IN SITU AND LCSM STUDY OF MECHANICAL STRESS IN RUBY INDUCED BY SWIFT HEAVY IONS

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Accumulation of mechanical stress in Al_2O_3:Cr crystals induced by high energy heavy ions has been studied by using *in situ* ionoluminescence and postradiation laser confocal scanning microscopy measurements. It was found that lattice defects created via high level electronic excitation lead to generation of high level compressive stresses even at moderate ion fluences, less than 1×10^{12} cm^{-2}. The results obtained are discussed in the frame of model suggesting interaction of non overlapping highly stressed track regions with individual "Cr^{3+} piezosensors".

Introduction

Lattice damages produced by swift heavy ions are concentrated within small volume, surrounding ion trajectory. This inevitably results in generation of local mechanical stress, which in own turn may affect final defect structure. The knowledge about of such a high energy heavy ion track-assisted stress is of considerable practical value in view of simulation of fission product impact in radiation resistant oxides and ceramics, as candidate materials for nuclear waste management (inert matrix fuel hosts) and prediction of their long-term radiation stability [1].

To monitor the evolution of stress in real-time during ion bombardment one can use the well-known piezospectroscopic method, utilizing the relationship between the stress and changes in optical spectra, in particular, in the ion beam induced luminescence (IBIL) spectra. Recently, we have reported the first estimates of the mean stress level in ruby crystals generated by (3÷7) MeV/amu Ar, Kr and Bi ions in the fluence range of 10^9 -10^{12} cm^{-2} for Ar and Bi and 10^9 -1×10^{13} cm^{-2} for Kr [2]. In particular, there was shown, that mechanical stresses are registered for bismuth ions only, when the average electronic stopping power, determined as total ionizing energy losses normalized to projected range, was about 29 keV/nm. Contrary, no significant changes in the stress state have been detected for Ar and Kr ions at the same radiation damage dose levels, as those for Bi ions. This strongly implies that lattice disorder induced by collective electronic excitations plays a dominant role in the observed effect. The purpose of this study is to continue the examination of stress accumulation in Al_2O_3:Cr crystals under 710 MeV Bi ion irradiation in the extended ion fluence range, up to 2×10^{12} cm^{-2} and obtain information about spatial distribution of stress field in irradiated material.

Experimental

The c-oriented ruby crystals used in our study were in form of thin platelets (5x5x0.5 mm) with chromium concentration approximately 0.05% Cr_2O_3 in weight. Before irradiation, the specimens were mechanically polished and annealed in air at 1150°C during 8 hours in order to remove the residual surface polishing defects. The IBIL spectra were registered by using experimental set-up on the ion beam line for applied research on the U-400 cyclotron at FLNR, JINR [3]. Spectra were acquired with time step of 5 s using Oriel MS260i™ Spectrograph employing a cooled CCD array. *In situ* examination was followed by laser confocal scanning microscopy (LCSM) measurements on the irradiated specimens. The LCSM technique (SOLAR TII set-up) has been applied to acquire the depth-resolved photostimulated R-lines spectra and to determine the residual stress profile through the heavy ion irradiated layer.

Regarding ruby, the piezospectroscopic effect consists of in stress-induced shift of the Raman and *R*-line luminescence positions. The latter data, as well as linewidths and intensities were determined by fitting spectra to double pseudo-Voigtian profiles by the least-squares method. The frequencies of the R-lines were found with accuracy of 1.0 cm^{-1}.

Since the positions of *R*-lines are temperature dependent, it was very important to separate the effects of stress and temperature on the luminescence wavelength shift. First of all, we diminished possible temperature contribution by applying very low ion beam input thermal power. In all our IBIL measurements the input power was not higher than 0.025 Wcm^{-2}. For comparison, the temperature increase of 120 K for 2 mm thick ruby crystal, irradiating with 330 keV protons at 300 K, was detected by piezospectroscopic method at input thermal power of 3 Wcm^{-2} [4]. This value exceeds those employed in our experiments more than two orders of magnitude. The temperature instability of the target holder with irradiating specimen was in the range ± 0.5 K. Possible dispersion in the *R*-lines positions due to such instability is no more than + 0.2 cm^{-1}.

Results and discussion

An example of the *R*-line luminescence spectra in ruby detected under 710 MeV Bi ion bombardment at 80 K is given in Fig. 1 for several ion fluences. As was expected, the lattice disorder produced by high energy heavy ions results, first of all, in gradual decrease of the luminescence signal with ion fluence. Another evident effect consists in the shift of positions and following splitting of the *R*-lines. Such a prominent changes in R-lines positions, as seen from Fig. 1, correspond to hydrostatic compressive stress level in the irradiating target about 2 GPa at ion fluence ~ 1.6×10^{12} cm^{-2}. The stresses in ruby were determined through the shift of the R_2-line luminescence, $\Delta\nu_2$, since this parameter has been shown to be rather insensitive to the deviatoric stress component in compression and are dependent only on the overall magnitude of hydrostatic stress component, σ_h.

