Fundamental luminescence of ionic crystals with high ionization levels

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A new form of intrinsic luminescence of ionic crystals, which is drastically different from all exciton luminescences and predominates for short excitation pulses, high temperatures and ionization levels, was observed. The intraband luminescence has been observed for the first time.

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The luminescence was excited through use of a nanosecond electron accelerator with adjustable beam parmeters: 1–25 nsec, 1–2000 A/cm^2, 0.3–0.5 MeV.[1–3] We noticed a luminescence whose properties are slightly dependent on the impurities. We have tentatively given it the name " plasma luminescence (PL)," whose properties are presented below.

1. The *luminescence spectra* of KI are shown in Fig. 1(a). At low temperatures the luminescence of self-captured excitons predominates—singlet (SEL) and triplet (TEL). At 77 K the maximum of the SEL band lies at 4.13 eV, and the decay time is equal to 1.4 nsec. The values for TEL are 3.31 eV and 6 μsec, respectively. The intensity of the exciton luminescence decreases rapidly with temperature and the PL remains the same [Fig. 1(b)]. It has a very broad spectrum which extends from the IR to the UV, region where it is cut off by the intrinsic absorption of the crystal [Fig. 1(a)]. A comparison of the luminescence and absorption spectra of the KI, KBr, RbCl, KCl, and NaCl crystals during the irradiation pulse shows that the observed spectrum structure is the result of a superposition on the PL of the exciton-luminescence (including hot luminescence) band and the light absorption by the color centers that are formed during irradiation. The NaCl and RbCl crystals were found to be convenient for observing the PL spectra. The formation of color centers in them is inefficient. An RbCl crystal is generally devoid of SEL[5,6] and NaCl has a low quenching temperature (42 K), in contrast to KI (82 K).[5] At 600 K the SEL is quenched in KI by a factor of 10^2, and in NaCl by more than a factor of 10^3. Therefore, the PL spectrum in NaCl and RbCl can be observed in almost pure form (Fig. 2). This spectrum is almost without a structure. A slight increase in the number of emitted photons per unit energy ($N_{\hbar\omega}$) is observed in the region 1.5–6.5 eV. The two small dips are the result of absorption of the PL by holes and F centers, respectively.

2. The *temperature independence* of plasma luminescence sets it apart from all forms of emission of localized electrons in the optical band. For example, the SEL intensity in KI decreases by a factor of $\sim 10^2$ in the range 77–500 K, but the PL is temperature independent up to the melting point of the crystal [Fig. 1(b)]. A shift of the short-wave edge of the PL spectrum directly mirrors the known[4] behavior of the

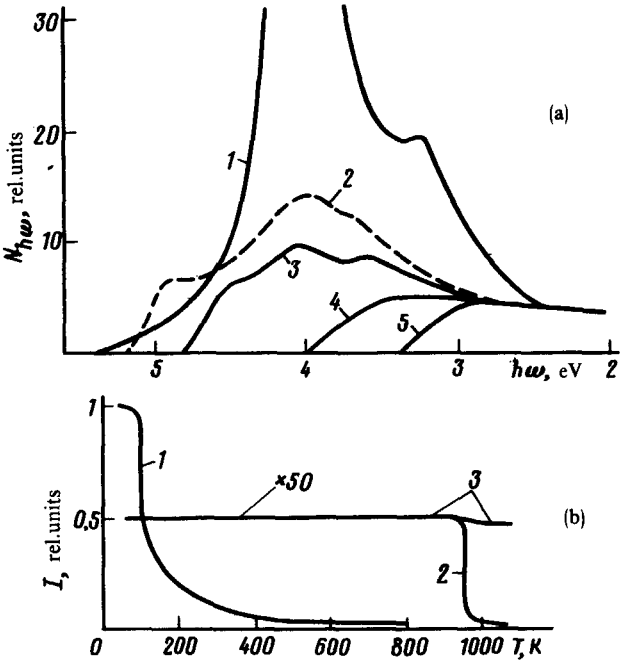

FIG. 1. (a) Luminescence spectra of KI for pulsed irradiation with dense electron beams: 1, Single crystal at 77 K; 2, at 200 K; 3, at 600 K; 4, at 900 K; 5, melt at 970 K; (b) temperature dependence of self-captured exciton luminescence intensity (1) and plasma luminescence intensity in the 3.5-eV region (2) and in the 2.5-eV region (3); below 959 K—KI single crystal, above 959 K—melt.

intrinsic absorption edge. Up to the melting point it shifts continuously toward longer wavelengths, and at the melting point it suddenly jumps by 0.5–0.7 eV [Fig. 1(a)]. As a result of melting, the PL intensity decreases by almost a factor of 10^2 in the short-wave region of the spectrum and remains essentially unchanged in the long-wave region [Fig. 1(b)].

