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Ion-velocity dependence of high-density electronic excitation effects in oxide superconductors

N. Ishikawa [a,*], Y. Chimi [a], O. Michikami [b], T. Hashimoto [b], T. Kambara [c], R. Neumann [d], A. Iwase [e]

[a] *Department of Materials Science, Japan Atomic Energy Research Institute, Tokai Establishment (JAERI-Tokai), Tokai-mura, Ibaraki 319-1195, Japan*
[b] *Iwate University, Morioka, Iwate 020-0066, Japan*
[c] *The Institute of Physical and Chemical Research (RIKEN), Wako, Saitama 351-0198, Japan*
[d] *Gesellschaft für Schwerionenforschung (GSI), Planckstrasse 1, D-64291 Darmstadt, Germany*
[e] *Osaka Prefecture University, Gakuen-cho, Sakai-shi, Osaka 599-8570, Japan*

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Abstract

Thin films of $EuBa_2Cu_3O_y$ oxide superconductor have been irradiated with high energy heavy ions (80 MeV I, 125 MeV Br, 1.1 GeV Mo and 3.5 GeV Xe) having same electronic stopping power, S_e, in order to investigate the ion-velocity dependence of the electronic excitation effects under the constant electronic energy deposition. Although S_e is constant, a strong reduction in the irradiation effect on lattice parameter with increasing ion-velocity is observed in the low ion-velocity region around $E \sim 1$ MeV/nucleon, while the ion-velocity dependence is hardly observed in the high ion-velocity region of $E > 10$ MeV/nucleon. If the observed velocity-dependence is assumed to be due to the change in the fraction of S_e contributing to defect creation, the fraction in the low velocity region ($E \sim 0.6$ MeV/nucleon) is estimated to be about two times larger than that in the high velocity region ($E > 10$ MeV/nucleon).

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Keywords: Ion-irradiation; Electronic excitation effect; Oxide superconductor; Coulomb explosion; Thermal spike

1. Introduction

It is established that atoms can be displaced as a result of high-density electronic excitation in a lot of materials irradiated with high energy heavy ions. The effects of high-density electronic

* Corresponding author. Tel.: +81 29 282 5472; fax: +81 29 282 6716.
E-mail address: ishikawa@popsvr.tokai.jaeri.go.jp (N. Ishikawa).

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excitation depend on target materials and the various aspects of the electronic excitation effects have been studied extensively; track formation [1], lattice expansion [2–4], phase transition [5–7], swelling [8] and so on. The electronic stopping power, S_e, is defined as a linear density of electronic energy deposition and has been considered as an important parameter that determines the electronic excitation effects. However, in some ion-irradiated oxide materials such as $Y_3Fe_5O_{12}$ (YIG), although S_e is the same, smaller track diameter has been observed for high ion-velocity regime (E = 5–38.7 MeV/nucleon) than for low ion-velocity regime (E = 0.85–3.6 MeV/nucleon) [9]. The velocity dependence observed for the electronic excitation effects is often called "*the velocity effect*". The velocity effect has been observed also for the lattice expansion [3,4] and the electrical resistivity change [10] in oxide superconductors. However, there is little number of studies focusing on the ion-velocity dependence of the electronic excitation effects. It should be noted that S_e just defines a linear density of initial energy deposition to electron system. The appearance of the velocity effect under constant S_e is probably related to the radial distribution of initial energy deposition and/or the energy sharing within the electron system. In this sense, the precise investigation of the velocity effect is important especially for understanding the initial stage of the energy deposition.

As far as we know, the only studies focusing on the precise velocity dependence is those of Szenes [11,12]. He has analyzed the ion-velocity dependence of efficiency, g, for YIG and $LiNbO_3$, where g is defined as the fraction of S_e covering the thermal energy which contributes to track formation. He has assumed that the velocity effect appears as the result of change in efficiency g which varies as a function of ion-velocity. The efficiency is defined based on the thermal spike concept which assumes that a part of S_e contributes to a temperature increase leading to lattice melting. Here, it should be noted that the efficiency g is estimated by assuming a thermal spike concept and it is not the direct consequence of track diameter data. In this study, we have investigated the ion-velocity dependence of lattice expansion by performing several ion irradiations having same S_e but different ion-velocity and we have tried to estimate by another approach the ion-velocity dependence of the fraction of S_e contributing to the defect creation.