Accordingly to [5], the R_2-line shift and approximate value of hydrostatic stress are connected by empirical relation:

$$\sigma_h\,(\mathrm{GPa}) \approx \Delta\nu_2\,(\mathrm{cm}^{-1})/7.61 \qquad (1)$$

Evidently, that residual stresses in the irradiated layer are not hydrostatic, therefore the expression (1) could be applied for estimates of mean stress level only and for comparison of different ion species impact.

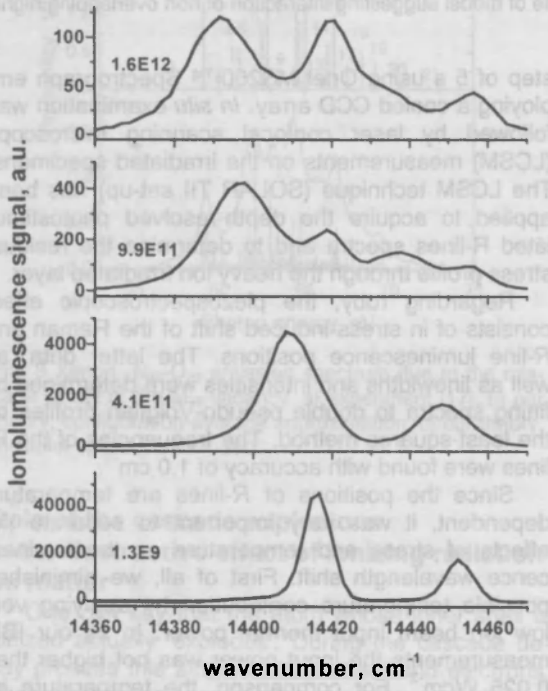

Fig. 1. Evolution of *R*-lines spectra versus 710 MeV Bi ion fluence. Irradiation temperature - 80 K. The frequency shift toward lower energies means that irradiating target layer is under compression.

Important information about stress tensor components can be found from different response of the R_1 and R_2-lines shift on the applied stress. If stresses are purely hydrostatic, $\Delta\nu_2$ and $\Delta\nu_1$ are equal. In our case $\Delta\nu_1 - \Delta\nu_2 = 0.5$ cm^{-1} already at ion fluence 3×10^{11} cm^{-2}. Detail piezospectroscopic studies of ruby stressed along different crystallographic axes gave the following relations [6,7]:

$$\Delta\nu_1 = 3.26(\sigma_{11}+\sigma_{22})+1.53\sigma_{33} \qquad (2)$$

$$\Delta\nu_2 = 2.73(\sigma_{11}+\sigma_{22})+2.16\sigma_{33} \qquad (3)$$

Here σ_{ii} (i = 1, 2, 3) – stress tensor comoonents, oriented along ***a***, ***m*** and ***c*** axes, respectively. The assumption of isotropic piezospectroscopic properties in the basal plane ($\sigma_{11}=\sigma_{22}$) enables the tensor components to be expressed as:

$$\sigma_{33} = (\Delta\nu_2 - 0{,}83\Delta\nu_1)/0.88 \qquad (4)$$

$$\sigma_{11} = (\Delta\nu_2 - 2{,}16\sigma_{33})/5.46 \qquad (5)$$

Fig. 2 shows the σ_{11} and σ_{33} values as a function of ion fluence under irradiation with Kr and Bi ions at 80 and 300 K calculated by using expression (4) and (5). As follows from this figure, the Bi ion irradiation leads to generation of compressive stresses in the basal plane and tensile stresses in the ***c***-axis direction.

