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Magnetic Substates Populated by Double Electron Capture

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We have made the first measurements of relative cross sections for the population of individual M_L substates in a double electron capture collision. This information was extracted from measurements of the angular distribution of Auger electrons emitted in the decay of the doubly-excited $1s2l2l'\ ^2L$ states of C^{3+} formed in collisions of C^{5+} ions with He. Large anisotropies were observed for the $1s2p^2\ ^2D$ and $1s[2s2p\ ^1P]\ ^2P^o$ terms which varied markedly with projectile velocity in the range 0.29 to 0.50 a.u. Strong velocity variation was also observed for the total L-subshell relative cross sections. Our results suggest strong rotational coupling at small internuclear separation and show the impact of alignment on the determination of total cross sections from measurements made at a single angle.

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This Letter reports the first determination of magnetic substate population distributions created in a double electron capture collision, and thus represents the most complete characterization of the projectile final state following this simplest multiple electron transfer process. This work follows on the vast literature of slow (velocities less than 1 a.u.) ion-atom collisions which has established the dominance of electron capture in these encounters. Photon, Auger and projectile energy gain spectroscopy have established the preponderance of single electron capture into principal quantum states which nearly match the binding of the active electron in the target atom and have resolved, in several cases, partial cross sections for population of individual orbital angular momentum states.[1] These observations are in reasonable agreement with the predictions of the classical over barrier model (CBM).[2,3] Fluorescence and Auger angular distributions have been measured in a small number of cases for slow singly-charged projectiles.[4-9] Recently, projectile Auger electron spectra have been used to identify the prominent reaction channels in two or more electron transfer events and to establish energy level structure in highly excited species.[10,11]

In this work we report a study of the process

$$\mathrm{C}^{5+}(1\mathrm{s}) + \mathrm{He}(1\mathrm{s}^2) \rightarrow \mathrm{C}^{3+}(1\mathrm{s}2\mathrm{l}2\mathrm{l}'\ {}^2\mathrm{L}) + \mathrm{He}^{++}$$

by observing Auger electron spectra at nine angles from the decay

$$\mathrm{C}^{3+}(1\mathrm{s}2\mathrm{l}2\mathrm{l}'\ {}^2\mathrm{L}) \rightarrow \mathrm{C}^{4+}(1\mathrm{s}^2\ {}^1\mathrm{S}) + \mathrm{e}^-(\epsilon\mathrm{L}).$$

This has been done at four collision velocities from 0.29 to 0.50 a.u. (energies from 25 to 75 keV). The C^{5+} ions were produced by the LBL ECR (Electron Cyclotron Resonance) ion source; the measurements were made using a portion of the joint LBL/LLNL atomic physics facilities at LBL's 88-inch Cyclotron. The beam was collimated to a 3mm x 3mm cross section with a divergence of ~5 mrad, after which it entered a magnetically shielded chamber where it intersected a He gas jet on the axis of a rotatable turntable which carried a 45 degree parallel plate electron spectrometer. Low gas density was used to ensure single collision conditions. Grids in front of the spectrometer entrance were used to decelerate the Auger electrons to energies typically between 40 and 60 eV to improve the resolution of the instrument. Collimated beam currents varied from ~2 nA at 25 keV to ~12 nA at 75 keV.

