Tracks induced by swift heavy ions in semiconductors

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InSb, GaSb, InP, InAs, and GaAs single crystals were irradiated with Pb ions in the range of 385–2170 MeV. The samples were studied by transmission and high-resolution electron microscopy and Rutherford backscattering in channeling geometry. The energetic ions induced isolated tracks in all crystals but GaAs. The thermal spike analysis revealed that the variation of the damage cross section with the ion energy is considerably weaker than in insulators. The widths of the thermal spike $a(0)$ was estimated. The analysis was extended to recent C_{60} experiments on Ge and Si. A quantitative relation was found between $a(0)$ and the gap energy E_g: $a(0)$ is reduced with increasing E_g, and its lowest value is close to that found in insulators.

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I. INTRODUCTION

By irradiating insulators with energetic ions, amorphous tracks can be induced with a diameter up to 25 nm, and the size of these tracks can reliably be predicted.[1] Ion irradiation may become a tool to create tracks in semiconductors, with a good control of track density, diameter, and length. Presently, we do not know the main controlling parameters of this process in semiconductors. Developments in this field may have importance for basic and applied research, because of the growing interest in the methods of production and in the properties of quantum wells and quantum dots.

In this paper we present and discuss experiments in which isolated tracks are induced in a crystalline semiconducting matrix. All previous attempts to induce such tracks with monoatomic beams were unsuccessful except one: Vetter *et al.* produced amorphous tracks in GeS, in a highly anisotropic layered crystal.[2] Recently, cylindrical tracks were observed in Si (Refs. 3 and 4) and Ge (Ref. 5) single crystals after C_{60} irradiation. A systematic investigation and considerably more experimental data are necessary to reveal how the electron properties affect the localization of the energy deposition. In this paper we report on the first results of our research.

II. EXPERIMENT

In Table I we listed the irradiated crystals, which all had a (001) orientation and a 0.5-mm thickness. The first irradiation was performed in the GANIL (Caen, France) by a Pb beam with an initial energy of 4.17 MeV/nucleon, which we reduced to 1.85 and 0.85 MeV/nucleon by Al foils. A second irradiation was performed in the UNILAC in GSI (Darmstadt, Germany) by a Pb beam with an initial energy of 11.4 MeV/nucleon. This time the ion energy was reduced by Al foils to 4.0, 5.7, and 10 MeV/nucleon, to study the variation of the damage cross section with the ion energy. The maximum fluences varied between 3.9×10^{11} ions/cm^2 (InSb) and 1.6×10^{12} ions/cm^2 (InP). The maximum flux was 2×10^{8} ions/cm^{2}s to avoid beam heating. The fluences were given with a relative error of 15%. The ion beams were scanned over a surface of about 12 cm^2, and up to 16 samples could be simultaneously irradiated. Parallel samples were placed at the center and edges of the sampleholders to check the homogeneity of the fluences, and no systematic deviations were observed. The irradiations were performed at room temperature. According to our estimate the temperature increase was within a few K during irradiations.

The irradiated samples were studied by Rutherford backscattering in channeling geometry (c-RBS), that has been extensively used in studies of radiation damage induced by swift heavy ions.[6,7] In this method the backscattered yields are measured in random and aligned orientations and compared to the yield of an aligned virgin single crystal. The damaged fraction of the irradiated crystal F_d is calculated from the results by a standard method. The average track radii R_e are determined from the variation of F_d with the fluence of the bombarding ion. In our experiments the backscattered yields of the virgin samples were 2.8% and 6.2% in InP and InSb, respectively. The energy spectra of the backscattered $^4He^+$ ions of 1-MeV incident energy were collected by a blind (i.e., not light sensitive) semiconductor particle detector of 50-mm^2 sensitive area mounted at 147° scattering angle at a distance of 30.5 mm from the sample.

TABLE I. Parameters of semiconductors in track experiments; ρ, density; c, specific heat; T_m, melting point; T_{ir}, irradiation temperature; $T_o=T_m-T_{ir}$; E_g, gap energy; $a(0)$, Gaussian width of the thermal spike (p, present study; a, monoatomic beams).

