$S_e$-scaling of lattice parameter change in high ion-velocity region ($v \geq 2.6 \times 10^9$ cm/s) in ion-irradiated EuBa$_2$Cu$_3$O$_y$

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Abstract

Swift heavy ions ($^{35}$Cl–$^{238}$U) with wide energy range of 80 MeV–3.84 GeV have been irradiated to EuBa$_2$Cu$_3$O$_y$ oxide superconducting films, and the lattice parameter change due to electronic excitation has been measured. In the high ion-velocity region ($v \geq 2.6 \times 10^9$ cm/s), the change in crystallographic $c$-axis lattice parameter per unit ion-fluence varies as the 4th power of $S_e$. However, in the low ion-velocity region ($v \leq 1.7 \times 10^9$ cm/s), the deviation from the 4th power dependence is observed. The $S_e$ scaling in the high ion-velocity region cannot be explained by the thermal spike model that is based on a radial distribution of energy deposition by secondary electrons. The change in $c$-axis lattice parameter per unit ion-fluence varies as the 4th power of the primary-ionization rate, $dJ/dx$, in the whole ion-velocity region. The result supports that the Coulomb explosion triggers the atomic displacements. © 2002 Elsevier Science B.V. All rights reserved.

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1. Introduction

In a lot of oxide materials, when irradiated with high-energy heavy ions, continuous linear defects are produced along ion-paths. The defect production is attributed to the high-density electronic excitation. The electronic stopping power, $S_e$, has been considered as a parameter that determines the electronic excitation effects [1–3]. However, in some ion-irradiated oxide materials such as $\text{Y}_3\text{Fe}_5\text{O}_{12}$, although the electronic stopping power is the same, larger electronic excitation effects have been observed for irradiation with slower ions ($v \leq 2.6 \times 10^9$ cm/s) than that with faster ions.
(\(v \geq 3.0 \times 10^9\) cm/s) [4]. This velocity-dependence has been often expressed as the “velocity effect”. In our previous study [5], the velocity effect was also observed in the electrical resistivity change in ion-irradiated superconductors EuBa\(_2\)Cu\(_3\)O\(_y\) (EBCO).

The electronic stopping power is a total energy density transferred to electron system, but it does not necessarily mean that all of the transferred energy is used for atomic displacements. The transferred energy can be divided into two parts; the energy that is carried as the kinetic energy of secondary electrons and the energy that is stored as the electrostatic energy of target atoms ionized by passing ions. One of the thermal spike models assumes that former part of the energy is a cause of local melting of lattice which results in production of defects [4,6–8]. On the other hand, the Coulomb explosion model assumes that the latter part of the energy transforms to the kinetic energy of ionized atoms. There is still a controversy on which part of the energy triggers target atoms to move. It is expected that a careful investigation of the velocity effect gives us a clue to solve this problem. In this study, a detailed analysis of the velocity effect is done by performing ion-irradiation with a wide range of ion energy (80 MeV–3.84 GeV) and a wide range of ion mass (\(^{35}\)Cl, \(^{58}\)Ni, \(^{79}\)Br, \(^{127}\)I, \(^{132}\)Xe, \(^{136}\)Xe, \(^{181}\)Ta, \(^{197}\)Au and \(^{238}\)U).

2. Experimental procedure

Thin films of oxide superconductors EBCO about 300 nm thick were prepared on MgO substrates by dc magnetron sputtering method. The films were c-axis oriented; the c-axis direction corresponded to the direction of film thickness. Irradiating ions were as follows: 120 MeV \(^{35}\)Cl, 90 MeV \(^{58}\)Ni, 200 MeV \(^{58}\)Ni, 125 MeV \(^{79}\)Br, 80 MeV \(^{127}\)I, 200 MeV \(^{127}\)I, 120 MeV \(^{197}\)Au and 200 MeV \(^{197}\)Au from the tandem accelerator at JAERI-Tokai and 3.54 GeV \(^{136}\)Xe and 3.84 GeV \(^{181}\)Ta ions from the ring cyclotron at RIKEN. In addition to these irradiations, we have recently performed the irradiations with 1.51 GeV \(^{132}\)Xe and 2.71 GeV \(^{238}\)U by using UNILAC accelerator at GSI. When analyzing the data, we used the values of nuclear stopping power, \(S_n\), and the electronic stopping power, \(S_e\), calculated by using the SRIM-98 computer code [9]. All the irradiations were performed from the direction of film thickness at room temperature. As the ion ranges for irradiating ions were much larger than the film thickness, a possibility of ion-implantation can be ruled out. X-ray (CuK\(\alpha\)) diffraction pattern was measured, and peak positions of (001) to (00\(\bar{1}0\)) were used to estimate the change in c-axis lattice parameter as a function of ion-fluence.

