Effect of x-ray irradiation on the magnetoplastic effect in NaCl crystals

V. I. Al'shitz, E. V. Darinskaya, and O. L. Kazakova
Institute of Crystallography, Russian Academy of Sciences, 117333 Moscow, Russia

(Submitted 20 July 1995)
Pis'ma Zh. Éksp. Teor. Fiz. **62**, No. 4, 352–357 (25 August 1995)

A new method is proposed for investigating the effect of irradiation with x-rays on the spectrum of point defects in crystals. The method is based on studying the magnetoplastic effect in the irradiated crystals. The main information is contained in the measurements of the average travel distance of dislocations as a function of the rotational frequency of the sample in a magnetic field. Experimental studies of NaCl crystals demonstrated that the method is uniquely sensitive to small irradiation doses (starting at 10^3 rad). It was found that the measured frequency dependences of the dislocation displacements respond to a change in the state of the existing impurity centers and to the production of intrinsic radiation-induced defects. The kinetics of the regeneration of radiation-induced defects and metastable states of impurity complexes as a result of illumination of the irradiated samples by a tungsten lamp for t_l= 15–60 min was analyzed. © *1995 American Institute of Physics.*

The existence of the hardening effect of x-ray irradiation on the plasticity of crystals has been well known for a long time[1,2] The mechanism of this effect is usually linked with the modification of existing potential barriers to the motion of dislocations and to the production of new point defects which stop dislocations. We are generally dealing with appreciable irradiation doses (for alkali-halide crystals, starting with doses of the order of 10^5 rad). Existing methods usually cannot identify the different stoppers that pin dislocations. We shall demonstrate here a new method for studying radiation-induced defects in crystals. As we shall show below, on the one hand, this method is highly sensitive to low doses of x-ray irradiation (starting at 10^3 rad) and, on the other, it makes it possible to follow the change in the state of impurity centers and the production of new radiation-induced defects.

The method is based on the investigation of the effect of x-ray irradiation on the magnetoplastic effect (MPE) in nonmagnetic crystals. This crux of this phenomenon, first discovered in NaCl crystals,[3] is that dislocations are displaced under the action of a static magnetic field in the absence of a mechanical load. In subsequent investigations performed in our group,[4–12] the same effect was observed in other alkali-halide crystals and in the metals Zn and Al. Other authors have also made independent contributions to the theoretical and experimental study of the magnetoplastic effect.[13–17]

The basic features of the phenomenon are as follows. The average displacement of dislocations, l, in a magnetic field is proportional to the magnetic "treatment" time t_m of the sample and to the squared magnetic induction ($l \propto B^2 t_m$). Dislocations which are oriented parallel to the magnetic field are insensitive to the latter quantity. The dislocation displacement l decreases with increasing concentration C of paramagnetic impurities ($l \propto 1/\sqrt{C}$). The effect is "killed" if diamagnetic impurities are predominant in the crystal. The temperature dependence of the dislocation displacement is very weak: l is virtually identical at 4.2 K and 77 K, and as the temperature increases to room temperature, the displacement increases by only 15–20%. The density of mobile dislocations increases with the magnetic induction B and increasing time t_m, and it can reach 100% of the density of freshly introduced dislocations. For high values of $B^2 t_m$ the displacement l also saturates — at a level corresponding to the average distance between the dislocations in a "forest." Rotation of the sample in a magnetic field gives rise to a critical frequency ν_c ($\nu_c \propto B^2$), above which the displacement l drops sharply to the background levels which have no connection with the magnetic field.

The physical model describing the phenomenon is based on the idea of detachment of dislocations from paramagnetic impurity centers as a result of spin-dependent electronic transitions which are stimulated in the system impurity + dislocation by a magnetic field whose role reduces to removing the spin forbiddenness on the processes which breakdown local barriers. The subsequent motion of the dislocations occurs in the field of the long-range internal stresses from other dislocations and, naturally, stops when the displacements reach values of the order of the average distance between dislocations.