Crystal	$\rho c T_o$ J/cm^3	E_g eV	track[a] yes/no	$a(0)$ nm
InSb[p]	607	0.17	y	13.0±1.2
GaSb[p]	995	0.67	y	11.2±1.0
InAs[p]	1370	0.36	y	9.8±0.9
InP[p]	1677	1.27	y	7.95±0.72
Ge (Ref. 5)	1713	0.67	n	9.85±0.9
GaAs[p]	2249	1.35	n	
Si (Ref. 3)	2890	1.1	n	7.2±0.65

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TABLE II. Track parameters, deduced from *c*-RBS measurements; R_e, and g are the effective track radius and the efficiency, respectively, E is the specific ion energy in MeV/nucleon.

	R_e (nm)				g	
E	InP	InSb	GaSb	InAs	InP	InSb
1.85	3.5	8.6	1.8		0.092	0.092
4.0	3.9	9.2		2.2	0.082	0.079
5.7	3.4	8.6			0.075	0.074
10	3.05	5.2			0.074	0.066

To minimize the damage caused by He ions, the intensity of the measuring ion beam of $\sim$1-mm^2 cross section was kept as low as 0.1 nA, and the applied fluence was about 100 nC. The error of the measurements of F_d was below 10%. According to our estimate the relative error of the measurement of the track diameters was always lower than 10%. The results are shown in Table II.

Transmission electron microscopy (TEM) measurements were made on InSb, InP, and GaSb samples with a Philips CM20 electron microscope. Plane-view TEM specimens of (001) surface were prepared prior to the ion irradiation to avoid the formation of artifacts. The number of tracks in the TEM pictures was in reasonable agreement with the fluence. No systematic deviation from the circular shape was observed in any compound. In InSb the TEM studies revealed a stress field with fourfold symmetry around the tracks [Fig. 1(a)]. High-resolution electron microscopy (HREM) was also applied, and we obtained $d = 11$ nm for the mean track diameter. In Fig. 1(b) the track is completely recrystallized. The lack of strain field in the picture may be a consequence of the selected high-resolution imaging conditions. An abrupt change of the orientation occurs inside the recrystallized track, which can be characterized by a 45° rotation along the [001] direction coinciding with the zone axis of the crystal. This internal subgrain shows a more faceted (octagonal) shape, with a diameter of about 5.5 nm. Although the {220} lattice fringes are slightly shifted upon crossing the track boundary, the crystallographic phases inside the track and in the subgrain are the same as in the bulk of the sample.

Previously, recrystallized tracks were also observed in irradiated UO_2, which is an insulator.[8] We calculated the radii of the cylindrical melt and found good agreement with the track sizes.[9] We assume that the two sizes are close in InSb, as well.

In InSb the *c*-RBS method provides considerably larger track radii than HREM. As InSb samples were irradiated simultaneously with other compounds, we can rule out the possibility of an incorrect fluence measurement. We attribute the deviation to the effect of the large deformation field around the tracks in the (001) plane [Fig. 1(a)]. By comparing the HREM and the *c*-RBS results a correction factor was determined and we used $R_e(c\text{-RBS})/1.55$ for the true track radii in the analysis.

HREM measurements revealed amorphous tracks in prethinned GaSb samples after the Pb ion irradiation with $E = 4$ MeV/nucleon. However, no tracks were found at $E = 0.85$ MeV/nucleon, which we attribute to the 30% reduction of the electronic stopping power S_e.