Figure 1 shows the data obtained at a collision energy of 50 keV, after transformation to a frame moving with the emitting projectile. The $1s2p^2\ ^2S$ line played an important role in this work, since it must be isotropic; by normalizing other peaks to the $1s2p^2\ ^2S$ we removed corrections for variation of the overlap of the viewing angle of the spectrometer with the beam/jet intersection, small changes in gas jet density, varying transmission of the decelerator/spectrometer combination, drifts in detector gain or electronic thresholds etc. The spectra were well fit by a set of gaussian line shapes, of the same width, at each angle. The resolution varies with angle and is understood to be a combination of a fixed instrument resolution of about 3% of the decelerated electron energy and kinematic broadening due to the finite acceptance angle for viewing the moving emitters. This latter effect is maximum near 90°. Perturbation of the Auger line shape by the post-collision interaction (PCI) of the outgoing electron with the target is not significant in this work in part because of the resolution of the instrument, and the relatively high energy of the electrons. This effect has been observed[12] at 0° with a higher resolution instrument on the $1s2s^2\ ^2S$ line. Deflection[13] of electrons by the residual target ion might be observable at angles close to 180°. The lifetimes are:[14] $\tau(1s[2s2p\ ^1P]\ ^2P^o)=1.8\times10^{-14}$s, $\tau(1s2p^2\ ^2D)=1.25\times10^{-14}$s, and $\tau(1s2s^2\ ^2S)=8.1\times10^{-14}$s; thus the average separation between the excited projectile and the He^{++} ion at the time of Auger emission ranges from 150 a.u. for the $1s2p^2\ ^2D$ line at a collision velocity of 0.29 a.u. to over 1600 a.u. for the $1s2s^2\ ^2S$ line at a velocity of 0.50 a.u. A Coulomb scattering calculation for the Auger electron in the field of the He^{++} ion shows, for the worst case ($1s2p^2\ ^2D$ line, collision velocity 0.29 a.u., laboratory emission angle 160°), a deflection of less than 0.3° at 150 a.u. separation. Confirmation that PCI is negligible comes from the observed angular distributions, which are symmetric about 90°.

Cylindrical symmetry about the ion beam axis dictates that the population of M_L substates can depend only upon the absolute value of M_L; that is, the $C^{3+}(1s2l2l'\ ^2L)$ state is *aligned.* The angular distribution, in the emitter frame, is given by[15] $I(\theta) = F\sigma_L W(\theta)$ where $W(\theta) = [1+\sum A_{2k}P_{2k}(\cos\theta)]$, σ_L is the cross section for producing the $1s2l2l'\ ^2L$ state, F is a factor including the target density, beam flux, detector solid angles, and other geometric factors; θ is measured from the beam direction, and the sum extends up to k=L and thus for S states $W(\theta) = 1$. Note that the anisotropy coefficients A_{2k} are *completely* determined by the M_L-substate cross sections provided that the ion is left in an S state following the Auger decay, as is the case here.

A small correction to $W(\theta)$ must be made to account for the decrease in alignment due to mixing of M_L substates by the fine structure (fs) interaction during the interval between the collision and the Auger emission. Mehlhorn and Taulbjerg[15] have shown that the net effect is to multiply each A_{2k} by a de-alignment factor D_{2k} which depends upon the ratios of the fs splittings to the natural widths of the doubly-excited levels. Using theoretical fs intervals and natural widths supplied by Chung,[14] we calculate D_2=0.9637 for 1s[2s2p ^{1}P] ^{2}P^o and D_2=0.9863, D_4=0.9544 for 1s2p^2 ^{2}D.

From the anisotropy coefficients A_2 and A_4 one can obtain[15] values for the ratios of cross sections, σ_{LM_L}, for populating the $|L,M_L>$ states. For P states

$$\frac{\sigma_{11}}{\sigma_{10}} = \frac{1-A_2/2}{1+A_2},$$

and for D states

$$\frac{\sigma_{21}}{\sigma_{20}} = \frac{1+A_2/2-2A_4/3}{1+A_2+A_4}, \qquad \frac{\sigma_{22}}{\sigma_{20}} = \frac{1-A_2+A_4/6}{1+A_2+A_4}.$$

