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Electron correlation in an Auger process

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Coincident 205-eV electrons resulting from the double ionization of argon by electron impact have been measured for incident energies from 400 to 2200 eV. The two 205-eV electrons are produced by knockout of an argon $2p$ electron followed by 205-eV LMM Auger decay of the $2p$ vacancy. The cross section near threshold is an order of magnitude larger than at higher energies. In addition, an angular correlation is observed for low incident energies. The two electrons are emitted preferentially at a large angle to each other. The results suggest strong electron correlation in this process.

Recently, $(e,3e)$ and $(\gamma,2e)$ experiments have been proposed as methods to study the correlation of two electrons in the valence shells of atoms and molecules.[1] The interpretation of such experiments in this context requires that the two electrons be ejected in a direct knockout with energies many times their binding energies. Unfortunately, this process is expected to have a very small cross section and is difficult to observe although an $(e,3e)$ process leading to the ejection of a pair of relatively low-energy electrons has been reported recently.[2] In the present work we have undertaken an investigation of double ionization events involving the Auger process which lead to a pair of relatively fast electrons. Although inner-shell electron impact ionization followed by Auger decay of the vacancy is normally viewed as occurring in two steps and no correlation is expected between the knocked-out and Auger electrons, our results show an angular correlation between the two electrons at low incident energies.

Previous work[3] using coincidence techniques to study angular correlations between Auger electrons and other electrons produced in the excitation process has concentrated on large-impact-parameter collisions at incident energies of 1 and 8 keV. These measurements involved the ejection of the $2p$ electron from argon. The results demonstrate that when the energy transferred to the $2p$ electron is just sufficient to remove it from the atom, there is a clear postcollision interaction (PCI) of the Auger electron with the slow ejected electron. This PCI gives rise to a small shift in the energy of the Auger electron.[4] Under these conditions it is not possible to interpret the results in terms of a two-step, first Born approximation model. There is also some evidence for a direct double-excitation process from the valence shell in these experiments.

In contrast to the above work, the present experiments involve events in which an Auger electron from argon near 205 eV is collected in coincidence with a second electron of the *same* energy. Creation of 205-eV Auger electron begins with the ejection of an electron from the $2p$ subshell. The final state is doubly ionized argon with two vacancies in the $3p$ subshell. The energetics of the process are best understood by treating the final state as doubly ionized argon plus two electrons with 205-eV kinetic energy. Since removal of two electrons from the $3p$ subshell requires 43.4 eV (the sum of the first and second ionization potentials), the threshold for a process in which two electrons are emitted, each with energy E_a, will be $2E_a+43.4$ eV. The same final state can also be produced by direct double ionization of two electrons from the $3p$ subshell. The two possibilities are illustrated schematically in Fig. 1. The third electron, not observed in either of these processes, has a final energy $E_0-(2E_a+43.4 \text{ eV})$, where E_0 is the energy of the incident electron.

The coincidence rates for 205-eV electrons were measured for incident electron energies in the range 400–2200 eV. Measurements were made using a modified version of

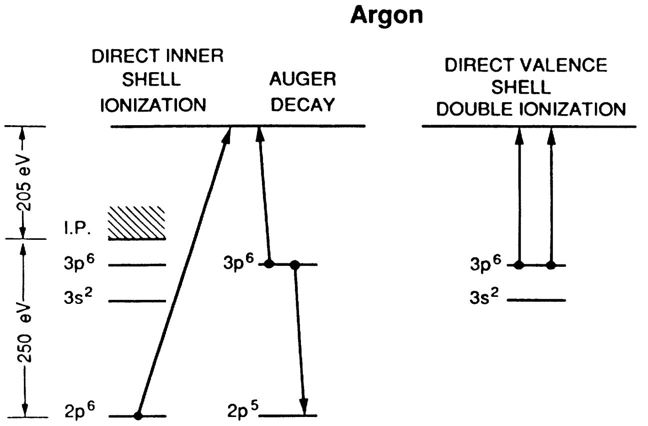

FIG. 1. Diagram showing how two electrons of equal energy can be produced via $2p$ subshell ionization and Auger decay or via direct $3p$ double ionization.

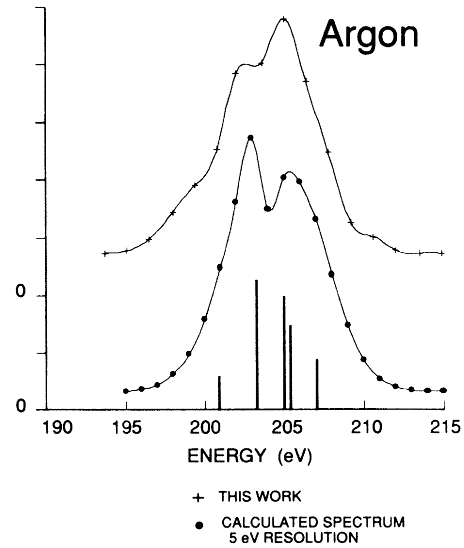

FIG. 2. Measured Auger spectrum compared with intensities obtained from Ref. 6 convoluted with a 5-eV half-width Gaussian.