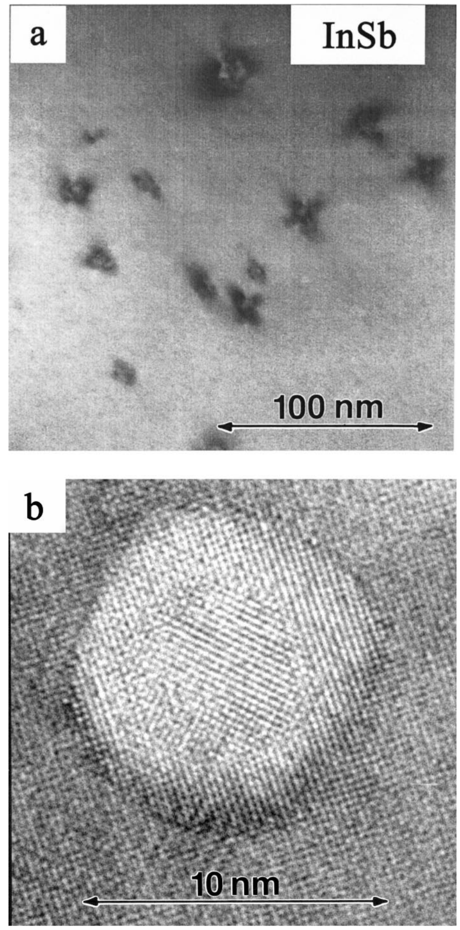

FIG. 1. Transmission electron micrograph of an irradiated InSb ($E = 1.85$ MeV/nucleon) (a) bright-field image with deformation contrast; (b) high-resolution electron micrograph of a single track (taken at 400 kV in a JEOL 4000 EX).

In Fig. 2, tracks in InP are shown in a sample tilted by 45° in the electron microscope. The tracks exhibit an intermittent contrast. A similar effect was observed by Chadderton in MoS_2,[10] but we do not deny the change of the track radii with the depth. In plane-view pictures a thin cylindrical stress field appears and the tracks slowly disappear during a prolonged exposition to the electron beam. This points to an

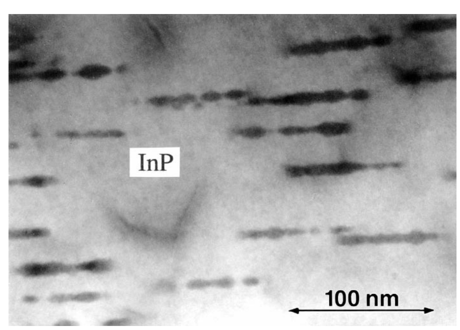

FIG. 2. Transmission electron micrograph of an irradiated InP ($E = 1.85$ MeV/nucleon). The sample is tilted in the electron microscope by 45°.

amorphous structure. The mean track diameter deduced from TEM measurements $2R_e=6.5$ nm is in good agreement with the c-RBS results ($2R_e=7.0$ nm).

III. DISCUSSION

We perform the analysis of tracks according to our thermal spike model.[1] The main assumption of the model is that there is a Gaussian temperature distribution in the ion-induced spike, which is characterized by its initial width $a(0)$ and the efficiency g (gS_e is the fraction of the deposited energy transferred to the thermal spike).[1] We make a similar assumption for semiconductors, as well, and apply Eqs. (1) and (2) derived in Ref. 1,

$$S_{et}=\pi\rho c T_o a^2(0)/g, \tag{1}$$

$$R_e^2=a^2(0)\ln\frac{gS_e}{\pi\rho c T_o a^2(0)} \quad \text{for} \quad 1<S_e/S_{et}<2.7, \tag{2}$$

where ρ, c, and T_o denote the density, the average specific heat, and the difference between the melting point T_m and the irradiation temperature T_{ir} and S_{et} is the threshold value of S_e for track formation. The efficiency $g(E)$ has close values in various insulators both at low ($E<2$ MeV/nucleon) and at high specific ion energies ($E>7$ MeV/nucleon)[1,11,12] and $a(0)=4.5$ nm for $0.1<E<20$ MeV/nucleon without exception.[1,13] In agreement with the experimental data, the model predicts that the $R_e^2-S_e/\rho c T_o$ curves coincide at low and high ion energies.[1,12]

In Table I some parameters of those semiconductors are shown, which were used in our and previous experiments. The compounds were selected to have low values of $\rho c T_o$, and a suitable range of parameters to check various correlations.

