. 6, 252–255 (25 March 1984)

Nonequilibrium absorption spectra of bismuth iodide crystals in the region of the inverse hydrogen-like series (IHS) under strong optical excitation are investigated. Successive dissociation of lines in the IHS, beginning with the upper long-wavelength terms, is observed. The results are interpreted in terms of the screening of the IHS, thereby confirming the common nature of the lines.

The existence of bound Coulomb states near the singularities of the energy bands of a crystal is determined by both the type and parameters of the actual critical points and the magnitude of the external magnetic field.[1] In a favorable case a bound state of two particles with like charge can appear.[2]

At low temperatures a series of lines with an effective Rydberg constant of 0.2473 eV, converging on the long-wavelength side to the limit 1.9809 eV, is observed in the absorption by $BiI_3 \rightarrow BiI_3$. This inverse hydrogen-like series (IHS) is interpreted as the spectrum of a bielectron (bihole)—a system consisting of two electrons (holes) with a reduced negative mass and a positive total effective mass bound by Coulomb forces.[2,3] One of the particles can be localized on an impurity. The effect of external actions on the spectrum of the IHS agrees with its proposed origin.[3,4] Other models also exist,[4,6] but they cannot explain all the results obtained to date and, moreover, lines of the IHS and lines whose energies are close to lines of a different origin are not distinguished in Refs. 5 and 6. The rarity of the observation of the IHS in semiconductors, as compared with exciton effects, is explained by the strict requirements imposed on the energy spectrum of the crystal, the sign and magnitude of the effective masses, and the parameters of the impurity states. The recent observation of an IHS in a ZnP_2 crystal[7] showed that the BiI_3 spectrum is not unique in this regard, and it has increased the interest in this problem.

The BiI_3 specimen was submerged in liquid helium and excited by laser pulses with quantum energy $h\nu_L = 1.17$ eV. The strong absorption edge in BiI_3 begins at 2.0116 eV, so that the crystal is excited by a two-photon method. In this case the small coefficient of two-photon absorption k_2 assures that the specimen is excited uniformly over its depth and that surface effects are insignificant. The light source used to investigate absorption at the time of excitation was the wide band of radiation of the rhodamine-6G dye, which was pumped by the second harmonic of the same laser pulse. The spectrum was recorded using the pulsed synchronous detection scheme.

In the experiment we investigated the dependence of the nonequilibrium absorption spectrum of the IHS on the intensity J_0 of the exciting light. For $J_0 = 7.5$ MW·cm^{-2} the intensity of the $n = 6$ line decreases by a factor of 2–3, and the intensity of the $n = 5$ line also decreases (Fig. 1a). As J_0 is increased up to 15 MW·cm^{-2}, the $n = 6$ line essentially disappears; the intensities of the $n = 5$ and $n = 4$ lines decrease by a factor of 4 and 2, respectively; and, the continuous absorption is amplified, especially on the long-wavelength side of the IHS limit. In this case the crystal remains transparent in the region of IHS lines with large n.

The successive weakening and vanishing of lines and the appearance of continuous absorption are not related to thermal dissociation. The temperature coefficient of the displacement of lines in the IHS is equal to 1.6×10^{-4} eV·deg^{-1}, so that the displacement of lines with maximum excitation (Fig. 1b) correspond to heating of the crystal from 4 K by not more than 5 K. At this temperature the contours of IHS lines change little (Fig. 2). The absorption edge at 2.0116 eV also changes little when the specimen is heated up to 10 K, while continuous absorption below the IHS limit does not appear at least up to 30 K.

We assume that the successive weakening and vanishing of lines in the IHS in our experiment is a result of screening of bielectronic states in the presence of a high concentration of photocarriers. In experiments on screening of excitons, for example, in CdS and Cu_2O,[8,9] dissociation of lines is accompanied by narrowing of the forbidden band and amplification of the continuous absorption tail due to an increase of the energy of interaction of carriers. It is possible that the absorption appearing beyond

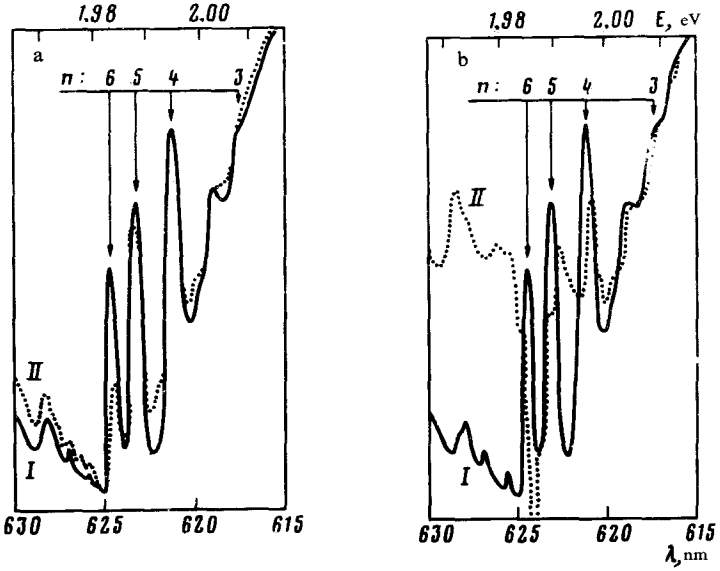

FIG. 1. The equilibrium (I) and nonequilibrium (II) absorption spectra of BiI_3 for different intensities of photoexcitation J_0 with quantum energy 1.17 eV. 1) $J_0 = 0$; IIa) $J_0 = 7.5$ MW·cm^{-2}; IIb) $J_0 = 15$ MW·cm^{-2}, $T = 4$ K, $n = 3$–6 are the lines in IHS.

the limit of the IHS with strong excitation of BiI_3 has an analogous origin for bielectrons.