2. Experimental procedure

Thin films of oxide superconductors $EuBa_2Cu_3O_y$ (EBCO) which have about 0.3 μm thickness were prepared on MgO substrates by the dc magnetron sputtering method. The films were c-axis oriented; the c-axis direction corresponded to the direction of film thickness. As-sputtered films were expected to have an oxygen content of y = 7, since superconducting transition temperature for these films was around 89 K. The irradiations with 80 MeV I, 125 MeV Br, 1.1 GeV Mo and 3.5 GeV Xe ions were performed at room temperature. All of the irradiations have almost the same S_e (S_e = 26.2–28.5 (MeV/(mg/cm^2))) as demonstrated in Fig. 1. The electronic stopping power is estimated by using SRIM2003 [13,14]. As long

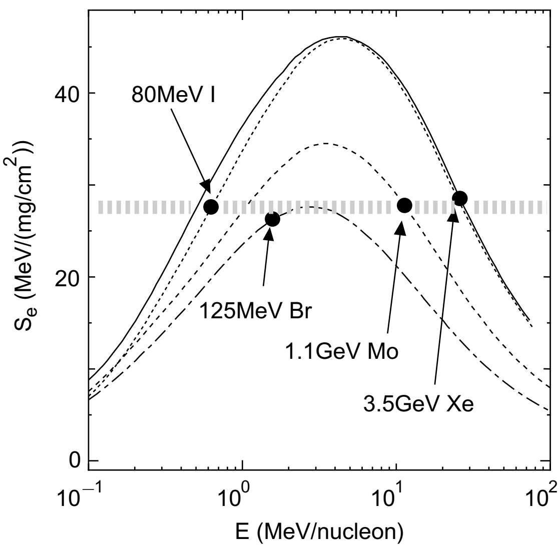

Fig. 1. Electronic stopping power, S_e, plotted as a function of energy. The electronic stopping power is estimated using SRIM2003 [13,14]. The S_e values for the present irradiations are indicated by full circles. The gray dotted line is to demonstrate that values of S_e for the present irradiations are almost the same.

Table 1
Irradiation parameters for the present study

Ion	E (MeV/nuleon)	S_e (MeV/(mg/cm^2))	S_n (MeV/(mg/cm^2))	Projected range (μm)
80 MeV I	0.6	27.6	3.2×10^{-1}	8
125 MeV Br	1.6	26.2	6.9×10^{-2}	11
1.1 GeV Mo	11.4	27.7	1.9×10^{-2}	54
3.5 GeV Xe	26.0	28.5	1.4×10^{-2}	142

The values of S_e, S_n and the projected range are estimated using SRIM2003 [13,14].

as single element target is concerned, SRIM2003 program can calculate the stopping power with the average accuracy of 6% for heavy ions [15]. The additional uncertainty for S_e estimation emerges by adopting the Bragg's additivity rule. But it is already demonstrated that for many of heavy compounds containing elements with atomic numbers greater than 12 there is no measurable deviations from Bragg's rule [14]. Since the present compound ($EuBa_2Cu_3O_7$) contains heavy elements, we believe the error related to Bragg's rule is very small.

The energy ranges from 0.6 to 26 MeV/nucleon. The irradiation parameters are listed in Table 1. The irradiations with 80 MeV I and 125 MeV Br ions were performed by using the tandem accelerator at JAERI-Tokai. The irradiation with 1.1 GeV Mo was performed by using the UNILAC accelerator at GSI and the irradiation with 3.5 GeV Xe ions by using the ring cyclotron at RIKEN. As all ion ranges were much larger than the film thickness, a possibility of ion-implantation can be ruled out. X-ray (Cu Kα) diffraction pattern was measured before and after irradiation. Fluence dependence of c-axis lattice parameter was investigated.

