Spontaneous migration of liquid inclusions in an irradiated crystal

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(Submitted 10 December 1986)
Pis'ma Zh. Eksp. Teor. Fiz. **45**, No. 3, 144–146 (10 February 1987)

A spontaneous random walk of liquid inclusions has been observed in a crystal with a uniform distribution of radiation defects. An explanation is offered for the motion itself and for the fact that its direction is random. The thermodynamic basis for the observed effect is a decrease in the defectiveness of a crystal as inclusions move in it.

A liquid inclusion in a single crystal (e.g., an inclusion of the mother solution which is trapped during growth) can move as a whole.[1] This motion occurs through a dissolution of the crystal material at one surface of the inclusion, a diffusion of this material through the inclusion, and its deposition at the opposite surface. The motion of the inclusion occurs in the direction opposite the direction in which the crystal material flows through the bulk of the inclusion. The driving force for this process is a gradient of the chemical potential of the crystal atoms, itself due to either an external force field or a nonuniform distribution of defects or stresses in the part of the crystal with the inclusion.[1–3] We have detected a different type of motion of inclusions, which occurs when the crystal is uniformly defective, so that in the initial state there is no force which is capable of driving the motion.

We observed this other type of motion of inclusions in irradiated KCl crystals with inclusions of a saturated aqueous solution up to 100 μm in size. The crystals are bombarded with 10-MeV electrons in a dose of 10^{17} cm^{-2}. Spectral studies of the irradiated crystals revealed uniformly distributed color centers, primarily F and V_3 centers, with concentrations comparable in magnitude, reaching 2×10^{17} cm^{-3}.

After the bombardment, the crystals were held under isothermal conditions at room temperature. Here we observed a spontaneous random motion of the inclusions. The brightened tracks left behind the inclusions testify to the reality of this motion and show its path. Figure 1 is a typical illustration of the effect. In these experiments it was found that the velocity at which the inclusions move depends on their size (Fig. 2).

The motion of an inclusion is a consequence of a directed transport of crystal material through the inclusion, driven by a difference between the chemical potentials of the atoms on the bow and stern surfaces of the inclusion. This difference arises upon a very slight displacement of an inclusion, because of a fluctuation or some briefly applied external agent, because the crystal contains color centers near the bow surface of the inclusion, while an uncolored crystal forms at the stern surface. This nonequivalence of the structural states of the bow and stern surfaces of the inclusion persists as the inclusion moves, and it maintains the existing difference in chemical potentials. The system draws the energy required for the motion of an inclusion from the energy of the color centers, which disappear in the course of the motion.

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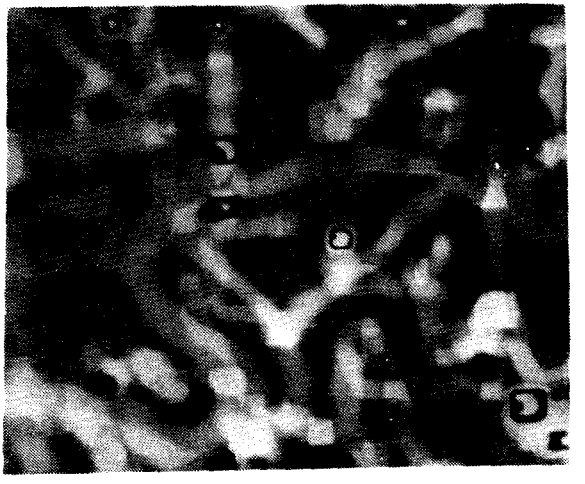

FIG. 1. Migration of inclusions in an irradiated crystal, 30 days after irradiation. Magnification is 100x.

The observed dependence $v(R)$ is explained in the following way. For small inclusions, the motion of an inclusion is known[4] to be a threshold effect, by which we mean that inclusions with a size below a certain threshold are immobile in a given force field. The velocity of a moving inclusion is proportional to the gradient of the chemical potential of the atoms in the interior.[1] A distinctive feature of our case is that the difference between the chemical potentials at the bow and stern surfaces does not depend on the size of the inclusion. Consequently, the gradient of the chemical potential and thus the velocity of the inclusions decrease with increasing size of the inclusions. This is the dependence which we observe for large inclusions.

The fact that the motion of the inclusions is a random walk is explained in the following way. The dissolution of crystal material occurs at that surface of the inclusion which has the most active dislocation source of dissolution steps. As the inclusion moves, its surface is intersected by new, randomly positioned dislocations. The role of guide is transferred from one dissolution source to another, so that the inclusion starts to move in a different direction.

This effect might be utilized to study the dissolution of irradiated crystals and to find the energy characteristics of radiation defects.

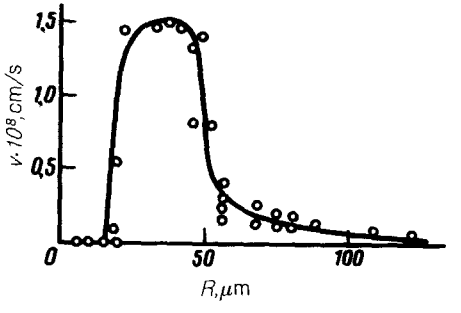

FIG. 2. Velocity of the inclusions versus their size.

We are deeply indebted to Ya. E. Geguzin for useful comments in the course of a discussion of this study and to Yu. V. Vladimirov, who irradiated the samples.

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Translated by Dave Parsons