Our purpose is to estimate the two parameters of the model, $a(0)$ and g, from the experimental data. At least two independent track sizes are necessary for the analysis. In this experiment, however, only a single track size could be determined at each ion energy. Therefore, $a(0)$ and g can be estimated only with additional assumptions. We assume that the basic mechanisms of track formation are similar in insulators and semiconductors. The behaviors of the $a(0)$ and g parameters exhibit common features in various insulators; therefore, we expect that similarities also exist in semiconductors. Actually, we assume that $a(0)$ does not depend on E, as in insulators, but that it may have different values in various semiconductors. In insulators tracks have been studied both by monoatomic and cluster ions. We found that $g(E)$ has close values at $E=0.05$ MeV/nucleon and $E=1$–2 MeV/nucleon.[11] We assume that this holds in our semiconductors, as well.

In Fig. 3 we plot the results reported in Refs. 3 and 5. Tracks were induced in Ge and Si crystals by C_{60} ions of 20–40-MeV energy. The track diameters were measured by TEM. We applied Eqs. (1) and (2) for the analysis and obtained $a(0)=9.85$ nm, $g=0.097$ for Ge and $a(0)=7.2$ nm and $g=0.087$ for Si. We note that $g=0.4$ was obtained in $Y_3Fe_5O_{12}$ and $LiNbO_3$ in similar experimental conditions.[12] This means that at equal values of S_e the energy of the spike is about four times lower in Ge and Si than in insulators. If this high-g value were valid for Ge and Si, S_{et} would be as low as 8.3 and 7.4 keV/nm, respectively. In the following analysis we will use the mean value $g_{av}=0.092$, and assume that in our semiconductors the efficiency has the same value at $E=1.85$ MeV/nucleon, as well.

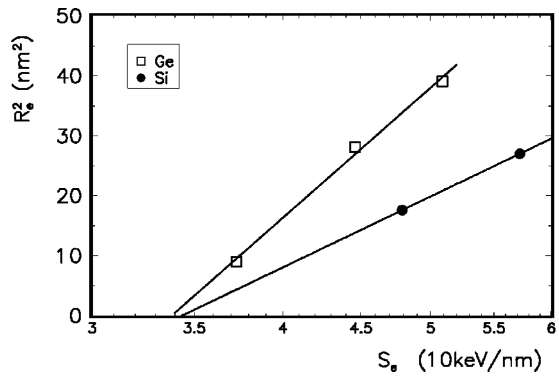

FIG. 3. Track evolution in Ge (Ref. 5) and Si (Ref. 3). The lines are fits to Eq. (2).

We have data sets for InP and InSb, which were measured by c-RBS at $E=1.85$, 4.0, 5.7, and 10 MeV/nucleon (in insulators the change of g between 8 and 10 MeV/nucleon is negligible). The calculation was performed according to Eq. (2). The $a(0)$ and $g(E)$ values are shown in Tables I and II. The experimental error of R_e and S_e were not given in Refs. 3 and 5; thus we cannot estimate the uncertainty of g_{av}. We note that presently the additivity of S_e is an open question for the individual atoms in the cluster ions. Another problem is the strong tendency of the tracks in Ge and Si to recrystallize. Since the uncertainty of g_{av} affects all results of our calculation, a systematic error may modify the derived g and $a(0)$ values. Therefore, we do not concentrate on the absolute values, rather on the relative variation of these parameters, which are practically not affected by the actual value of g_{av}.

We estimate the relative standard deviations $\Delta a(0)/a(0)$ and $\Delta g/g$ keeping only the first derivative of the logarithmic function in Eq. (2). We obtain

$$\left(\frac{\Delta g}{g}\right)\approx\left[\left(\frac{2R^2}{a^2(0)}\right)^2\left(\frac{\Delta R_e}{R_e}\right)^2+\left(\frac{\Delta S_e}{S_e}\right)^2\right]^{1/2}, \tag{3}$$

where R denotes the mean value of R_e. Equation (3) is valid when we estimate the variation of g versus E at $a(0)=\text{const}$ for InSb and InP. A slightly different expression for $\Delta a(0)/a(0)$ provides an even lower error when $a(0)$ is estimated for InSb, InP, GaSb, and InAs at $g=g_{av}$. In our experiments $R/a(0)$ varies between 0.2 and 0.45. Thus when we are interested only in the relative variation of the $a(0)$ and g parameters, the error is not sensitive to the accuracy of the measurements of R_e, if R_e is not close to $a(0)$. The S_e values were calculated by the TRIM code. When we take $\Delta S_e/S_e=0.1$, Eq. (3) provides $\Delta g/g\approx 0.12$ and $\Delta a(0)/a(0)\approx 0.09$.

