Complex Substate Amplitudes Formed in Double Electron Capture

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Complex amplitudes and their dependence upon the projectile scattering angle have been measured for $|L, M\rangle$ substates of doubly excited $1s[2s2p\ ^1P]^2P$ and the $1s2p^2\ ^2D$ terms of C^{3+} formed in double electron capture by 25 keV C^{5+} ions from He atoms. These are extracted from Auger electron anisotropy measured with respect to the incident beam and scattering plane by a coincident electron–scattered ion technique. The results are the most complete experimental description of a low energy double capture collision.

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Electron capture is the dominant inelastic process in slow ($v < 1$ a.u.) collisions of multiply charged ions with atoms, and, nearly always, the transferred electrons populate excited states of the product ion. In single electron capture the ion relaxes radiatively and the photon spectra, polarization, and/or angular distribution can be used to identify and characterize the excited state (see, e.g., Refs. [1,2]). This includes determination of the sense of rotation (orientation parameter) and angular properties of the charge density (e.g., alignment angle) of the captured electron [3]. A similarly detailed description of the state formed in a multiple electron capture collision has been lacking, despite its expected bearing on, e.g., understanding the role of the interelectron interaction during the collision and the competition between pathways among the transient quasimolecular levels leading to the final state [4]. This Letter describes the first such detailed study of a double electron capture collision. This was done on a system which is both theoretically (see, e.g., Ref. [5]) and experimentally tractable; C^{5+} on He. We have measured the scattering angle dependence of the complex amplitudes a_M for populating the $|L, M\rangle$ substates of the excited terms $1s[2s2p\ ^1P]^2P$ and $1s2p^2\ ^2D$ in C^{3+} by transfer of both He electrons through analysis of anisotropy in the subsequent Auger electron emission from the decay of these states. This is analogous to studies of photon anisotropy in single electron capture studies, however, Auger decay is not bound by electric dipole selection rules and the final ion state is the S state ($C^{4+}\ 1s^2$) for all excited $C^{3+}\ 1s2l2l'$ terms, including ^2D.

To determine the relative phases of the a_M, the anisotropy must be measured with respect to the collision plane; this requires use of a coincidence technique. We measure the $1s2l2l'$ Auger spectrum (see Ref. [6]) emitted into a small solid angle at polar angle θ_e, with respect to the C^{5+} beam direction, in coincidence with two dimensional position detection of the scattered C^{4+} final state ions. A transformation of the coincident scattered projectile positions associated with each Auger line gives the azimuthal coordinate, ϕ_e of the electron emission with respect to the scattering plane, and, θ_I, the projectile ion scattering angle. A previous noncoincidence study [6] provided values of the $|a_M|$ averaged over all scattering angles by measuring the θ_e dependence of the Auger emission rate integrated over all ϕ_e. Though different in significant ways, our approach bears similarity to that used in studies of He and Ne states excited by ion impact [7,8].

The C^{5+} ions were produced by the LBL Electron Cyclotron Resonance ion source and transported via the joint LBL/LLNL beam line facilities. The collimated beam (1×1 mm) entered a chamber containing a He jet target and an electron spectrometer. The spectrometer viewed the intersection of the C^{5+} beam with the He jet at angles, θ_e ranging from $\approx 35°$ to $\approx 105°$ with respect to the beam direction. At 0.75 m down-stream from the jet, the beam entered parallel plates charged to deflect the product C^{4+} ions 17° onto a 40-mm-diam position sensitive microchannel plate (MCP) ion detector located 1.05 m from the deflection plates. The electron spectrometer includes a 25-mm-diam MCP detector (similar to the ion detector) mounted near the exit plane. The Auger electrons from the $1s2l2l'$ terms have energies near 240 eV in the emitter frame. We decelerate these to ≈ 70 eV at entrance to the spectrometer; this provides resolution (≈ 0.7 eV) adequate to resolve and spread the lines across the detector face. A data collection system records 5 paramaters for each event: the position of the electron hit from the spectrometer detector (x_e, y_e), the scattered ion position (x_I, y_I) and the output from a time-to-amplitude converter (TAC) started by the electron and stopped by the ion signals. Position sensitive detection of the electron spectrum and the scattered ions was essential to achieve a usable coincidence rate, however, an additional wrinkle was required.