3. Results and discussion

The fluence dependence of c-axis lattice parameter is shown in Fig. 2. Here $\Delta c/c_0$ is the change in c-axis lattice parameter, Δc, normalized by the c-axis lattice parameter before irradiation, c_0. Linear increase in c-axis lattice parameter is observed for all the irradiations suggesting that each ion create defects that contribute to lattice expansion. The slope of lattice expansion is defined as $(\Delta c/c_0)/\Phi$, where Φ is the fluence. The error of the absolute value of the fluence has been confirmed to be typically within about 20% by TEM observation. We believe the relative error between each fluence value for the same beamtime is very small. We assume here that the lattice expansion is attributed to the sum of defect creation via elastic displacements and that via electronic excitation. The contribution of electronic excitation can be estimated to be $((\Delta c/c_0)/\Phi)_{\text{electronic}} = (\Delta c/c_0)/\Phi - ((\Delta c/c_0)/\Phi)_{\text{elastic}}$, where $((\Delta c/c_0)/\Phi)_{\text{elastic}}$ is the contribution of elastic displacements which is estimated from the previous irradiation experiment using ~1 MeV ions [2]. In the case of the present irradiations, we find that $((\Delta c/c_0)/\Phi)_{\text{elastic}}$ is only 1.0–1.3% of $(\Delta c/c_0)/\Phi$ and that $((\Delta c/c_0)/\Phi)_{\text{electronic}}$ is almost the same as $(\Delta c/c_0)/\Phi$.

The energy dependence of $((\Delta c/c_0)/\Phi)_{\text{electronic}}$ is plotted in Fig. 3. Since the energy is indicated in the unit of MeV/nucleon, it is proportional to the square of ion-velocity. Although S_e is constant, the lattice expansion per unit fluence decreases rapidly as a function of ion-velocity especially in the low velocity region of around $E \sim 1$ MeV/nucleon. In the high-velocity region of around $E > 10$ MeV/nucleon, the velocity dependence is absent or very small, if any.

We have already demonstrated that $((\Delta c/c_0)/\Phi)_{\text{electronic}}$ is scaled by S_e in the high ion-velocity region of $E > 3.4$ MeV/nucleon [4] as shown in Fig. 4 in which recent data is added. The recent data of 1.1 GeV Mo irradiation also lie on the scaling line. The present result confirms that there is hardly ion-velocity dependence of electronic excitation effect in high ion-velocity region. An interesting characteristics found by the figure is that $((\Delta c/c_0)/\Phi)_{\text{electronic}}$ is proportional to S_e^4 for the high ion-velocity region. The origin of the power

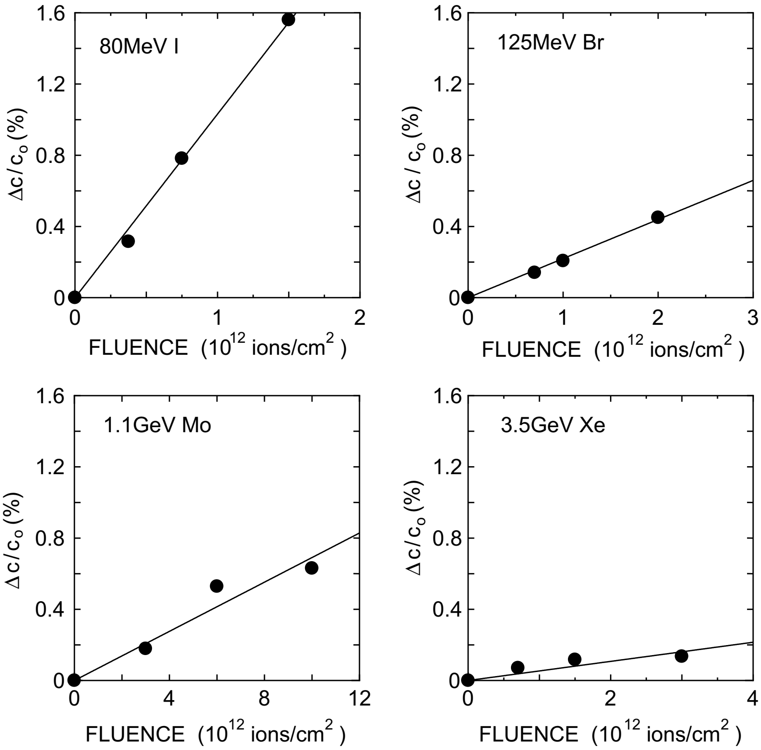

Fig. 2. Fluence dependence of normalized lattice expansion, $\Delta c/c_0$, for the irradiations having same S_e and different ion-velocity. The solid lines are to demonstrate the c-axis lattice parameter increases linearly with increasing ion-fluence.

