Energy and Angular Correlations of the Scattered and Ejected Electrons in the Electron-Impact Ionization of Argon

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Momentum distributions have been measured separately for $3p$ and $3s$ electrons in argon. The measured summed energy spectrum shows the presence of a peak which is analyzed in terms of electron-electron correlations.

In order to obtain full information on the single ionization of an atomic system by an incident electron it is necessary to determine the kinematics of the two outgoing electrons separately. Such "$(e, 2e)$" experiments provide the most sensitive tests of the various theoretical approximations to the ionization process[1] and open the possibility of accurately measuring the momentum distribution of the electrons in different states of atoms, molecules, and crystals[2] as well as testing the validity of the independent-particle model of the atom and the residual ion.[3]

Measurements of angular correlations between the outgoing electrons in electron-impact ionization of atoms have recently been reported by Ehrhardt *et al.*[4] and Camilloni *et al.*[5] The former group has restricted itself to low incident energies and the very asymmetric coplanar situation in which the "scattered" electron is emitted with nearly all of the available energy at a small angle while the "ejected" electron is emitted with low energy at a relatively large angle to the incident direction. Such experiments, characterized by low momentum transfer to the "ejected" electron, provide sensitive tests of ionization theories at low energies. This group has made some measurements on argon, although their main emphasis has been placed on using helium as a testing ground for theories of ionization. The work of Camilloni *et al.*[5] on carbon was restricted to very high incident energies (9 keV) and coplanar symmetric geometry, in which the outgoing electrons had the same energy and made the same angle with the incident beam, resulting in high momentum transfer to the "ejected" electron. The use of a solid carbon target and very high incident energies resulted in limited energy resolution, preventing the separation of the outer $2p$ and $2s$ single-particle states. However the correlation obtained for the inner $1s$ state, when analyzed in the framework of the impulse approximation, gave the first direct $(e, 2e)$ measurement of the momentum distribution of an electron in an atomic single-particle state.

In this Letter we report the angular and energy correlations obtained for electrons emitted from single-particle states in the outer shell of argon for the symmetric noncoplanar situation, in which the angular variable is the azimuth φ of one of the outgoing electrons, measured from the plane defined by the incident electron and the other outgoing electron. Both electrons are emitted at an angle θ of 45° relative to the incident direction. This includes the first data on p-state electron momentum distributions as well as the first evidence from $(e, 2e)$ measurements of electron-electron correlations in atomic states. The present communication is limited to results obtained for argon with incident energies near 400 eV. Full details of the apparatus and measurements at other energies will be presented in a subsequent paper. The apparatus is shown schematically in Fig. 1. The target gas was admitted to the interaction region through a fine nozzle. A set of deflection plates was used to center the electron beam in a 2.5-mm aperature in a guard electrode placed in front of a Faraday cup. The pressure in the interaction region was estimated to be of the order of 10^{-3} Torr, with a pressure rise in the vacuum chamber of 10^{-5} Torr compared with the base pressure of less than 10^{-6} Torr.

The effective size of the interaction region was limited by the width of the electron beam (approximately 1 mm) and by two cylindrical 1-mm slits of radius 6 mm mounted coaxially with the electron beam. The electrons leaving these slits passed through the entrance apertures of the two cylindrical mirror analyzers mounted coaxially with the electron beam. These entrance apertures were circular when viewed at $\theta = 45°$ with a full width of 5°. The aperture on the larger analyzer could be rotated in azimuth by $\pm 75°$. Great care was taken to reduce stray electric

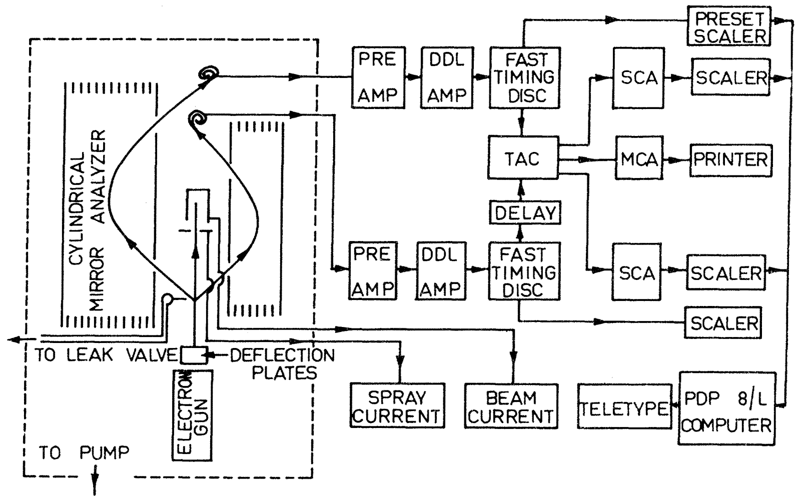

FIG. 1. Schematic diagram of the apparatus. For details see text.

fields. Earth's magnetic field was reduced to less than 50 mG by the use of Helmholtz coils. Pulses from the two-channel electron multipliers were processed as shown in Fig. 1. The two single-channel analyzers (SCA) at the output of the time-to-amplitude converter (TAC) were used to determine the coincidence counting rates. The multichannel analyzer (MCA) was used to monitor the random coincidence counting rates and the time resolution, typically 12 nsec.