Because single electron capture produces C^{4+} ions of essentially the same energy (25 keV) as those from double

electron capture followed by Auger decay (the ions of interest) and the cross section for single capture exceeds that for double capture, the double capture products are submerged in a larger stream from single capture. Use of a C^{5+} current (e.g., 25 pA) giving an acceptable Auger electron rate produces a continuous C^{4+} stream from single capture that swamps the ion detector forcing reduction of the beam, and thus an unusable coincidence rate. To ameliorate this problem we use an ion sorting technique in which the deflector field is set to over-deflect all C^{4+} products beyond the ion detector. Upon detection of an Auger electron, the field is reduced before the ion reaches the deflector, held for the period of transition through the plates at a value which directs the ion onto the detector face, then restored to its quiescent, over-deflected, condition. This scheme allowed data collection at real coincidence rates of 10 to 15 per minute.

Data are analyzed using a right-hand coordinate system with the z axis along the beam and the y axis in the collision plane parallel to the scattered ion's transverse momentum. Conservation of positive reflection symmetry with respect to the scattering ($y - z$) plane requires that $a_M = a_{-M}$. For the P state, the coincidence rate S_P for ions scattered into a range $\Delta\theta_I$ at angle θ_I and emission of Auger electrons into a small solid angle $\Delta\Omega_e$ at angles θ_e, ϕ_e is

$$S_P(\theta_I, \theta_e, \phi_e) = K \frac{d\sigma_P}{d\theta_I} \frac{3}{4\pi} \times [|a_1|^2 \sin^2\theta_e(1 - \cos 2\phi_e) + |a_0|^2 \cos^2\theta_e - |a_1||a_0| 2^{1/2} \sin 2\theta_e \sin\phi_e \sin\Delta\beta_{01}],$$

where the amplitudes for the $M = 1, 0$ substates have magnitudes $|a_{1,0}|$ and relative phase $\Delta\beta_{01} = \beta_0 - \beta_1$, and $d\sigma_P/d\theta_I$ is the cross section for creation of the P state; all of these are functions of θ_I. K is a factor including $\Delta\Omega_e$, $\Delta\theta_I$, the beam intensity, target thickness, and detector efficiencies. Including the normalization relation $2|a_1|^2 + |a_0|^2 = 1$, the P state anisotropy depends upon two parameters, i.e., one of the $|a_M|$ and $\Delta\beta_{01}$. In the more complex expression for S_D, the coincidence rate from the $1s2p^2\,{}^2D$ term, the anisotropy depends upon four parameters. At selected angles θ_e, the anisotropy depends upon a subset of the $|a_M|$ and $\Delta\beta_{M'M}$. For example, at $\theta_e = 90°$, one has

$$S_D(\theta_I, \theta_e, \phi_e) = K \frac{d\sigma_D}{d\theta_I} \frac{5}{16\pi} \times [6|a_2|^2 + |a_0|^2 - 2 \times 6^{1/2}|a_2||a_0| \cos 2\phi_e \cos\Delta\beta_{02}].$$

We have collected data for S_P and S_D at electron polar angles (emitter frame), $\theta_e = 45°$, 54.7°, and 90°, from which we extract (by fitting the ϕ_e dependences for separate regions of θ_I) the $|a_M|$ and $\sin\Delta\beta_{01}$ (for the P state) and $\sin\Delta\beta_{12}$ and $\cos\Delta\beta_{02}$ (for the D state). Figure 1 shows a sample of data from decay of the ^2D term observed at $\theta_e = 54.7°$, and describes the analysis process. Figures 2 and 3 summarize the results of all measurements.

