Anomalous Population of Deep Capture States of Fast Ions Emerging from Solid Foils

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The first comparison of core-state populations of initially bare Kr^{36+} ions emerging from gaseous and solid targets is presented. Another new important feature of this experiment lies in the fact that for such fast heavy ions, the single-collision condition is fulfilled even in solid targets. Our results can be explained by a wake-field–induced Stark mixing of the substates in a solid. Further coherence angles and field strength measurement are discussed.

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Several indications of the existence of a wake following a fast ion moving in a solid target have been given in the last few years.[1,2] The wake-induced electric field at the projectile nucleus has been shown to account for its energy loss in matter,[3] and to be strong enough ($\sim 10^8 - 10^9$ V/cm) to produce measurable energy splitting of the $n=2$ manifold of light hydrogenlike ions.[4] Possible related effects on elementary collision processes in solids have never been observed. We report in this Letter new effects of the wake field on the substate population of heavy hydrogenlike ions produced by electron capture in solids.

Anomalous features in the electron-capture process in solids have so far been reported only for the case of electron capture to the continuum or for the production of highly excited ("Rydberg") states.[5,6] All the quoted experiments belong to the case when multicollision effects are of importance even for core states, and it is still not clear whether the excited (or continuum) states are formed by direct ("last layer") capture or relate to electron loss or excitation of various charge-state fractions in the bulk of the target. Especially, *nl-nl'* intrashell excitation may have large effects on the population of highly excited states,[7] but has been seldom considered in the case of ions in solids.[8] Finally, we also not that proposed explanations have ignored a possible effect of the wake-field–induced Stark *mixing* of excited states.

In order to get clear evidence for possible solid effects, new experimental conditions have been achieved in our measurements with fast projectiles of high atomic number, Z_p, where *single-collision conditions* (SCC) are satisfied even in solid targets. Indeed, for projectiles' velocities $v \approx Z_p$ (a.u.),[9] cross sections for ionization or n-n' excitation roughly scale as $n^2 Z_p^{-4}$, intrashell l-l' excitation as $n^4 Z_p^{-4}$, and radiative deexcitation $[\sigma=(nv\tau)^{-1}]$ as $Z_p^3 n^{-3}$. SCC signify large mean free path for ionization or excitation as compared to that for radiative deexcitation or to the target thickness. These conditions are all the better satisfied for heavy projectiles ($Z_p \gg 1$) and low excited states (small n). This led us to study *deep* capture-state population ($2 \le n \le 4$) of 33.2-MeV/u Kr^{36+} ions ($Z_p=36$, $v=35.6$ a.u.). The actual situation is illustrated in Table I. Our values for ionization and n-n' and nl-nl' excitation mean free paths have been calculated by use of the plane-wave Born approximation (with screening and antiscreening effects accounted for)[8]—a procedure giving predictions consistent with available experimental data.[10] When these values are compared with typical target thicknesses of few micrograms per square centimeter (easy to get for carbon targets), it is clear that SCC are achieved in $n=2$ and $n=3$ levels, and that only l-l' excitation may

TABLE I. Mean free path (μg/cm^2) for various processes in the carbon target for the first three excited states of krypton ions with $v=36$ a.u. λ_c, capture; λ_i, ionization; $\lambda_{n\text{-}n'}$, total $n \to n'$ excitation; $\lambda_{np\text{-}nd}$, p-d excitation; λ_{rad}, radiative deexcitation. The lower lines give the same information for 125-MeV sulfur ions (Ref. 5).

Ion	n	λ_c	λ_i	$\lambda_{n\text{-}n'}$	$\lambda_{np\text{-}nd}$	λ_{rad}
Kr^{35+}, 2.8 GeV	2	10000	440	1000	340[a]	15
	3	30000	160	800	90	50
	4	50000	100	700	25	110
S^{15+}, 125 MeV	2	5	12	30	8[a]	130
	3	7	4	22	2	450
	4	12	3	19	0.6	1000

[a] $\lambda_{np\text{-}ns}$.

lead to small effects in $n \geq 4$. In contrast, large deviations from SCC occurring even for core states with lighter projectiles are illustrated by the case of 125-MeV sulfur[5] ($Z_p = 16$).

