Growth phenomenon in amorphous solids irradiated with GeV heavy ions: Electronic-energy-loss dependence of the initial growth rate

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Electronic excitation $(dE/dx)_e$ due to swift-heavy-ion irradiation induces giant plastic deformation in amorphous solids. *In situ* electrical resistance experiments performed on Fe-B ribbons irradiated with different tilting angles with respect to the ion beam provide quantitative information about the deformation phenomenon. In particular, dimensional variations occurring in the early stages of the process can be evaluated. At medium $(dE/dx)_e$ (irradiation with Xe ions) the growth starts with a zero rate within experimental uncertainties, whereas an initial nonzero rate is observed at high $(dE/dx)_e$ (irradiation with Pb or U ions). Results are discussed in light of a recent model based on irradiation-induced thermal spikes. [S0163-1829(96)00146-4]

I. INTRODUCTION

From the beginning of the 1980s it is known that the electronic slowing down of swift heavy ions, hereafter referred to as $(dE/dx)_e$, induces atomic displacements in metallic targets. For instance, damage creation, phase transformations, and amorphous track formation were reported[1] in pure metals or metallic compounds irradiated with GeV heavy ions. Furthermore, a dramatic effect due to $(dE/dx)_e$, which does not occur in a target bombarded with low-energy ions, was observed in amorphous materials:[2,3] a huge macroscopic and anisotropic deformation of the irradiated sample.

The plastic deformation phenomenon of amorphous solids submitted to severe electronic excitation exhibits the following characteristics: (i) in a large majority of materials there exists an incubation fluence Φ_C; (ii) above Φ_C, the irradiated sample shrinks in the direction parallel to the ion beam and expands in the directions perpendicular to it (without volume change), and the rate of the deformation remains constant up to the highest fluences used in the experiments; (iii) below Φ_C, the growth rate is smaller, and it is often assumed that the growth starts with a zero rate at the beginning of the irradiation though direct proofs are scarce; (iv) it is believed that below Φ_C irradiation leads to short-range order modifications or pointlike defect creation (giving rise to a resistivity increase which saturates at high fluence, as it is the case for low-energy ions[4,5] or electrons[6]); (v) the growth rate strongly depends on the irradiation temperature (it generally decreases as the temperature increases). Most of the features listed above were learned by either the measurement of the dimensions of the sample after irradiation,[2,7,8] or electrical resistance experiments *in situ* throughout irradiation.[9,10] The former technique offers the advantage to provide a direct measure of the effect but presents technical limitations, such as data recorded at scarce fluence values, large relative uncertainties below the incubation fluence. Moreover, these experiments were restricted to maximum $(dE/dx)_e$ values of 35 keV nm^{-1} (irradiation with hundreds MeV Xe ions). Electrical resistance experiments take advantage of the possibility of extracting both resistivity and dimensional variations from the data, provided that samples with different tilt angles with respect to the ion beam direction are irradiated at the same time. Nevertheless, this technique is indirect and requires a few assumptions for the data analysis.

This paper reports an attempt to revisit the growth phenomenon in the light of new electrical resistance results[11] and recent progress in the theoretical description of the process.[12–15] Section II presents a critical discussion of the way used to reduce electrical resistance data recorded in online experiments. In Sec. III an analysis of the results obtained in electrical resistance experiments performed on the irradiated Fe-B system is presented, as well as a discussion about the implications of these results concerning the plastic deformation process.

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II. DATA REDUCTION IN ELECTRICAL RESISTANCE EXPERIMENTS

In a typical electrical resistance experiment performed *in situ* during irradiation, the samples consist of ribbons (length L, width w, thickness t) tilted by an angle θ with respect to the beam direction in the plane parallel to both L and t. The electrical resistance is measured all along irradiation during beam stops with the current flowing along L. If one assumes that the dimensional variations are isotropic in the directions perpendicular to the ion beam and if one restricts the description to small deformations, the relative variation of the three dimensions of the ribbons upon irradiation can be written

$$\Delta L/L = \Delta y/y \ \cos^2\theta + \Delta x/x \ \sin^2\theta,$$

$$\Delta t/t = \Delta y/y \ \sin^2\theta + \Delta x/x \ \cos^2\theta,$$

$$\Delta w/w = \Delta y/y. \tag{1}$$

In these equations $\Delta x/x$ and $\Delta y/y$ represent the relative dimensional variations in the directions, respectively, parallel and perpendicular to the beam axis. In the geometry described above, the relative variation of the ribbon resistance is

