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Characterization of Charge States of Energetic Ions in Solids from Associated K X-Ray Production*

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The strong projectile–charge-state dependence of K x-ray production in gases establishes a charge-state scale for ions penetrating solids. Si K x-ray cross sections in gaseous SiH_4 were compared to those in solid Si for 40-MeV O (6+ to 8+) and 86-MeV Ar (6+ to 16+) projectiles. For Ar an effective charge of 11 ± 1 is found compared to an emergent charge of 14.8 ± 0.5. The result is discussed in terms of alternate models for steady-state excitation in solids.

The states of energetic ions penetrating solids have long been a subject of considerable study. In 1951 Lassen[1] found that the charge states of heavy ions emerging from solids were considerably higher than those emerging from gaseous targets. However, it is also found that the electronic stopping powers in solid media are essentially the same as those measured in gases, and the "effective charge" derived from stopping powers in solids is that which is observed for ions in gases.[2,3] The first of two solutions to this disparity suggests that dynamical screening by electrons in the solid tends to neutralize the excess charge on the ion moving in the solid.[4,5] However, at velocities $v \gg v_0$ the effect of screening should be small, and recent experiments with channeled oxygen ions indicate that screening electrons are ineffective in altering the stopping power of an ion in a given charge state.[6] An alternative explanation was proposed by Betz and Grodzins[7] who suggested that the actual charge state in the solid is much like that in the gas but that many of the bound electrons are in highly excited states and are lost by Auger events after emergence. Hence, comparison of charge states inside the solid with emergent charge states provides an experimental test of these theories. Related studies of exit-channel effects[8] in quasimolecular excitation and of x-ray satellite structure generated by low-energy ions in solids[9] yield information concerning average *excitation* states but do not permit direct determination of effective *charge* states in solids.

We utilize the observation that x-ray production cross sections for high-velocity (1–4 MeV/nucleon) ions are highly sensitive to the charge state of the projectile ion.[10] We reasoned that comparing the x-ray production cross sections in solid and gaseous targets containing the same target element would provide a measure of the charge state of the penetrating ion.[11] The results of these experiments are also critical to interpretation of measured heavy-ion–induced x-ray cross sections in solid targets.

Beams of 86-MeV Ar^{6+} from the Oak Ridge isochronous cyclotron and 40-MeV O^{6+} from the

Oak Ridge tandem Van de Graaff accelerator[11] were passed through foils for further stripping. A pure-charge-state beam was then magnetically selected and entered a differentially pumped windowless gas cell. The cell could be filled with SiH_4 gas at pressures of ~20 mTorr, and x rays produced were counted by a Si(Li) detector viewing the collision region at 90° to the beam. The maximum estimated charge exchange in the gas was always $\lesssim 5\%$ so that single-collision conditions with ions of known charge state were obtained. The measurement techniques have been fully described previously.[10] The solid targets consisted of thin Si films (7–50 $\mu g/cm^2$) deposited on a 150-$\mu g/cm^2$ plastic backing which could be positioned at the center of the evacuated gas cell. The Si surface faced the beam and was tilted at an angle of 36° so that the Si(Li) detector could view the Si surface directly. In the Ar experiments an additional Si foil could be inserted about 30 cm upstream of the cell. An entrant charge-state distribution characteristic of that emerging from solid Si could thus be prepared. The particle current was alternatively determined by Rutherford scattering from a gold foil placed behind the cell, or by a separated pumped, electrically and magnetically guarded, Faraday cup.

Charge-state distributions for the oxygen ions emerging from Si were measured by using electrostatic analysis and a position-sensitive detector. The charge fractions F_i were 0.2, 0.7, and 0.1 for 6+, 7+, and 8+, respectively. The charge-state distributions for Ar ions emerging from a Si target were obtained by measuring the current at the focus of a magnetic analyzer following a Si stripper foil. The charge fractions were 0.11 (13+), 0.27 (14+), 0.34 (15+), and 0.20 (16+), to an estimated accuracy of better than ±0.03.

