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Electronic excitation effects in ion-irradiated high-T_c superconductors

N. Ishikawa [a,*], Y. Chimi [a], A. Iwase [a], H. Maeta [b], K. Tsuru [c,1], O. Michikami [d], T. Kambara [e], T. Mitamura [f], Y. Awaya [e,1], M. Terasawa [f]

[a] Advanced Science Research Center, Japan Atomic Energy Research Institute, Tokai-mura, Ibaraki-ken 319-11, Japan
[b] Department of Materials Science and Engineering, Japan Atomic Energy Research Institute, Tokai-mura, Ibaraki-ken 319-11, Japan
[c] Device Physics Research Laboratory, NTT Basic Research Laboratories, Tokai-mura, Ibaraki-ken 319-11, Japan
[d] Faculty of Engineering, Iwate University, Morioka-shi, Iwate-ken 020, Japan
[e] Atomic Physics Laboratory, The Institute of Physical and Chemical Research (RIKEN), Wako-shi, Saitama-ken 351-01, Japan
[f] Faculty of Engineering, Himeji Institute of Technology, Himeji-shi, Hyogo-ken 671-22, Japan

Abstract

We have measured the fluence dependence of the *c*-axis lattice parameter in $EuBa_2Cu_3O_y$ (EBCO) irradiated with various ions from He to Au over the wide energy range from 0.85 MeV to 3.80 GeV. We have observed a linear increase of the *c*-axis lattice parameter with increasing fluence for all irradiations. The slope of *c*-axis lattice parameter against fluence, which corresponds to the defect production rate, is separated into two contributions; the effect via elastic displacement and the effect via electronic excitation. The former contribution exhibits a linear increase against the nuclear stopping power, S_n. The latter contribution is scaled by the primary ionization rate, dJ/dx, rather than by the electronic stopping power, S_e, and is nearly proportional to $(\mathrm{d}J/\mathrm{d}x)^4$. © 1998 Elsevier Science B.V.

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1. Introduction

Irradiation effects on oxide superconductors have been studied extensively [1], as the introduction of lattice defects is a useful method to increase the critical current density (J_c). Especially, columnar defects introduced by irradiation with high energy heavy ions have drawn attention for their effectiveness in increasing J_c. Since columnar defects are expected to be created due to electronic excitation induced by ion-irradiation, it has been considered that electronic stopping power, S_e, may be an important parameter in describing the production of columnar defects. Several works

* Corresponding author. E-mail: ishikawa@popsvr.tokai.jaeri.go.jp.

[1] Present address: NTT Integrated Information & Energy Systems Laboratories, Musashino-shi, Tokyo 180, Japan.

[2] Present Address: Musashino Art University, 1-726, Ogawa-cho, Kodairashi Tokyo 187, Japan.

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[2–6] have shown the relation between the diameter of columnar defects and S_e, but its physical meaning has not been clarified yet for high-T_c superconductors. For other oxide materials such as $Y_3Fe_5O_{12}$, when the value of S_e is fixed, the lower ion-velocity results in a larger diameter columnar tracks [6]. The velocity dependence of irradiation effect is often called the "velocity effect" [7]. For high-T_c superconductors, however, the velocity effect has not been clearly observed [6]. It is necessary to clarify whether the velocity effect can be observed and to study which defect production mechanism most consistently explains the irradiation effects in ion-irradiated high-T_c superconductors.