Thus the behavior of IHS in BiI_3 under the conditions of photoexcitation indicates the common serial origin of the lines.

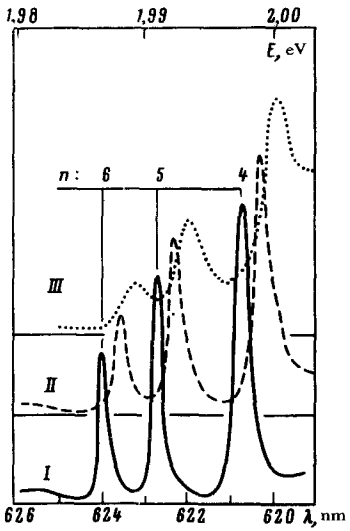

FIG. 2. Temperature dependence of the absorption spectrum of BiI_3. $T = 9$ K (I), 15 K (II), 28 K (III). The region of the IHS with lines $n = 4$, 5, and 6 is shown.

The realization of screening conditions permits estimating the lifetime τ_0 of the nonequilibrium carriers under the assumption that τ_0 is much less than the duration of the photoexcition pulse $\tau_L = 7\times10^{-9}$ sec. It may be assumed that

$$\tau_0 = \frac{2h\nu_L}{k_2 J_0^2}(1 + k_2 J_0 l)n_0 , \tag{1}$$

where n_0 is the carrier concentration, with pumping J_0, that screens the state with quantum number n and binding energy E_n, and l is the thickness of the specimen 0.015 cm. The binding energy E_n and the concentration n_0 are related by Mott's relation

$$E_n = \frac{1}{2}\frac{e^2}{\epsilon_0}\kappa(n_0), \tag{2}$$

where ϵ_0 is the static dielectric constant, and $\kappa(n_0)$ is the inverse screening radius. According to Ref. 10, the screeniung of a discrete state at low temperature is characterized by a sharp decrease of the intensity of the corresponding line in a narrow range of concentrations of free carriers, so that a two-fold decrease of the intensity may be viewed as corresponding to condition (2). In Fig. 1a it is evident that the $n = 6$ line is screened with $J_0 \approx 7.5$ MW·cm^{-2}. Using criterion (2), the known value of E_6 and the dependence of κ on n_0 in the Debye approximation, we obtain from (1) $\tau_0 = 5\times10^{-12}$ s to within a factor of several units, after first measuring $k_2 = 2$ cm·MW^{-1}. Calculations for conditions corresponding to screening of the lines $n = 5$ and $n = 4$ give close values of τ_0. If we take into account that under strong excitation the electronic temperature can exceed the lattice temperature, then τ_0 would be slightly higher.

The authors thank A. V. Malov for assistance in the experiment.

[1]É. I. Rashba and V. M. Édel'shtein, Pis'ma Zh. Eksp. Teor. Fiz. **9**, 475 (1969) [JETP Lett. **9**, 287 (1969)].
[2]E. F. Gross, V. I. Perel', and R. I. Shekhmamet'ev Pis'ma Zh. Eksp. Teor. Fiz. **13**, 320 (1971) [JETP Lett. **13**, 229 (1971)].
[3]E. F. Gross, N. V. Starostin, M. P. Shepilov, and R. I. Shekhmamet'ev, Izv. Akad. Nauk. SSSR, Ser. Fiz. **37**, 885 (1973).
[4]N. O. Krylova, R. I. Shekhmamet'ev, and M. Yu. Gurgenbekov, Opt. Spektrosk. **38**, 947 (1975).
[5]M. P. Lisitsa and F. V. Motsnyĭ, Kvant, Elektron. (Kiev) No. 17, 1979, p. 26.
[6]T. Karasawa, T. Komatsu, and J. Kaifu, Solid State Commun. **44**, 323 (1982).
[7]A.V. Sel'kin, I. G. Stamov, N. N. Syrbu, and A. G. Umanets, Pis'ma Zh. Eksp. Teor. Fiz. **35**, 51 (1982) [JETP Lett. **35**, 57 (1982)].
[8]V. G. Lysenko and V. I. Revenko, Fiz. Tverd. Tela **20**, 2144 (1978) [Sov. Phys. Solid State **20**, 1238 (1978)].
[9]V. F. Agekyan, N. N. Vasil'ev, and Yu. A. Stepanov, Pis'ma Zh. Eksp. Teor. Fiz. **33**, 16 (1981) [JETP Lett. **33**, 14 (1981)].
[10]R. Zimmerman, K. Kilimann, W. D. Kraft, D. Kremp, and G. Ropke, Phys. Status Solidi B **90**, 175 (1978).

Translated by M. E. Alferieff
Edited by S. J. Amoretty