Wake effects, on the other hand, have very short response time ($\tau_w = 2\pi\omega_p^{-1} \simeq 2\times10^{-16}$ s in carbon, where $\omega_p \sim 25$ eV is the plasma frequency[3]). From theoretical calculations,[3] the wake field at the projectile nucleus has been estimated to be $\sim 10^9$ V/cm in a carbon target, in good agreement with a measured energy loss of 16 MeV/(mg/cm^2). This field induces Stark splitting of the excited states large enough to allow for time evolution of these states in the range of thicknesses considered here, as will be shown in the following.

The experiment was performed on the LISE facility (Ligne d'Ions Super Epluchés: beam line of highly stripped ions) the Grand Accélérateur National d'Ions Lourds (GANIL). 33.2-MeV/u fully stripped Kr^{36+} ions were produced with an efficiency of more than 50% by post stripping of the incident 34.5-MeV/u Kr^{26+} ions in a 7-mg/cm^2 carbon foil. Charge-state selection was insured by the achromatic double magnetic spectrometer of the LISE line. We used for gaseous targets an effusive jet of known profile.[11] Solid targets were unsupported carbon or carbon-backing–supported, vacuum-evaporated targets. Typical thicknesses of 5 μg/cm^2 on 10–20-μg/cm^2 carbon have been used. Precise determination of the target thicknesses and purity has been made by 1-MeV α-particle backscattering measurements. Lyman lines from the projectile ($np \rightarrow 1s$ transitions) have been recorded with a Si-Li detector positioned at a polarization-insensitive angle of 120° with respect to the beam direction. Lyman α, β, and γ lines were easily resolved, leading to very small uncertainties (<2%) on their measured relative intensities. Highly accurate absolute cross sections have been extracted from the data.[11]

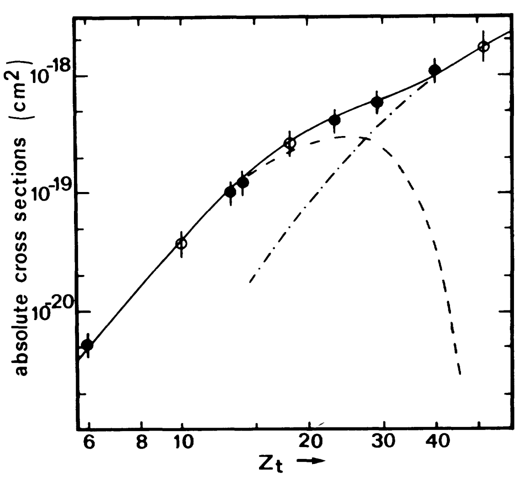

FIG. 1. Total Lyman-line emission cross section as a function of the target's atomic number. The solid line corresponds to CDW calculations times a constant scaling factor of 0.8. Dashed line: contribution from the K shell; dot-dashed line: from other shells of the target. Open and closed dots are for gaseous and solid targets, respectively.

Our measured total Lyman-line emission cross sections[11] are given in Fig. 1. The new and important result here is that no systematic deviation between gaseous and solid targets measurements is observed: These cross sections, equivalent to *total* capture cross sections in excited states (except $2s$),[11] are found to be the same in ion-atom and ion-solid collisions. They also are all in good agreement, within our experimental uncertainty ($\sim$20%), with theoretical predictions (including cascades)[12] based on continuum distorted-wave (CDW) calculations[13] of capture cross sections to excited states ($2 \leq n \leq 10$) and tabulated[14] lifetimes and branching ratios for excited states of hydrogenlike ions. By contrast, large differences are observed in the *relative* populations of $2p$, $3p$, and $4p$ substates as reflected by the Lyman α-, β-, and γ-line relative intensities (Fig. 2). Very good agreement with theory is observed for ion-atom (gaseous targets) collisions, but large discrepancies appear for ion-solid measurements. Next, the x-ray yield variation with target thickness has been explored for carbon targets (Fig. 3). The observed small decrease in total yield for increasing target thickness is mainly due to electron loss from highly excited states ($n \geq 6$), and is very well accounted for on the basis of known electron-loss cross sections (Fig. 3). By contrast, the rapid change in relative Lyman α-, β-, and γ-line intensities within 30–40 μg/cm^2 cannot be explained by this type of multicollision effect.

