Production of Highly Excited States of Fast Ions Emerging from Solid Targets

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Production of projectile Rydberg states in fast-ion–foil collisions is shown to exhibit a pronounced target-thickness dependence, proving that Rydberg states are not predominantly formed by direct capture of *target* electrons. Instead, it is proposed that ionized *projectile* electrons can undergo transition back to Rydberg states when the ion emerges from the foil.

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It has been known for a long time that projectile ions directed through thin foil targets may emerge in a variety of electronic excitation states. Beam-foil spectroscopy and related fields have utilized radiative decays of foil-excited atomic beams as a light source for some decades and the availability of heavy-ion accelerators enabled similar research with respect to inner-shell transitions in few-electron ions with high nuclear charge Z. Despite this wide-spread interest it is not yet possible to predict the degree of projectile excitation obtained as a result of ion-solid collisions. Especially for fast collisions, when projectile velocity v exceeds the first Bohr velocity $v_0 = e^2/\hbar$, it is not even clear what kind of mechanisms are effectively producing the observed highly excited states.

Our interest is directed to the formation of Rydberg states rather than core states. In this context, core states are defined by their orbital dimensions which are required to be smaller than the lattice spacing (~2 Å in carbon targets) so that processes involving these states can be described similarly in both binary ion-atom and ion-solid encounters. By contrast, more highly excited states are assumed not to be formed inside a solid because of screening of the target atoms and the rapid collision frequency. As a consequence, it is safe to assume that the actually observed highly excited (Rydberg) states are formed asymptotically during and after passage of the projectiles through the exit surface of the foil target.

For slow collisions, an often proposed model[1,2] assumes that excited states which are prohibited inside a solid are formed by Coulomb capture of target electrons from the last layer of atoms in the solid just when the ion exits into the vacuum. Evidence for the validity of such a "last-layer" model for slow collisions has been frequently presented[2,3] but decisive questions remain: Projectile excitation to higher Rydberg states in the last layer, for example, has not been considered although this process might be of particular importance. It is purpose of this Letter to demonstrate that Rydberg states of fast ions are not formed predominantly by direct capture of *target* electrons. We find, instead, a connection between *projectile* core electrons and final Rydberg states.

125-MeV sulfur ions were obtained from the Munich tandem Van de Graaff accelerator, passed through a prestripper to yield primary charge states 16^+ and 15^+, magnetically analyzed, and directed onto carbon targets with thicknesses x ranging from 2 to 220 $\mu g/cm^2$. A Si(Li) detector with energy resolution of 150 eV (full width at half maximum at 5.9 keV) was positioned at various distances, $s = 2$, 6, 8, and 200 cm, behind the foil target and detected sulfur K x rays emitted at 90° with respect to the beam direction. Detector resolution was sufficient to separate heliumlike and hydrogenlike $K\alpha$ and $K\beta$ transitions in sulfur which have energies from ~2.4 to ~3.5 keV. The vacuum in the beam line was good enough ($<10^{-6}$ Torr) to ensure a negligible amount of beam excitation due to collisions with residual gas atoms. Beam intensity was monitored by means of a Faraday cup.

Figure 1 presents the measured target-thickness dependence of three K-shell x-ray transitions observed 2 cm behind the foil: prompt Ly-α and Ly-β transitions which occur as a result of cascades of long-lived Rydberg states[5] to the $2p$ and $3p$ level, respectively, and the two-photon decay ($2s \rightarrow 1s$) of the metastable $2s$ core state. It has been verified in separate experiments[6,7] that the $2E1$ intensity decays exponentially and reflects the $2s$ population of the ions induced by collisions throughout the target, and that the intensities of the Lyman lines exhibit a power-law dependence, $I_{\alpha,\beta} \propto s^{-a}$, and signify the decay of long-lived Rydberg states. Increasing the observation distance s allows detection of decays