In our previous work [8], we suggested that $(\Delta c/c_0)/\Phi$ is a good quantity to investigate the defect production rate, where Δc is the increment of c-axis lattice parameter, c_0 the c-axis lattice parameter for the sample before irradiation and Φ the fluence. We showed that the defect production rate consists of two contributions; one is the effect due to elastic displacement and the other is the effect due to electronic excitation. When S_e is high enough, the former effect can be neglected. However, if the both effects are comparable, separation of the two becomes necessary for the accurate estimation of both effects. In this study, we have performed irradiation of the $EuBa_2Cu_3O_y$ (EBCO) superconductor with various ions from He to Au changing the energy from 0.85 MeV to 3.80 GeV and measured the value of $(\Delta c/c_0)/\Phi$. For low energy ($\leqslant 2$ MeV) ion irradiation, $(\Delta c/c_0)/\Phi$, which corresponds to the defect production rate, is proportional to the nuclear stopping power, S_n. On the other hand, for high energy (80 MeV–3.80 GeV) ion irradiation, the contribution of electronic excitation to $(\Delta c/c_0)/\Phi$ value is precisely estimated by assuming that the observed $(\Delta c/c_0)/\Phi$ value is the sum of the contribution of elastic displacement and that of electronic excitation. The latter contribution depends not only on the electronic stopping power, S_e, but also on the ion velocity, exhibiting the velocity effect. In order to explain this velocity effect, we propose the adoption of the primary ionization rate, dJ/dx, as a parameter replacing S_e to consistently describe all the data. The contribution of electronic excitation of the $(\Delta c/c_0)/\Phi$ value is found to be proportional to $(dJ/dx)^n$, where n is nearly 4.

2. Experimental procedure

The films of EBCO oxide superconductor were prepared by RF magnetron sputtering [9,10]. The thickness of the films was about 300 nm. The superconducting transition temperature, T_c, ranged from 80 to 89 K. We performed the irradiation with low energy ($\leqslant 2$ MeV) ions and with high energy (80 MeV–3.80 GeV) ions. The irradiating ions and energies for low energy ion irradiation were as follows; 0.85 MeV ^{4}He, 1.0 MeV ^{12}C, 0.95 MeV ^{20}Ne, and 2 MeV ^{40}Ar from 2 MV van de Graaff accelerator at JAERI-Tokai. The irradiating ions and energies for high energy ion irradiations were as follows; 120 MeV ^{35}Cl, 90 MeV and 200 MeV ^{58}Ni, 125 MeV ^{79}Br, 80 MeV and 200 MeV ^{127}I, 120 MeV ^{197}Au from the tandem accelerator at JAERI-Tokai, and 3.54 GeV ^{136}Xe and 3.80 GeV ^{181}Ta from the ring cyclotron at RIKEN. Samples were irradiated parallel to the c-axis at room temperature and under vacuum. We measured the X-ray (Cu Kα) diffraction pattern both before and after the irradations. The peak positions of (0 0 1) to $(0\,0\,\underline{10})$ were used to estimate the c-axis lattice parameter. The c-axis lattice parameter for sample before irradiation, c_0, ranged from 11.73 to 11.76 Å. When analyzing the data, we used the values of nuclear stopping power (S_n) and electronic stopping power (S_e) calculated using the TRIM-92 computer code [11]. The calculated results are listed in Table 1. In the table are also shown the ion-velocity and the projected range. By using the ions listed in the table we can investigate the effect of various combination of S_e and ion-velocity on the defect production in EBCO. All of the values of the projected range are much larger than the film thickness (0.3 μm). This means that defects are uniformly introduced along the sample thickness, and we can exclude the effect of ion implantation.

3. Results and discussion

Fig. 1(a)–(c) shows $\Delta c/c_0$ plotted as a function of ion fluence Φ for high energy ion irradiations. The figures show that all irradiations exhibit a linear Φ-dependence of $\Delta c/c_0$. Assuming that $\Delta c/c_0$

Table 1
Irradiation parameters. An ideal value of sample density for EBCO, 6.9 g/cm^3, is adopted to calculate the ion range