$$\frac{\Delta R}{R_0} = R - L(1-3\sin^2\theta), \tag{2}$$

where

$$R = \Delta\rho/\rho_0 - \Delta v/3v_0, \tag{3}$$

$$L = \Delta x/x - \Delta v/3v_0. \tag{4}$$

In Eqs. (3) and (4) $\Delta\rho/\rho_0$ and $\Delta v/v_0$ are the resistivity and volume variations, respectively. By assuming that the volume of the sample remains constant during irradiation,[16] Eq. (2) can be reduced to the usual expression[9,10]

$$\Delta R/R_0 = \Delta\rho/\rho_0 - (1-3\sin^2\theta)\Delta x/x. \tag{5}$$

Equation (5) allows one to discriminate between resistivity and dimensional contributions to the total resistance variation in electrical resistance data.

An example of the analysis described above is presented in Fig. 1. Figure 1(a) shows raw electrical resistance data recorded on amorphous $Fe_{85}B_{15}$ ribbons irradiated at 80 K with 5.2 GeV Pb ions with different tilt angles θ between the normal to the sample surface and the ion beam direction. Figure 1(b) presents the electrical resistance variation of the ribbons as a function of $\sin^2\theta$ for some typical Pb fluences. A clear linear variation is obtained in all cases, in agreement with Eq. (2). The fits to these data allow one to derive values of R and L, which are both plotted in Fig. 1(c) as a function of the ion fluence.

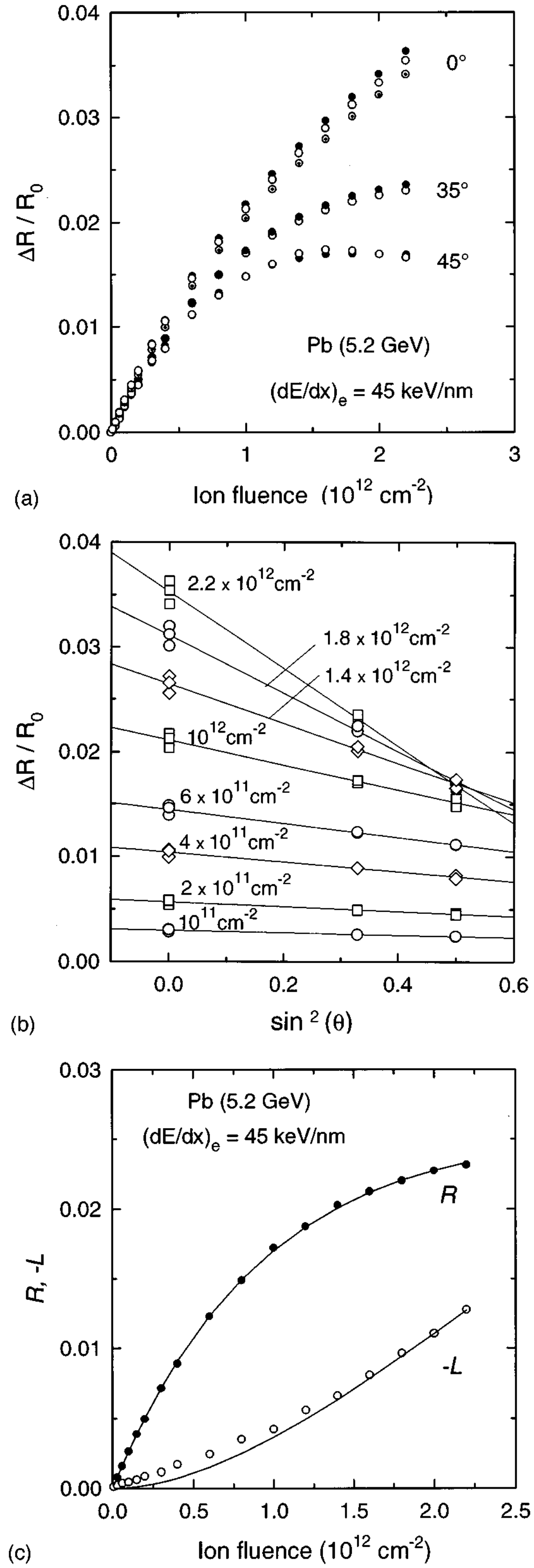

FIG. 1. (a) Ion fluence dependence of the relative electrical resistance of $Fe_{85}B_{15}$ samples irradiated with 5.2 GeV Pb ions. The value of the tilt angle is indicated on the figure. (b) Tilt angle (θ) dependence of $\Delta R/R_0$ deduced from the data of (a) for typical irradiation fluences. Solid lines are fits to the data with Eq. (2). (c) Ion fluence dependence of R and L deduced from the data of (b). Solid lines are fits to the data with Eqs. (6) and (7).