an $(e,2e)$ spectrometer that has been described previously.[5] The instrument consists of an electron gun, scattering cell, and spherical condenser electrostatic energy analyzer whose configuration allows the transmission of only those electrons that leave the collision region with equal energies at polar angles near 80° with respect to the incident electron direction. The spectrometer was operated at reduced angular and energy resolution and increased collection efficiency; the angular and energy acceptances of the detectors were 0.01 sr and 5 eV, respectively. Detectors were positioned at azimuthal angles of 180°, 120°, and 60° near the focal plane of the analyzer. The included angles between pairs of detected electrons were therefore 160°, 117°, and 59°. The energy width of the incident beam was approximately 1 eV, beam currents were a few tenths of a microampere, and target densities were of the order of 10^{15} cm^{-3}. (No difference in results was noted at a factor of 10 lower density.)

Noise rejection is vital in experiments of this type since the cross sections are very small. Important features of the instrument in this regard are a completely enclosed path for the electrons and a minimum number of slits and apertures from which electrons can be scattered. In addition, a screen biased a few hundred volts positive with respect to the electron gun cathode surrounds the spectrometer and attracts a large fraction of the stray electrons.

The argon *LMM* Auger spectrum has been obtained by a number of investigators at high energy resolution. Figure 2 shows a comparison of the Auger spectrum measured in our apparatus with that obtained by convoluting the individual components of a previously measured spectrum[6] with a Gaussian profile of 5-eV half-width. The comparison shows satisfactory agreement. Since our experiment did not resolve individual Auger peaks, the detection energy was set to 205 eV, at the peak of our measured spectrum. Under these conditions, the threshold for emission of two 205-eV electrons will be 453.4 eV corresponding to zero energy for the third, undetected, electron; at an incident energy of 658.4 eV, all three electrons will have equal energy.

For each of the three detection geometries, single runs of 8–60 h were made for a given incident energy, gas pressure, and beam current. Coincidence counts and background counts due to accidental coincidences were recorded. Since the true coincidence rate is proportional to time, pressure, and incident electron current, while the background rate is proportional to the product of time and the square of the pressure-current product, relative coincidence rates can be determined by normalizing to either the product of time, pressure, and current or to the product of the background and the square root of the time. At any one angle, the two methods gave consistent results although the latter method appeared superior and was used for the data presented here.

Since little angular correlation is expected between ejected and Auger electrons at the higher incident electron energies, we made the assumption of no correlation between the directions of Auger and ejected electrons at 1000 eV and normalized data at all three included angles to the same value at this energy. Differences in coincidence rates for the three angles at other energies therefore depend explicitly on this normalization. Results are shown in Fig. 3 and in tabular form in Table I. When presented in this way, several features are apparent: (a) No coincidences are observed below the threshold value of 453 eV as expected. At 160° included angle, the coincidence rate rises sharply to a peak at 459 eV and then falls off gradually between this energy and about 650 eV. (b) At the higher incident energies, from 700 to 2200 eV, the coincidence rate at 160° is relatively constant and at least an order of magnitude smaller than the peak value. (c) The coincidence rate near threshold decreases as the included angle between the two 205-eV electrons decreases indicating a definite angular correlation between the two 205-eV electrons.

To verify that our observed coincidence rates at high incident electron energies are reasonable, we calculated the

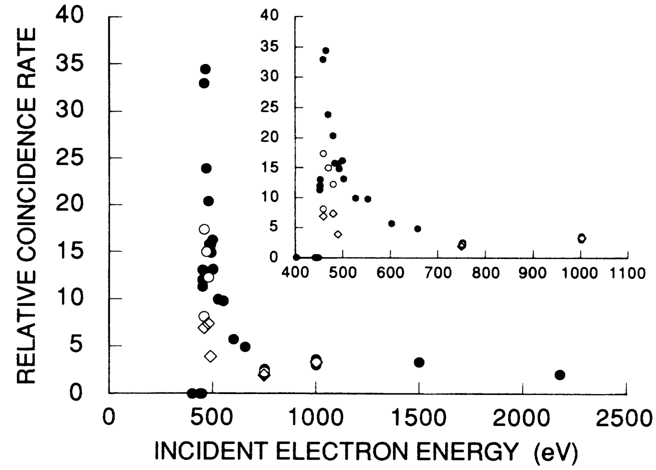

FIG. 3. Relative coincidence rates for the production of 205-eV electrons vs incident energy for relative angles of 160° (●), 117° (○), and 59° (◇).