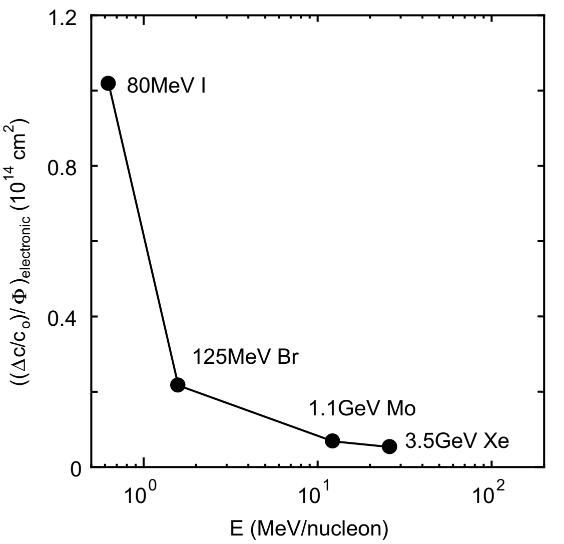

Fig. 3. Contribution of electronic excitation to lattice expansion per unit fluence, $((\Delta c/c_0)/\Phi)_{electronic}$, plotted against energy per nucleon.

law is still unknown, but it can be explained [16] by the combination of kinetic energy due to Coulomb repulsion between ionized atoms [17,18] and the atomic displacements due to thermal energy [19,20] converted from the kinetic energy of atoms. Here we make a following assumption; $((\Delta c/c_0)/\Phi)_{electronic}$ is always proportional to $(kS_e)^4$, where efficiency k is the fraction of S_e contributing to the defect creation or lattice expansion. We believe this assumption is natural because $((\Delta c/c_0)/\Phi)_{electronic}$ is proportional to S_e^4 in the high ion-velocity region and because the velocity effect appeared in the low ion-velocity region is probably due to the change in efficiency k. From this assumption, efficiency k can be estimated as $k \sim (C_e/S_e^4)^{1/4}$, where C_e is $((\Delta c/c_0)/\Phi)_{electronic}$. Of course, this assumption is influenced by the validity of the S_e^4-scaling mentioned above and it depends on the uncertainty of S_e calculated by SRIM2003 and the experimental error of lattice

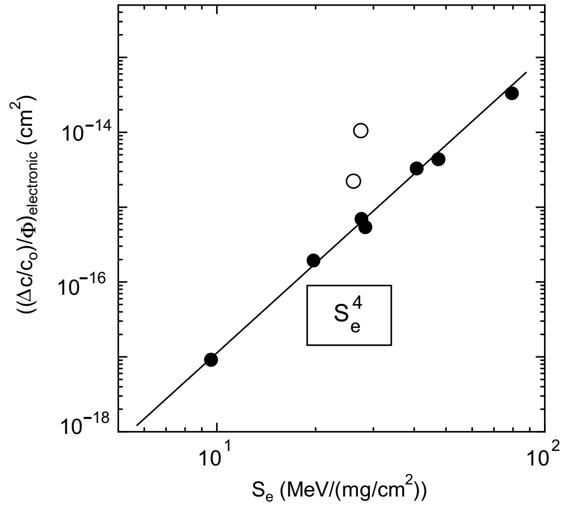

Fig. 4. S_e dependence of $((\Delta c/c_0)/\Phi)_{electronic}$ for the irradiations with high-velocity ions with the energy of $E > 3.4$ MeV/nucleon (closed circles). Most of the data are quoted from [4]. A solid line demonstrates that $((\Delta c/c_0)/\Phi)_{electronic}$ is proportional to S_e^4 [4]. In the figure, the result for the irradiations with 80 MeV I ($E = 0.6$ MeV/nucleon) and 125 MeV Br($E = 1.6$ MeV/nucleon) is also presented (open circles), demonstrating that the data for the low-velocity ions do not participate in the S_e^4-scaling, while for the irradiations with 1.1 GeV Mo ions($E = 11.4$ MeV/nucleon) and 3.5 GeV Xe($E = 26.0$ MeV/nucleon) the data participate in the S_e^4-scaling.

