

## Electron-Impact Ionization and Energy Loss of 27-MeV/u Xe<sup>35+</sup> Incident Ions Channeled in Silicon

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We have measured the emerging charge-state distribution of 27-MeV/u Xe<sup>35+</sup> beams channeled through a thin Si single crystal, and deduced electron-impact-ionization cross sections for Xe<sup>35+</sup> to Xe<sup>45+</sup> by 14.7-keV electrons. They are  $\approx 2$  to 4 times higher than predicted by usually accepted empirical estimations. We have also measured the energy loss versus the emerging charge state. For hyperchanneled Xe<sup>Q+</sup> ions, the stopping power depends only on the mean (and not on the actually sampled) density of valence electrons and compares well with the prediction of the electron-gas model.

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Recent experiments have demonstrated that channeling is a powerful tool for radiative-electron-capture (REC) studies and electron-impact-ionization (EII) cross-section measurements,<sup>1,2</sup> the usually dominant role of target nuclei on charge exchange being suppressed. We report here on EII and energy-loss measurements for 27-MeV/u Xe<sup>35+</sup> beams sent through a thin Si crystal. When these ions are well channeled, the distribution of charge states  $Q_{\text{out}}$  at emergence,  $F_c(Q_{\text{out}})$ , results mainly from EII (REC cross sections are very small here). This distribution is thus steadily displaced towards higher  $Q_{\text{out}}$  for increasing crystal thicknesses, in sharp contrast with the random case where  $F_R(Q_{\text{out}})$ , arising from the balance between the charge-exchange processes induced by interactions with target nuclei, gets rapidly equilibrated around  $Q_{\text{out}}=50$ . In our experiment, the crystal thickness was such that the most probable  $Q_{\text{out}}$  in the alignment condition was 44, and we have determined electron-impact single-ionization cross sections for Xe<sup>35+</sup> to Xe<sup>45+</sup> by 14.7-keV electrons (kinetic energy of Si valence electrons in the rest frame of the ion) by fitting  $F_c(Q_{\text{out}})$  via a Monte Carlo calculation. The experiment also provides the emergent energy distribution  $G(E_{\text{out}}|Q_{\text{out}})$  for each  $Q_{\text{out}}$  value.  $Q_{\text{out}}$  tends to increase with the electronic density  $\rho_e$  sampled by each ion along its path and thus with its transverse energy  $E_{\perp}$ . Thus, the fit of  $G(E_{\text{out}}|Q_{\text{out}})$  by Monte Carlo calculations connects  $E_{\perp}$ ,  $\rho_e$ , and the stopping power quite precisely and sheds light on local and nonlocal aspects of energy-loss processes.

The experiments were performed at GANIL on the LISE beam line.<sup>3,4</sup> We used the same (111) 17- $\mu\text{m}$ -thick Si single crystal as in Ref. 1. The thickness was measured in various places by infrared interferometry and the nonuniformity was found to be less than  $\pm 3\%$ . The experimental setup is described in Ref. 1. The beam currents were 0.1 to 1 nA. The channeling axis was [110] and thus the path length in the crystal was 20.8  $\mu\text{m}$ . The charge-state and energy distribution of the transmitted beam was analyzed with a magnetic dipole followed by a wire chamber. The optics was set in such a way that angular divergence of the outgoing beam had a negligible influence on the wire-chamber response. The overall energy resolution was  $\approx 20$  keV/u and the counting dynamics of the wire chamber was  $\approx 10^6$ .

