Quasilocal states in indium-activated alkali-halide crystals

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Two-step spectroscopy is used to observe absorption from the lower excited state of In$^+$ ions in alkali-halide crystals. This absorption is attributed to the presence of quasilocal states of the activator in the conduction band of the crystal. The positions of the quasilocal levels relative to the ground state are estimated.

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It was suggested in Refs. 1–3 that photoionization from a relaxed excited state (RES) of mercury-like ions (MI) in alkali-halide crystals (AHC), which is observed with intense excitation ($I \geqslant 1$ MW/cm^2) in the A band,[4,5] is related to the high-energy states of MI in the conduction band of the crystal (quasilocal states).[6] From this point of view, it is interesting to study experimentally the spectral dependence of photoionization of MI from RES.

In this work, using two-step spectroscopy, we observed and investigated absorption from the excited sate of In$^+$ ions in AHC. The experiments were performed using the setup described in Refs. 4 and 5. We used an excimer XeCl laser (quantum energy $E = 4$ eV, pulse energy $P = 2 \times 10^{-2}$ J, pulse duration $\tau = 1.5 \times 10^{-8}$ s) as the excitation source. The absorption from the excited state was probed with a collimated light beam from a mercury arc lamp.

We observed absorption for indium-activated KCl, KBr, and KI crystals, whose damping duration coincides with the spontaneous decay time of the lower excited state of the activated ions.

It is well known that different impurity and characteristic defects,[4,5] which having wide absorption bands, are formed under intense excitation in the A band of MI in a AHC. For this reason, the absorption observed by us can be attributed not only to absorption from the excited state but also to the change in concentration of defects due to thermal and photoionization, as well as to tunneling recombination.

In order to clarify the nature of the absorption observed, we investigated the dependence of the damping time on temperature ($T = 100$–600 K), excitation intensity (2–30 MW/cm^2) and activator concentration ($C = 10^{17} - 10^{19}$ cm^{-3}). The temperature studies showed that the change in the damping time of the absorption coincides exactly with the temperature changes in the lifetimes of the excited states of the activator.[7] Figure 1 shows the damping curves for absorption and luminescence of In$^+$ ions in KBr and KCl at $T = 100$ K and $T = 293$ K.

It was also established that the duration of damping does not depend on the intensity of excitation and the activator concentration.

Analysis of the experimental data leads to the following conclusions. 1) The observed absorption cannot be attributed to thermal ionization of defects, since the prob-
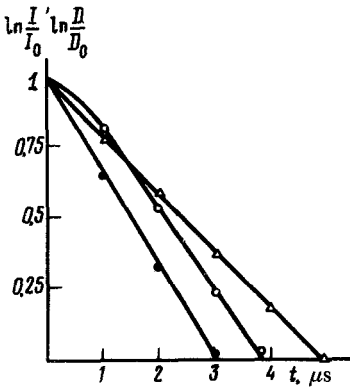
FIG. 1. Luminescence (solid-straight lines) and absorption damping times. $\Delta - KCl - In(T = 100 - 293$ K); $O - KBr - In(T = 100$ K); ● $- KBr - In(T = 293$ K).

ability of thermal ionization depends exponentially on temperature. 2) Since the absorption time does not depend on the excitation intensity, photoionization of electrons in color centers (In^0 type) by luminescence photons can be ignored. In this case, the absorption damping time would depend on the excitation intensity. 3) It was shown in Ref. 8 that the tunneling probability depends exponentially on the distance between the activator ions. Tunneling processes cannot be a reason for the observed absorption, since the absorption damping times do not depend on the activator concentration. 4) The experiments performed permit interpreting the observed absorption as absorption from RES of In$^+$ ions.

Using the spectral lines of a mercury arc lamp, we investigated the spectra of absorption from RES, which are presented in Fig. 2. It is evident from Fig. 2 that at $T = 100$ K, the absorption spectra have distinct maxima.

The fact that in the spectral region where maxima are observed in absorption from RES the cross sections for photoionization from RES are close to the cross sections of the allowed transitions[2,3] supports the idea of resonance photoionization,[1-3] i.e., optical transition from RES occurs into a quasilocal activator level. It is possible
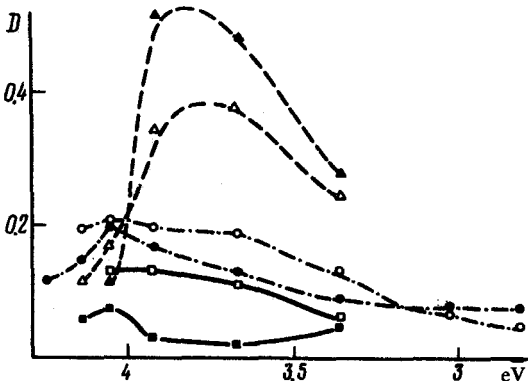

FIG. 2. Spectra of absorption from excited state of In$^+$ ions in AHC ($\Delta - KBr$, $O - KI$, $\square - KCl$; $T = 293$ K), (▲ $- KBr$, ● $- KI$, ■ $- KCl$; $T = 100$ K).

TABLE I.

Crystal	S-levels (eV)	D-levels (eV)	quasilocal levels (eV)
KCl – In	7,3	7,5	7,8
KBr – In	7,1	7,4	7,4
KI – In	6,5	7,0	7,3

that the spectrum of absorption from RES is affected by peaks in the density of states of the conduction band of AHC, but the RES absorption spectrum and its temperature dependence do not correspond to the spectrum of interband transitions in pure AHC.

Transitions to the upper S and D levels of the In^+ ion should have the highest probability for transitions from the $^3P_1$ level. Using the data in Refs. 9 and 10, we estimated the energy of transitions in AHC from the ground state of the In^+ ion to the S and D levels and to the observed quasilocal levels from maxima in absorption from RES, taking into account the Stokes shift (see Table I).

Since the position of the S and D levels is estimated approximately using the linear approximation,[9] the observed maxima in RES absorption spectrum can be attributed to S as well as D states of the free In^+ ions.

In conclusion, we note that the cross sections of phototransitions between RES and quasilocal states of In^+ ions, estimated according to the optical absorption density, agree well with the cross sections measured according to the change in the relative quantum yield of intracenter luminescence.[2,3]

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