Recent theoretical [3,9–11] and experimental [2,12] work has demonstrated the usefulness of a propensity rule for single electron capture, favoring population of the substate with $M = -L$ in the natural frame (reached by a +90° rotation of our laboratory frame about the y axis). However, our measurements are not sensitive to the orientation of the normal to the scattering plane. That is, in terms of Fig. 1(a), the experiment cannot distinguish between x, y axes as shown and the alternative with $x, y \rightarrow -x, -y$. Thus, the expectation value of the angular momentum perpendicular to the scattering plane (orientation parameter), $L_\perp$, for the P state (in units of $\hbar$) is $L_\perp = -2 \times 2^{1/2}|a_0||a_1|\cos\Delta\beta_{01}$, and measurement of $\sin\Delta\beta_{01}$ leaves the sign of $\cos\Delta\beta_{01}$ and, hence, of $L_\perp$ undetermined. Our knowledge of $L_\perp$ is summarized in Fig. 4, which also shows the scattering angle dependence of the angle γ (alignment angle) between the major axis of the P state charge cloud and the z axis; unlike $L_\perp$, γ is

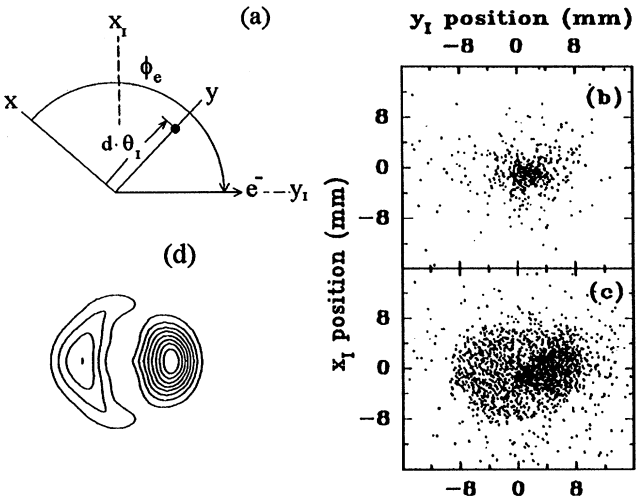

FIG. 1. Scattered C^{4+} ion positions on the projectile detector, associated with Auger electron emission from C^{3+} $1s2p^2\,{}^2D$ at $\theta_e = 54.7°$. The panels view the detector from the beam (z) direction. (a) Relationship between the detector fixed coordinates (x_I, y_I) and the event determined coordinates (x, y). The distance to the event from the origin is $d\theta_I$; d is known constant. (b) Events not coincident with electron emission (randoms, i.e., outside the TAC peak), mostly products of single electron capture. (c) Events coincident with (i.e., in the TAC peak) electron emission together with randoms. The analysis subtracts (b) from (c) and fits $S_D(\theta_I, 54.7°, \phi_e)$ to the resulting pattern. (d) Contour plot representing (c) and (b) constructed from the results of the fit. The ions were deflected in the x_I direction for charge analysis. The offset of the random pattern (b) from the origin is due to the difference in the exothermicity for single and double capture (x_I offset) and recoil from the (≈240 eV) electron emission (y_I offset).

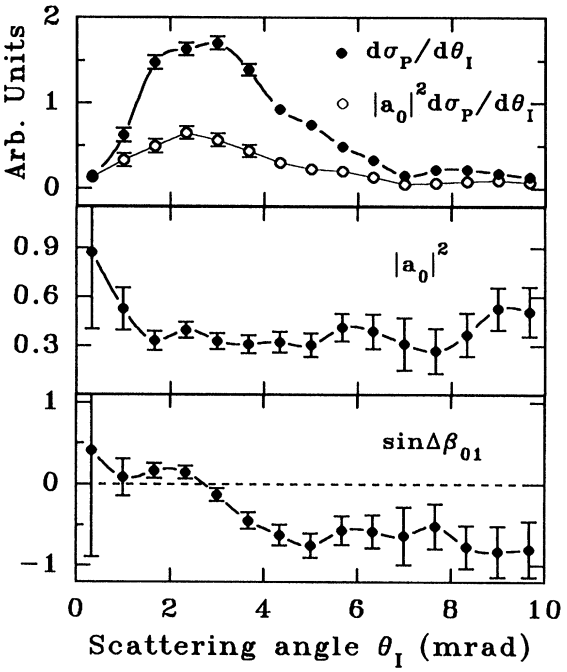

FIG. 2. Results from analysis of the coincident Auger anisotropy from the C^{3+} $1s[2s2p\,^1P]^2P$ term.

