Electron correlation effects near threshold for electron-impact ionization of helium*

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Using a modification of the trapped-electron technique, we have measured the total intensity of electrons inelastically scattered from helium as a function of both incident and final energies near the ionization threshold. We find that a "cusp," which appears in the scattered electron intensity at the ionization threshold for electrons of near-zero final energy, decreases in visibility as the energy of the scattered electrons increases. For scattered-electron energies greater than about 2.0 eV, the intensity is smooth through the ionization threshold. The physical reason for the cusp is the formation of states with two excited and strongly correlated electrons. These experiments define the range of excess energies above the ionization threshold for which correlation effects are important, and hence measure the energy range of validity of the Wannier law of threshold ionization.

Despite the two decades since the formulation of the Wannier law of threshold ionization,[1] the energy range of validity of this law remains indeterminate theoretically. Recently Cvejanovic and Read[2] convincingly verified experimentally that the Wannier $E^{1.127}$ threshold ionization law is valid for electron ionization in He to at least 1.7 eV above the ionization threshold for scattered energies of less than 0.02 eV. In this paper an alternative method is presented for determining the energy range of validity of the Wannier law based on observations of an electron correlation effect at the ionization threshold. Cvejanovic and Read[2] measured the intensity of electrons scattered from He with near-zero final energy (<0.02 eV) as a function of the incident electron energy E, and observed a series of well-resolved inelastic peaks as E traversed successive inelastic thresholds below the ionization threshold. For values of E well above or below the ionization threshold I, they found the *average* intensity was roughly independent of E. However, for incident energy $E=I$, a dip or "cusp" appeared in the intensity of scattered electrons. The "cusp" at the ionization threshold has been interpreted by Fano[3] as a manifestation of the formation and decay of states with two excited and strongly correlated electrons. If correlation effects are important, as implied by the Wannier calculation and suggested by Rau[4] and Fano,[3,5] then clearly the threshold "cusp" should disappear for scattered energies in excess of the range of validity of the Wannier theory, as suggested by Fano.[3]

The technique of Cvejanovic and Read[2] is constrained to measurements of threshold spectra, i.e., measurement of electrons with near-zero final energy. The technique used in the present experiment does not have this limitation, and we make the first observations of a gradual decrease in the visibility of the cusp as the excess electron energy increases. The average intensity becomes smooth across the ionization threshold when the excess energy is greater than about 2 eV. Thus the threshold data of Cvejanovic and Read[2] are confirmed using a different technique, the suggestion of Fano[3] regarding the disappearance of the cusp is confirmed, and an alternative method to that of Cvejanovic and Read is provided for determining the energy range of validity of the Wannier threshold law.

THEORETICAL BACKGROUND

Classical[1] and quantum-mechanical[3-6] theories of ionization show that for energies E only slightly above the ionization threshold I, ionization can only occur if the positions of the incident and excited electrons remain tightly correlated (i.e., moving in opposite directions at the same speed) up to a large distance from the ion core. This critical distance, known as the "Wannier radius," r_c, is defined by $3e^2/2r_c = E - I$. Once past the critical radius, the residual kinetic energy of the electrons predominates and ionization proceeds.

While the angular correlation is stabilized by electron repulsion, the radial correlation is inherently unstable. If one electron achieves a higher escape velocity, it will draw ahead of the other and experience a lesser attraction by the core. For a large loss of correlation, one electron will remain within this Wannier radius, trapped in a highly excited bound orbit, instead of reaching ionization. Instability in the radial correlation thus *reduces* the probability of ionization near threshold from a rate proportional to $(E-I)$ expected from a simple phase-space volume, to the Wannier rate, proportional to $(E-I)^{1.127}$. Similarly, below the ionization threshold where only bound states can be excited, the excitation of levels very close to the incident energy E can only occur if the electron motion remains correlated in the same manner as required for ionization near threshold. This correlation can extend up to a Wannier-type radius r_c, now defined by $3e^2/2r_c=(I-E)$, at which distance the kinetic energy of both electrons is fully expended.

Again, however, instabilities in the radial correlation make it unlikely that the electron pair will remain correlated up to the limiting radius r_c. Thus, the probability of excitation to levels near the maximum available energy E should be *depressed* when E approaches the ionization threshold I.

The above theoretical argument, due to Fano,[3] satisfactorily explains the cusplike structure observed by Cvejanovic and Read[2] at the ionization threshold to be a manifestation of electron correlation.

