Statistical Nature of Atomic Disorder in Irradiated Crystals

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Atomic disorder in irradiated materials is investigated by means of x-ray diffraction, using cubic SiC single crystals as a model material. It is shown that, besides the determination of depth-resolved strain and damage profiles, x-ray diffraction can be efficiently used to determine the probability density function (PDF) of the atomic displacements within the crystal. This task is achieved by analyzing the diffraction-order dependence of the damage profiles. We thereby demonstrate that atomic displacements undergo Lévy flights, with a displacement PDF exhibiting heavy tails [with a tail index in the $\gamma = 0.73$–0.37 range, *i.e.,* far from the commonly assumed Gaussian case ($\gamma = 2$)]. It is further demonstrated that these heavy tails are crucial to account for the amorphization kinetics in SiC. From the retrieved displacement PDFs we introduce a dimensionless parameter f_D^{XRD} to quantify the disordering. f_D^{XRD} is found to be consistent with both independent measurements using ion channeling and with molecular dynamics calculations.

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Ion beams are nowadays used for the development of advanced materials with tailored physical properties. This includes materials for spintronics [1], nanophotonics [2], or for the development of materials for the space and nuclear industry [3]. The understanding of irradiation effects in solids is therefore a fundamental issue to be addressed.

The slowing down of a charged particle with a low velocity, as compared to the root-mean-square velocity of its own electrons, passing through matter can be essentially described by elastic (so-called nuclear) collisions between the projectile and the (screened) nuclei of the target. During this interaction, part of the projectile kinetic energy is transferred to the target atoms. If this transferred energy is larger than a threshold displacement energy, the primary knock-on atom may be ejected with a velocity sufficient to induce collective displacements of target atoms; this phenomenon is referred to as a collision cascade [4]. After this collisional stage, an energy dissipation phase occurs, followed by a diffusional phase, and defect recombination may occur. At the end of this multistep process, the target atoms may remain, slightly or significantly (*i.e.,* far from their initial lattice site) displaced, inducing a residual disorder. Increasing the irradiation dose usually leads to defect clustering and the formation of extended defects or even amorphous clusters.

The need for understanding the exact mechanisms of defect generation and disorder accumulation in irradiated materials has fueled a huge number of both experimental and computational studies over the last decades (for a recent review, see Ref. [5]). Experimentally, transmission electron microscopy is an invaluable tool for defect imaging, although obtaining both atomic-scale and statistically relevant information is extremely challenging in irradiated, damaged crystals. Spectroscopic techniques such as Raman spectroscopy, positron annihilation spectroscopy, and Rutherford backscattering spectroscopy in channeling mode (RBS/C) are widely used to evaluate the disorder level in irradiated materials [6]. Most of the time, they rely on a phenomenological approach where the disorder is quantified through a simple parameter such as the weakening of the Raman lines, the fraction of positron annihilations with core or shell electrons, or the fraction of displaced atoms determined from the backscattering yield in RBS/C.

Another widespread technique is x-ray diffraction (XRD). Being an interferometric technique, XRD is highly sensitive to the atomic displacement field within the crystal, and numerical simulations of XRD data permit us to retrieve the lattice strain consecutive to ion irradiation, as well as the level of disorder which is estimated through the determination of the so-called static Debye-Waller (DW) factor [7–12]. The DW factor is a dimensionless parameter that affects the diffracted intensity: perfect crystalline regions diffract at their nominal value (DW = 1), whereas damaged regions are characterized by a lower diffracted intensity (DW < 1), with amorphous regions being characterized by vanishingly small values of DW. Despite this apparent simplicity, the DW factor cannot be used as a general measure of disorder since it is dependent on experimental parameters (such as the choice of diffracting planes) and its definition relies on assumptions which we will prove to be incorrect in the case of irradiated materials.