fully determined by these measurements:

$$\gamma = \frac{1}{2}\pi + \frac{1}{2}\tan^{-1}\left(\frac{2(2)^{1/2}\,|a_0|\,|a_1|\sin\Delta\beta_{01}}{2|a_1|^2 - |a_0|^2}\right).$$

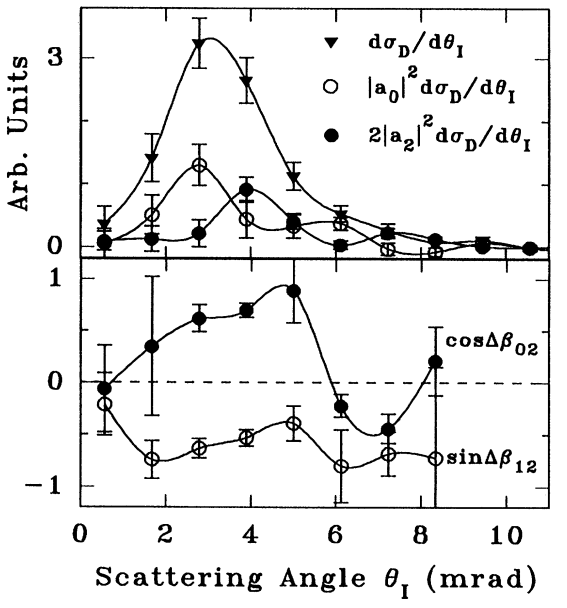

FIG. 3. Results from analysis of the coincident Auger anisotropy from the C^{3+} $1s2p^2\,^2D$ term.

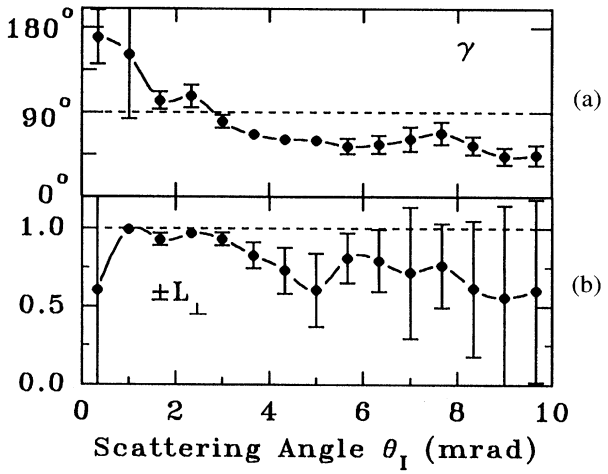

FIG. 4. (a) Alignment angle γ and (b) magnitude of the angular momentum perpendicular to the scattering plane $|L_\perp|$ for the ^2P term, versus scattering angle.

For no scattering ($\theta_I = 0$) one expects $\gamma = 180°$ since, for such a distant "collision," the electron motion remains coupled to the internuclear axis while it rotates one half turn. At nonzero scattering angles the electron motion lags this rotation by an amount reflecting the strength of rotational coupling during the collision; this coupling is responsible for population of substates with $M \neq 0$. Near the scattering cross-section maximum, γ passes through 90° and for larger scattering angles is approximately constant near 50°. $|a_0|^2$ is near unity for θ_I approaching zero, but fixed at $\approx 1/3$ over the range of significant scattering. $|L_\perp|$ for the P state approaches unity in the region of the maximum in $d\sigma_P/d\theta_I$, falling slowly at larger scattering angles. This behavior is consistent with a propensity rule favoring $M = -1$ in the collision frame and theoretical and experimental studies of orientation [2,3,9–12] in single electron capture collisions. To the extent that the captured electrons are properly described by the single particle configuration $2s2p$, only one of them contributes to M. However, whereas treatments of single capture [2,3,11,12] show $L_\perp$ to be negative near the peak of the differential cross section, with a sign change at larger scattering angles (or smaller impact parameters), our observations do not show $\pm L_\perp$ reaching zero, and, hence, do not support a sign change.