Three possible mechanisms can be considered to explain our results: (i) The primary capture process itself could be affected inside solids. However, it is very likely

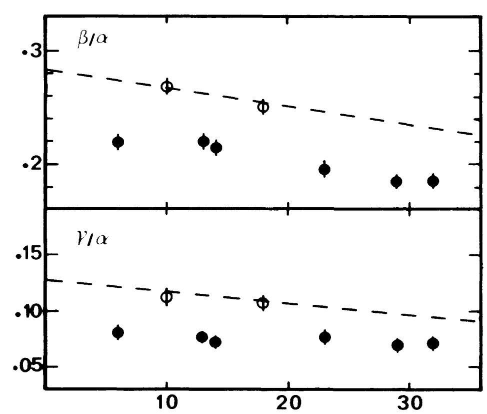

FIG. 2. Relative intensities of Lyman α, β, and γ lines as a function of the target's atomic number. Broken line is from CDW calculations.

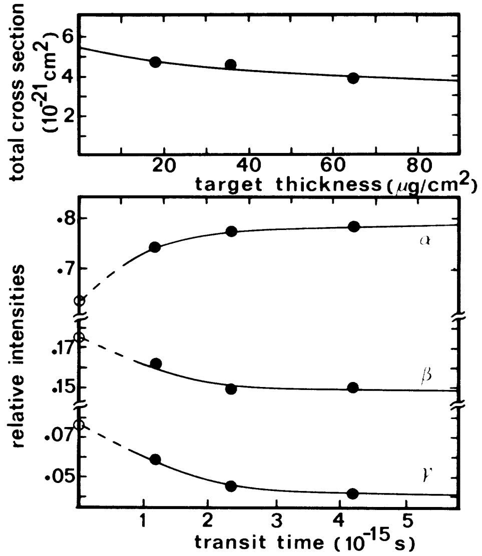

FIG. 3. Total Lyman-line x-ray yield (upper curve) and relative intensities of Lyman α, β, and γ lines as functions of carbon target thickness. Solid curve through total x-ray yield data results from a simple model accounting for electron loss in excited states. Curves through relative intensities are to guide the eyes. Zero-thickness values are extrapolated from gaseous-target results.

that some change of total cross sections should also be observed, which is not the case. (ii) A possible excess of population in highly excited states due to capture of convoy electrons at the exit of the target has been previously considered,[5] but it can easily be checked that cascades from these states would not affect our Lyman α, β, and γ intensities very much. Moreover, any similar "last layer" effect should also manifest itself as an excess of total yield whose contribution decreases for increasing target thickness. Again, none of these effects is observed (Figs. 1 and 3). (iii) Finally, some large-scale effects mechanism leading to a rapid change in the l distribution following electron capture must be considered. In particular, an increase in the *relative* population of high angular momentum states, which mostly decay by cascading through the $2p$ substate, could explain both the observed enhancement of Lyman α intensity and the reduced Lyman β and Lyman γ intensities. CDW cross sections[13] and branching ratios[14] show that such explanations can be found only if the $n=3$ shell is already affected. This new result then differs from observations of Betz, Röschenthaler, and Rothermal[5] relative to very high-n (Rydberg) states.

Looking for a possible explanation of such an increase of high-l substate population, we first investigate the effect of intrashell nl-nl' excitation process. The change in relative intensities appears to saturate much too fast to be accounted for by this process only (see Table I). Moreover, its predicted effects at equilibrium on relative intensities do not agree with experiment. We know, indeed, from experiments on excitation of Rydberg atoms,[7] that a statistical population $[P(l)=(2l+1)/n^2]$ must be reached. In our thickest target, such populations can be assumed for all excited states $n\geq 3$ (Table I). A simple calculation[8,15] based on a statistical redistribution over n,l substates of each n (≥ 3) population gives α/β and α/γ ratios equal to 8.0 and 28.6 instead of the experimental values 5.1 and 16.4 (uncertainty $\sim 2\%$). In this model calculation, we used for $3\leq n \leq 10$ theoretical CDW cross sections and an n^{-3} scaling law for $n>10$. These values indeed reproduce our results very well in the case of gaseous targets (Figs. 1 and 2). Cascades from all excited states, radiative deexcitation inside the target, and electron loss in highly excited states (see Fig. 3) have been considered. Finally, we note that by consideration of either a larger ($n\geq 2$) or a smaller ($n\geq 4$) number of equilibrated states, no improvement in the comparison is obtained.

