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Surface structure changes of InP and GaAs single crystals irradiated with high energy electrons and swift heavy ions

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Abstract InP and GaAs crystal structure changes under the influence of swift Kr and Bi ions have been studied by means of scanning electron microscopy, atomic force microscopy and selective chemical etching. The previous disordering of samples by electron irradiation has shown to lead to macrodefect formation in the form of cracks and breaks, at the depths near the ion end-of-range, and on the crystal surface. A possible explanation of the observed effects is proposed.

Key words InP crystals • GaAs crystals • preliminary disorder • swift ion irradiation • surface topography • macrodefect formation

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Introduction

The formation of nanoclusters and nanowires which can modify electronic and optical properties of solids provide new possibilities for optoelectronic and nano-electronic development. One of the ways of nanometer structuring of solids is their irradiation with swift heavy ions. Under such an irradiation the track could be formed in the form of a nanometer cylinder or cluster chain with a modified structure embedded into the undamaged matrix. The mechanisms of the track formation under superhigh levels of the crystal electronic subsystem excitation are not clarified in detail until now.

The thermal spike model is often used to explain track formation [6]. Part of the energy used for the thermal spike formation (heating of the possible track region) is determined by electron-phonon coupling constant g characterizing the efficiency of the energy transfer from the excited electrons to the lattice atoms. The track formation in insulators and metals is observed that the electronic energy loss is exceeding a certain threshold value $(dE/dx)_{ethr}$ determined by an irradiated material type and its structural features. For semiconductors the value of $(dE/dx)_{ethr}$ usually exceeds the corresponding values for insulators and can be more than 30 keV/nm. However, we have demonstrated that track formation in previously disordered InP is possible at $(dE/dx)_e \sim 13$ keV/nm [4, 5]. It is very likely that disordering can modify electronic properties of the material thus changing the value of the electron-phonon coupling constant. Modification of electronic properties of the crystal due to the controllable embedding of the damages by light particles irradiation could be one of the ways of $(dE/dx)_{ethr}$ reducing.

Table 1. Parameters of electron interaction with GaAs and InP crystals: electron energies E_e, fluences Φ_e, defect formation cross sections σ_e, damage rates $D_e = \sigma_e \Phi_e$, maximum energies E^{AIII}_{max} and E^{BV}_{max} of displaced atoms and their mean energies E^{AIII}_{cp} and E^{BV}_{cp}.

Crystal	E_e [MeV]	$\Phi_e \times 10^{17}$ [e^-/cm^2]	$\sigma_e \times 10^{-22}$ [(dpa·cm²)/e⁻]	D_e [dpa]	E^{AIII}_{max} [keV]	E^{BV}_{max} [keV]	E^{AIII}_{cp} [keV]	E^{BV}_{cp} [eV]
GaAs	23	3.0 ± 0.5	3.1	9.3×10^{-5}	16.8	15.7	124	124
GaAs	4	4.0	1.45	5.8×10^{-5}	0.61	0.57	59.6	58.5
InP	23	3.0 ± 0.5	3.12	9.4×10^{-5}	10.2	37.9	127	136

Table 2. Parameters of ion interaction with GaAs and InP crystals: ion ranges R_p, specific inelastic energy losses in the near-surface layer $S_e = -(dE/dx)_{inel}$, defect formation cross sections near the surface σ ($x = 0$) and in the Bragg peak region σ ($x \approx R_p$), damage rates D ($x = 0$) and D ($x \approx R_p$), and sputtering coefficients S^{Kr}_{AIII} and S^{Kr}_{BV}.