EXPERIMENT

The experimental technique used for these measurements is an improved version of the "double retarding-potential-difference" (DRPD) technique used by Knoop and Brongersma,[7] and only a brief description of the apparatus, shown schematically in Fig. 1, will be given here. Electrons are emitted by a thoria-coated iridium filament F, and a monoenergetic beam of electrons is selected by the trochoidal monochromator (TM).[8] The electron beam is accelerated to an energy $e(V_a+W)$ into the gas-filled collision chamber C, where electrons which have lost energy between $e(V_a+W)$ and eV_a in an inelastic collision remain trapped by the potential well W and are collected by the mantle M. However, by modulating the well depth W by a small voltage ΔW (about 0.025 V in the present experiments), and measuring the ac component of the current at M with a lock-in amplifier (as used in the DRPD technique),[7] only electrons whose final energy is equal to W (within about ΔW) are *detected*. This technique differs from the usual trapped electron technique[9] in that electrons with final energy less than W are *not* detected. As Cvejanovic and Read[2] have verified that the energy distribution of ejected electrons is flat to at least $E-I\approx 1$ eV, our detection efficiency is independent of E at least up to $E-I\approx 1$ eV, i.e., we have equal probability of detecting *one* electron to at least $E-I\approx 1$ eV. Fixing W and increasing V_a produces a spectrum of inelastic peaks whose magnitudes are proportional to the total inelastic cross section (i.e., cross sections integrated over all angles) at an energy eW above their respective thresholds (i.e., the excess energy of the detected inelastically scattered electrons is eW). Setting $W\approx 0$ and increasing V_a produces a threshold electron spectra similar to that obtained by Cvejanovic and Read,[2] who use an entirely different technique.

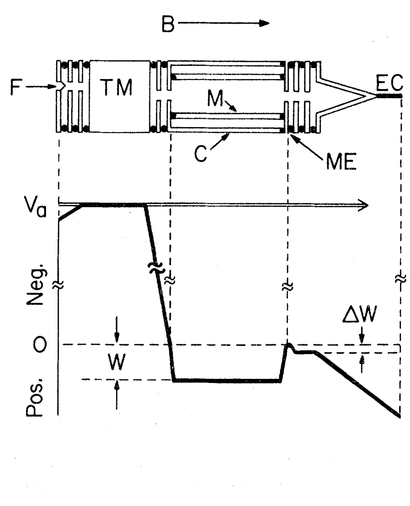

FIG. 1. Schematic diagram of the tube and the potential distribution along the axis of the electron beam.

In the present experiments successive scans of incident electron energy are made, and the signal in the lock-in amplifier is stored in a 512-channel analyzer to improve signal/noise ratios. Gas pressure in the collision chamber is typically 5×10^{-4}.

Primary electron beam currents are typically a few 10^{-9} A. Below the ionization threshold, the measured signals are proportional to the total inelastic cross sections. Above the ionization threshold, the signal is proportional to a partial ionization cross section, $\sigma^i_{\text{partial}}$. This occurs because only a part, ΔE, of the energy distribution (which is flat[2]) is detected. Above the ionization threshold, the measured current is proportional to $\sigma^i_{\text{partial}}=[\Delta E/(E-I)]\sigma^i_{\text{total}}$. Now $\sigma^i_{\text{total}}\propto(E-I)^n$, where n is the Wannier exponent. Hence, above the ionization threshold, the measured signal is proportional to $(E-I)^{n-1}$ within the range of excess energies for which the Wannier law is applicable.