rate from the experimental parameters, assuming a clear distinction between the scattered and ejected electrons, an isotropic distribution of Auger electrons and a distribution of ejected electrons similar to that produced by photoionization. Our singles rate for the detection of 205-eV electrons at an incident electron energy of 1000 eV was typically 200 Hz. The observed coincidence rate can be estimated from the singles rate by assuming that most of the detected electrons are Auger electrons. Since for every Auger electron there is an ejected $2p$ electron, the coincidence rate is given by the singles rate times the fraction of ejected electrons that falls within the energy and angular window of the second detector. The ejected electrons can have energies from 0 to 750.9 eV, and $\approx 0.8\%$ are within the energy acceptance of the analyzer. The solid angle subtended by the detector is 0.01 sr, and the fraction of ejected electrons within this solid angle is 0.001. This latter estimate is based on a theoretical estimate[7] of the angular distributions of the electrons. The calculated coincidence rate is approximately 1.6×10^{-3} Hz, somewhat larger than the observed rate.

At lower incident energies, near threshold, the unobserved electron will have less energy than the two 205-eV detected electrons and the distinction between "scattered" and "ejected" electron is lost; thus the large rise in coincidence rate just above threshold may be related to the characteristic energy sharing observed in near-threshold ionization.[8] Experiments have shown that for electron impact ionization, the energy of the incident electron is shared with the target electron in such a way as to leave most of the energy with one of the electrons. The probability is a minimum for both electrons to leave the ionized atom with the same energy. With our experimental apparatus, where we observe a 205-eV electron in coincidence with the 205-eV Auger electron, the optimum incident electron energy for production of the 205-eV non-Auger electron is the near-threshold value of 453.4 eV. At higher incident energies, the probability for 205-eV electron production decreases, and reaches a minimum at 658.4 eV, the energy corresponding to the production of three 205-eV electrons.

TABLE I. Relative coincidence rates vs incident energy. The incident energy scale is subject to an estimated uncertainty of approximately 0.5 eV. Uncertainties in the relative rates are 1σ statistical estimates.

Energy (eV)	Relative rate (160°)	Relative rate (117°)	Relative rate (59°)
402.7	0.0 ± 0.5		
441.9	0.0 ± 0.4		
446.0	0.0 ± 0.9		
449.0	0.0 ± 0.8		
452.0	11.3 ± 1.2		
452.0	12.0 ± 1.6		
452.8	12.0 ± 1.8		
452.9	13.0 ± 2.7		
459.3	32.9 ± 3.9	8.1 ± 3.1	6.9 ± 0.5
459.3	32.9 ± 3.2	17.3 ± 1.8	
465.1	34.4 ± 3.3		
469.2	23.9 ± 1.8		
470.2		14.9 ± 1.6	
480.0	20.4 ± 2.7	12.2 ± 1.4	7.4 ± 0.6
483.7	15.8 ± 2.2		
490.0	15.7 ± 1.9	3.9 ± 0.4	
492.8	14.9 ± 2.9		
499.8	16.2 ± 3.3		
502.4	13.1 ± 1.9		
527.3	9.9 ± 1.1		
552.3	9.8 ± 1.3		
602.3	5.7 ± 1.5		
658.2	4.9 ± 1.5		
750.0			1.9 ± 0.7
752.0		2.3 ± 0.6	
753.0	2.6 ± 0.7		
753.0	2.1 ± 1.3		
1002.0	3.6 ± 0.6		
1002.0	3.0 ± 1.0		
1003.0		3.3 ± 1.2	3.3 ± 0.8
1003.2			3.3 ± 2.2
1500.0	3.3 ± 0.7		
2180.0	2.1 ± 0.7		

To investigate the possible contribution of direct double ionization, we set the spectrometer to transmit 225-eV electrons (20 eV above the Auger energy) and recorded the coincidence rate. For an incident energy of 600 eV and the 160° geometry, the coincidence rate was at least an order of magnitude smaller than for Auger electrons, just enough above the background level to be detectable. We believe that these coincidence events were the result of direct double ionization. They most certainly are not the origin of the effects we observe, but are useful in setting an upper limit on direct double ionization cross sections. It is clear that the study of direct double ionization will require an instrument with at least an order of magnitude increase in sensitivity and noise rejection over the present one.

While the rapid rise of the relative cross section just above threshold may be explained by energy sharing arguments, the angular correlation we observe between the two 205-eV outgoing electrons near threshold is surprising.

Although a detailed explanation of our results will require further theoretical and experimental work, the angular correlations may well be related to the kind of PCI effects referred to above. On the other hand, Rau[9] has pointed out that near-threshold electrons in neutral and singly ionized atoms are expected to be highly correlated in angle and escape in opposite directions since the slow moving ejected electron can provide significant core shielding; however, it is not clear whether these effects are strong enough to produce the correlation observed in our experiment.

Although we have sampled only a very small fraction of the energies and angles associated with double ionization, the information obtained has shown that electron correlation, while a subtle phenomenon, can nevertheless give rise to rather dramatic effects. This gives us confidence that direct measurements of electron correlation are indeed possible using electron scattering coincidence techniques.

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