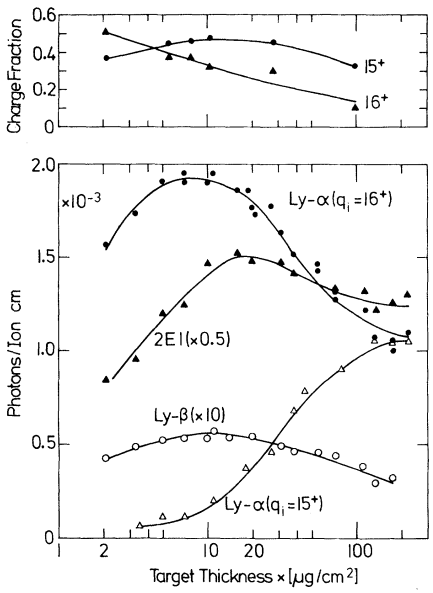

FIG. 1. Absolute K-shell x-ray transition intensities (bottom) and charge-state fractions (Ref. 4) (top) for 125-MeV sulfur ions, as a function of the thickness of the traversed carbon foils. Incident ions were fully stripped (16^+); Ly-α yields are also shown for incident charge state 15^+. X-ray observation took place 2 cm behind the foil, a distance which corresponds to 1.2×10^5 decay lengths of the prompt $2p \rightarrow 1s$ transition. $2E1$ denotes the two-photon transition $2s \rightarrow 1s$ in hydrogenlike sulfur. For convenience, actual $2E1$ and Ly-β intensities have been multiplied by 0.5 and 10, respectively.

from progressively higher initially excited quantum states. For example, lifetimes of $50p$ and $50f$ states in S^{15+} are 0.36 and 2 ns and correspond to decay lengths of ~1 and ~5 cm, respectively. It is obvious, therefore, that quite high-lying Rydberg states manifest themselves at centimeter distances behind the foil.[6,7]

Collisions at a high beam velocity, $v = 2.73$ cm/ns, lead to efficient production of few-electron sulfur ions and, as a result of small cross sections for processes involving projectile K electrons, to a relatively large equilibrium thickness for charge states. Figure 1 shows that charge-state equilibrium is not attained below 200 $\mu g/cm^2$ carbon.

When we follow the concept that hydrogenlike Rydberg states are formed via direct capture of last-layer target electrons by bare sulfur ions one would expect an x-ray yield $I_{\alpha,\beta}$ which varies in proportion to the fraction of bare ions, Y^{16+}, which are available at the end of the foil, $I_{\alpha,\beta} \propto Y^{16+}\sigma_c \Delta x_L$. Here, σ_c and Δx_L denote capture cross section and last-layer target thickness, respectively. Inspection of Fig. 1, however, gives no evidence for a proportionality between $Y^{16+}(x)$ and $I_{\alpha,\beta}(x)$. In fact, for small target thicknesses $I_{\alpha,\beta}$ increases while Y^{16+} decreases. An increase of $I_{\alpha,\beta}(x)$ for $x \lesssim 10$ $\mu g/cm^2$ was also found for observation distances $s = 6$, 8, and 200 cm. This finding rules out the conjecture that the dominant part of highly excited projectile states is formed by direct capture of *target* electrons, a conclusion which is independent of the assumed kind of capture process and the precise effective layer thickness Δx_L.

There are two further arguments against electron capture from the last layer, based on absolute population and distribution of angular momentum, l, of Rydberg levels. The expected initial (H-like) Rydberg-state population may be estimated by $N_c(n) = Y^{16+}\sigma_c(n)\Delta x_L$, where n is the principal quantum number of the final state. Using a last-layer thickness $\Delta x_L = 2.2 \times 10^{15}$ atoms/cm^2 (~2 Å) and, for the present system, a cross section for capture of carbon $1s$ electrons $\sigma_c(1s \rightarrow n \gg 1) \cong 1.3 \times 10^{-16} n^{-3}$ cm^2,[8] we obtain $N_c \cong 0.3 n^{-3} Y^{16+}(x)$. Cascading of this initial population gives rise to a delayed x-ray intensity I_β which is more than 2 orders of magnitude smaller than observed.[6] A similar discrepancy was found for fast oxygen ions emerging from carbon.[9] Furthermore, electron capture in fast-ion–atom collisions is expected to populate mainly final states with low l, but an analysis of experimental decay curves $I_{\alpha,\beta}(s)$ reveals that high-l states are heavily populated.[6,7]

Another last-layer effect which has received little attention in the past consists of bound-bound excitation of projectile core electrons in collisions with last-layer target atoms. Excitation cross sections have been calculated for bound-bound transitions $n_i \rightarrow n$ of hydrogenlike atoms[10] and can be evaluated for the present system: $\sigma_{exc}(n_i \rightarrow n \gg 1) \cong 7 \times 10^{-19} n_i^4 n^{-3}$. The relevant core-state fraction $Y_{n_i}{}^{15+}$ can be roughly estimated from the balance of capture and loss inside the target whereby we assume $n_i \leq 5$ (corresponding to orbital sizes less than ~2 Å) and take into account that no more than ~40% of all projectile ions are in 15^+ states (Fig. 1). The resulting Rydberg-state population is given by

$$N_{exc} = \sum_{n_i} Y_{n_i}{}^{15+}\sigma_{exc}(n_i \rightarrow n \gg 1)\Delta x_L \simeq 0.2 n^{-3},$$

a population which is approximately of the same magnitude as last-layer capture N_c from above and, thus, much too small to explain the ob-

served cascade decays of Rydberg states. Furthermore, dipole excitation of (low-n) core states does not give access to the high-l states observed as a result of fast beam-foil interaction.[6,7]

Our data suggest a correlation between projectile core electrons and final asymptotic Rydberg states: Projectile core electrons are ionized inside the target and become continuum electrons with respect to the moving ion. When the projectile leaves the solid a correlated continuum electron ends up as either a (free) convoy[11-14] or a (bound) Rydberg electron.

