Electron-beam-induced cyclic structural changes in twinned crystals

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Electron microscopy reveals that an electron beam induces cyclic changes in the twinning structure in twinned zinc sulfide and silicon crystals. The typical periods of these cyclic changes are 10 s for zinc sulfide and 0.1–0.5 s for silicon.

Cyclic structural changes have been observed previously[1] during illumination of a sample at a constant light intensity in the region of a first-order ferroelectric phase transition.[1] Shmyt'ko *et al.*[1] attribute the cyclic nature of the phase transition to a change in the nonlinear characteristics of the ferroelectric material during the conversion. Another case of cyclic structural changes caused by constant excitation was described by Negriĭ and Osip'yan,[2] who detected periodic optical "scintillations" of various polarizations in cadmium sulfide. In other words, they observed a pumping of intensity from one polarization to another. The effect occurred in a "grazing" band during continuous illumination.

In the present letter we report electron-microscope observations of cyclic changes in an atomic structure which are induced by a continuous electron beam in zinc sulfide and silicon crystals at temperatures far from any possible phase transitions.

The samples were zinc sulfide microtwinned crystals and silicon crystals containing growth twins. The crystals were cut in the $\{110\}$ plane.

The cyclic nature of the structural changes in the zinc sulfide sets in after a threshold current (10^{-7} A) of the electron beam is reached. The effect can be described as follows: Additional reflections appear and disappear periodically in time on the microdiffraction image of the twinned matrix for certain regions of the crystal (Fig. 1, a and b). Several types of additional reflections can be distinguished. The reflections of type II (Fig. 1c) appear as brief bursts and do not form anything resembling a regular system of points belonging to a plane lattice. The reflections of type I "flare up" nonuniformly with respect to each other, but they ultimately form a regular lattice.[1)] The oscillation period of this lattice changes from cycle to cycle and lies in the interval 8–10 s. This process was also detected in the diffraction-contrast regime. It was found that the oscillations of the image contrast arise at boundaries of a general type against the background of a system of coherent twins. In this case we observe a wavelike motion of the shadows of the contrast, similar to those which are customarily observed during the motion of dislocations in dynamic electron-microscopy experiments.[3]

The structural changes in silicon are slightly different. These changes are observed only in regions of the crystal containing nonequilibrium growth twins, which become saturated with dislocations by virtue of a preliminary deformation of the crystal. In contrast with the case of zinc sulfide, reflections corresponding to a new crystal

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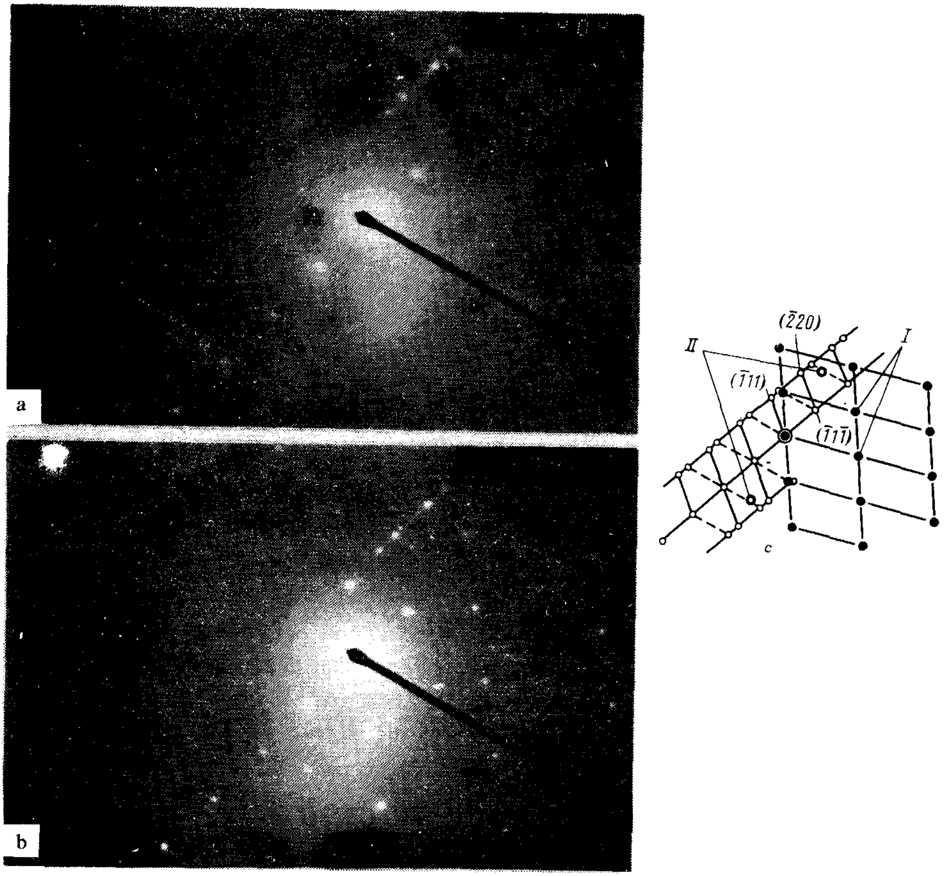