In this Letter, we reinvestigate the well-established interpretations of the static DW factor and demonstrate that the actual probability density function (PDF) of atomic

displacements can be retrieved from the analysis of XRD data. We thereby show that, in irradiated materials, atomic displacements exhibit Lévy flights which are expected to play an important role in the irradiation-induced structural modifications (such as amorphization, as addressed hereafter, or in ion-beam mixing phenomena, for example [13]). Finally, with the computed cumulative distribution functions of the atomic displacements, we introduce a dimensionless parameter that takes into account the statistical nature of the atomic displacements and which can be used to quantify the disorder in irradiated materials.

We use irradiated 3*C*-SiC single crystals as a model system of a material undergoing an amorphization under irradiation. SiC exhibits attractive electronic and structural properties [14,15] and its behavior under various irradiation conditions is now very well documented thanks to decades of studies [16–23]. {001}-oriented 3*C*-SiC single crystals (with lattice parameter $a_0 = 4.359$ Å) were irradiated with 100 keV Fe ions with ion fluences in the 4×10^{13}–2×10^{14} cm^{-2} range, which corresponds to damage doses of ~0.072 to ~0.36 dpa (displacement per atom) at the damage peak, as determined with the SRIM code [24] using threshold displacement energies of 20 and 35 eV for the *C* and Si sublattices, respectively [25,26]. Details regarding the irradiation conditions can be found in [22,27].

High-resolution XRD experiments were carried out at the BM02 beamline of the European Synchrotron Radiation Facility (ESRF, Grenoble, France). Longitudinal θ–2θ scans were recorded around the 002 and 004 reflections of 3*C*-SiC. Experimental details and scans are given in [28]. Depth-resolved strain and damage profiles were retrieved from the XRD data using the RaDMaX program [11,32] and are given in Fig. 1.

Let us briefly discuss the strain (e_z) depth profiles, Fig. 1(a). For the lowest damage doses (0.072 and 0.144 dpa) the simulation of the 002 and 004 reflections yields very similar profiles, and for the highest damage doses (0.29 and 0.36 dpa), the profiles are indistinguishable, which clearly confirms the validity of the simulations (given in [28]). The strained region extends over 90 nm, with a tail spreading to 120 nm, and a maximum strain located 20–30 nm below the surface of the crystals (in good agreement with damage profiles obtained by ion channeling experiments [22]). The overall strain increases with increasing fluence, and the maximum strain increases from 1.2% to extremely high values of 8.4%.

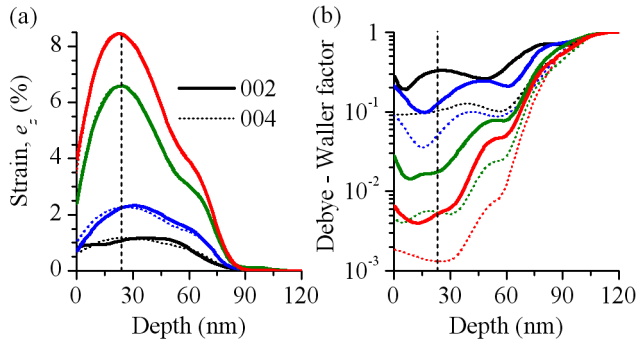

FIG. 1. Strain (a) and DW (b) depth profiles obtained from the simulation of XRD curves presented in [28]. The results obtained from the 002 and 004 reflections are plotted as solid and dotted lines, respectively. The following color scheme is used: black-blue-green-red for increasing damage dose.

Figure 1(b) shows the depth-resolved DW factor, for the different ion fluences, for the 002 and 004 reflections. It can be observed that, for increasing fluence, the DW factor significantly decreases, pointing to an increase in atomic disorder, consistent with the expected eventual amorphization of SiC. Concomitantly, as mentioned above, the elastic strain in the crystalline regions increases with the ion fluence. Therefore, these two findings indicate that the volume of the strained, crystalline regions decreases with irradiation dose, with the concomitant formation of amorphous regions (in perfect agreement with the results presented in [27] where only the 004 reflection was probed).