After transforming our data into the emitter frame, we formed the ratio of the areas of the lines shapes fit to the 1s2p^2 ^{2}D and 1s[2s2p ^{1}P] ^{2}P^o peaks to the 1s2s^2 ^{2}S area at each angle. The anisotropy coefficients and the ratio σ_L/σ_0 were adjusted to fit the observed ratios. The results are shown in Figures 2 and 3, which present the angular distributions for the 1s2p^2 ^{2}D and 1s[2s2p ^{1}P] ^{2}P^o lines. The coefficients and cross-section ratios are presented in Table I. Figure 4 displays the variation of the population fractions with collision energy. Our results show a dramatic variation of the magnetic substate populations over a narrow range of collision velocities, and predominant population of $M_L=\pm1$ substates at higher velocities. Mann[16] showed that the 1s2l2l$'$ levels are directly populated by two electron transfer at a quasimolecular crossing at an internuclear separation of ~2.9 a.u. Comparison with the CBM and Landau Zener (LZ) models showed better agreement with the latter. In addition, LZ theory predicts variation of the crossing separation with collision energy; it decreases by 0.4 a.u. from 25 to 100 keV. Given the small size of the internuclear separation and its dependence upon collision velocity, it is perhaps not surprising to observe large velocity dependence for the M_L populations. Changes of M_L by ±1 are induced by the rotational coupling interaction,[17] $<\psi_2|\frac{\partial}{\partial t}|\psi_1> = -i\dot{\phi}<\psi_2|L_y|\psi_1>$, which accounts for deviation from the Born-Oppenheimer approximation due to rotation of the quasimolecule. This interaction is large at small separations because of the $\dot{\phi}$ factor and the radial overlap integral in the L_y matrix element. We expect that a molecular orbital calculation in which a single Σ–Π coupling dominates may explain the main features of our observations.

The variation of the L-subshell population fractions has been seen previously by Mack and Niehaus[18] at 50° laboratory angle and by Mann[16] at 0°. Our angular distributions clearly demonstrate that one should not infer total cross sections from measurements at a single angle. For $1s2p^2\ ^2D$, $W(0^\circ)$ decreases by a factor of 3.3 from 25 to 75 keV. In the case of P states one might avoid this problem by observing at the "magic angle" $\cos^{-1}(1/\sqrt{3})$; however, for a state with $L>1$ it is essential to measure at a minimum of $L+1$ different angles.

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TABLE I. Anisotropy coefficients, A_{2k}, and cross-section ratios.

State	E(keV)	σ_L/σ_0	A_2	A_4	σ_{L1}/σ_{L0}	σ_{L2}/σ_{L0}
$1s2p^2\ ^2D$	75.0	3.66(11)	0.32(6)	-0.82(9)	3.39(76)	1.07(29)
	50.0	2.98(2)	0.33(1)	-0.67(2)	2.43(9)	0.85(4)
	37.5	2.40(6)	0.35(5)	-0.40(7)	1.51(17)	0.61(9)
	25.0	1.31(4)	0.60(8)	0.19(12)	0.66(9)	0.24(6)
$1s[2s2p\ ^1P]\ ^2P^o$	75.0	7.90(25)	-0.28(5)		1.58(13)	
	50.0	5.08(8)	-0.20(3)		1.38(6)	
	37.5	3.19(9)	-0.17(5)		1.30(11)	
	25.0	1.09(5)	0.12(10)		0.84(12)	

Figure Captions

FIG. 1. Auger spectra for $C^{3+}(1s2l2l'\ {}^2L) \rightarrow C^{4+}(1s^2) + e^-$, with least-squares fits to gaussian lineshapes. ${}^2S_a \equiv 1s2s^2\ {}^2S$, ${}^4P^o \equiv 1s2s2p\ {}^4P^o$, ${}^2P_a^o \equiv 1s[2s2p\ {}^3P]\ {}^2P^o$, ${}^2P_b^o \equiv 1s[2s2p\ {}^1P]\ {}^2P^o$, ${}^2D \equiv 1s2p^2\ {}^2D$, ${}^2S_b \equiv 1s2p^2\ {}^2S$.

FIG. 2. Auger angular distribution for $C^{3+}(1s2p^2\ {}^2D) \rightarrow C^{4+}(1s^2) + e^-$ with least-squares fits to $W(\theta)=1+D_2A_2P_2(\cos\theta)+D_4A_4P_4(\cos\theta)$.