Figure 2 shows the energy correlation which is observed when the emitted electrons each have a

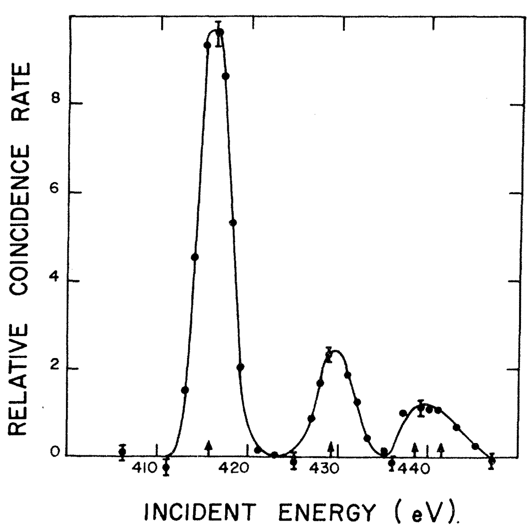

FIG. 2. Coincidence rate versus energy of the incoming electron for $\varphi = 10°$, $\theta_1 = \theta_2 = 45°$, and $E_1 = E_2 = 200$ eV. The arrows indicate the positions of the ground state and various excited states in Ar II discussed in the text.

fixed energy of 200 eV and with $\varphi = 10°$. Three distinct peaks are observed at incident energies of 415.7, 429.3, and approximately 440 eV. The first two correspond to the binding energies of 15.76 and 29.3 eV of the $3p$ and $3s$ single-particle states of argon, leaving the ion in its ground ($3s^2 3p^5\, {}^2P°$) and first excited ($3s^1 3p^6\, {}^2S_{1/2}$) states, respectively.

The group of electrons with an energy loss of 40 eV does not correspond to any single hole state in the closed-shell configuration of argon. Auger processes and autoionization can be ruled out as sources of this group of coincident electrons since the angular correlation shown in Fig. 3(b) is peaked for coplanar geometry, with the coincidence counting rate decreasing rapidly as the azimuth φ increases from 0° to 30° out of the plane ($K \approx \alpha_0^{-1}$). Such peaking is to be expected for fast two-body collisions,[1] whereas the angular correlation expected for more complicated processes, such as those proceeding through relatively long-lived states, should be more nearly isotropic.[6]

States involving shell-model configuration with excited valence electrons can be excited if there are correlations in both the initial atomic and final ionic states.[3] It is interesting to note that Luyken, de Heer, and Baas,[7] in their optical study of the excitation of the $3s3p^6\, {}^2S_{1/2}$ level of Ar II by electron-impact ionization of Ar, used configuration interaction in the final state in order to explain the difference between their measured cross sections and calculated values.[8] From optical data these authors found that they

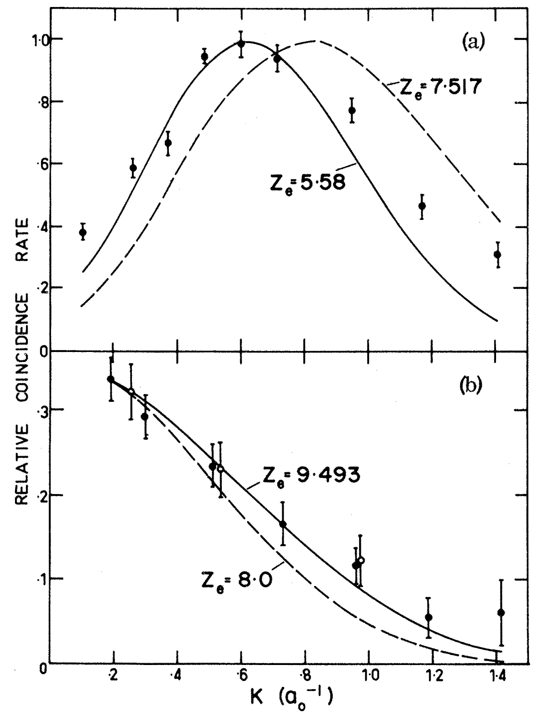

FIG. 3. The momentum distributions observed for (a) the $3p$-state and (b) the $3s$-state electrons in argon. In (b) the solid circles are the points obtained for the peak at 429.3 eV whereas the open circles are those obtained for the peak at approximately 440 eV after multiplication by a factor of 1.3. The solid and dashed curves are the predictions obtained by using screened hydrogenic wave functions with different values of the effective charge.

required considerable mixing of the $3s3p^6\,{}^2S_{1/2}$, $3s^23p^4(^{\prime}D)3d\,{}^2S_{1/2}$, and the $3s^23p^4(^{\prime}D)4d\,{}^2S_{1/2}$ configurations in order to describe the $3s$ hole state. According to their eigenfunction expansions, these latter two states, indicated respectively by arrows at 438.5 and 441.2 eV in Fig. 2, should be excited with a summed probability of 0.38, whereas that of exciting the $3s3p^6\,{}^2S_{1/2}$ state should be 0.62. By integrating over the experimental angular correlations for the 40- and 29.3-eV peaks we find that the relative cross sections for these peaks are 0.42 ± 0.04 and 0.58 ± 0.04, respectively. Although these values and the summed energy spectrum are consistent with the excitation of the states suggested by Luyken, de Heer, and Baas, our energy resolution is inadequate to permit positive identification of these states. In this respect it is interesting to note that the $3s^23p^44s\,{}^2S_{1/2}$ state is not excited to any marked degree since this would lead to a peak centered about 436.5 eV in Fig. 2.