expansion. The uncertainty of SRIM calculation is discussed in the previous section. The experimental error of lattice expansion measurement is discussed below. The maximum error may be caused by a sample dependence, although the samples are prepared with the same experimental procedure. The quantification of this error is very difficult, but the independently performed irradiation experiment is helpful to infer the error. We have prepared two kinds of samples with different thicknesses (300 nm and 1000 nm) and the difference in lattice expansion per unit fluence is estimated for these samples respectively for the irradiations with 120 MeV Au, 200 MeV I and 200 MeV Ni ions. The lattice expansion per unit fluence for 300 nm sample exhibit 13%, 18% and 25% difference compared with that for 1000 nm sample for 120 MeV Au, 200 MeV I and 200 MeV Ni respectively. The maximum error was 25%, when the sample thickness is varied from 300 nm to 1000 nm. Since the present experiment adopts only the samples with 300 nm thickness, we believe the error due to sample dependence *for the present study* is even smaller. Furthermore, since all of the samples for one beamtime are cut from a same batch of the sample, the error due to sample dependence is further reduced. In addition, the small error for one beamtime is already demonstrated by the linearity of the lattice expansion as a function of ion-fluence. In conclusion, the error due to sample dependence is expected to be comparable to the size of closed circles in Fig. 4. Here we are not going to discuss the physical implication of fourth power which is still unknown, but we use this power law just for the estimation of the efficiency.

Fig. 5 shows the velocity dependence of the efficiency k which is estimated by $k \sim (C_e/S_e^4)^{1/4}$. The efficiency for low ion-velocity ions ($E = 0.6$ MeV/nucleon) is 2.2 ± 0.1 times higher than that for high velocity ions ($E = 26$ MeV/nucleon) when the errors of S_e and the lattice expansion are taken into account. It is interesting to note that the ratio of the efficiency which is independently estimated by Szenes [11,12] from the thermal spike concept

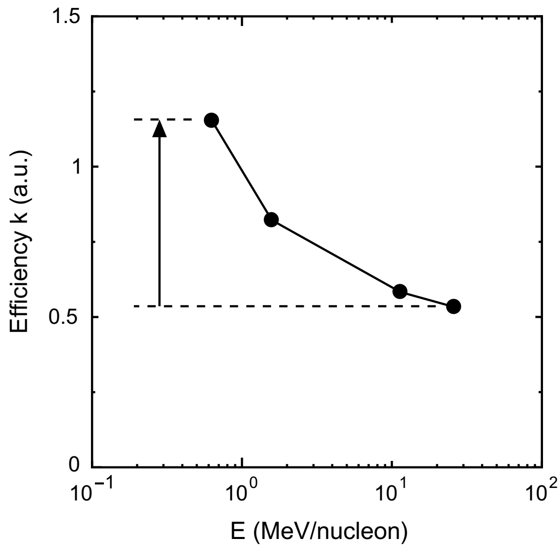

Fig. 5. Efficiency plotted against energy per nucleon. Here the efficiency is defined as $k = (C_e/S_e^4)^{1/4}$, where $C_e = ((\Delta c/c_0)/\Phi)_{electronic}$. An arrow is to show that the efficiency for $E = 0.6$ MeV/nucleon is 2.2 times higher than that for $E = 26$ MeV/nucleon.

is twice which is close to our result. The present result does not directly support any defect creation model, but we believe that the present result of the velocity dependence of the electronic excitation effect or the efficiency can be one of the important criteria for judging the validity of the models accounting for the electronic excitation effects.

4. Summary

Thin films of $EuBa_2Cu_3O_y$ oxide superconductors have been irradiated with high energy heavy ions (80 MeV I, 125 MeV Br, 1.1 GeV Mo and 3.5 GeV Xe) under the condition of constant S_e and different ion-velocities in order to investigate the ion-velocity dependence of efficiency of S_e to create lattice defects. A strong reduction in the electronic excitation effect with increasing ion-velocity is observed around $E \sim 1$ MeV/nucleon, although S_e is constant, while in the high velocity region of $E > 10$ MeV/nucleon the velocity dependence is hardly observed. By assuming that the velocity effect is caused by the change in the fraction of S_e contributing to defect creation, the fraction for the low velocity ions ($E = 0.6$ MeV/nucleon) is found to be about two times larger than that for the high velocity ions ($E > 10$ MeV/nucleon).

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