3. The *luminescence relaxation time* was measured with a time resolution of 0.5 nsec by using a 31 ELU-F photomultiplier. The measured relaxation time of 1.4 nsec for SEL in KI at 77 K agrees well with the known data.[5] However, the PL relaxation time turned out to be less than the time resolution, i.e., $\tau_{PL} < 0.5$ nsec.

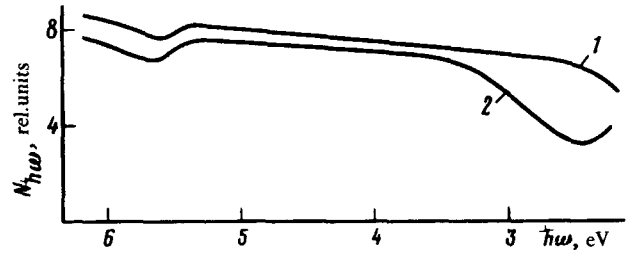

FIG. 2. Plasma luminescence spectrum of RbCl (1) and NaCl (2) produced by nanosecond pulses of electron irradiation at 600 K.

4. *PL yield* was measured by using two methods. The first method used as the standard the SEL of a KI crystal at 77 K with a known yield.[5] The second method used as the standard a TRSh-2850-3000 ribbon-filament incandescent lamp with a known equilibrium emission temperature. Both gave approximately the same results: the absolute energy yield of the plasma luminescence is equal to $(0.5\text{–}0.7)\times 10^{-3}$ of the absorbed beam energy.

5. *Model.* In terms of the spectrum and certain other properties, the plasma luminescence is similar to Cerenkov radiation (CR). However, the yield of the latter is much less. In our case it is equal to $(1\text{–}3)\times 10^{-5}$. A direct experiment showed that the CR contribution is insignificant. The PL was excited by 0.11-MeV electrons producing no CR, since their velocity was less than the phase velocity of light in the crystal. According to all the data, the plasma luminescence is a radiation of nonlocalized (quasi-free and band) electrons and holes. Their energy-relaxation rate ($-dE/dt$) can be estimated, from their yield and the PL spectrum. The average PL photon energy is 4–5 eV. An electron radiates such energy in 10^{-9}–10^{-8} sec (the characteristic time of optical dipole transitions). Since the PL yield is $\sim 10^{-3}$, the electron loses this energy primarily without radiation in 10^{-12}–10^{-11} sec. Hence, $-dE/dt = 10^{12}$–10^{13} eV/sec. At high ionization levels[1)] there is only one part of the energy spectrum of free electrons and holes in which their energy relaxation rate has this order of magnitude. It is the "electron-hole passive band (EH-band),"[2,3] that consists of two regions. The first region, electronic, is in the conduction band; the second region, hole, is in the valence band. The width of each region is smaller or equal to the gap between these bands (E_g). The high-energy electrons and holes produce EH-pairs, excitons, plasmons, etc. beyond the limits of the EH-band. In this case $-dE/dt = 10^{15}$–10^{16} eV/sec. As soon as they enter the EH band, they lose their ability to excite electrons and give up their energy to the lattice, causing the production of phonons. In this case $-dE/dt = 10^{12}$–10^{13} eV/sec, i.e., a factor of 10^3 lower than that outside the limits of the EH band. Its population density exceeds that of other regions of the free electron spectrum by this factor. The data obtained make it possible to assume that the plasma luminescence is an intraband luminescence (IBL) of the EH band, i.e., this luminescence arises during direct and indirect radiative transitions between the subbands within the conduction band or within the valence band. In the UV it becomes band-to-band luminescence. The model of PL as intraband luminescence of the EH band provides a simple explanation of the experimental data. In fact, according to this model the spectrum width is approximately equal to E_g; the relaxation time is 10^{-11}–10^{-12} sec, the energy yield is $\sim 10^{-3}$; the luminescence intensity depends weakly on the temperature up to the melting point, since $-dE/dt$ of the electrons in the EH band is determined by a temperature-independent, spontaneous phonon emission. We observed no PL threshold in the range of beam densities of 1 to 2000 A/cm^2, consistent with the model.

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[1)]The ionization level (E_I) is equal to the maximum average energy of an electron-hole pair. It is considered to be high if $E_I > 2E_g$. In our case E_I almost coincides with the energy of the beam electrons.

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