TABLE I. Irradiation parameters. E is the mean ion energy inside the sample; $(dE/dx)_e$ is the ion energy loss; T is the irradiation temperature; $(dL/d\Phi)_0$ and $(dL/d\Phi)_{ss}$ are the L rate [see Eqs. (2) and (4)], respectively, at the beginning of the irradiation and at high fluence; Φ_c is the incubation fluence. Data for Xe and U ion irradiations are deduced from Ref. 10.

Ion	E (GeV)	$(dE/dx)_e$ (keV/nm)	T (K)	$-(dL/d\Phi)_0$ (10^{-15} cm^2)	$-(dL/d\Phi)_{ss}$ (10^{-15} cm^2)	Φ_c (10^{12} cm^{-2})
Xe	2.7	25	20	0.0±0.2	3±1	5±3
Pb	5.2	45	80	5.0±0.2	9.3±0.5	0.9±0.1
Pb	5.2	45	80	4.0±0.5	8.2±0.3	0.7±0.1
U	2	66	80	10±2	20.5±0.7	0.25±0.10

III. PLASTIC DEFORMATION OF AMORPHOUS Fe-B

A phenomenological model has been developed[10] to account for the data obtained in electrical resistance experiments on amorphous Fe-B ribbons irradiated with swift heavy ions. This model assumes a two-hit phenomenon: (a) an ion impinging in a virgin part of the target creates disorder all along its track and does not lead to significant dimensional variations; (b) a subsequent ion impact in a disordered region induces anisotropic atomic movements leading to the sample growth. Three parameters are involved: D_0, Φ_c, and G_{ss}, which are, respectively, the rate of the resistivity increase at the beginning of the irradiation, the incubation fluence, and the growth rate in the steady state. This so-called ''two-hit model'' allows one to derive two equations which describe the resistivity and dimensional variations of the irradiated sample:

$$\frac{\Delta\rho}{\rho_0}=D_0\Phi_c\left[1-\exp\left(-\frac{\Phi}{\Phi_c}\right)\right], \tag{6}$$

$$\frac{\Delta x}{x}=-G_{SS}\left\{\Phi-\Phi_c\left[1-\exp\left(-\frac{\Phi}{\Phi_c}\right)\right]\right\}. \tag{7}$$

Equation (6) implicitly assumes that the resistivity of a ''grown'' region is identical to that of a virgin sample. In former irradiations,[10] Eqs. (6) and (7) reproduced well the plastic deformation process, namely the saturation behavior of the resistivity variation at high fluence and the zero (constant) rate of the growth far below (above) the incubation fluence. Moreover, it has to be noted that if no volume changes are induced by irradiation, D_0 and G_{ss} should have the same values as $dR/d\Phi$ at zero fluence and $-dL/d\Phi$ at high fluence, respectively.

Electrical resistance data obtained on amorphous Fe-B ribbons irradiated with swift heavy ions under various incidences, i.e., by varying the angle between the normal to the sample surface and the ion beam direction, were fitted with Eqs. (6) and (7). Equation (6) reproduces nicely the fluence dependence of R whatever the value of $(dE/dx)_e$ considered. However, whereas for low $(dE/dx)_e$ (Xe irradiation) the fit of Eq. (7) to L data is good, this equation fails to reproduce L data at low fluence for high $(dE/dx)_e$ (Pb or U irradiation). Figure 1(c), which presents the case of Pb irradiation [for which $(dE/dx)_e$ is 45 keV nm^{-1}], illustrates this feature. As a matter of fact, the experimental nonzero initial rate observed in the figure is in disagreement with the predictions of the two-hit model. Table I lists the L rates measured at the beginning and at the end of irradiation [$(dL/d\Phi)_0$ and $(dL/d\Phi)_{ss}$, respectively], as well as the incubation fluence Φ_c for the various irradiations performed.[17] The values of $(dL/d\Phi)_0$ and Φ_c obtained provide a demonstration of the above statements.