We now reinvestigate the interpretation of the DW factor. For the thin subsurface region of the crystal damaged by the ion beam, the diffracted x-ray amplitude can be written

$$E = \sum_i f_i \exp(i\mathbf{Q} \cdot \mathbf{r}_i), \tag{1}$$

where f_i is the atomic scattering factor of the atom at the ith lattice site in the crystal with position vector $\mathbf{r}_i$, and $\mathbf{Q}$ is the diffraction vector ($Q = 4\pi \sin\theta/\lambda$, where θ is the diffraction angle). The displacement field consecutive to random collision events can be conveniently described with the vector $\mathbf{u}$, where the displacement $\mathbf{u}_i$ at a given site i is determined by the PDF, $p(\mathbf{u})$, of the random variable $\mathbf{u}$. The diffracted amplitude is then given by the ensemble average over $p(\mathbf{u})$:

$$\langle E \rangle = \int d\mathbf{u} \sum_i p(\mathbf{u}) f_i \exp[i\mathbf{Q} \cdot (\mathbf{r}_i + \mathbf{u}_i)] \tag{2}$$

and the coherent part of the diffracted intensity, which is considered in this Letter, is given by $I = \langle E \rangle \langle E^* \rangle$. If all atoms share the same displacement PDF, Eq. (2) can be written as $E \times \mathrm{DW}$ [33], where [7]:

$$\mathrm{DW} = \int d\mathbf{u}\ p(\mathbf{u}) \exp(i\mathbf{Q} \cdot \mathbf{u}). \tag{3}$$

This well-known result has been briefly rederived here to highlight an overlooked property of the DW factor: this term is the Fourier transform of $p(\mathbf{u})$, *i.e.,* its *characteristic function* [34]. $p(\mathbf{u})$ captures all the statistical information regarding atomic displacements and is therefore of fundamental importance for the understanding of the physics underlying the radiation behavior of materials.

The displacement PDF is often assumed to be Gaussian shaped. In the case of *thermal* disorder, lattice dynamics in the isotropic harmonic regime predicts a Gaussian PDF [35], which gives rise to the well-known result

$\mathrm{DW} = \exp(-Q^2\langle u^2\rangle/2)$. In the case of *static* disorder, a Gaussian PDF can be justified by the central limit theorem, which states that the sum of identically distributed random variables (*i.e.,* the displacement fields from individual defects in our case), with finite variance, converges to a Gaussian distribution. Using the assumption of a Gaussian PDF for the 002 reflection, and considering the DW value for the lowest fluence at the 20 nm depth [Fig. 1(b)], a root-mean-squared displacement $\langle u^2\rangle^{1/2} = 0.52$ Å is obtained, which in turns gives a DW factor of ~0.01 for the 004 reflection. However, the observation of Fig. 1(b) reveals that the actual DW factor determined from the 004 reflection is 0.1, *i.e.,* 10 times higher. This simple observation indicates that a Gaussian PDF is not appropriate to describe the disorder in irradiated SiC, and most likely in other irradiated materials either.

As mentioned earlier, the process of energy dissipation of low-velocity projectiles leads to atomic disorder that is due to both atoms close to equilibrium lattice sites and atoms significantly displaced from a regular site. This situation where some displacements are statistically much larger than the "average" displacement, are known as Lévy flights [34]. Besides this qualitative argument, several theoretical works suggested that the displacements in a collision cascade and in the subsequent disordered crystal actually exhibit Lévy-stable distributions [13,36,37] in which the atomic displacement PDF exhibits heavy tails, *i.e.,* Lévy flights. Interestingly, Lévy-stable distributions are a natural consequence of the *generalized* central limit theorem, where the condition of finite variance is dropped (hence allowing for large deviations from the average value) [34]. The characteristic function of Lévy-stable distributions is known [38] and can be used to solve Eq. 3. In the case of 00l reflections the diffraction vector is $\mathbf{Q} = (0, 0, Q)^T$ and the DW factor can be written

$$\mathrm{DW} = \exp\left\{ iQ\mu_u - \frac{1}{2}|Q|^\gamma \sigma_u^\gamma \left[1 - i\eta \frac{Q}{|Q|}\omega_\gamma(Q)\right]\right\}, \qquad (4)$$