The phenomena which are determined by spin-dependent electronic transitions in a magnetic field are well known in physics.[18] These are, for example, the effect of a magnetic field on the rates of chemical reactions,[19] on the photocurrent in semiconductors,[20] and on the viscosity of amorphous alloys.[21] In all cases the role of the magnetic field reduces to removing the spin forbiddenness on the electronic transitions followed by a change in the configuration of the system, so that the interaction energy in the system can change substantially and it can even change sign while holding the total energy virtually constant. A necessary condition for the existence of such effects is that the spin-dependent transition time must be small compared with the spin-lattice relaxation time τ_{sl} of the experimental system.[18] We note that for systems of interacting defects in alkali-halide crystals τ_{sl} can be quite large. For example, according to the measurements performed in Ref. 22 , the spin-lattice relaxation time of associations of F-centers in KCl crystals at $T = 50$ K in a magnetic field $B = 0.34$ T is equal to 10^{-2} s. In our experiments the detachment time of dislocations from a paramagnetic center, estimated to be $\sim 10^{-2} \nu_c^{-1}$, is usually an order of magnitude smaller.

The critical frequency ν_c, which is the fundamental characteristic of an elementary act of detachment of a dislocation from a point defect, does not depend on the temperature, on the concentration of defects, and on the magnetic treatment time. It is determined solely by the magnetic field and the type of paramagnetic centers.[9] The parameter ν_c is therefore a convenient characteristic for analyzing the results of x-ray irradiation on crystals. This is the basis of the proposed method.

The investigations were performed on vacuum-grown NaCl crystals with a yield strength $\tau = 15$ g/mm^2 and Ca impurity concentration $C = 0.5$ ppm. The preannealed samples which were prepared for the test were irradiated in an IRIS-M x-ray apparatus with a molybdenum source and wavelength $\lambda = 0.7$ Å . The voltage on the x-ray tube and

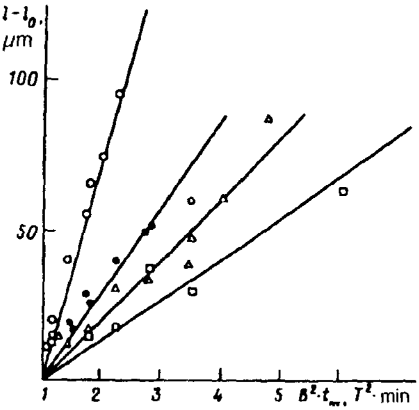

FIG. 1. Average dislocation displacement measured from the background value l_0 (with $B=0$) as a function of the holding time t_m in a magnetic field and the squared magnetic induction B^2 for different irradiation times t_r. ○ — $t_r=0$; ● — $t_r=5$ s; △ — $t_r=10$ s; □ — $t_r=20$ s.

the current were, respectively, $U=45$ kV and $I=35$ mA. The irradiation time was $\tau_r=5-60$ s. For the sample thicknesses investigated the measured through-transmission coefficient for x-rays was 70–80%. No visible coloring of the crystals was observed at such low irradiation doses. Fresh dislocations were introduced into the sample after irradiation, after which the crystal was subjected to magnetic treatment for a time $\tau_m=2-20$ min in a uniform static field $B=0.2-0.7$ T of an electromagnet. A magnetic field with a variable direction was produced by rotating the sample with frequency $\nu=0-200$ Hz in a constant magnetic field. In experiments on the decay of radiation-induced defects the irradiated sample was de-excited under a tungsten incandescent lamp within 15–60 min. The dislocations were then introduced into it, and it was subjected to the standard magnetic treatment. The displacement of dislocations was monitored by the method of selective chemical etching. All experiments were performed at room temperature. Displacement of the edge and screw dislocations in a magnetic field was observed in the irradiated crystal. However, representative histograms for determining the average dislocation displacements were obtained only for edge dislocations. The transmission spectra of the irradiated samples were investigated on an IFS-113v Fourier spectrometer for the IR range ($\lambda=2-25$ μm) and on a Hitachi spectrometer for the visible and UV ranges. A deuterium lamp ($\lambda=185-360$ nm) was used for the UV range and a tungsten lamp ($\lambda=364-2500$ nm) was used for the visible range.