The results presented here rely on good and controlled channeling conditions. This is illustrated in Fig. 1, where two angular scans across the [110] axis are presented. The broad one (HWHM  $\Psi_{1/2}=1.01$  mrad) corresponds to Lyman- $\alpha$  photons emitted by Xe ions in the crystal. For Xe<sup>35+</sup> incident ions such an emission requires the creation of a K-shell vacancy which, at our energy, can result only from close encounters with target nuclei. The corresponding measured minimum yield,  $\chi_{\text{min}}=(2\pm 0.2)\%$ , is very low, in agreement with Monte Carlo simulations.<sup>5</sup> This confirms the high quality of the crystal and shows that beam angular divergence is small compared to  $\Psi_{1/2}$ . The sharp peak in Fig. 1 represents the angular dependence of the  $Q_{\text{out}}=37$  fraction ( $1\times 10^{-3}$  for perfect alignment). Its very low HWHM,

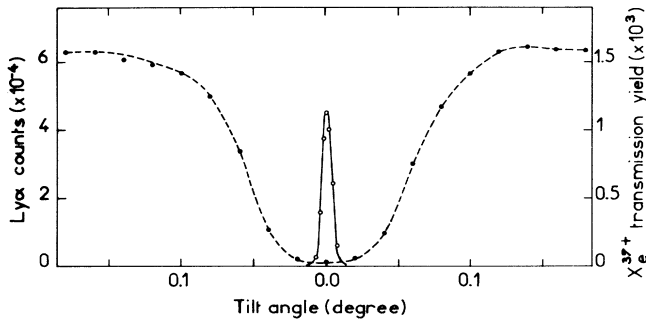


FIG. 1. Angular scans across the [110] axis. Solid circles: Xe Lyman- $\alpha$  photon yield. Open circles: fraction of emerging Xe<sup>37+</sup>. The line through the open circles is the result of a Monte Carlo simulation.

$\Psi'_{1/2} = 0.075$  mrad  $= 0.075\Psi_{1/2}$ , demonstrates that the considered ions are hyperchanneled. Tilting the crystal by 0.12 mrad reduces the 37+ fraction by 1 order of magnitude, while the electronic density sampled by the best-channeled ions is only increased by  $\approx 20\%$  (as calculated by the Monte Carlo method). This scan thus shows that selecting low outgoing charge states is a formidable tool for selecting low transverse energies.

The outgoing charge-state distributions for a random crystal orientation and for a [110]-aligned geometry are displayed in Fig. 2. In the random case, an equilibrium distribution is obtained with  $Q_{\text{out}} = 49.6$ , close to the value  $Q_{\text{out}} = 49.3$  found for 25.5-MeV/u incident Xe<sup>53+</sup>.<sup>1</sup> The distribution  $F_c(Q_{\text{out}})$  obtained with the aligned beam is very broad, ranging from  $Q_{\text{out}} = 35$  (i.e., without any charge exchange) to  $Q_{\text{out}} = 53$  (H-like ion). The  $Q_{\text{out}} \geq 50$  part of the distribution corresponds mainly to the nonchanneled part of the beam. Most of the ions with  $47 \leq Q_{\text{out}} \leq 49$  are still channeled as shown by their energy loss. However, they have rather high mean transverse energy and may also lose electrons by interaction with strongly thermally displaced Si nuclei (the ratio between electron- and nucleus-impact-ionization cross sections is  $\approx 3 \times 10^{-3}$ ). For  $Q_{\text{out}} \leq 46$ , neglecting nucleus-impact ionization is a good approximation and the  $35 \leq Q_{\text{out}} \leq 46$  part of the distribution was fully analyzed by using a Monte Carlo simulation that we describe now.

We used electronic wave functions of isolated Si atoms to determine the core electronic density  $\rho_c(\mathbf{r})$  ( $K$  and  $L$  shells) and the Fourier coefficients from the nonlocal pseudopotential calculations of Ref. 6 to calculate the valence electron density  $\rho_v(\mathbf{r})$ . The potential  $U(\mathbf{r})$  for a unit charge was then calculated with these electronic densities. Both  $\rho_e = \rho_c + \rho_v$  and  $U$  were averaged along the [110] direction, leading to  $\rho_e(\mathbf{r}_\perp)$  and  $U(\mathbf{r}_\perp)$  maps.<sup>7</sup> In the simulation, a statistical equilibrium was assumed to be rapidly reached. The accessible transverse space for an ion of given  $E_\perp$  and the mean electronic density  $\langle \rho_e(E_\perp) \rangle$  it experiences may then be determined from