Crystal	R_p [μm]	S_e [keV/nm]	σ ($x = 0$) [(dpa·cm²)/ion]	σ ($x \approx R_p$) [(dpa·cm²)/ion]	D ($x = 0$) [dpa]	D ($x \approx R_p$) [dpa]	S^{Kr}_{AIII} [atom/ion]	S^{Kr}_{BV} [atom/ion]
^{209}Bi (710 MeV, 1.0×10^{12} ion/cm²)								
GaAs	30.6	35.2	1.96×10^{-16}	6.8×10^{-15}	1.96×10^{-4}	6.8×10^{-3}		
^{86}Kr (253 MeV, 1.3×10^{14} ion/cm²)								
GaAs	23.3	15.8	6.7×10^{-17}	3.2×10^{-15}	8.7×10^{-3}	4.2×10^{-1}	0.016	0.017
InP	25.5	12.6	5.3×10^{-17}	2.8×10^{-15}	6.9×10^{-3}	3.6×10^{-1}	0.017	0.03

The aim of the present work was to study the influence of preliminary disordering by MeV electron irradiation on the effects of the swift ion interaction with InP and GaAs crystals.

Experimental

The samples of (100)-oriented n-type GaAs and InP single crystals of 1 × 1 cm² sizes and thickness of ~ 300 μm were irradiated with 4 MeV electrons to a fluence of 2×10^{17} e⁻/cm². After that, the virgin and previously irradiated with electrons the samples were bombarded with 710 MeV ^{209}Bi ions to a fluence of 1.0×10^{12} ion/cm².

Other series of the same GaAs and InP samples were irradiated with 23 MeV electrons to a fluence of (3.0 ± 0.5) × 10^{17} e⁻/cm². Then a half of the area of the virgin and irradiated samples was covered by a nickel foil with the thickness exceeding ion projected range, and the samples were irradiated with 253 MeV ^{86}Kr ions to a fluence of 1.3×10^{14} ion/cm². The temperature of both the series of samples did not exceed 50°C during the electron and ion irradiations.

Surface topography was investigated in a scanning electron microscope S-806 (Hitachi) and an atomic force microscope "Femtoscan 001".

Depth distribution of the damage was revealed by the treatment of the crystals cleaved perpendicular to the sample surface in AB etchant (CrO_3:H_2O:HF:$AgNO_3$) [10] with following observations in an optical microscope Leica INM-100.

The parameters characterizing ion interactions with GaAs and InP were calculated using the TRIM-98 program [2].

Results and discussion

The main calculated parameters of the process of ion and electron interactions with GaAs and InP are reported in Tables 1 and 2.

As our AFM data show, the electron irradiation does not change surface topography of InP and GaAs samples (not shown in figures). The ^{86}Kr ion irradiation leads to a some smoothing of the surface relief of the virgin and electron irradiated crystals. At the same time, the "ripples" appear on the irradiated part of the surface, and (in the case of InP) cones and needles arise (Fig. 1). It seems to be due to the non-uniform sputtering during the irradiation.

The authors of [3, 8, 9] have studied processes of the sputtering of GaAs and InP crystals, and $Si_{0.84}Ge_{0.16}$ epitaxial layers due to the low-energy ^{40}Ar ion bombardment. After the irradiation "ripples" and a needle relief appear on the surface of the investigated samples similar to our samples. But, noticeable surface changes start at a fluence of $\Phi_{Ar} \sim 10^{15}$ ion/cm². We have calculated sputtering coefficients and the mean thicknesses of sputtered layers using an elastic sputtering model for Ar ion bombardment with energy of 5 keV and $\Phi_{Ar} = 10^{15}$ ion/cm². The calculated mean thicknesses of sputtered layers are ~ 0.84 nm for InP and 1.0 nm for GaAs.

The thicknesses of layers sputtered by ^{86}Kr ions InP and GaAs calculated for our experimental conditions are 9.8×10^{-4} and 9.6×10^{-4} nm, correspondingly. These values are too small to explain the observed surface topography changes. We conclude that inelastic sputtering [1] takes place during the 253 MeV krypton irradiation due to the high inelastic energy losses S^{Kr}_e