FIG. 3. Auger angular distribution for $C^{3+}(1s[2s2p\ {}^1P]\ {}^2P^o) \rightarrow C^{4+}(1s^2) + e^-$ with least-squares fits to $W(\theta)=1+D_2A_2P_2(\cos\theta)$.

FIG. 4. Collision-velocity dependence of (a) L-subshell fractional populations, (b) M_L-substate fractional populations for $1s2p^2\ {}^2D$, and (c) M_L-substate fractional populations for $1s[2s2p\ {}^1P]\ {}^2P^o$.

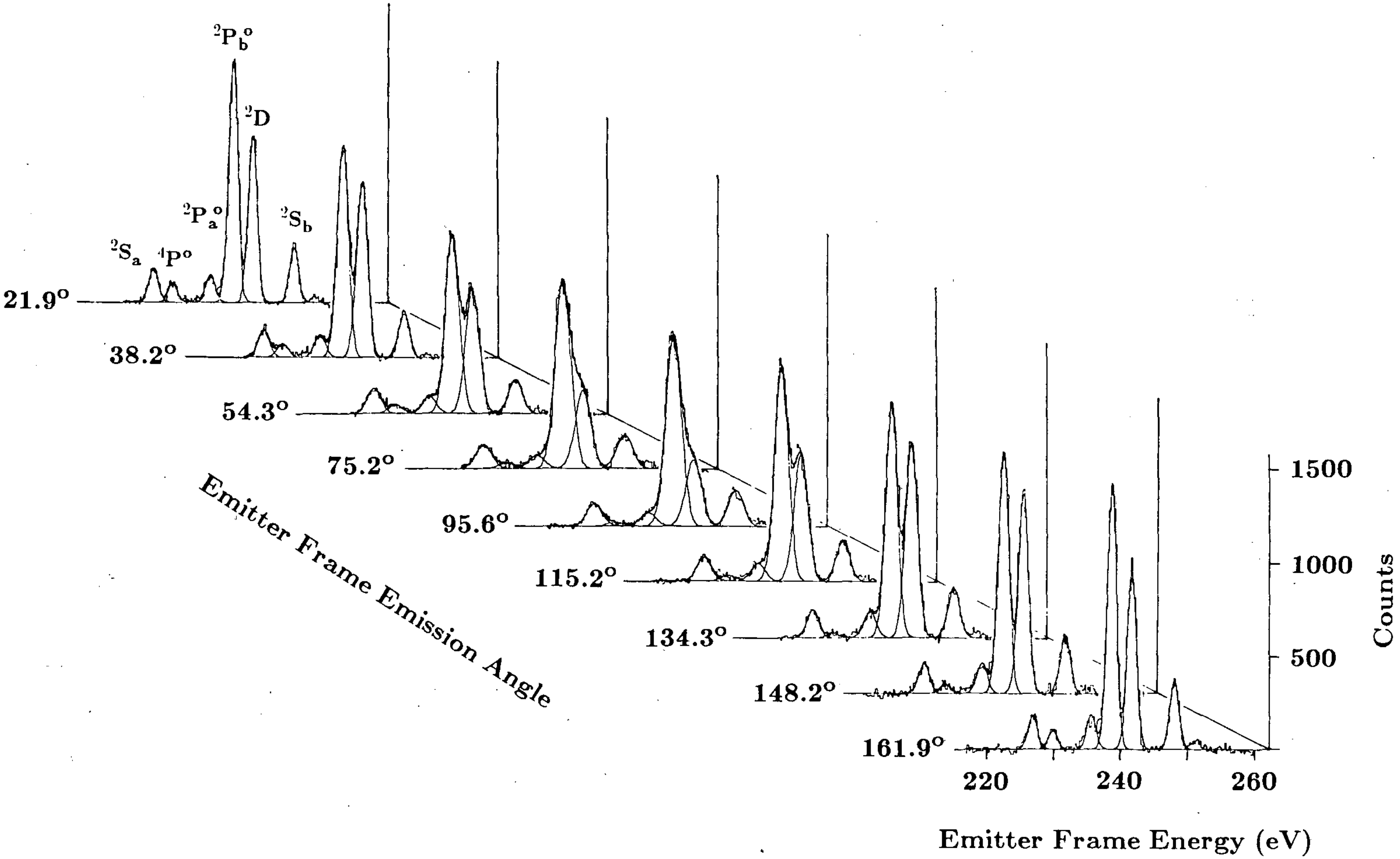

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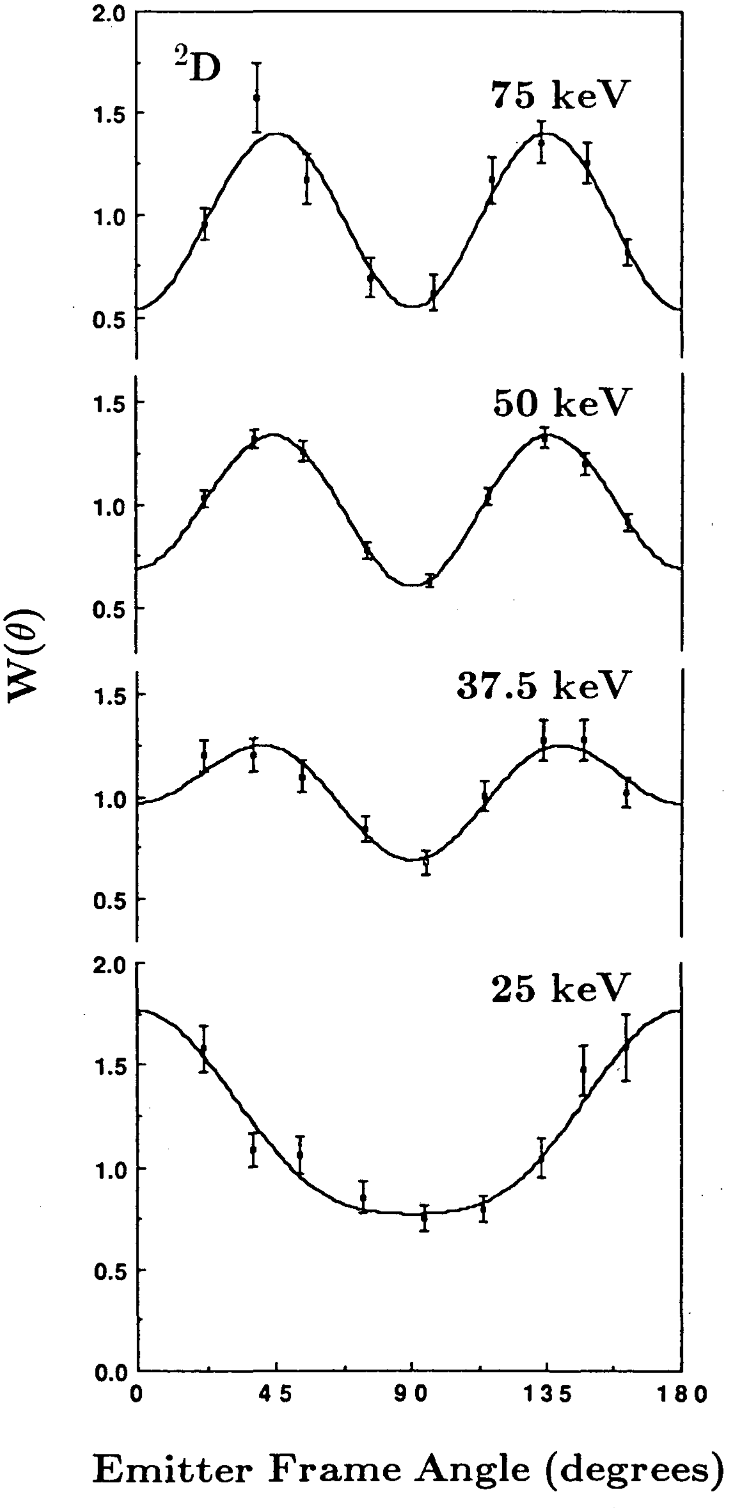
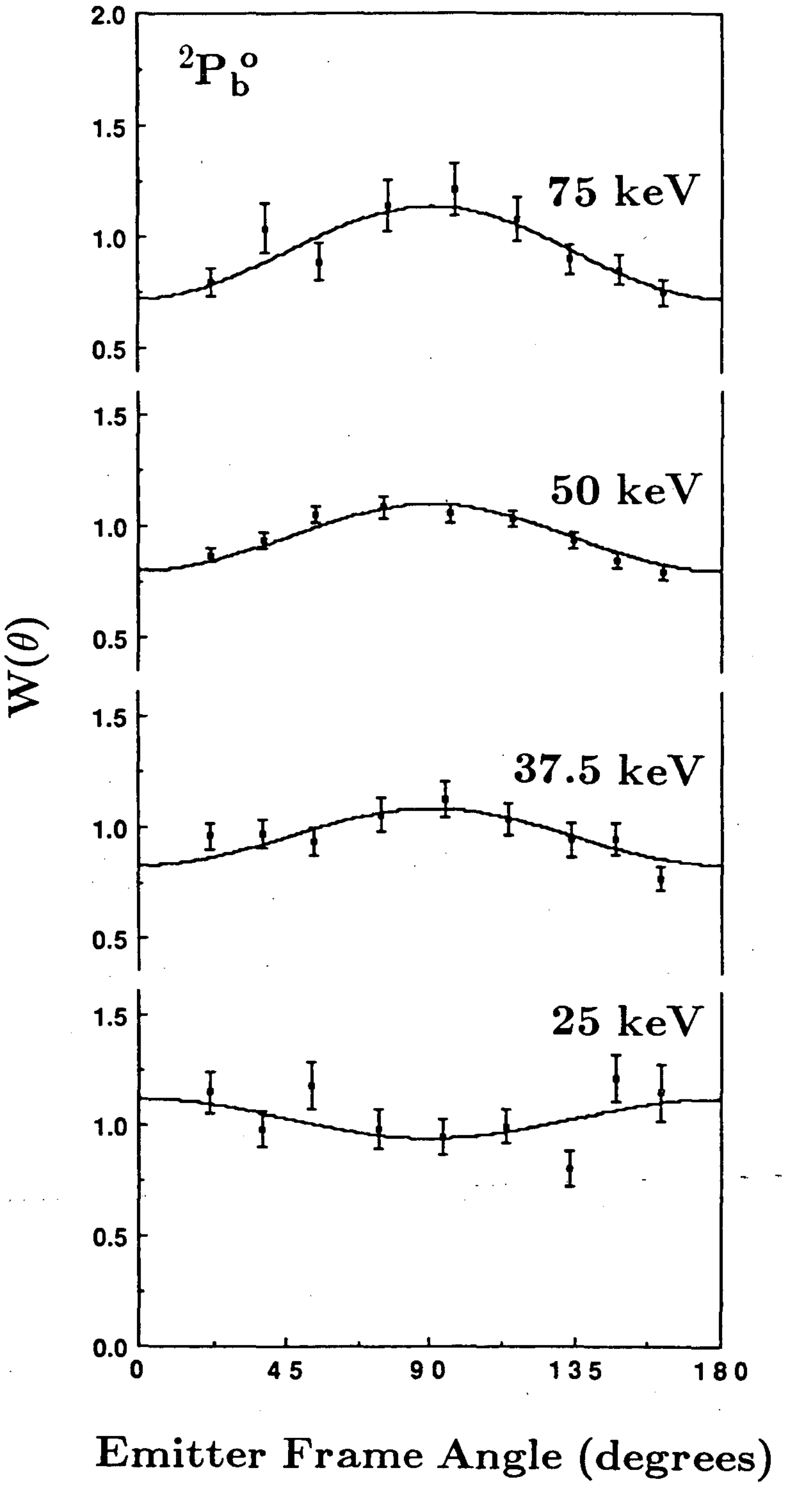
Figure 1.