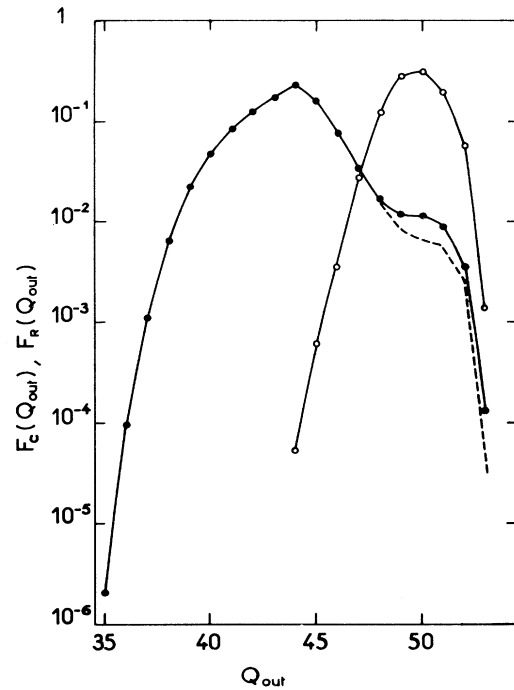


FIG. 2. Emerging charge-state distributions. Open circles: random conditions; solid circles: [110] alignment. For  $Q_{\text{out}} \geq 48$ , the specific contribution of channeled ions is also represented (dashed curve).

the  $U(\mathbf{r}_\perp)$  and  $\rho_e(\mathbf{r}_\perp)$  maps. For  $L$ - and  $M$ -shell Xe electrons, EII is a close-encounter process; thus the ionization probability for a particle of given  $E_\perp$  was assumed to be proportional to  $\langle \rho_e(E_\perp) \rangle$ . Consequently, the precise determination of the EII cross sections requires a good knowledge of the  $E_\perp$  distribution. A transverse energy was assigned for each incoming particle through its entrance coordinate and direction with respect to the axis. The beam angular profile was not directly measurable in the experiment; the only indication is provided by the narrow scan in Fig. 1. Thus two extreme assumptions were made: (i) The profile is Gaussian with  $\text{HWHM} = 0.06$  mrad  $\approx \Psi'_{1/2}$ ; this probably underestimates the overall angular beam spread and consequently the mean electron density sampled by the beam, leading to an overestimation of the EII cross sections. (ii) 20% of the beam has, as above, a narrow Gaussian distribution, the remaining 80% being in wide Gaussian wings ( $\text{HWHM} = 0.35$  mrad). This wide angular spread leads ultimately to an underestimation of the EII cross sections. Both hypotheses give still satisfactory fits of the two scans in Fig. 1, but the energy-loss spectra  $G(E_\perp | Q_{\text{out}})$  are much better adjusted with assumption (ii). For each particle,  $E_\perp$  changes along their path induced by multiple scattering from target electrons have been taken into account.

For well-channeled ions ( $Q_{\text{out}} \leq 46$ ), electron capture

is negligible. Thus, fitting  $F_c(Q_{out}=35)$  gives the EII cross section  $\sigma(35 \rightarrow 36)$ . With this value known, the fit of  $F_c(Q_{out}=36)$  provides  $\sigma(36 \rightarrow 37)$ . The procedure is iterated and leads to the two sets of  $\sigma(Q \rightarrow Q+1)$  presented in Fig. 3, corresponding, respectively, to assumptions (i) and (ii). The EII cross section determined corresponds to both direct and indirect (i.e., starting by excitation) ionizations. We have neglected double ionization. The sharp decrease observed between  $\sigma(43 \rightarrow 44)$  and  $\sigma(44 \rightarrow 45)$  corresponds to a shell effect:  $Xe^{44+}$  has an empty  $M$  shell. In Fig. 3 we compare our results to those of Donets,<sup>8</sup> to the predictions from the Lotz empirical formula,<sup>9</sup> and to a calculation using the Thomson formula, with a cutoff for energy transfer below the binding energy. For the two latter comparisons,  $Q$ -dependent binding energies were used.<sup>10</sup> Our cross sections are 2 to 4 times larger than the predicted ones, and 1 to 3 times higher than those measured by Donets. The latter difference may be due to multistep ionization taking place in our experiments, which, due to the low electron flux in crossed-beam measurements, cannot happen in Donets' experiment. In Ref. 2, the authors, assuming, from REC measurements, that the mean electron density  $\langle \rho_e \rangle$  sampled by the channeled ions is 6.2 electrons per Si atom, found cross sections in reasonable agreement with the Lotz theoretical predic-