where $\omega_\gamma(Q) = \tan(\pi\gamma/2)[(\sigma_u|Q|)^{1-\gamma}/2^{1/\gamma} - 1]$ if $\gamma \neq 1$, and $\omega_\gamma(Q) = (2/\pi)\ \ln(\sigma_u|Q|/2)$ if $\gamma = 1$. In Eq. (4), μ_u is the mode of the distribution and $\eta \in [-1, 1]$ is a dimensionless parameter describing the asymmetry of the distribution. The tail index $\gamma \in (0, 2]$ determines the shape of the distribution: $\gamma = 2$ corresponds to the Gaussian distribution, and distributions with $\gamma < 2$ exhibit heavy tails (asymptotically behaving as $\sim 1/|u|^{1+\gamma}$), like, for instance, the Lorentzian distribution ($\gamma = 1$) [34]. σ_u is the characteristic width of the distribution [39], whose definition varies depending on the value of γ; for instance, it corresponds to the standard deviation for $\gamma = 2$, and to the full width at half maximum for $\gamma = 1$. In the following, to avoid any confusion, we use the width at half maximum (u_{HM}) of the distribution as a measure of atomic disorder for all cases.

In the presence of lattice strain, the mode of the displacement distribution is given by $\mu_u = \int_0^z d\zeta\ e_\zeta \zeta$, where z is the depth below the surface and e_z is the strain distribution given in Fig. 1(a). In order to compare crystals with different levels of strain, only the relative deviation from the average displacement must be considered in the following, *i.e.,* $u - \mu_u$. Additionally, in a collision cascade there is no preferential direction for the displacements, indicating that the displacement PDF is symmetrical, *i.e.,* $\eta = 0$. With these considerations, the DW factor reduces to

$$\mathrm{DW} = \exp\left(-\frac{1}{2}|Q|^\gamma \sigma_u^\gamma\right). \qquad (5)$$

It appears that ln(DW) is a linear function of Q^γ, so that the characteristic width of the distribution and, more importantly, the exact shape of the displacement PDF can be retrieved by analyzing the Q dependence of ln(DW). This analysis is presented below using the 002 and 004 diffraction orders.

The inspection of Fig. 1(b) reveals that some of the DW profiles exhibit oscillations which reflect the (moderate) uncertainty of the determination of this parameter. In order to limit the influence of these oscillations we focus on the region of maximum damage and compute the average DW factor in the 10–30 nm range. The computed values of γ and u_{HM} as a function of the damage level are displayed in Figs. 2(a) and 2(b), respectively, along with the evolution of the maximum strain, $e_z^{(\max)}$. As noted above, we observe a neat correlation between the maximum strain and the level of disorder. More interesting is the evolution of the tail index, Fig. 2(a). For the lowest irradiation dose, its value ($\gamma = 0.73$) is close to the Lorentzian limit, but decreases to extremely low values (0.37) for the highest damage dose. This result leads to two important conclusions regarding the irradiation-induced atomic disorder in SiC: (i) it is actually highly non-Gaussian, (ii) its nature changes with increasing fluence, where large displacements become more likely to occur, probably as a consequence of the increasing fraction of amorphous (*i.e.,* completely disordered) regions [27].

To illustrate the importance of these findings we generated disordered structures of 3*C*-SiC by applying random displacements to all atoms in the crystals, Figs. 2(d)–2(g). Figures 2(d) and 2(e) correspond to the disorder level for the lowest and highest fluences ($u_{\mathrm{HM}} = 0.033$ and 0.14 nm, respectively) in the case of the Gaussian displacement PDF. Figures 2(f) and 2(g) correspond to the same level of disorder but in the case of the Lévy-stable distributions obtained in this work ($\gamma = 0.73$ and 0.37, respectively). The generation of Lévy-stable random numbers was performed using an algorithm detailed in Ref. [40]. In the Gaussian case, even for the highest disorder level, atomic rows are still visible [Fig. 2(e)], which is in contrast with the actual microstructure of the samples at this fluence, as previously shown in [22,27]. In the case of Lévy-stable distributions, the lowest level of disorder [Fig. 2(f)] gives a similar structure as that for the highest fluence of the Gaussian case [Fig. 2(e)], indicating that the presence of heavy tails in the displacement PDF allows for