Results for O ions are shown in Fig. 1. With the gas target the Si *K* x-ray cross section varies by a factor of ~2.5 as the incident ion is changed from 6+ to 8+. A smaller variation (~60%) is observed when a solid target is used. This effect of incident charge state in thin solid targets has been recently reported for O ions in Al by Brandt *et al.*,[5] who found that the effect diminishes as the foil thickness is increased, and attributed this charge-state effect to dynamic screening. An alternative explanation could lie in the inequilibrium of the ion charge state. Capture and loss cross sections for 40-MeV O ions in Ar, for example, are on the order[12] of 2×10^{-17} cm². Thus the Si target thickness (~2 $\times 10^{17}$ atoms/cm²) should be insufficient to attain equilibrium.

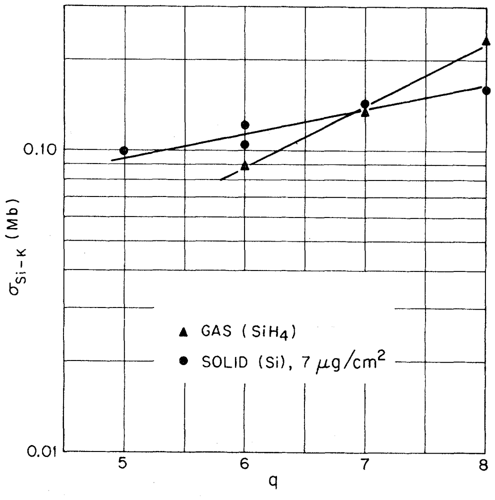

FIG. 1. Si *K* x-ray production cross section in megabarns produced by 40-MeV O-ion bombardment of SiH_4 and solid Si as a function of incident-ion charge state.

The observed charge-state dependence is expected to evolve with increasing target thickness from the rising straight line observed for thin, gas targets to a horizontal straight line at equilibrium thickness. The two lines representing the extremes in density should intersect at the projectile charge corresponding to the effective equilibrium charge in the solid. The emergent equilibrium charge-state distribution of 40-MeV O is strongly peaked at 7+. At equilibrium, the input-charge dependence of the x-ray cross section would be a horizontal line intersecting the SiH_4 line at charge 7+, if the effective charge in the solid were equal to the most probable emergent charge. Because the cross sections for leaving a charge state of 7+ ($\sigma_{7,6}$ and $\sigma_{7,8}$) should be lower at this energy than those for attaining a charge of 7+ (i.e., $\sigma_{8,7}$ and $\sigma_{6,7}$), one would expect near coincidence with the 7+ value of the gas target for incident 7+ ions even before charge-state equilibrium is attained. Arguments based on dynamic screening in which the number of bound electrons are not considered lead to almost the same expected effective-charge state. Here

$$q_{\mathrm{scr}}=Z_{\mathrm{core}}(1-\gamma v_{\mathrm{F}}/v), \tag{1}$$

where v_{F} is the target-electron Fermi velocity ($\sim v_0$), Z_{core} is the core-electron-screened nuclear charge, and γ is an electron "enhancement

TABLE I. Summary of the data. Bracketed values are for 40-MeV O; unbracketed values are for 86-MeV Ar.

	X-ray data	Measured distribution	Semiempirical theory[a]
Mean charge emerging from solid	14 ± 0.5	14.8 ± 0.3 [6.9 ± 0.2]	13.7 [7.6]
Mean charge emerging from gas			12.5 [7.4]
Effective charge state in solid	11 ± 1 [7.0 ± 0.25]		

[a] Ref. 14.

factor"[5] ~1. For 40-MeV O, $q_{\text{scr}} = 7.2$.

For relatively low-Z ions such as oxygen only a slight dependence of the emergent charge state on the medium (solid or gas) is expected.[13] The semiempirical calculations of Dmitriev and Nikolaev[14] give 7.4 and 7.6 for the expected emergent charge (Table I). From our charge-distribution measurements we obtain $\bar{q} = 6.9$, and the effective charge state (q_{eff}) indicated by the measured x-ray cross section is 7 ± 0.25. Hence, the oxygen results are consistent with expectations and support the validity of our method.

The different situation for Ar ions is shown in Fig. 2. The cross sections for Si K x-ray emission vary by a factor of ~7 for Ar charges ranging from 6+ to 16+. The cross section for Si K x-ray emission from solid Si was found to be 0.095 ± 0.005 Mb and was essentially independent of input charge and target thickness. This cross section corresponds to an effective charge state of 11 ± 1 and is to be compared with our measured emergent mean charge of 14.8. The corresponding predictions of Dmitriev and Nikolaev[14] are 13.7 for solids and 12.5 for gases.