We found that in insulators $0.4>g(E)>0.17$ in the range $1<E<20$ MeV/nucleon.[1,12,13] This is the so-called damage cross-section velocity effect.[7] In high-T_c superconductors (HTCS's) $g(E)$ has close values at high and low projectile velocities.[14] Until now, no information has been obtained about the velocity effect in semiconductors. Initially, we assumed that at low ion velocity g is also high in semiconductors. This was the reason why the first irradiation was performed at $E=1.85$ and 0.85 MeV/nucleon. Our analysis shows that in InSb and InP the efficiency g only slightly varies with E in the range of $2<E<10$ MeV/nucleon. If the velocity effect is characterized by the ratio of the efficiencies G at $E=2$ and 10 MeV/nucleon, then $G=2.35$ in insulators and only 1.24 and 1.4 in InSb and InP, respectively. If $G=2.35$ were valid for InP and InSb, no tracks would be formed at $E=10$ MeV/nucleon according to Eq. (2).

Colder *et al.* applied Pb ions with $E=4.8$ MeV/nucleon ($S_e=38.3$ keV/nm), which did not induce tracks in Ge, while a C_{60} beam ($S_e=37.3$ keV/nm) created tracks with $d=6$ nm.[5] This behavior is in agreement with our estimate of $g(E)$, as the slight reduction of the efficiency to $g=0.078$ (see Table II) is sufficient to increase S_{et} from 34 to 40 keV/nm. Tracks are not formed since $S_{et}>S_e$. We note that neither the correction of the InSb track radii by 1.55, nor the choice of the efficiency value at $E=1.85$ MeV/nucleon, modifies the relative variation of the $g(E)$ function; only its absolute value changes. The low value and the slight variation of $g(E)$ is markedly different from the behavior of insulators, and it is closer to the results obtained in HTCS's.

It is difficult to account for these considerable differences if the electron-phonon scattering is the sole relaxation mechanism. We assume that several mechanisms contribute to the energy of an ion-induced thermal spike. At a given energy E, these contributions may depend on whether the target is an insulator, a semiconductor, or a HTCS. The track experiments are important because the parameters of the thermal spike can be deduced from the data, providing information about the physical processes. We believe that systematic studies of $g(E)$ in various solids in a broad range of ion energies are useful for the separation of these contributions, and that they can provide important information about the mechanisms of energy relaxation.

Recently, Gaiduk, Komarov, and Wesch reported on ion-induced tracks in InP at $S_e>13$ keV/nm after predamaging with high ion fluences. No tracks were observed without preirradiation, and the number of ions was much higher than the number of tracks.[15] Our opinion is that the predamaging of the InP samples modified the electron properties and reduced the value of the $a(0)$ parameter. The complex depth dependence of the types and concentration of various defect structures was the result of variations of S_e and $a(0)$ with the depth. Without predamaging, Eq. (1) provides $S_{et}=23.7$ keV/nm for the beams in Ref. 15; we believe that no isolated tracks form in a virgin InP single crystal at $S_e<S_{et}$.

While in insulators the problem is why the width of the thermal spike $a(0)$ is constant, in semiconductors we have to explain the reason for the changes in $a(0)$. In Fig. 4 we depict $a(0)$ versus $E_g^{-1/2}$ and the results of the analysis of experiments on Si (Ref. 3) and Ge (Ref. 5) also fit well to the line. Since GeS has highly anisotropic transport properties that strongly affect the formation of tracks[16] it was omitted in this analysis. The estimate from Fig. 4 is $a(0)=7.4$ nm for GaAs. Equation (1) provides only a slightly higher value [$a(0)=7.8$ nm] when $S_{et}=S_e$ is assumed at $E=1.85$ MeV/nucleon. The results show that in GaAs the formation of tracks by monoatomic ions is almost possible. Previously, tracks could be induced by monoatomic beams neither in Ge nor in Si, both having high values of $\rho c T_o$. Thus the results in Table I demonstrate that low values of $\rho c T_o$ are advantageous for track formation. According to the figure, the lowest possible value in these semiconductors is $a(0)=3.5$ nm. This is in reasonable agreement with that observed in insulators.