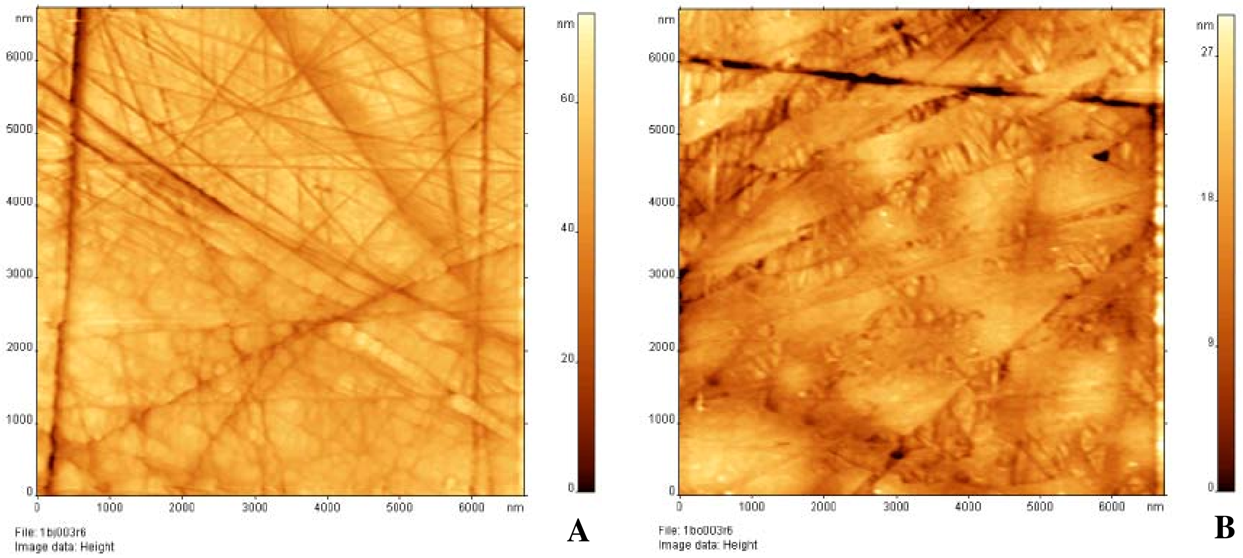

Fig. 1. Unirradiated (A) and irradiated InP surfaces with ^{86}Kr ions (E = 253 MeV, Φ_{Kr} = 1.3 × 10^{14} ion/cm^2) and electrons (E = 23 MeV, Φ_e = 3.0 × 10^{17} e$^-$/cm^2) (B). AFM topographic image.

equal to 12.6 keV/nm for InP and 15.8 keV/nm for GaAs.

For GaAs crystals bombarded with electrons and ^{209}Bi ions, the beginning of the flaking phenomenon have been fixed in our experiments. The region with strong mechanical damages in the form of microcracks has been revealed on the cleave of the double-irradiated sample at depths exceeding the ion projected range (Fig. 2).

We have revealed also that double irradiation of InP and GaAs with electrons and ^{86}Kr ions leads to swelling of double-irradiated crystal parts, and the formation of the microcracks on the surface and breaks and cleaves in the near-surface layers (Figs. 3, 4). The swelling effect has been observed in [7] for the ionic crystals (LiF and CaF_2) and covalent crystals (SiO_2, $Y_3Fe_5O_{12}$ and $Gd_3Ga_5O_{12}$) influenced by high-energy ions, where at fluences of about 10^{13} ion/cm^2 sometimes the irradiated layer of insulators cleaved out of the underlying undamaged crystal. In our experiment the level of mechanical stresses in the semiconductors InP and GaAs irradiated with much higher fluence Φ_{Kr} = 1.3 × 10^{14} ion/cm^2, is found to be insufficient to destroy the material. However, the preliminary disordering of crystals by electron irradiation facilitates destruction processes during the following swift ion irradiation. The observed effects could be explained by an increase of the volume of double-irradiated parts due to the amorphization process and by the mechanical stresses appearance in the region of swift ions stopping. Parts of the samples covered with a metal foil were not influenced by Kr ions, and in the irradiated parts the ions have been stopped in the crystal at the projected range depth. The free expansion of ion-irradiated volume was limited by the neighboring undamaged region, and the irradiated parts are swelled out over the sample surface. The mechanical stresses appear to lead to the cracking of double irradiated material regions. As Tables 1 and 2 show, the rate of damage due to electrons for InP and GaAs crystals is much lower