tions for  $L$ -shell ionization and markedly higher than predicted for  $K$ -shell ionization. Our simulations yield  $\langle \rho_e \rangle$  values 2 to 2.5 times smaller, which are in fair agreement with the REC yields measured in Ref. 1.

As for energy losses, we have measured the random value  $\langle \Delta E_R \rangle$  in the crystal (1.32 MeV/u; i.e., a stopping power of  $35.2 \text{ MeV cm}^2 \text{ mg}^{-1}$ ) which agrees within 3% with tables.<sup>11</sup> In Fig. 4 we present the measured mean energy loss in channeling as a function of  $Q_{out}$ ,  $\langle \Delta E_C(Q_{out}) \rangle$ . In particular,  $\langle \Delta E_C(35) \rangle = 0.21 \langle \Delta E_R \rangle$ . The increase of  $\langle \Delta E_C \rangle$  with  $Q_{out}$  is due to (i) the stopping-cross-section increase with the ion charge state and (ii) the increase of energy loss with encountered electronic density. In order to isolate the latter effect, we show also in Fig. 4 the reduced mean energy loss  $\epsilon_C(Q_{out}) = \langle \Delta E_C(Q_{out}) \rangle (35)^2 / \langle Q^2(Q_{out}) \rangle$ , where  $\langle Q^2(Q_{out}) \rangle$  is the mean square charge of Xe ions inside the crystal for a given  $Q_{out}$  (calculated by simulation).  $\epsilon_C$  is a good parameter since the  $Xe^{Q+}$  are nearly point charges.  $\epsilon_C(Q_{out})$  is found to be constant for  $35 \leq Q_{out} \leq 43$  ( $\epsilon_C = 0.4 \epsilon_R$ ) and is very close to the theoretical energy loss of a 27-MeV/u  $Xe^{35+}$  point charge in a uniform free electron gas with  $\rho_e = \rho_v = 0.2 \text{ e \AA}^{-3}$  (plasmon energy  $\hbar \omega_p = 16.6 \text{ eV}$ ). For each  $Q_{out}$ , our simulation provides the conditional mean values  $\langle E_{\perp}(Q_{out}) \rangle$  and  $\langle \rho_e(Q_{out}) \rangle$ . With assumption (ii), for the ions with constant  $\epsilon_C$ ,  $\langle \rho_e(Q_{out}) \rangle$  varies between  $0.2 \rho_v$  (for  $Q_{out}=35$ ) and  $0.9 \rho_v$  (for  $Q_{out}=43$ ): Energy