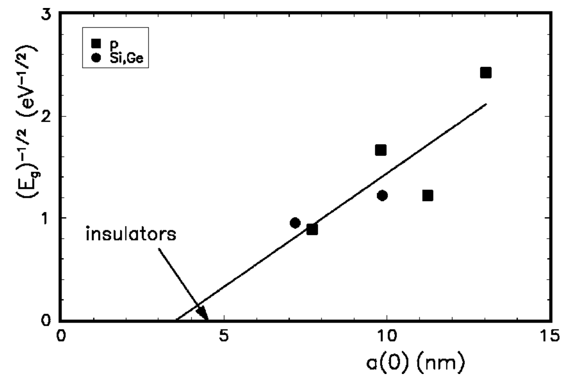

FIG. 4. Variation of the Gaussian width of the thermal spike $a(0)$ with the gap energy E_g; p, present study. The data are given in Table I.

In insulators the thermal diffusivity D does not affect $a(0)$.[17] In semiconductors the spike is broadened [$a(0)>4.5$ nm], and $a(0)$ may have different values. In semiconductors the charge carriers are mobile, and we assume that the thermal diffusivity may contribute to $a(0)$. Thus $a(0)$ becomes a function of $(\tau D)^{1/2}$, where τ is the energy relaxation time. In the high-temperature region of the spike the majority of lattice atoms are ionized. The average ion concentration is roughly proportional to E_g^{-1}; thus τ increases with E_g. On the other hand, a high gap energy impedes the spatial expansion of the electron excitation.[18] The two effects are opposite; therefore, the dependence of $a(0)$ on E_g is expected to be weak. We did not find correlation between $a(0)$ and the room-temperature values D_{rt}. Obviously, in the conditions of a thermal spike D_{rt} is not appropriate. We note that we could scale the R_e^2-S_e track evolution curves without the E_g values[1,12,14,16] for those tracks, which were recently analyzed by Toulemonde *et al.*[19] For all those solids $a(0)=4.5$ nm as in insulators. Thus our approach provided no E_g dependence for those tracks, in contrast with the conclusion in Ref. 19. For our semiconductors scaling is possible only when $a(0)$ is considered as a variable, and this introduces a dependence on E_g.

Our opinion is that the effective mass m^* may also play an important role in the energy transfer process as D and τ both depend on the effective mass. The relevance of m^* is also supported by our analysis of track formation in anisotropic crystals (HTCS's, GeS, MoS_2, mica), where the importance of the anisotropy of m^* was obvious.[16] In this experiment we could not observe the effect of m^* as $m^* \sim E_g$ for our semiconductors.

The low number of reports on track formation in semiconductors is not due to the lack of interest. Rather, it is a consequence of the uncertainty of experimentators in the formation of tracks in their irradiation experiment. We believe that Fig. 4 and Eqs. (1) and (2) may be useful for a rough prediction of whether tracks can be induced or not in the given experimental conditions. Obviously, the description of the formation of tracks in terms of E_g and m^* is oversimplified. A considerably larger set of experimental data is necessary to include further parameters in the analysis. Additional experiments are also needed to evaluate how general our conclusions are regarding $g(E)$ and the $a(0) \sim (E_g)^{-1/2}$ relation.

IV. CONCLUSIONS

In summary, isolated, cylindrical tracks were induced in InSb, InAs, GaSb, and InP by Pb ion irradiation, and no tracks were found in GaAs. The results confirmed that the term $\rho c T_o$ affects track formation in semiconductors. Recent results on tracks in Si and Ge induced by C_{60} ions were also included into the analysis. Compared to insulators, a considerably lower fraction of the projectile energy loss is transferred to the thermal spike at low ion velocity, and the variation of $g(E)$ is small in the range of $2 < E < 10$ MeV/nucleon. The expression $a(0) = b + c(E_g)^{-1/2}$ (b and c are constants) provides a good approximation for the width of the ion-induced thermal spike in our semiconductors and in Ge and Si.

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