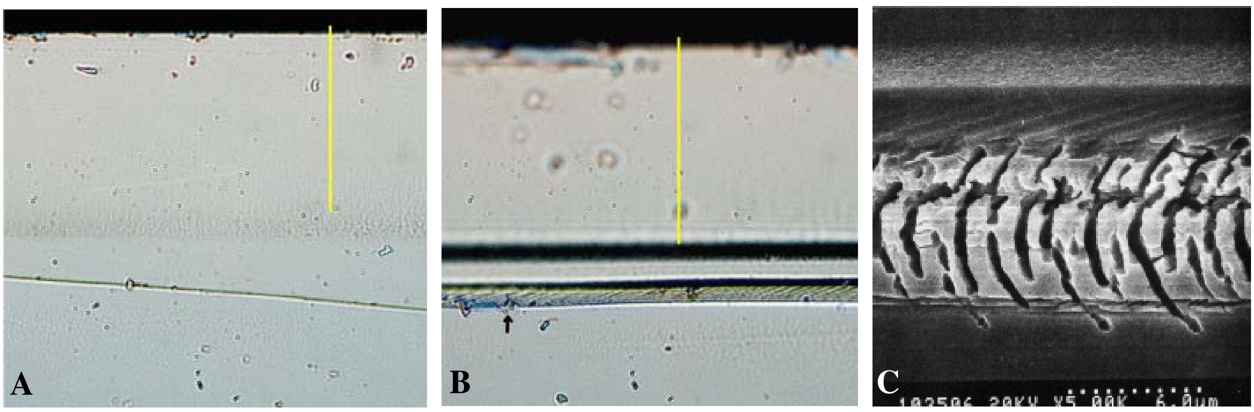

Fig. 2. Radiation damages revealed by selective chemical etching of GaAs samples cleaved perpendicularly to the sample surface: A – sample irradiated with ^{209}Bi ions (E = 253 MeV, Φ_{Bi} = 1 × 10^{12} ion/cm^2). B – sample irradiated with electrons (E = 4 MeV, Φ_e = 4.0 × 10^{17} e$^-$/cm^2) and ^{209}Bi ions (E = 253 MeV, Φ_{Bi} = 1 × 10^{12} ion/cm^2). C – image with a higher magnification of the microcracks region indicated by arrow in Fig. B. Vertical bars in Figures A and B show the ion projected range R_p^{Bi} = 30.6 ± 1.0 µm.