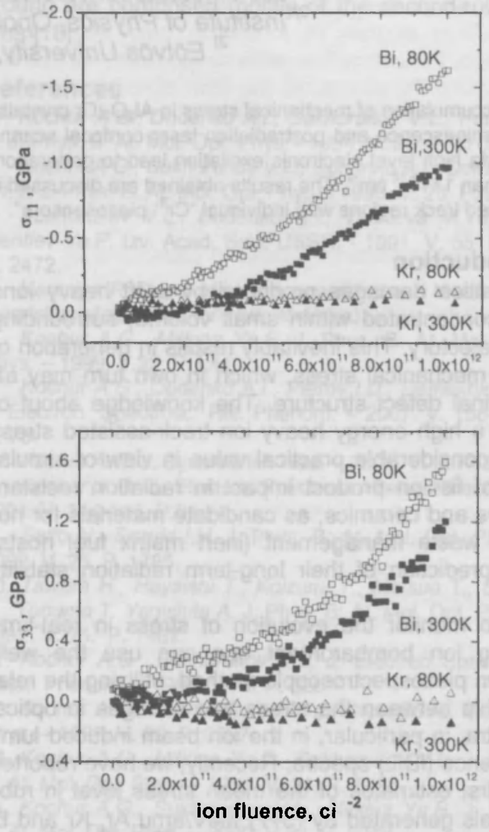

Fig. 2. Stress tensor components determined for ruby specimens irradiated with 710 MeV Bi and 245 MeV Kr ions at 80 and 300 K

Non-uniform energy deposition along swift heavy ion path suggests the complicated dependence of luminescence efficiency on target thickness. Under *in situ* measurements we register the integrated *R*-line luminescence signal influenced by radiation defects and mechanical stresses in total probed volume. Therefore, the residual stress profiles have been found in postradiation measurements by using confocal microscopy technique. Fig. 3 demonstrates the *R*-lines spectra as a function of depth, registered on ruby specimen irradiated with 710 MeV Bi ions under 60^0 ion beam incident angle at 80 K to fluence of 1.6×10^{12} cm^{-2} (the same specimen as was used in the IBIL experiment).

The ion projected range under such a tilted irradiation is 21.2 μm. The corresponding hydrostatic stress profile is given in Fig. 4 together with ionizing energy loss profile. As can be seen from the above figures, the stresses are maximal in the range 0-10 μm, where the electron stopping power varies from 41 to ~ 20 keV/nm. This proves the dominant role of electronic excitation in the build up of mechanical stress under Bi ion bombardment.

Since the 710 MeV bismuth ion track diameter in Al_2O_3 is within 3÷4 nm [8], our experimental conditions, namely, relatively low ion fluences used in our experiments correspond to non track overlapping irradiation regime.

Thus, the detected residual stresses could be considered as contribution of separate disordered regions, having strongly damaged core surrounded by a shell of stressed material. In own turn, the IBIL and photoluminescence signals are formed by superposition of the characteristic emission of chromium atoms, which act as individual piezosensors in definite point of the irradiated layer. Therefore, under non track overlapping regime the spectra will consist of emission from both intact and damaged parts of the target. If Cr atom is located in vicinity of track region, its radiative de-excitation is affected by stress field due to track-associated radiation defects. With increasing of ion fluence, the number of piezosensors in stressed regions will increase, thus enhancing the shift and splitting of the *R*-lines.

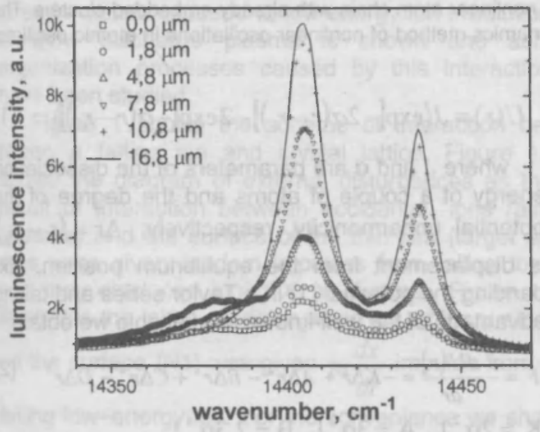

Fig. 3. R-lines spectra as function of depth in ruby specimen irradiated with 710 MeV Bi ions at 80 K. Measurement temperature - 300 K.

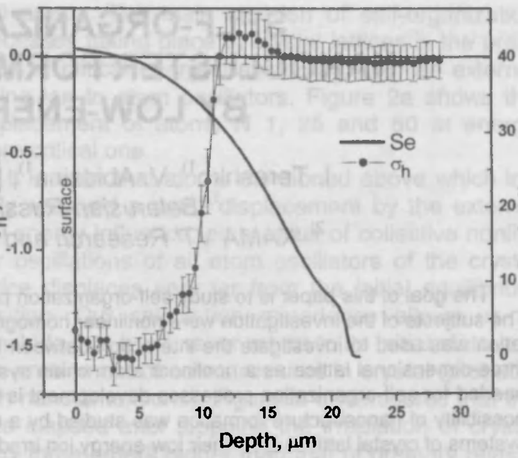

Fig. 4. The stress and ionizing energy loss profiles in ruby Irradiated with 710 MeV Bi ions.

Acknowledgements

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