Energy (MeV)	Ion	amu	Velocity (10^9 cm/s)	S_n (MeV/(mg/cm^2))	S_e (MeV/(mg/cm^2))	Range (μm)
0.85	He	4	0.41	1.4×10^{-3}	0.81	1.8
1.0	C	12	0.16	2.4×10^{-2}	2.3	0.9
0.95	Ne	20	0.09	9.3×10^{-2}	2.3	0.7
2.0	Ar	40	0.10	2.7×10^{-1}	3.9	0.9
120	Cl	35	2.57	8.9×10^{-3}	10	17
90	Ni	58	1.73	4.6×10^{-2}	22	9
200	Ni	58	2.58	2.3×10^{-2}	20	16
125	Br	79	1.75	6.9×10^{-2}	27	10
80	I	127	1.10	3.2×10^{-1}	27	7
200	I	127	1.74	1.5×10^{-1}	39	13
3540	Xe	136	7.09	1.4×10^{-2}	27	148
3800	Ta	181	6.37	3.1×10^{-2}	48	106
120	Au	197	1.08	6.8×10^{-1}	37	8

value is proportional to the concentration of irradiation induced defects, we can regard that $(\Delta c/c_0)/\Phi$ is proportional to the defect production rate. It should be noted that a linear Φ-dependence of $\Delta c/c_0$ is also observed for EBCO irradiated with low energy ($\leqslant 2$ MeV) ion. As can be seen in Fig. 2, $(\Delta c/c_0)/\Phi$ for low energy ion irradiation is a linear function of S_n [8], i.e.,

$$(\Delta c/c_0)\Phi = aS_n, \tag{1}$$

where $a = 3 \times 10^{16}$ mg/MeV. This means that the defect production rate is proportional to the energy transferred from the ion to the sample through elastic collisions. Therefore, we can conclude that elastic displacement is the dominant process of defect production for low energy ($\leqslant 2$ MeV) ion irradiation. This is consistent with other experiments showing a linear S_n-dependence of $dT_c/d\Phi$ [12] and change rate of normal-state resistance [13]. As the S_e value become higher, the effect of electronic excitation becomes prominent. Fig. 3 shows $(\Delta c/c_0)/\Phi$ for high energy ion irradiation as a function of S_n. The results for low energy ion irradiations are also shown as a dotted line. All of the $(\Delta c/c_0)/\Phi$ values for high energy ion irradiation are higher than those for low energy ion irradiation, implying the existence of the contribution of electronic excitation to $(\Delta c/c_0)/\Phi$ value. As elastic collision and electronic excitation occur independently in the sample, the observed $(\Delta c/c_0)/\Phi$ value can be expressed by

$$(\Delta c/c_0)/\Phi = ((\Delta c/c_0)\Phi)_{\text{elastic}} + ((\Delta c/c_0)/\Phi)_{\text{electronic}}. \tag{2}$$

Since the former effect is expressed by Eq. (1), we can readily estimate $((\Delta c/c_0)/\Phi)_{\text{electronic}}$ by subtracting aS_n from the observed $(\Delta c/c_0)/\Phi$ value. In Fig. 3, $((\Delta c/c_0)/\Phi)_{\text{electronic}}$ is indicated by arrows. As a first step in the analysis, we focused on the S_e-dependence of $((\Delta c/c_0)/\Phi)_{\text{electronic}}$ which is shown in Fig. 4. This figure shows that $((\Delta c/c_0)/\Phi)_{\text{electronic}}$ depends not only on S_e but also on the ion-velocity. When S_e is fixed to, for example, 27 MeV/(mg/cm^2), we can find $((\Delta c/c_0)/\Phi)_{\text{electronic}}$ increases as ion-velocity decreases, indicating the presence of the velocity effect. Since S_e alone can not account for the observed value of $((\Delta c/c_0)/\Phi)_{\text{electronic}}$, it is necessary to introduce a quantity replacing S_e in order to account for the velocity effect. As one of the available parameters which can explain the present results, we propose the adoption of primary ionization rate, dJ/dx, where J is the number of ions induced along the projectile trajectory, and x the length of projectile path. The introduction of this quantity is based on the idea that the number of ionized atoms may be responsible for defect production through Coloumb explosion process. Fleischer et al. [14] have shown that the track-registration threshold in some insulators, above which track are observed, can be defined by using dJ/dx and not S_e. They have estimated the value of dJ/dx based on the Bethe formula [15]:

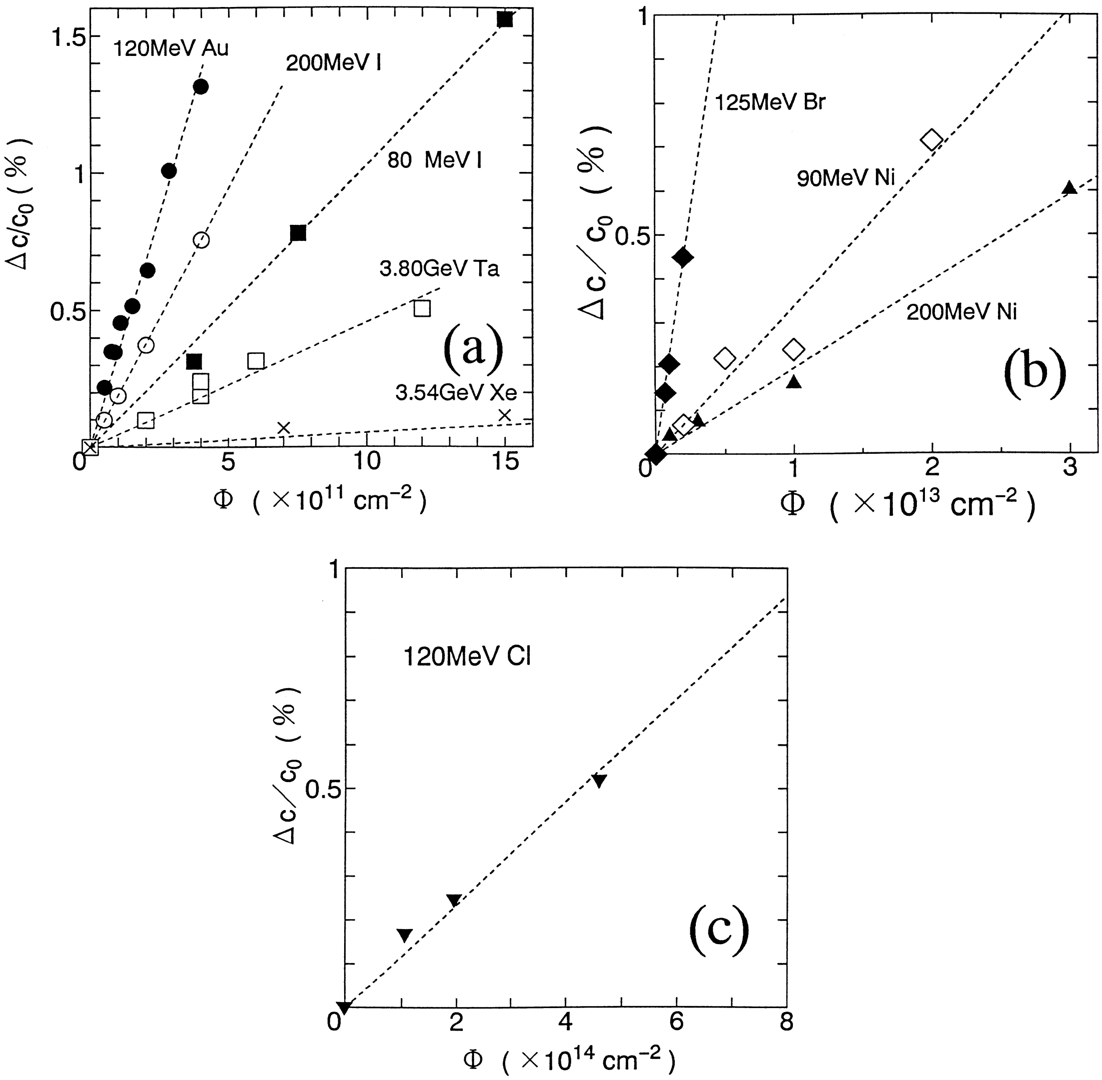

Fig. 1. The $\Delta c/c_0$ values as a function of fluence for EBCO irradiated with high energy ions.