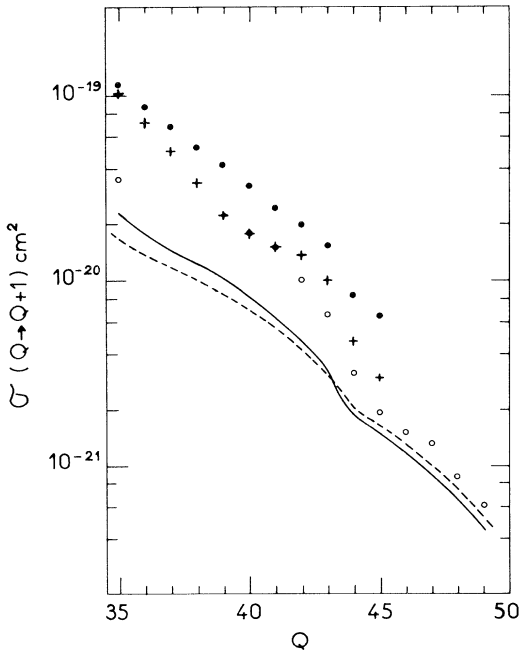


FIG. 3. Electron-impact-ionization cross sections  $\sigma(Q \rightarrow Q+1)$  of  $Xe^{Q+}$  ( $35 \leq Q \leq 45$ ) by 14.7-keV electrons. Solid circles: our measurements [assumption (i)]; crosses: our measurements [assumption (ii)]; open circles: experimental results by Donets (Ref. 8); solid curve: calculation from Lotz formula (Ref. 9); dashed curve: calculation from Thomson formula (see text).

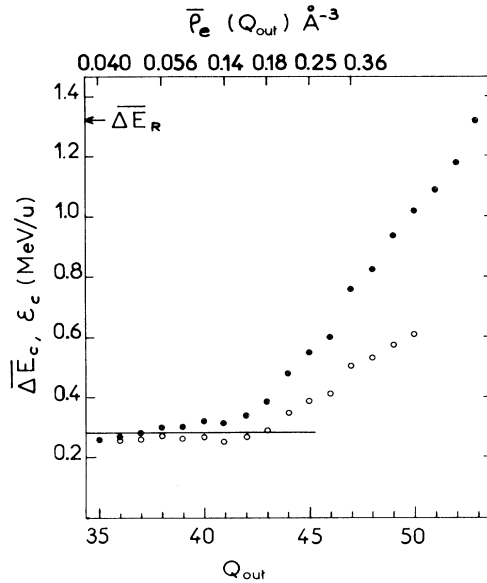


FIG. 4. Mean energy loss for [110] alignment (path length  $20.8 \mu\text{m}$ ) vs charge state at emergence ( $Q_{out}$ ). Solid circles: experimental results  $\langle \Delta E_C(Q_{out}) \rangle$ . Open circles: reduced mean energy loss  $\epsilon_C = \langle \Delta E_C(Q_{out}) \rangle (35)^2 / \langle Q^2 \rangle$ . The mean encountered electron density  $\langle \rho_e(Q_{out}) \rangle$  for each  $Q_{out}$  is also given on the upper scale [assumption (ii)]. The theoretical  $\epsilon_C$  in an electron gas of uniform density  $\rho_v$  corresponds to the solid horizontal line.

loss to valence electrons is thus essentially nonlocal. This may appear surprising: Since roughly half the energy loss is due to binary collisions with energy transfer  $> \hbar\omega_p$ ,<sup>12</sup> the stopping power is expected to depend on the locally sampled electron density. An explanation may be that the impact parameter  $P_c$  for an energy transfer  $\hbar\omega_p$  to a free electron is very high ( $P_c = 1.2 \text{ \AA}$ ). It is then reasonable to assume that even hyperchanneled ions may have significant "binary" interactions with all the electrons from the valence gas.<sup>13</sup> We have already seen that the charge state reached by an ion is determined by the local electron density it experiences. For hyperchanneled ions, this density thus has an influence on  $\langle \Delta E_C \rangle$ , via the  $\langle Q^2(Q_{\text{out}}) \rangle$ , if not on  $\epsilon_C$ . For  $Q_{\text{out}} \geq 44$ , the steady increase of  $\epsilon_C(Q_{\text{out}})$  with  $Q_{\text{out}}$  (and then with  $E_{\perp}$ ) is explained by increasing interactions with  $L$ -shell Si electrons: Exciting these electrons requires impact parameters lower than  $0.5 \text{ \AA}$  and the mean transverse energy ( $E_{\perp}(Q_{\text{out}}=44)$ ) corresponds to a distance of approach to the strings of  $0.7 \text{ \AA}$ ; considering thermal vibrations and the spatial extension of the  $L$  Si electron cloud, one understands that ions with  $Q_{\text{out}}=44$  may have excited these core electrons. A more detailed analysis of the full energy-loss distributions as a function of  $Q_{\text{out}}$  will be presented in a forthcoming paper.

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