Dramatic intensification of diffusion of interstitial particles in KCl crystals at high densities of electrons and holes

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A dramatic intensification of the diffusion of interstitial particles has been observed for the first time. This intensification results from the charge exchange of these particles at the high densities of electrons and holes which arise in the track of a heavy charged particle or during pulsed bombardment of a crystal by an electron beam.

Ionizing radiation produces interstitial particles of two types in an ionic crystal of the KCl type: atoms ($\mathrm{Cl}_{\mathrm{in}}^{0}$) and ions ($\mathrm{Cl}_{\mathrm{in}}^{-}$). These particles diffuse by a classical jumping mechanism with activation energies $E_a = 0.07$–0.08 eV for $\mathrm{Cl}_{\mathrm{in}}^{0}$ atoms and $E_i = 0.03$–0.05 eV for $\mathrm{Cl}_{\mathrm{in}}^{-}$ ions (Refs. 1–3). The interstitial atoms $\mathrm{Cl}_{\mathrm{in}}^{0}$ arise as parts of defect pairs: $\mathrm{Cl}_{\mathrm{in}}^{0} + F$-center. All types of ionizing radiation produce pairs of this sort in KCl, with a huge yield (5–10 pairs per 100 eV), because each electron-hole pair or exciton is capable of producing a Frenkel defect pair in a defect-free lattice (see the review by Lushchik *et al.*[4]). When a crystal is bombarded by a low-intensity electron beam or x-ray beam, the density of free electrons and holes is low, and an interstitial particle diffuses in a neutral charge state ($\mathrm{Cl}_{\mathrm{in}}^{0}$). At high densities of free electrons, the probability for a charge exchange is significant, and as an interstitial atom diffuses it captures an electron, becoming an ion: $\mathrm{Cl}_{\mathrm{in}}^{0} + e^{-} = \mathrm{Cl}_{\mathrm{in}}^{-}$. The classical diffusion coefficient is increased by factor $\exp[(E_a - E_i)/kT]$, or 10^1–10^5, depending on the temperature.

This phenomenon has been observed for the first time in the present experiments. The diffusion rate of the interstitial particles is determined from the yield of the high-temperature dissociation of F_2-centers.[1] The dissociation of an F_2-center, i.e., the division of this center into two well-separated, immobile F-centers, requires the displacement of a chlorine ion (or atom) from an anionic site and its transfer to one of two anionic sites occupied by an F_2-center. Accordingly, the relative yield of the dissociation of F_2-centers is proportional to the velocity at which the interstitial anions move. We know that at temperatures above 250 K, anionic vacancies (V_a^+) are mobile in KCl, and ionizing radiation causes not only a buildup of F-centers but also their coalescence in F_2-centers: $F+p^+=V_a^+$, $V_a^+ + F = F_2^+$, $F_2^+ + e^- = F_2$, where p^+ is a mobile hole (a band hole or a self-localized hole).[2] At temperatures below 250 K, the anionic vacancies are relatively immobile, and the coalescence yield falls off by a factor of 10^2–10^3. The low-temperature dissociation of F_2-centers is therefore irreversible.

In the experiments, KCl crystal wafers are bombarded at a temperature in the interval 290–360 K until the desired concentration of F_2-centers is attained. The wafers are then held at the same temperature until all of the F_2^+-centers have decayed. The sample is then cooled to a certain temperature below 250 K, and the bombardment is resumed. During this second bombardment, we observe the destruction of F_2-centers in its pure form—undistorted by a buildup. The concentration of these centers can thus be lowered by a factor of 10^2 (Ref. 5).

Figure 1 shows the decay of the relative number of F_2-centers (n/n_0) with increasing dose (D) during proton bombardment of KCl. Since the ordinate axis has a logarithmic scale, the plot consists of two exponential functions. The first, "short," component reaches saturation. This component is associated with the reversible low-temperature destruction of F_2-centers: $F_2 + p^+ = F_2^+$, $F_2^+ + e^- = F_2$. The second component does not reach saturation. It is associated with an irreversible dissociation of F_2-centers. We compared the temperature dependence of the relative yield of the dissociation of F_2-centers, $g(T)/g(250\text{ K})$, for three types of bombardment of the KCl crystals: 1) low-intensity beams of electrons and x rays; 2) protons with an energy of 1–10 MeV; 3) intense electron beams with parameters 0.3 MeV, 5 nsec, and $1-10^3$ A/cm^2 from a high-current accelerator. We immediately see that in all cases the increase in the sample temperature caused by the beam is $\Delta T < 10$ K. The results are

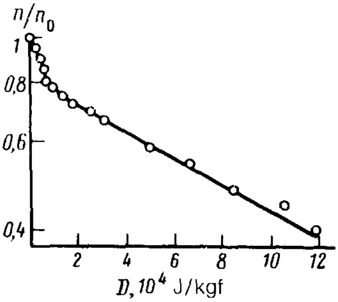

FIG. 1.

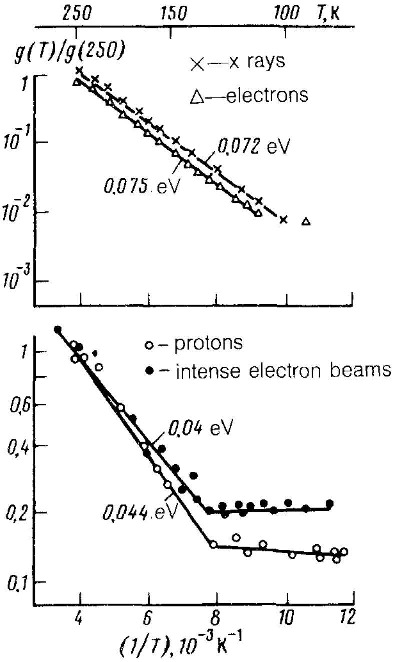

FIG. 2.

shown in Fig. 2. In the first case, the activation energy of 0.072–0.075 eV corresponds to the diffusion of interstitial Cl^0_{in}; in the second and third cases, the activation energy of 0.04–0.044 eV corresponds to the diffusion of interstitial ions Cl^-_{in}. The dissociation of F_2-centers by low-intensity x-ray and electron beams was originally studied in Refs. 2 and 3. The mechanism for the low-temperature dissociation at low densities of electrons and holes was identified: $e^- + p^+ = F + Cl^0_{in}$, $Cl^0_{in} + F_2 = f$. Those studies revealed the pure result: the dissociation of an F_2-center into two F-centers. The results of the experiments reported by us here show that the mechanisms for the dissociation of F_2-centers are different in tracks of protons and during pulsed bombardment by an intense electron beam, where there is a high density of electrons and holes: $e^- + P^+ = F + Cl^0_{in}$, $e^- + Cl^0_{in} = Cl^-_{in}$, $p^+ + F_2 = F_2^+$, $Cl^-_{in} + F_2^+ = F$. In other words, after an interstitial atom captures an electron, it converts into an ion; in this charged state it traverses the greater part of the distance from its point of creation to an F_2-center. Consequently, at high densities of electrons and holes the diffusion rate of interstitial species and, correspondingly, the observed relative yield of the dissociation of F_2-centers increase by a factor of 10–100 at low temperatures (Fig. 2). At high densities of electrons and holes, temperature-independent dissociation mechanisms involving the recombination of holes and electrons directly at F_2-centers become significant below 150 K (Ref. 6). This circumstance also prevented us from observing an increase in the yield of F_2-center dissociation by a factor of more than 10^2.

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