$$dJ/dx = \sigma N = (\alpha Z^{*2}/I_0\beta^2)[\ln\{2mc^2\beta^2/(1-\beta^2)I_0\} - \beta^2 + 3.04], \quad (3)$$

where σ is the ionization cross section, N the number density of target atoms, Z^* the equilibrium charge of the irradiating ion in the target [16], β the ion-velocity divided by the velocity of the light, c, I_0 the ionization energy of most loosely bound electron of a target material, and m the electron mass. Eq. (3) was originally constructed to account for the ionization cross section of hydrogen atoms irradiated with electrons, where $Z^* = 1$. As the possibility of the excitation of the most loosely bound electrons is much higher than that for all other electrons, the target atoms can be treated as hydrogen-like, and Eq. (3) can be used approximately for the heavy ion irradiations of EBCO. In Eq. (3), the parameters that depend on target materials are α and I_0. We adopt the value $I_0 = 10$ eV, which is the average value of the first ionization energy for atoms composing EBCO. Since there

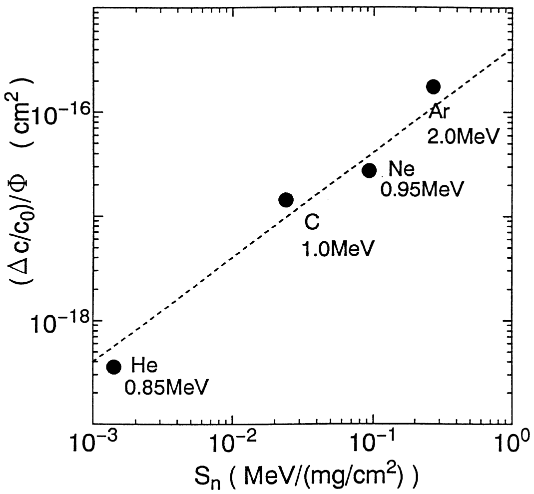

Fig. 2. The values of $(\Delta c/c_0)/\Phi$ as a function of S_n for EBCO irradiated with low energy ions. The straight dotted line is a linear function of S_n. (see Ref. [8]).

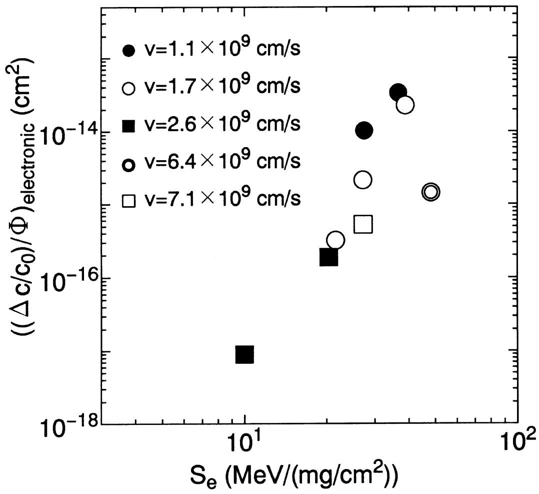

Fig. 4. $(\Delta c/c_0)/\Phi_{\mathrm{electronic}}$ as a function of S_e. The velocities of the incident ions are shown for reference.