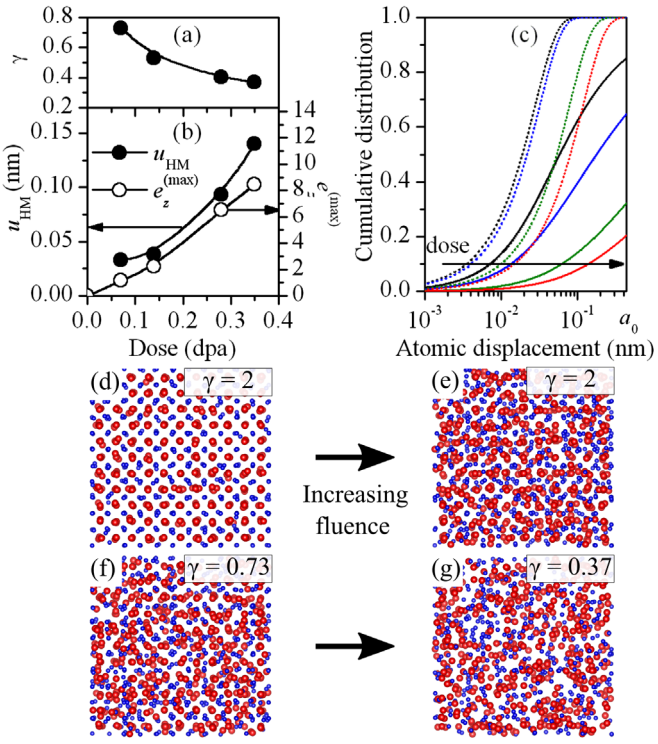

FIG. 2. Variation of the tail index (a) and of the atomic disorder and maximum strain (b) for increasing damage. (c) Cumulative distribution functions for increasing dose (black-blue-green-red) assuming Lévy-stable distributions (solid lines) and Gaussian distributions (dotted lines). (d),(e) Simulated structures of 3C-SiC in the case of Gaussian disorder for the lowest and highest disorder, respectively. (f),(g) Simulated structures of 3C-SiC in the case of Lévy statistics for the lowest and highest disorder, respectively (red spheres, Si; blue spheres, C). The structure is viewed along the [100] direction.

a faster disordering. In the high disorder case, no long-range order can be (visually) detected, suggesting a nearly complete amorphization of the crystal.

In a more quantitative approach, Fig. 2(c) displays the displacement cumulative distribution functions, computed from the values of γ and σ_u, for increasing damage and for both the Gaussian and Lévy-stable distributions. It is remarkable that in the Gaussian case, even for the highest irradiation dose, the maximum displacement does not exceed the length of the unit cell (a_0). Conversely, in the case of the Lévy-stable distribution, displacements larger than a_0 concern 15% of the atoms at the lowest fluence.