If one assumes that the changes of the volume of the ribbons during irradiation are negligible, it is possible to rewrite Eq. (7) in the limit of high fluences ($\Phi \gg \Phi_c$) as

$$\frac{L}{(dL/d\Phi)_{ss}\Phi_c}=\frac{\Phi}{\Phi_c}-1. \tag{8}$$

Equation (8) allows a direct comparison of the data concerning the various irradiations of amorphous Fe-B ribbons. Figure 2, which presents $L/[(dL/d\Phi)_{ss}\Phi_c]$ as a function of Φ/Φ_c, exhibits a similar behavior of all data at high fluence and a clear difference between Xe and Pb (or U) data at low fluence. Actually this figure shows that the initial growth rate is zero only in the case of 20 K Xe irradiation[18] and that it increases with increasing $(dE/dx)_e$.

Let us now discuss possible artifacts which could alter the analysis of electrical resistance data. First, Eq. (1) assumes that the dimensional variations are isotropic in the plane perpendicular to the ion beam direction. However, amorphous

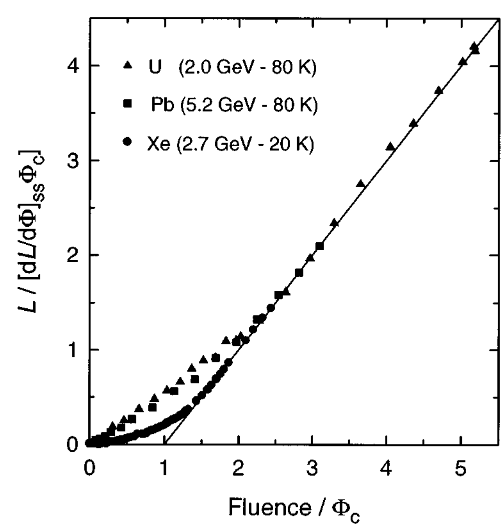

FIG. 2. Ion fluence dependence of L for several irradiation conditions. The solid line stands for Eq. (8). $L/[(dL/d\Phi)_{ss}\Phi_c]$ and Φ/Φ_c are dimensionless parameters defined in the text.

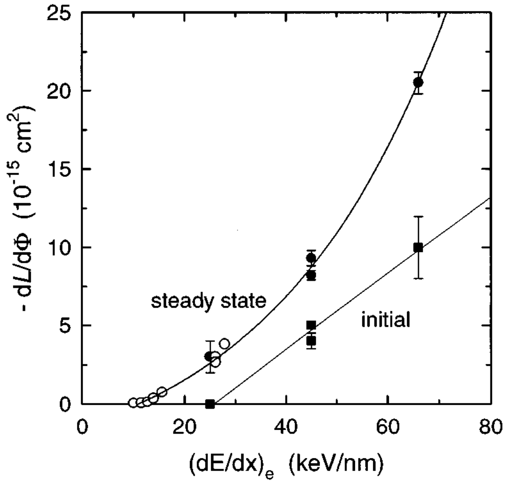

FIG. 3. $(dE/dx)_e$ dependence of the initial (squares) and steady-state (circles) growth rates. Solid and open symbols stand for data derived from electrical resistance data collected on samples irradiated with different tilt angles [according to the deconvolution procedure of Eq. (2)] or on samples irradiated under normal incidence (Ref. 10), respectively. Solids lines are guides for the eyes.

ribbons are prepared by the melt-spinning technique which induces (mainly planar) internal stresses. Such quenched-in stresses could influence the deformation behavior of the ribbon during irradiation. In order to check this point, some of the samples have been annealed at 200 °C during 30 min prior to irradiation. Although the annealing process has induced an important structural relaxation of the sample and thus has certainly suppressed quenched-in stresses, the behavior of annealed ribbons was found identical upon irradiation to that of unannealed ones. A second possibility of obtaining a wrong analysis of electrical resistance data is to consider an eventual alteration of the surface of the irradiated sample. As a matter of fact, very recently swift Xe ion irradiation has been shown to induce important surface modifications in an amorphous $Fe_{40}Ni_{40}B_{20}$ alloy.[19] These surface modifications, which strongly depend on the beam incidence angle, might alter the ribbon shape and thus modify the measured ribbon resistance. However, the above phenomenon has only been observed on thick samples with a beam spot area less than the sample surface area. In such experimental conditions, the stress relaxation can only occur in a limited region of the free surface, which certainly strongly enhances the observed effects. Moreover, very large fluences are required (more than 10^{14} cm^{-2}) in order to obtain significant surface modifications. Finally, surface sputtering could be considered, although very large sputtering rates ($Y \sim 10^6$) could only account for the measured value of $(dL/d\Phi)_0$.