In the course of the experiments it was found that in irradiated NaCl crystals the displacement of edge dislocations remains a linear function of the magnetic treatment time and a quadratic function of the magnetic induction (Fig. 1). In Fig. 1 and subsequent figures the dislocation displacement is measured from the background displacement l_0 observed in the absence of a magnetic field. The quantity $l_0 \sim (20-30)$ μm is determined by the weak relaxation of the dislocation structure as a result of the etching out of surface stoppers.[23] As one can see from Fig. 1, displacement distances are much shorter in irradiated samples than in unirradiated samples. This is related in an obvious manner to

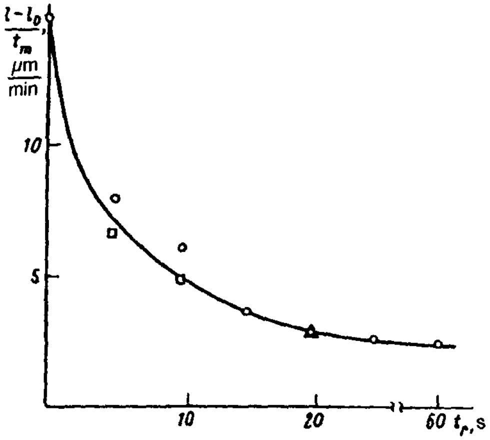

FIG. 2. FIG. 2. Effective dislocation velocity $(l-l_0)/t_m$ in a magnetic field $B=0.5$ T as a function of the irradiation time t_r of the samples for different magnetic treatment times t_m: ○ — $t_m=5$ min; □ — $t_m=10$ min; △ — $t_m=20$ min.

the increase in the concentration of point defects.[24,25] The effective velocity $(l-l_0)/t_m$ of dislocations in a magnetic field decreases monotonically with increasing irradiation time t_r (Fig. 2).

Figure 3 shows the average dislocation displacement curve plotted as a function of the rotational frequency of the sample in a magnetic field. As follows from this figure, the shape of the curve $l(\nu)$ is substantially different for the irradiated crystals: It changes from a one-step curve into a two-step curve. Therefore, instead of one critical frequency ν_c, there are two critical frequencies ν_{c1} and ν_{c2} ($\nu_c<\nu_{c1}<\nu_{c2}$), and the dislocation displacement decreases to zero only above ν_{c2}. Simply increasing the holding time of the samples in a magnetic field results only in an overall increase of the displacements on the

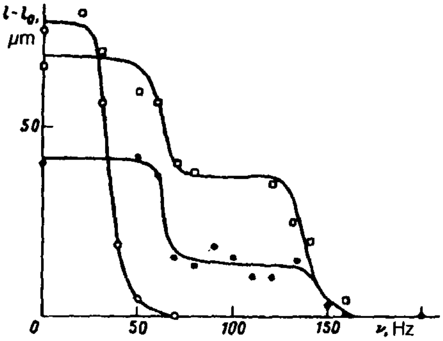

FIG. 3. Dislocation displacement $l-l_0$ as a function of the rotational frequency ν of the sample in a magnetic field $B=0.5$ T for different irradiation times and magnetic treatment times: ○ — $t_r=0$, $t_m=5$ min; ● — $t_r=5$ s, $t_m=5$ min; □ — $t_r=5$ s, $t_m=10$ min.

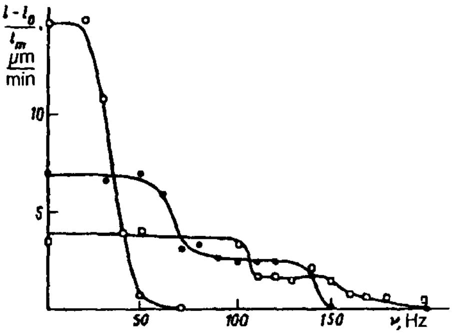

FIG. 4. Effective velocity $(l-l_0)/t_m$ as a function of the rotational frequency ν of a sample in a magnetic field $B=0.5$ T for different irradiation doses: ○ — $t_r=0$ s; ● — $t_r=5$ s; □ — $t_r=20$ s.

plateaus, with no change in the critical frequencies. The presence of two critical frequencies indicates that there exist two types of paramagnetic stoppers for dislocations in irradiated crystals. We shall call them type-1 and type-2 stoppers.

It is important that the first and second steps on the curve $l(\nu)$ respond differently to an increase in the irradiation dose. As one can see from Fig. 4, when the irradiation time increases from $t_r=5$ s to $t_r=20$ s, the frequency ν_{c1} increases substantially without any appreciable change in the frequency ν_{c2}. The intermediate dose corresponding to irradiation time $t_r=10$ s extends the first step, as if "smearing" it between the positions characteristic for $t_r=5$ s and $t_r=20$ s, but leaves the second step at virtually the same place (within the experimental errors).