Further evidence that the 40- and 29.3-eV peaks are related can be obtained from their respective electron momentum distributions shown in Fig. 3(b). The two distributions are very similar and agree very well with that expected from the impulse approximation for the ejection of our electron from a hydrogenic $3s$ state. A more detailed theoretical investigation on the information to be obtained on initial- and final-state electron correlations is being carried out.[9]

In the impulse approximation, the cross section for the simultaneous detection of emitted electrons with momenta $\vec{k}_1$ and $\vec{k}_2$ due to an incident electron with momentum $\vec{k}_0$ is given by

$$\sigma(\vec{k}_1, \vec{k}_2, \vec{k}_0) \propto (d\sigma_{ee}/d\Omega_1)|F_{if}(\vec{K})|^2,$$

where $\vec{K} = \vec{k}_1 + \vec{k}_2 - \vec{k}_0$ and $d\sigma_{ee}/d\Omega_1$ is the Mott scattering cross section. In the self-consistent field approximation, $F_{if}(\vec{K})$ is merely the momentum-space wave function of the ejected electron. For our symmetric noncoplanar geometry,

$$K = |(2k_1\cos\theta - k_0)^2 + 4k_1^2\sin^2\theta\sin^2(\tfrac{1}{2}\varphi)|^{1/2}.$$

Figure 3 shows the angular correlations obtained for the ejection of atomic $3p$ and $3s$ electrons compared with the predictions of the impulse approximation when hydrogenic $3p$ and $3s$ states are used. The instrumental angular resolution has been folded into the theoretical calculation. An increase in the effective charge increases the spread of the distribution proportionally. The values of effective charge which give the best fit can be compared with those of 7.517 ($3p$) and 9.493 ($3s$) obtained from Hartree-Fock calculations for argon and used by Omidvar, Kyle, and Sullivan[10] for the calculation of total inner-shell ionization cross sections. The measured distribution for the $3p$ electrons at low and high momenta is considerably greater than that predicted by this simple theoretical model. This could be due to distortion of the incoming and outgoing waves or due to the inadequacy of using simple hydrogenic wave functions to describe the motion of the $3p$ and $3s$ electrons. Theoretical calculations including both these effects are being pursued.[9]

Our values for the ratio of the cross section for ionization by emission of an electron from the s state to that from a p state is 0.47 ± 0.05. This value, for the symmetric situation in which the momentum transfer to the ejected electron is large, is in marked contrast with the value of approximately 0.1 of Luyken, de Heer, and Baas[7] and various theoretical estimates.[8,10,11] Since the latter are total ionization cross sections integrated over all angles and energies, the difference may be due to the smallness of the 3s cross section at small momentum transfer.[7]

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Surface States of He^3 in Dilute He^3-He^4 Solutions*

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We present in support of Andreev's model a fully microscopic calculation of the surface states of He^3 in dilute He^3-He^4 solutions. The surface structure of pure He^4 is first determined. We find a surface tension of 0.36 dyn/cm, in good agreement with experiment. For He^3 there is just one bound surface state, at an energy 1.6°K lower than the chemical potential of He^3 in bulk solutions, centered on the Gibbs surface of the liquid.

The experiments by Esel'son and co-workers[1,2] first indicated that the presence of He^3 impurity in superfluid He^4 lowers the surface tension σ, and that the extent of lowering exceeds what one might expect from the usual rule of additivity. To interpret these results, Andreev[3] suggested that there exist surface states of He^3 with a minimum energy below that in the bulk. While at low concentrations the spectrum of He^3 at large distances from the surface can be described by

$$\epsilon(k) = -\mu_3 + \hbar^2k^2/2m^*, \tag{1}$$

it is modified near the surface to read

$$\epsilon_s(k) = -\mu_3 - \epsilon_0 + \hbar^2k^2/2m_s^*, \tag{2}$$

where ϵ_0 represents extra binding which drives He^3 atoms toward accumulation in the surface layer. Subsequent surface-tension measurements by Zinov'eva and Boldarev[4] determined ϵ_0 at 1.7 ± 0.2°K and m_s^* at $(0.9 \pm 0.1)m_3$. More recently, Guo *et al.*[5] carried out extensive work at lower temperatures and redetermined ϵ_0 at 1.95 ± 0.1°K and m_s^* at $(2.07 \pm 0.1)m_3$. The disagreement, mainly in m_s^*, is not surprising, since the latter experiment was carried out in the fully degenerate region where the He^3 monolayer acts like a two-dimensional Fermi liquid.

Andreev's model has found further support in the totally unrelated experiments on vortex-ring