FIG. 1. Dynamic microdiffraction patterns of zinc sulfide, (110) cut. a—Electron diffraction pattern of the initial stage of the formation of additional reflections (of type II in part c) (the main reflections correspond to the microtwin matrix); b—electron diffraction pattern of the state corresponding to a manifestation of the lattice of oscillating reflections (of type I in part c); c—relative arrangement of the twinned matrix and of the oscillating reflections.

structure do not appear, and there is a cyclic pumping of the intensity of the image of the reflections of one twinning orientation into the other. We estimate the period of these oscillations to be 0.5–1.0 s. In silicon, the process decays rapidly over time.

The relaxation period of the process varies from point to point in the sample in the interval 5–20 s. Another observation is that in silicon the structural changes are most obvious in twins containing boundaries with jogs (Fig. 2). The latter go into a more equilibrium state in the course of the process.

In interpreting these results we should seek an explanation both of the mechanism itself for the structural changes and of the cyclic nature of the process. We believe that the mechanism for the change in the crystal structure involves the changes in disloca-

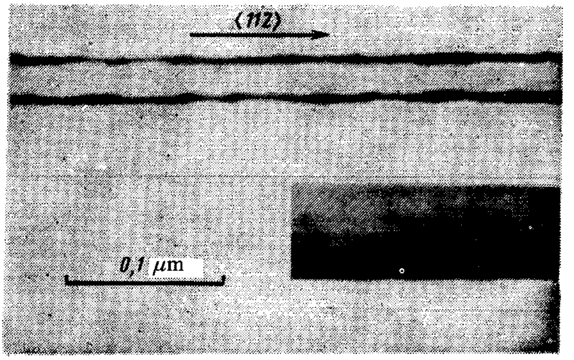

FIG. 2. Illustrative image of incoherent twinning regions in silicon in which oscillating structural changes are observed.

tion structure observed experimentally. Specifically, what we have in mind is that the relationship between the changes in the dislocation structure and those in the crystal structure may be caused by a repacking of atomic layers during the sequential motion of partial dislocations.[4] In our case the observed cyclic nature of the process requires that the motion of the dislocations be reversible. Precisely this behavior of dislocations has been observed during elastic detwinning processes.[5]

One candidate for the driving force that gives rise to the cyclic movement of dislocations may be the electrostatic repulsive force acting between charges which are produced by the incident beam and which accumulate near dislocation centers and near twinning boundaries. In this case the structural changes due to the motion of dislocations continue until the accumulated charge decreases, when a dislocation emerges from the zone affected by the beam, for example, or as a result of breakdown of the accumulated electrons to the mass of the crystal holder. In either case, a decrease in the repulsive force returns the dislocations to the equilibrium state and, correspondingly, restores the original structure. The process then repeats. If the defect structure in the crystal changes during the cyclic process, or if some of the dislocations become pinned or escape from the zone affected by the beam in each cycle, the spontaneous oscillations will decay over time, as is observed experimentally.

This model for the cyclic structural changes is only conjecture. Further research will be required to resolve all aspects of the mechanism for the structural changes.

The results of this study, like the results of Refs. 1 and 2, suggest that spontaneous oscillations are a rather common phenomenon in solids containing dislocations.

1)With only one cross section in the reciprocal lattice, we are not able to unambiguously identify the observed structure.

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[1] I. M. Shmyt'ko, V. I. Ivanov, V. Sh. Shekhtman, and S. S. Khasanov, Pis'ma Zh. Eksp. Teor. Fiz. **29**, 425 (1979) [JETP Lett. **29**, 386 (1979)].
[2] V. D. Negriĭ and Yu. A. Osip'yan, Pis'ma Zh. Eksp. Teor. Fiz. **35**, 484 (1982) [JETP Lett. **35**, 598 (1982)].
[3] S. Ikeno, Phys. Status Solidi **a36**, 317 (1976).
[4] I. M. Shmyt'ko, L. A. Matveeva, S. I. Bredikhin, V. Sh. Shekhtman, and S. Z. Shmurak, Fiz. Tverd. Tela

(Leiningrad) **26**, 2033 (1984) [Sov. Phys. Solid State **26**, 1233 (1984)].

[5] S. A. Omel'chenko, Issledovanie vliyaniya deformatsii na sturkturu kristallov sul'fida i selenida tsinka (Effect of Deformation on the Structure of Zinc Sulfide and Selenide Crystals. Author's Abstract, Candidate's Dissertation, 1984).

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