In light of this example, we propose to quantify the disorder with a parameter that gives a measure of the damage fraction, defined as follows: $f_D^{\mathrm{XRD}} = P(u > d_{\mathrm{th}})$, *i.e.*, the probability of finding atoms displaced by values larger than a threshold distance d_{th}. Let us first consider the usual Gaussian approximation of the displacement PDF [*i.e.*, $\gamma = 2$ in Eq. (5)]. In such a case, the damage fraction from XRD can be worked out analytically: $f_D^{\mathrm{XRD}(\gamma=2)} = 1 - \mathrm{erf}[d_{\mathrm{th}} Q_i / 2\sqrt{-\ln(\mathrm{DW}_i)}]$, where i refers to the 002 or 004 reflection. With the threshold value used above, $d_{\mathrm{th}} = a_0$, f_D^{XRD} is equal to zero for all fluences. A commonly used threshold distance is $d_{\mathrm{th}} = 0.57a_0$ which corresponds to the average of the first and second nearest neighbor distances. With this threshold distance the maximum damage is 0.025 at 0.36 dpa (see dotted line in Fig. 3). This observation definitely confirms that a Gaussian disorder cannot describe the amorphization process of SiC. Values of f_D^{XRD} in the case of Lévy-stable distributions are also plotted in Fig. 3, for cutoff distances $d_{\mathrm{th}} = 0.57a_0$ and $d_{\mathrm{th}} = a_0$. The variation of the disorder is identical for both cases, with slightly higher values for the $0.57a_0$ cutoff distance, which is expected since in this case smaller displacements are taken into account. It can be noted that at the highest dose (0.36 dpa), the damage fraction is 0.80, meaning that the irradiated layer is not, within this line, completely amorphous. A short discussion on how this approach relates to the generalized Lindemann criterion is given in [28].

This parameter can be compared with the damage fraction obtained by RBS/C, where f_D^{RBS} refers to the fraction of randomly displaced atoms (without any information regarding the magnitude of the displacement) [41]. The corresponding values for the actual irradiated crystals are plotted in Fig. 3. Up to half of the amorphization process (~0.2 dpa), the damage fractions determined by XRD and RBS/C are similar. At higher doses, full amorphization is observed for RBS/C ($f_D^{\mathrm{RBS}} = 1$) [22], contrarily to what is observed by XRD, indicating that even at this high dose, some (disordered) regions remain crystalline. This discrepancy is very probably due to the fact that in RBS/C all displacements, without any cutoff distance, are considered.

Despite this slight difference, it is remarkable that although the physical phenomena governing XRD and RBS/C are completely different, very similar trends of the damage fraction are obtained. Moreover, it must be borne in mind that, in the symmetric geometry used in this Letter, XRD probes out-of-plane displacements, whereas RBS/C is sensitive to in-plane displacements. The similar results obtained in both cases suggest an isotropic disordering upon irradiation. Finally, we also compared these damage fractions with the loss of long-range order (LRO) derived from independent molecular dynamics (MD) simulations of cascade overlapping in SiC (adapted from [19]). There is a remarkable agreement between f_D^{XRD} and the disorder

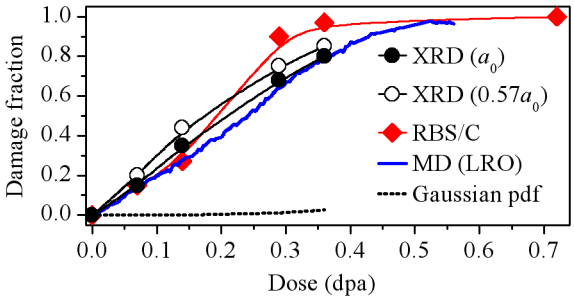

FIG. 3. Variation of the damage fractions as evaluated by XRD, RBS/C, and MD simulations.

obtained from the LRO parameter, which provides an experimental confirmation of the MD predictions of the amorphization scenario proposed in Ref. [19]. To finish, the similarity between the evolution of f_D^{XRD}, f_D^{RBS} and LRO with ion dose demonstrates the necessity to consider the existence of Lévy flights in the distribution of atomic displacements, since those play a crucial role in the amorphization kinetics of SiC.

Concluding, we have shown that XRD allows us to determine the probability density function of atomic displacements in irradiated materials. In the particular case of SiC, we have demonstrated that the atomic displacements exhibit Lévy flights and that heavy tails in the displacement PDF are required to accurately describe the amorphization kinetics of this material. From the atomic displacement distribution functions, we introduced a dimensionless parameter to quantify the disorder. We found that the variation of this parameter with irradiation dose agrees well with both the disorder parameter derived from molecular dynamics simulations of cascade overlapping in SiC and with the damage buildup determined from RBS/C experiments. The use of this dimensionless parameter could be generalized and could serve as a quantitative measure, using solely XRD data, of the disorder level in irradiated materials.

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