In the de-excitation experiment it was shown (Fig. 5) that illumination of the irradiated samples under a tungsten lamp for 15 min has no effect on the function $l(\nu)$. However, de-excitation for 30 min causes the second step to vanish. The first step then

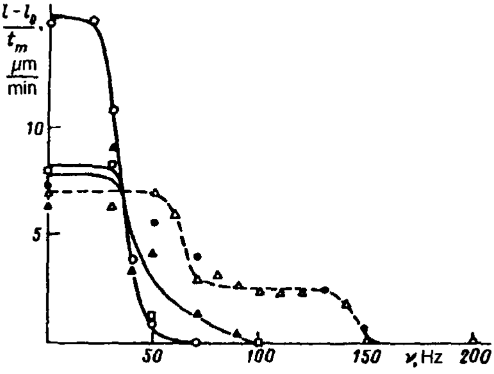

FIG. 5. Effective velocity $(l-l_0)/t_m$ as a function of the rotational frequency ν of a sample for holding time $t_m=10$ min and magnetic induction $B=0.5$ T for different de-excitation times t_l: ○ — $t_r=0$, $t_l=0$; △ — $t_r=5$ s, $t_l=0$; ● — $t_r=5$ s, $t_l=15$ min; ▲ — $t_r=5$ s, $t_l=30$ min; □ — $t_r=5$ s, $t_l=60$ min.

becomes smeared between the positions ν_c and ν_{c1}. Finally, illumination of the sample for 1 h restores the critical frequency ν_c characteristic of the unirradiated crystal. However, the dislocation displacement on the plateau (for $\nu < \nu_c$) does not return to the level corresponding to the unirradiated crystal.

The data presented above show that the stopper lifetimes are different. This agrees qualitatively with Refs. 26–29. Type-2 stoppers are short-lived. Their lifetime under our conditions of illumination is of the order of 15–30 min. This circumstance and the fact that they appear as a result of irradiation, and that their state does not change with the irradiation time suggests that these stoppers are radiation-induced intrinsic defects. We note that holding the irradiated samples for 1 day in black paper has no effect on the interaction of a dislocation with these stoppers in a magnetic field.

Type-1 defects are long-lived. They decay within 1 hour. The decay process starts approximately 30 min after de-excitation begins. It is logical to assume that type-1 stoppers are the Ca impurities which are altered in the process of irradiation. The three frequencies which were recorded in our experiments and which correspond to the first step on the functional dependence $l(\nu)$ — $\nu_c = 35$ Hz ($t_r = 0$), $\nu_{c1} = 65$ Hz ($t_r = 5$ s), and $\nu_{c1} = 105$ Hz ($t_r = 20$ s) — apparently reflect three states of the Ca impurity complexes — the ground state and two radiation-stimulated states. The latter states probably form as a result of trapping of electrons in metastable levels; they naturally decay in light, returning the complex to the ground state. Curiously, irradiation makes the impurity magnetically more sensitive, reducing the spin-dependent transition times ($\nu_{c1} > \nu_c$), and therefore increasing the probability for the dislocation detachment from the corresponding complexes.

For the spectral composition of the light from a tungsten lamp, not all radiation-induced defects are de-excited within 1 h. This is indicated by the low position of the plateau as compared to unirradiated crystals. The remaining defects are not paramagnetic, since they do not affect the critical frequency.

Optical investigations of the samples performed in a wide range of wavelengths (from IR to UV) showed that the devices are insensitive to the low irradiation doses which we used. The absorption bands corresponding to F centers and a small increase in the IR transmission (by 5–20%) could be observed only by increasing the irradiation dose of the sample by two orders of magnitude. At the same time, even at low irradiation doses which we used, and therefore the small structural changes that they cause the magnetoplastic effect is still sensitive to them. In our experiments a dislocation is thus a very fine and very sensitive tool which is responsive to very small changes in the structure of the crystal.

A combination of the method proposed here, together with the well-known method of deformation luminescence,[30-35] seems promising for achieving a definitive "interpretation" of the radiation-induced defects that appeared in our experiments.

We wish to thank V. L. Indenbom, A. F. Konstantinova, Ya. M. Soĭfer, and V. B. Timofeev for a helpful discussion of the results. We also thank A. V. Bazhenov, V. P. Kisel', L. A. Korostel', E. B. Rudneva, I. L. Smol'skiĭ, and V. M. Chernov for methodological assistance. This work was supported by the International Science Fund (grant M19300) and the Russian Fund for Fundamental Research (grant 95-02-03920-a).

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Translated by M. E. Alferieff