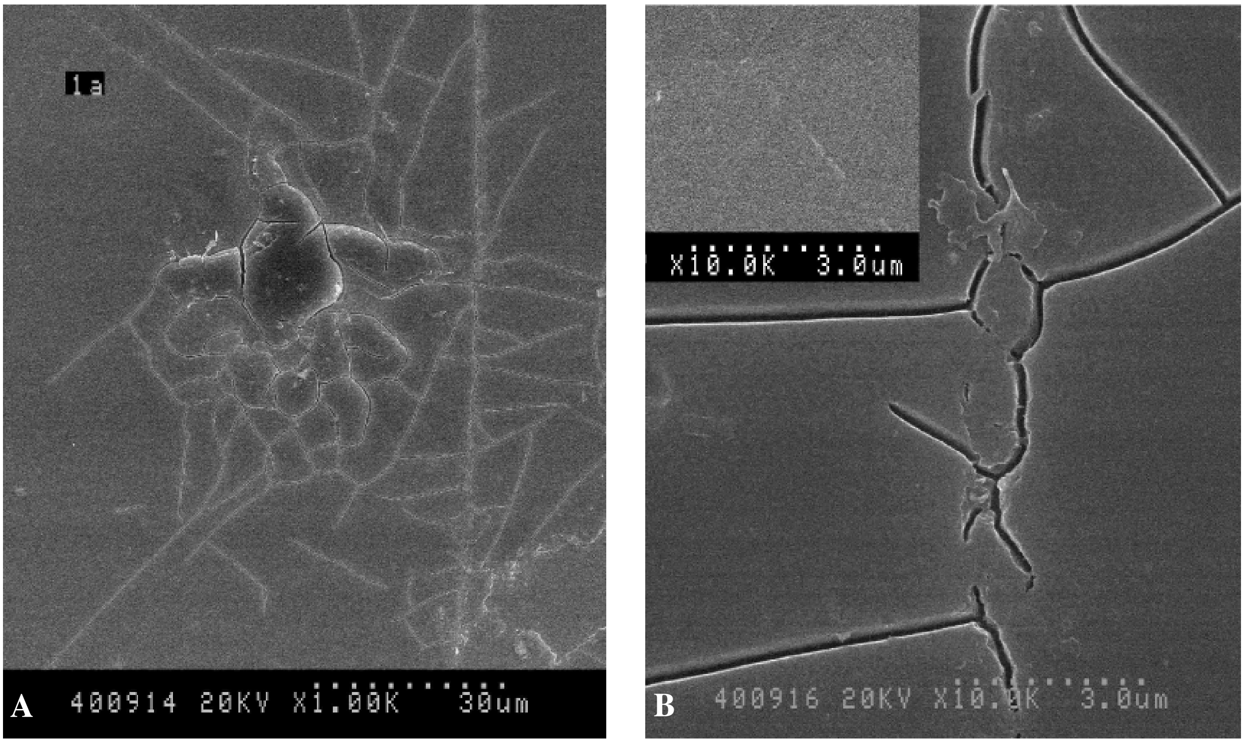

Fig. 3. Microcracks on the GaAs surface irradiated with electrons (E = 23 MeV, $\Phi_e = 3.0 \times 10^{17}$ e^-/cm^2) and Kr ions (Φ_{Kr} = 253 MeV, $\Phi_{Kr} = 1.3 \times 10^{14}$ ion/cm^2). Figures A and B are scanning electron microscopy images with different magnifications. Inset in Fig. B shows the sample surface irradiated with electrons only.

than due to ion irradiation. Nevertheless, the effects of swift ion bombardment of crystals previously irradiated with electrons are very considerable and unusual. To explain the influence of electron irradiation facilitating mechanical destroying of the samples, we have evaluated the sensitivity of the materials under consideration to the electronic energy losses. This property is defined by the parameter $\eta = Q/H_f$ [11]. Here ΔH_f is the latent heat of fusion, $Q = 0.63S_e/(\pi \cdot \lambda^2)$ is the mean energy density deposited by the ion in a cylinder of radius λ equal to the electron mean free path. If $\eta > \eta_{cr1} = 1.3$, the lattice is considered as being sensitive to inelastic energy losses, if $\eta < \eta_{cr2} = 0.7$, it is insensitive to inelastic energy losses. In the range of $\eta_{cr2} \le \eta \le \eta_{cr1}$, the lack of precision of used parameters does not allow any definite conclusion. The calculated mean distances between isolated point defects generated by electron irradiation were chosen as the electron mean free path. The wave