XBL 906-2278

Figure 2.



XBL 906-2279

Figure 3.

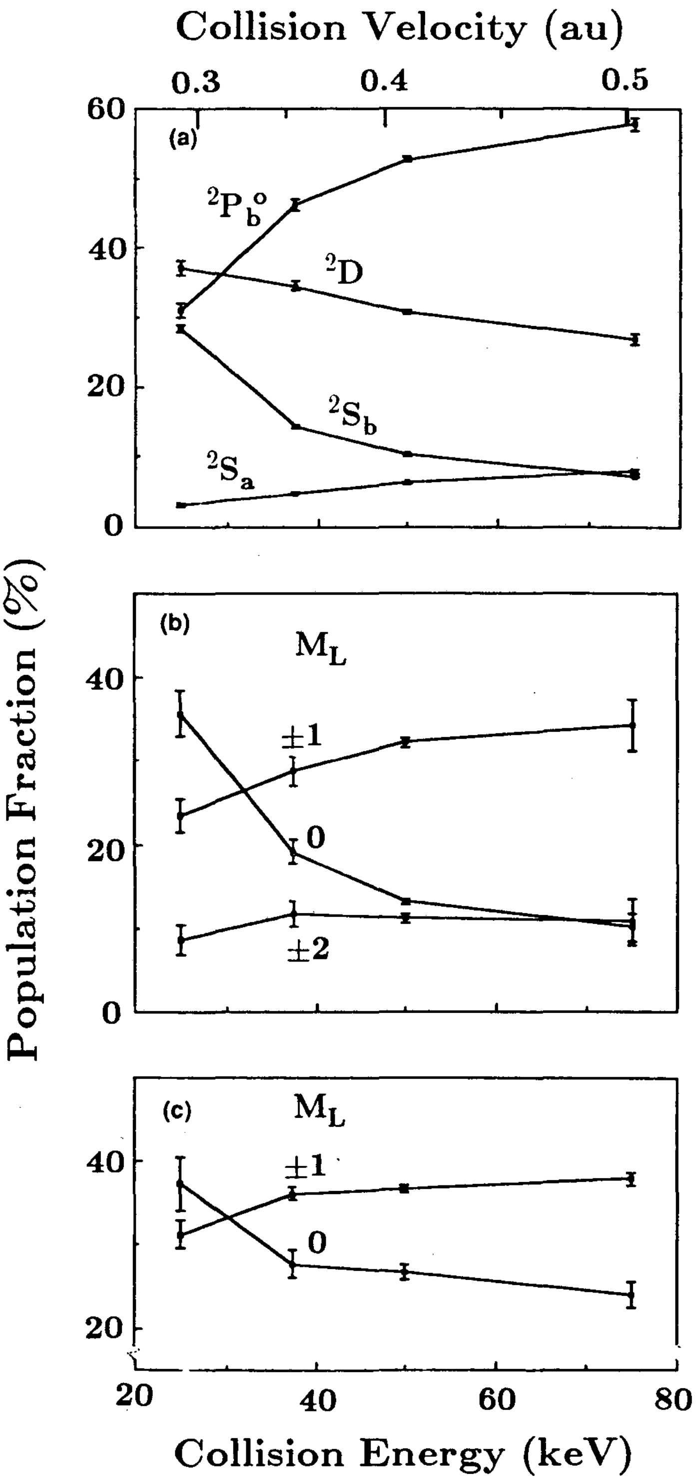


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Figure 4.

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