3. Results and discussion

Fig. 1 shows the examples of the change in c-axis lattice parameter as a function of ion-fluence. In the figure, \(\Delta c/c_0\) is the change in c-axis lattice parameter normalized by c-axis lattice parameter before irradiation, and it is plotted against ion-fluence, \(\Phi\). A linear increase is observed for all irradiations we have performed. The slope of the increase represents the irradiation effect per
unit ion-fluence. The effect originated from the electronic excitation is represented by 

\[ \frac{\Delta c}{c_0} = \frac{\Phi_{\text{electronic}}}{\Phi_{\text{elastic}}} \]

which is defined as 

\[ \left( \frac{\Delta c}{c_0} \right)_{\text{electronic}} = \left( \frac{\Delta c}{c_0} \right)_{\Phi} - \left( \frac{\Delta c}{c_0} \right)_{\Phi_{\text{elastic}}} \]

where \( \left( \frac{\Delta c}{c_0} \right)_{\Phi_{\text{elastic}}} \) is the effect expected from the elastic displacements [10]. It has already been confirmed that \( \left( \frac{\Delta c}{c_0} \right)_{\Phi_{\text{electronic}}} \) is much larger than \( \left( \frac{\Delta c}{c_0} \right)_{\Phi_{\text{elastic}}} \) for the present irradiations with high energy heavy ions [10].

As shown in Fig. 2(a) the irradiating ions can be classified into two groups; the ions located in the right side of the maximum in the \( S_e \) versus energy curves (the right-side ions) and the ions located in the left side of the maximum (the left-side ions). We can find that all the data for the right-side ion irradiation lie on the solid line in Fig. 2(b) and vary as the 4th power of \( S_e \). It is clear that all of the data for the high ion-velocity ions (\( v \geq 2.6 \times 10^9 \) cm/s) can be scaled with \( S_e \). Interestingly, data point for 90 MeV Ni, that has relatively low ion-velocity but is classified as the right-side ion, also participates in the \( S_e \)-scaling. Therefore, it is better to classify ions into right-side ions and left-side ions, rather than classifying only by their ion-velocities. The new finding from these figures is that the velocity effect is hardly observed, or very small if any, for the irradiations with the right-side ions, while for the irradiations with the left-side ions the prominent velocity effect appears as a deviation from the \( S_e \)-scaling. The clear difference in \( S_e \)-dependence between the right-side ions and the left-side ions gives us a criterion to judge which model is valid for explaining the defect production through the electronic excitation.

Since \( S_e \) is merely a line density of energy transferred from incident ions to electron system, there have been trials to explain the velocity effect by taking account of the radial distribution of energy carried by secondary electrons [4,6–8]. Based on this idea, thermal spike model has often been developed assuming that the energy carried by secondary electrons causes a rapid temperature increase of lattice system through electron-phonon coupling. Fig. 3 shows the example of the initial radial energy distribution calculated by the analytical formula developed by Waligórski et al. [11]. This figure indicates that slower ions deposit certain fraction of energy carried by the secondary electrons within a narrower radial distance. This means that higher volume density of energy deposition is expected for irradiation with slower ions, even when \( S_e \) is the same. This seems to ex-
plain the velocity effect. But, it also means that the velocity effect should be observed not only in the low ion-velocity region \( (v \leq 1.7 \times 10^9 \text{ cm/s}) \) but also in the high ion-velocity region \( (v \geq 2.6 \times 10^9 \text{ cm/s}) \). However, our results show that the velocity effect is absent in the high ion-velocity region. Therefore, the thermal spike model that is based on the radial energy distribution is ruled out for explaining the electronic excitation effect in ion-irradiated oxide superconductors.

A practical scaling parameter, we believe, to explain the present result is the primary ionization rate \[ \frac{dJ}{dx} \], where \( J \) is the number of atoms primarily ionized by an incident ion, and \( x \) the path length of the incident ion. Fig. 4 shows the electronic excitation effect plotted against \( \frac{dJ}{dx} \). The electronic excitation effect is scaled with \( \frac{dJ}{dx} \) over all ion-velocity regions. The reason why \( S_e \)-scaling of the electronic excitation effect is observed in the high ion-velocity region is merely because \( S_e \) has a similar ion-velocity dependence to \( \frac{dJ}{dx} \) in the high ion-velocity region. Since the primary ionization rate is a number density of ionized atoms, we conclude that the energy stored as the electrostatic energy rather than the energy carried by secondary electrons is the origin of the electronic excitation effects in the ion-irradiated superconductors. This result is consistent with the previous study [5] of \( \frac{dJ}{dx} \)-scaling of change in electrical resistivity due to the electronic excitation in ion-irradiated EBCO. There are several possible interpretations [15,16] for the \( (\frac{dJ}{dx})^4 \)-dependence. But, since this is not the focal point of this paper, the origin of this interesting behavior will be discussed elsewhere.

4. Summary

Change in \( c \)-axis lattice parameter due to electronic excitation is measured as a function of ion-fluence in EuBa2Cu3Ox oxide superconductor films irradiated with swift heavy ions \( (^{35}\text{Cl–}^{238}\text{U}) \) with wide energy range of 80 MeV–3.84 GeV. In the region of high ion-velocity \( (v \geq 2.6 \times 10^9 \text{ cm/s}) \), the electronic excitation effect is scaled with \( S_e \), and the power-law behavior is observed. However, in the region of low ion-velocity \( (v \leq 1.7 \times 10^9 \text{ cm/s}) \), the deviation from the power-law dependence is observed. The difference in \( S_e \)-dependence between the high ion-velocity ions and the low ion-velocity ions cannot be explained by the radial distribution...
of energy carried by secondary electrons. The electronic excitation effect is scaled with $dJ/dx$ in the whole ion-velocity region. The present result shows that Coulomb explosion triggers the atomic displacements in oxide superconductors.

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**References**