We instead suggest that the dominant effect is due to the wake-field–induced Stark splitting of the states. If we neglect for the moment the fine structure of the excited states of hydrogenlike krypton, the expected effects on the population of these states can be qualitatively understood on the basis of a simple model. Let $a_+\exp(i\phi_+)$ and $a_-\exp(i\phi_-)$ be the capture amplitudes in the two (parabolic) Stark states corresponding to $n=2$, $m=0$; ω_s be the (wake-field–induced) splitting of these states; and $\phi=\phi_+-\phi_-$. When capture occurs *inside* a solid, the Stark states will have different phase evolution during the time t between capture and exit from the target. Projection at time t on the spherical states gives therefore time-dependent "cross sections"

$$\sigma_s^t=\tfrac{1}{2}(a_+^2+a_-^2)-a_+a_-\cos(\omega_s t+\phi),$$

$$\sigma_{p0}^t=\tfrac{1}{2}(a_+^2+a_-^2)+a_+a_-\cos(\omega t+\phi).$$

This effect is germane to s-p coherence measurements in an external field.[16] In our case, however, capture can take place at any time between 0 and the dwell time t_d through the target. Integration over target thickness (or range of times t) leads us to predict a general increase ($2s$ quenching inside the solid), but also *damped oscillations*, of the Lyman α ($2p\rightarrow 1s$) line emission cross section as a function of target thickness. Indeed, part of the oscillatory behavior is kept for the thinnest targets when $t_d<2\pi\omega_s^{-1}$ ($\sim 10^{-15}$ s). Especially, the amplitude of the first oscillation is governed by the s-p coherence angle ϕ, and the time scale by the wake-field strength. Note also that fast ($\sim 10^{-17}$ s) periodic fluctuations of the mean wake field related to binary encounters in the solid would not alter this pattern deeply.

Our measurements indeed display an increase of Lyman α cross section, but the damped oscillations are further washed out by a significantly enlarged cascade contribution to Lyman α total intensity resulting from the l mixing in upper states. For large dwell times ($t_d \gg \omega_s^{-1}$) the contribution of the oscillatory term in the above equations becomes negligible, and the relative substate population in any shell tends in first approximation towards a random distribution, among all possible l values, of a given total m population. Assuming such a "complete l mixing" of the substates, we find an increase in the Lyman α relative intensity, corresponding now to values of 5.8 and 16.7 for the α/β and α/γ ratios, in much better agreement with experiment (5.1 and 16.4) than use of pure statistical populations in our model calculation (see above). Part of the small observed deviation ($\sim 15\%$) in the case of Lyman β intensity could also be accounted for by fine structure and QED effects. Only the splitting in the $n=2$, $m_j=\frac{1}{2}$ manifold has been evaluated, with the assumption of a wake field of $\varepsilon=10^9$ V/cm. This gives a characteristic time of 2.2×10^{-15} s in $n=2$ (and smaller or comparable times in upper levels), which is in good agreement with our experimental saturation time (Fig. 3).

In conclusion, use of high-Z projectiles gave us the opportunity to achieve an original comparison of the capture process in solid and gaseous targets under single-collision conditions. For the first time, evidence of anomalous l populations in *deep* excited states inside solids has been given. For these states, the *nl-nl'* excitation is a minor phenomenon, in contrast with Rydberg states which have been studied up to now.[5,6] A new wake-field–induced effect has been considered, and has been shown to give encouraging agreement with our results. Finally, we point out that subtraction of the cascade contribution to the feeding of lower levels[12] may lead in the future to quantitative determinations of coherence angles and wake-field intensities.

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