Most of the data presented in Table I and Fig. 2 (particularly those obtained for high values of the electronic stopping power) do not fit the phenomenological two-hit model described at the beginning of this section and leading to Eq. (6) and (7), which implicitly assumes a zero initial growth rate and the existence of a nonzero incubation fluence. This feature can be understood in the light of recent calculations based on shear stress relaxation within electronic excitation-induced thermal spikes by Trinkaus and Ryazanov[15] which predict that an incubation fluence would only be required at low $(dE/dx)_e$ and should disappear at sufficiently high $(dE/dx)_e$. In this model a value of Φ_C different from zero implies that structural changes are first required to modify the viscosity of the amorphous target in order to allow dimensional variations to occur, as it is the case for subcritical values of $(dE/dx)_e$. Figure 3 displays the $(dE/dx)_e$ dependence of the steady state and the initial growth rates for irradiated Fe-B ribbons. As for the resistivity variation[20] an electronic excitation threshold $[(dE/dx)_e^{\mathrm{th}}]$ is observed for both quantities. As a matter of fact, $(dL/d\Phi)_0$ data exhibit a linear dependence upon $[(dE/dx)_e-(dE/dx)_e^{\mathrm{th}}]$. This result is in agreement with the calculations of Trinkaus and Ryazanov[15] which predict that the initial growth rate should depend linearly on the part of $(dE/dx)_e$ spent into ''pronounced spikes,'' (S_e'), assuming that S_e' varies linearly with $[(dE/dx)_e-(dE/dx)_e^{\mathrm{th}}]$. Thus the two-hit model reproduces well experimental results obtained in the case where the electronic stopping power is subcritical for the growth phenomenon (Xe irradiation). In such a situation isolated tracks at the beginning of the irradiation will not cause any growth but will lead to structural modifications of the material along the ion path; ions impacting in the modified regions are supercritical for the plastic deformation and are responsible for the growth. For very heavy-ion irradiation (Pb or U) the electronic stopping power is already supercritical in the virgin material and the first hit already creates growth. Figure 3 also indicates that the threshold for $(dL/d\Phi)_0$ ($\sim$25 keV nm^{-1}) is higher than that for $(dL/d\Phi)_{ss}$ ($\sim$12 keV nm^{-1}). It is worth noting that this latter value is in agreement with the resistivity threshold deduced from *in situ* electrical resistance experiments in a $(dE/dx)_e$ range where a zero initial growth rate is obtained.[10] The clear difference obtained between the two threshold values mentioned above, which define the subcritical electronic excitation range, could also be understood by assuming that between 12 and 25 keV nm^{-1} discontinuous tracks are created. Indeed, revelation of continuous latent tracks by chemical etching of the samples[21] indicates that a $(dE/dx)_e$ value of 34 keV nm^{-1} is required for track formation in a similar amorphous metallic alloy,[22] which is even larger than the threshold for the supercritical regime.

IV. CONCLUSION

The electrical resistance experiments reported in this paper demonstrate that the existence of an incubation fluence for the plastic deformation of an amorphous solid irradiated with swift heavy ions depends on the amount of ion energy loss by electronic excitation in the target. The anisotropic growth starts with a nonzero rate (no incubation fluence) when irradiation is performed with very heavy ions (Pb or U), whereas a zero initial growth rate is observed in the case of irradiation with lighter ions (Xe). This feature is in qualitative agreement with a recent model based on the assumption of the creation of a thermal spike inducing a shear relaxation.[15]

The obtained results can be accounted for by the following description. At low $(dE/dx)_e$ (below $\sim$25 keV nm^{-1}), two (or even more) ion impacts are required for the growth process to occur. The first impact induces structural modifications of the target leading to a viscosity change necessary to the growth process (occurring under further ion impacts) in this subcritical electronic excitation regime. At high $(dE/dx)_e$, growth results from a direct ion impact mechanism (i.e., without incubation fluence) since the amount of electronic excitation is supercritical for the material considered.

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[17] In Table I Φ_C is the extrapolation at $L=0$ of the linear variation of L with Φ at high fluence.

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