Formation of high-energy states of mercury-like ions in alkali halide crystals during electron-hole recombination

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Transitions from highly excited $^1P_1$ states of impurity centers have been observed in the recombination-luminescence spectra of mercury-like ions in alkali halide crystals. These transitions indicate that a cascade mechanism of carrier capture operates in activated alkali halide crystals.

The most important practical application of alkali halide crystals activated with mercury-like ions (Tl^+, In^+, etc.) has been and remains their use as indicators of ionizing radiations, i.e., as scintillators. The final stage in the scintillation process is a recombination of carriers at an emission center, which puts the center in an excited state; then comes a radiative transition of the center to its ground state. As a rule, transitions from low-lying states of the center, $^3P_1$ (*A*-luminescence), are manifested in the spectra of the recombination luminescence of mercury-like ions.[1,2] There has been essentially no study of the role played by highly excited states of impurity centers in recombination processes in alkali halide crystals. In the one study which has been carried out, for a KI–Tl crystal, it has been shown that a low-lying $^3P_1$ excited state of the emission center forms during the recombination of an electron and a Tl^{2+} hole center.[1] In the present study we have observed luminescence bands in the recombination-luminescence spectra of certain activated alkali halide crystals (KI–Tl, KBr–Tl, and KCl–Sn). As was shown in Refs. 3–5, transitions from high-lying $^1P_1$ states of impurity centers are responsible for these luminescence bands (*C*-luminescence). We studied the recombination luminescence of the KI-Tl crystal in most detail.

In the experiments we used only optical excitation of the crystals. We used the UV light at the fourth harmonic from a Nd:YAG laser (30–40 MW/cm^2) for a stepwise photoionization of the activator[6] and to create hole (Tl^{2+}) and electronic (Tl^0) defects and *F* centers. The track of the color centers was subsequently bleached with an intense (50–100-MW/cm^2) single pulse ($\sim$30 ps) at the second harmonic from a Nd:YAG laser (or its stimulated-Raman shift in ethanol), and the flash of photostimulated luminescence of the activation was detected through a monochromator by a photomultiplier. The output signal from the photomultiplier was fed to an oscilloscope. The experiments were carried out at $T = 5$ K. The most likely mechanism for the excitation of the activator luminescence under these experimental conditions can be assumed to be the recombination of an electron which has appeared in the conduction band as a result of the photoionization of electronic centers and *F* centers with a hole center, Tl^{2+}. Figure 1 shows the recombination-luminescence spectra of a KI–Tl crystal after three successive photostimulation events involving the second harmonic from a Nd:YAG laser. The amplitude of the luminescence falls off systematical-

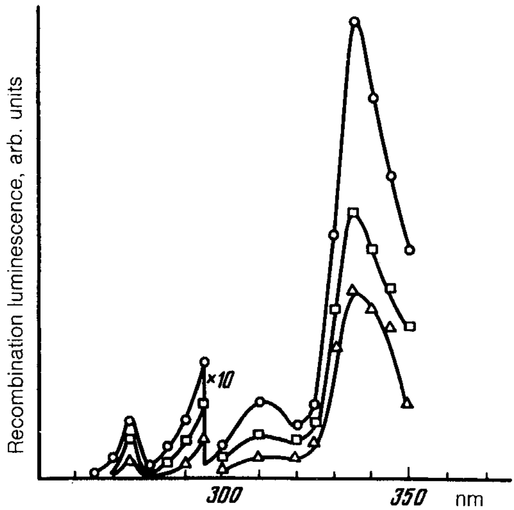

FIG. 1. Spectrum of the recombination luminescence of a KI–Tl crystal. ○—First photostimulation; □—second photostimulation; △—third photostimulation.

ly, as can be seen from the figure; this decrease is evidence that the luminescence is of a recombination nature. During the fourth, fifth, and subsequent photostimulations we observed no luminescence. The crystal was then exposed to an equal number of pulses (from three to ten, depending on the particular crystal and other experimental conditions) of the fourth harmonic from a Nd:YAG laser for the next cycle of measurements. The most important result of the experiments was the observation in the recombination-luminescence spectrum of the KI–Tl crystal of a luminescence band at 275 nm (Fig. 1), which is unambiguously assigned to a transition from a $^1P_1$ state (*C*-luminescence).[4] We can also clearly see a dip at 280 nm, which corresponds to the *A* absorption band in the KI–Tl crystal. The occurrence of *C*-luminescence in the recombination-luminescence spectra of alkali halide scintillators is evidence that a highly excited $^1P_1$ state of the mercury-like ion forms during the recombination of a band electron with a hole center; i.e., a cascade mechanism for carrier capture through the real levels of the impurity center operates in activated alkali halide crystals. Furthermore, since the existence of quasilocal states of mercury-like ions in alkali halide crystals can be regarded as essentially proved,[7] we do not rule out the possibility that the excited state of the activator may be formed even in an early stage of the recombination, while the electron is still in the conduction band.

We note in conclusion that the results of this study do not contradict the data obtained in Ref. 1, since (first) the experiments of Ref. 1 were carried out at liquid-nitrogen, rather than liquid-helium, temperatures, and (second) the excitation sources used there were not as powerful as pulsed lasers.

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