For the D term one has

$$L_\perp = -4|a_1|\cos\Delta\beta_{12}[|a_2| + (3/2)^{1/2}\,|a_0|\cos\Delta\beta_{02}] - 2\times 6^{1/2}|a_1|\,|a_0|\sin\Delta\beta_{12}\sin\Delta\beta_{02}$$

and, although the measurements do not determine the signs of $\sin\Delta\beta_{02}$ or $\cos\Delta\beta_{12}$, they are sensitive to the sign of their product; thus, as for ^2P, the sign of $L_\perp$ remains undetermined. Figure 5 shows values of $\pm L_\perp$ versus

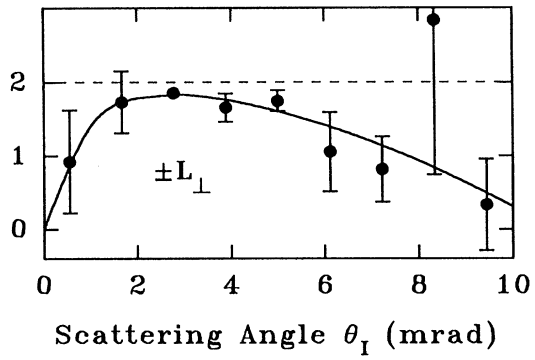

FIG. 5. $\pm L_\perp$ for the ^2D term versus scattering angle. The line is a polynomial fit to the points as a guide.

scattering angle for the D state. Note that $\pm L_\perp$ is near 2 in the vicinity of the maximum in the differential cross section, with no indication of a sign change within the full angular range. This is similar to the behavior seen for the ^2P term, and suggests applicability of the propensity rule favoring $M = -L$ to the $2p^2$ fully-equivalent electron configuration. Since the substate $|D, M = -2\rangle$ is well described as the product of $2p$ orbitals, each with $m = -l$, its population would follow from application of the single capture rule to each electron separately.

In summary, we have developed experimental methodology which determines the scattering angle dependence of the complex anisotropy in doubly excited states formed in multiple electron capture collisions. This provides a significantly more complete experimental description than previously available of the character of the excited two-electron wave function and its dependence upon the projectile scattering angle (i.e., impact parameter). The variation of the charge cloud orientation with scattering angle for the C^{3+} $1s(2s2p\ ^1P)^2P$ term demonstrates clearly the loss of electron coupling to the rapidly rotating internuclear axis. Values for the angular momentum, $L_\perp$, normal to the scattering plane suggest that a propensity rule favoring population of $M = -L$ substates in single capture collisions may also hold for double capture. The relevant crossings between diabatic states of $[C^{5+}, He]$ (initial channel), $[C^{4+}(1snl), He^+]$ and $[C^{3+}(1snln'l'), He^{++}]$ occur at internuclear separations less than 5 a.u. The duration of collisions reaching these crossings (at $v = 0.3$ a.u.) imply energy uncertainties ($\approx$10 eV or more) which exceed the separation of the $C^{3+}(1s2l2l')$ terms (3.1 eV between P and D). Thus it appears fruitful, at least for some systems, to regard double capture as the outcome of nearly independent single electron transfers with subsequent evolution into states more fully defined by the electron-electron interaction. A model based upon this approach has been described [13] for population of He-like $3l3l'$ and Be-like $1s^23l3l'$ terms from H_2 and He targets.

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