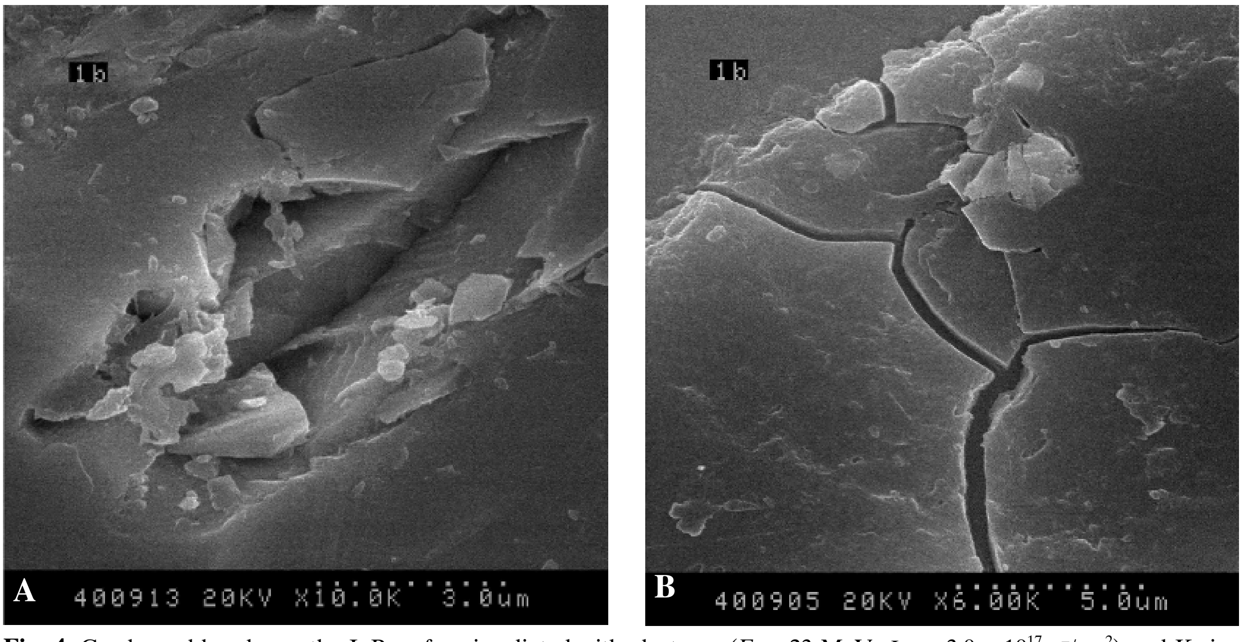

Fig. 4. Cracks and breaks on the InP surface irradiated with electrons (E = 23 MeV, $\Phi_e = 3.0 \times 10^{17}$ e^-/cm^2) and Kr ions (E_{Kr} = 253 MeV, $\Phi_{Kr} = 1.3 \times 10^{14}$ ion/cm^2).

functions of free electrons in an ideal crystal representing the translationally invariant Bloch functions [12]. The presence of vacancies breaks crystal periodicity, and this approximation seems to be reasonable. The mean distances between point defects generated by electron irradiations are equal to 12.1 and 12.5 nm for InP and GaAs, correspondingly. These values are comparable with electron mean free paths in metals [11]. As calculations show, for InP $\eta_{Kr} = 0.88$. For GaAs we have $\eta_{Kr} = 0.83$, $\eta_{Bi} = 1.34$. In all cases η exceeds a lower limit of crystal sensitivity to electronic energy losses $\eta_{cr} = 0.7$.

In the review [13], the results describing the isotopic disorder influence on kinetic coefficients of semiconductors and alkali-halide crystals (diamond-$^{12}C_{1-x}{}^{13}C_x$, $^{7}Li_{1-x}{}^{6}Li_xF$, $^{70}Ge_x^{nat}Ge_{1-x}$) and models for the calculation of these coefficients changes are presented. Moreover, ^{13}C concentration in a diamond increases from 0.001% up to 10% (four orders of magnitude) at room temperature (~300 K), the thermal conductivity coefficient decreases by four times. So, the significant thermal conductivity changes are caused by the change of an isotope atomic weight by one (^{12}C replacement with ^{13}C). In the case of electron irradiation at damage rate $D_e \sim 9.3 \times 10^{-5}$ dpa vacancies are formed in InP and GaAs crystals being the centers of excited electron scattering and affecting phonon propagation. This could reduce electron and lattice thermal conductivities and increase the lifetime of an overheated region around heavy ion trajectories, thus causing additional defects formation and amorphization of the irradiated layer of the crystal.

Summary

The influence of previous disordering by MeV electron irradiation on the effects of swift ion interaction with InP and GaAs crystals has been studied. It has been shown that previous electron irradiations lead to macrodefect formation during the following irradiation of InP and GaAs with such ions. For low ion irradiation fluences (^{209}Bi, $E_{Bi} = 710$ MeV, $\Phi_{Bi} = 1.0 \times 10^{12}$ ion/cm^2) the layer with macrodefects in the form of cracks is formed at the depth near the ion end-of-range. With increasing irradiation fluence (^{86}Kr, $E_{Kr} = 253$ MeV, $\Phi_{Kr} = 1.3 \times 10^{14}$ ion/cm^2) the macrodefects evolve, microcracks and breaks are formed on the surface of InP and GaAs crystals.

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