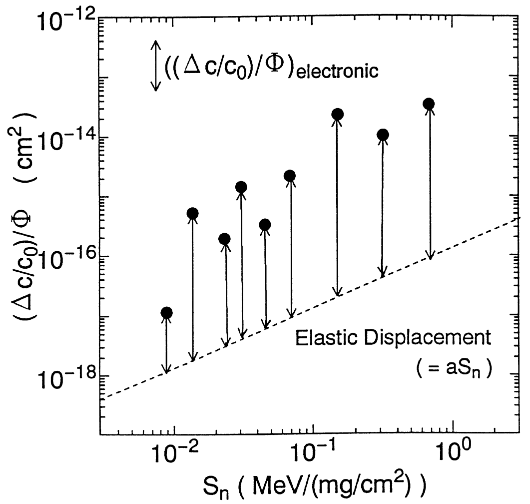

Fig. 3. The $(\Delta c/c_0)/\Phi$ value as a function of S_n for the high energy ion irradiation (solid circles). Data for the low energy ion irradiations are also shown as a dotted line.

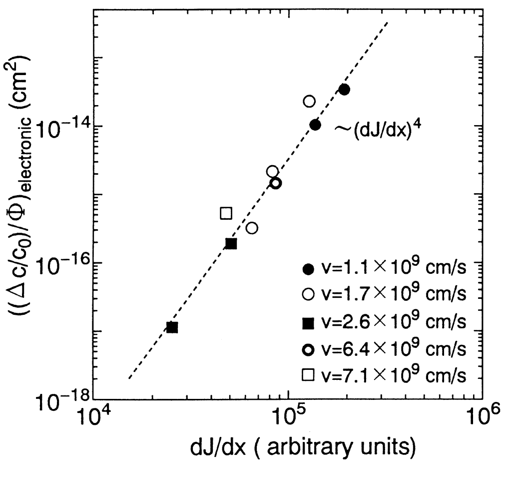

Fig. 5. $((\Delta c/c_0)/\Phi)_{\mathrm{electronic}}$ as a function of $\mathrm{d}J/\mathrm{d}x$. The dotted line shows the line proportional to $(\mathrm{d}J/\mathrm{d}x)^4$. The velocities of the incident ions are shown for reference.

is no means to determine the parameter α, the units of $\mathrm{d}J/\mathrm{d}x$ remain arbitrary. We can show, by using Eq. (3), that our data can be scaled by the single parameter $\mathrm{d}J/\mathrm{d}x$. As can be seen in Fig. 5, $((\Delta c/c_0)/\Phi)_{\mathrm{electronic}}$ varies as $(\mathrm{d}J/\mathrm{d}x)^n$, where n is nearly 4. It should be noted that the velocity effect is inherently involved in the $(\mathrm{d}J/\mathrm{d}x)^4$-dependence. When the deposited energy S_e is fixed, a higher ion-velocity results in lower value of $\mathrm{d}J/\mathrm{d}x$, because a higher ion-velocity leads to δ-ray with higher energy. Since $\mathrm{d}J/\mathrm{d}x$ is the number of atoms ionized per projectile path length and is not the energy deposited to the target, the above figure suggests that the line density of induced space charge is responsible for the defect production in EBCO.

The most plausible process of defect production via primary ionization, we believe, is that ionized atoms receive their recoil energy through the Coulomb explosion mechanism [14]. Consideration of spacial density of deposited energy may be necessary for further analysis. The reason for the $(\mathrm{d}J/\mathrm{d}x)^4$-dependence remains unknown. Nevertheless, it should be mentioned that there have been experiments which have shown that the yield of sputtering due to electronic excitation varies as $(\mathrm{d}J/\mathrm{d}x)^4$ [17,18].

4. Conclusion

We have measured fluence dependence of the c-axis lattice parameter in EBCO irradiated with various ions over the wide energy range from 0.85 MeV to 3.80 GeV. The effect of electronic excitation on c-axis lattice parameter is scaled by the primary ionization rate, $\mathrm{d}J/\mathrm{d}x$. The $(\mathrm{d}J/\mathrm{d}x)^n$-dependence on the defect production rate is observed, where n is nearly 4. This suggests that the density of induced space charge is responsible for the defect production through the Coulomb explosion mechanism.

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