A useful experimental check was made by preparing an equilibrium-distribution beam in the upstream Si foil referred to above. This equilibrium beam yielded a mean cross section σ_m in SiH_4 approximately equal to that anticipated on the basis of a sum of the single-charge-state cross sections weighted by the corresponding F_i. When the equilibrium beam was passed through a solid Si target, no difference was observed in the cross section derived from the results for single-charge-component beams. From this prefoil experiment alone it can be seen that the cross section in a solid target is a factor of ~2 lower than that anticipated for the measured emergent charge.

One can exclude three unrelated effects which might be expected to cause significant differences in the observed yields from solid and gaseous targets. The possibility of energetic Si recoils causing additional Si K vacancies in the solid can be assessed by using information available on Al-Al ($Z = 13$) collision-induced K x rays.[15] Here a threshold is observed at 150 keV; the cross section rises rapidly to 5×10^{-22} cm^2 at 300 keV. The impact parameter b required to impart $\Delta E = 200$ keV to a Si atom by an ion with energy $E \simeq 2$ MeV/nucleon can be estimated from

$$b = Z_1 Z_2 e^2 M_1^{1/2} (M_2 E \, \Delta E)^{-1/2}$$

to be ~4×10^{-12} cm, which clearly yields cross sections negligible compared with processes having cross sections on the order of 10^{-18} cm^2. One must also take into account possible fluores-

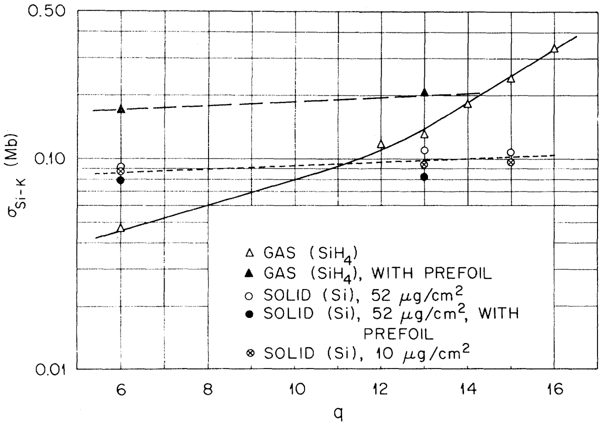

FIG. 2. Si K x-ray production cross section produced by 86-MeV Ar-ion bombardment of SiH_4 and solid Si as a function of incident-ion charge state. The "prefoil" points denote experiments in which the beam passed. through a Si foil before entering the target.

cent-yield differences for struck atoms recoiling in solids as opposed to vacuum, which are mainly determined by the corresponding L-shell electron configurations. That these differences are insignificant is indicated by the fact that the K-shell ionization is in the diabatic region (i.e., $\hbar v/I > r_K$) so that the impact-parameter dependence of the probability of ionization is peaked[16] near the K-shell radius ($r_K = 0.06$ Å). At this distance the energy transfer is only ~10 eV so that even interactions of the L shell of the recoiling Si with lattice atoms (which could affect the K fluorescence yield) are not possible. Finally, the effect of binding electrons (in the M shell) is not expected to be significant, especially since the Si valence electrons in both SiH_4 and solid Si are in the sp^3 configuration.

The results for O ions, exhibiting agreement with expectations, indicate that other possible solid-state effects on the Si K x-ray yield are small. The results for Ar at the same energy per nucleon clearly indicate a lower effective charge for the ion while in the solid than is observed in the emergent beam. The screening model[5] could be used to explain the effect but would require an unrealistically high value of $\gamma = 3$ with $Z_{\text{core}} = 16$ in Eq. (1). The model of Betz and Grodzins[7] would account for the difference by the loss of bound electrons upon emergence by Auger or autoionizing processes. However, the additional electrons present while the ion is in the solid must be in close enough proximity to the Ar nucleus (e.g., L shell) to act in reducing the Si atom's K ionization cross section; and, in the absence of K vacancies, there is no mechanism for autoionizing L electrons. Although the mechanism for charge-state change remains obscure, the work described herein constitutes direct evidence that high-velocity ions in solids have lower charge states than those observed in emergent beams.

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