Free-electron scattering data[15] suggest that the required correlation between electron and ion can be maintained for escape depths from the solid which amount to $\Delta x_e \lesssim 20$ Å, while experimentally observed charge-state variations $Y(x)$ (Fig. 1, top) and theoretical estimates of charge-changing cross sections[8,16] reveal that the mean free path for both capture into and loss from the involved excited states is of the order of 100 Å (2 μg/cm^2). The number of ionized electrons which escape fairly undisturbed from the solid is given by

$$N_e \cong \sum_{n_i} \sigma_i(n_i)\Delta x_e , \qquad (1)$$

where σ_i signifies reasonably well-known electron-loss cross sections[16] and the summation extends over all bound projectile electrons. Most of the produced continuum electrons have exceedingly small relative velocities in the frame of the ion and, thus, have a chance either to emerge as convoy electrons[12-14] (fraction N_{con}), or to undergo transitions into Rydberg states (fraction $N_{\mathrm{R}} = kN_e$), giving the approximate relation $N_{\mathrm{con}} + N_{\mathrm{R}} \lesssim N_e$. Along these lines, our data from Fig. 1 can be understood as follows.

The observed increase of $I_{\alpha,\beta}(x)$ for small x arises because of the buildup of states Y_{n_i} which contribute effectively to the production of continuum states and, thus, to final Rydberg states. For incident 16^+ ions we have $Y_{n_i}^{15+}(x) \cong Y^{16+} \times \sigma_c(1s \rightarrow n_i)x$, whereby capture proceeds mainly to excited states rather than to the $1s$ ground state.[8] In the case of incident 15^+ ions which, because of the long flight time to the target, are in the $1s$ state, the relatively small ionization cross section for these electrons ($n_i = 1$) prevents a significant contribution in Eq. (1) for small x.

Evaluation of Eq. (1) with excited-state fractions $Y_{n_i}^{15+}$ as used above yields $N_e \cong 2\%$ (per ion). This is in reasonable relation with the value $N_{\mathrm{con}} \cong 1\%$ which results from many measured yields of convoy electrons extrapolated[17] to the present system. In addition, the value $k = 0.5$ suggested by these yield numbers gives a Rydberg-state population $N_{\mathrm{R}} \cong 1\%$ which allows quantitative reproduction of our x-ray yields $I_{\alpha,\beta}$ provided that low- and high-l states are initially populated. The latter condition may not be an unexpected consequence of our model: The spatial distribution of continuum electron densities relative to the correlated ion will acquire some asymmetry with respect to the beam direction during passage through the solid. An angular asymmetry, though, requires inclusion of high-l states in the representation of the final wave function.[18]

A more thorough discussion of the formation of Rydberg states due to transitions of convoy electrons requires much more investigation and is beyond the scope of this Letter. We point, for example, to calculations which show that hypothetical wake-riding (convoy) electrons of 125-MeV sulfur in aluminum are captured into Rydberg states with probability $k \cong 0.7$.[18] In addition, three other mechanisms should be examined which may contribute to a transfer of convoy electrons to Rydberg states: (i) disappearance of the screening potential upon emergence from the solid and, thus, a sudden increase of binding energy by $\Delta E \lesssim Ze^2\omega_p/v$, where ω_p is the plasma frequency, possibly inducing free $\rightarrow$ bound transitions; (ii) radiative electron capture of convoy electrons,[19] and (iii) Coulomb capture in the field reaching out from the target surface. Incidentally, we expect a target-thickness dependence of N_{con} just as is found for I_α, provided that the targets used are thin enough to allow for nonequilibrium with respect to the formation of projectile core states. Such a dependence has not yet been found,[17] but we suspect that previously used targets were too thick.

Finally, it is illuminating to consider the ratio between primary continuum electrons and all those electrons, $N_L = \sum_n \{N_{\mathrm{exc}}(n) + N_c(n)\}$, which are transferred to excited projectile states by collisional last-layer effects. In a first-order approximation we get $N_e/N_L \cong \Delta x_e/\Delta x_L$,[7] i.e., N_e dominates over N_L quite generally in fast collisions, indicating that our present results may be relevant for a variety of other ion-solid collisions.

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