RESULTS

Figure 2 shows the spectra of scattered electrons from e-He collisions whose final energy is 0.07, 1.0, and 2.0 eV. Though the resolution in these experiments is considerably worse (0.200 eV) than in the experiments of Cvejanovic and Read[2] (0.03 eV), the average decrease in the intensity near the ionization threshold is clearly visible for $W = 0.07$ V. Above the ionization threshold for the curve $W = 0.07$ V, the intensity increases somewhat faster than $(E-I)^{0.127}$. This is purely an instrumental effect and is due to a monotonically increasing background above $E = I$, which could be observed by making $W = 0$, but has not been subtracted on this figure. This background, indicated by the dashed line for $W = 0.07$ V of Fig. 2, is a nonlinear function of the target gas pressure and may be due to trapping of electrons which have been scattered at large angles. Such electrons may have sufficient energy to escape the trap, but require multiple collisions in order for their velocity vector to be reoriented in the correct direction for escape. The effect of this "geometric trapping" in magnetically confined beams is an increase in the residence time,[9] and hence the effective path length of the scattered electrons in the collision chamber. This effect has been discussed in great detail for pure s-wave scattering by Burrow and Schultz,[10] and for p-, d-, and f-wave scattering by Spence *et al.*[11] Expressions for the fraction of scattering events leading to "geometric trapping" in magnetically confined beams have been given by Golden.[12] In general, calculation of the effect is difficult as the extent to which each partial wave contributes to the scattering process is usually not known. Consequently we have studied the effect of varying all the parameters that could have some effect on our observed spectral shapes. For a collision chamber of dimensions 10-cm length $(L) \times 4$-mm diameter (D), there is (somewhat surprisingly) no visible effect on the spectral shape when the exit hole of the collision chamber is increased from $\frac{1}{10}D$ to D. Measurements were made for magnetic field strengths between 200 and 1000 G. Decreasing the magnetic field causes a slight increase in the magnitude of the background mentioned above, as expected from calculations.[12] However, the observed spectral shapes are invariant to magnetic field strength above about 500 G and the data of Fig. 2 were taken at 750 G. The observed background is, however, strongly dependent on the gas pressure and was studied for pressures between 5×10^{-5} and 10^{-3} torr. Again spectral shapes are invariant below about 1 or 2×10^{-4} torr. At 10^{-4} torr the observed signal is consistent with an $(E-I)^{0.127}$ power law above the ionization threshold. Unfortunately, in this case, the signal-to-noise ratio that is achieved with a reasonable running time is rather poor, and this aspect was not the primary motivation for the present experiments. This background effect is negligible at higher values of W, where signal intensities become greater.

The curve for $W = 1.0$ V of Fig. 2 shows that for scattered energies of 1.0 eV, the cusp at the ionization threshold has largely disappeared, although a drop in the average intensity is still evident, and the shape of the curve above $E = I$ is consistent with the $(E-I)^{0.127}$ law. It is not until the excess energy of the scattered electrons is about 1.5–2 eV that the electron yield is essentially smooth through the ionization threshold, as shown in the curve for $W = 2$ V of Fig. 2. The background effects observed by varying the parameters mentioned above have no effect on the observed excess energy at which the cusplike structure disappears. The variation in the intensity through the ionization threshold is shown more clearly in Fig. 3, where we have superimposed several curves of different W. Though each of these curves represents vastly different

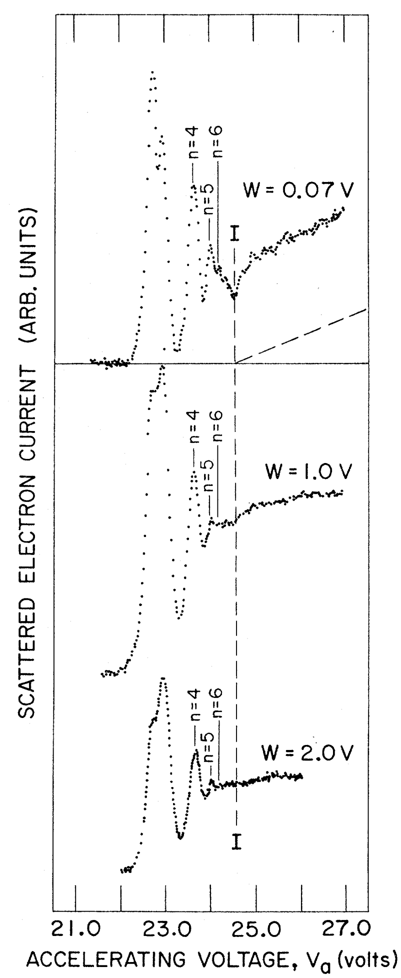

FIG. 2. Spectra showing total yield of scattered electrons from He with final energies eW as a function of the incident energy $e(V_a + W)$. The ionization threshold is marked I and the target pressure is $\approx 5 \times 10^{-4}$ torr.

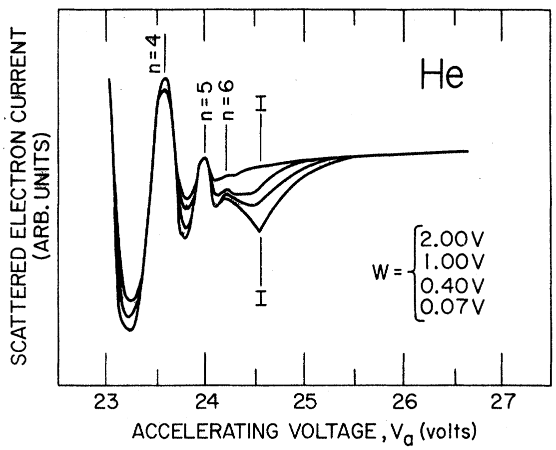

FIG. 3. Comparison of spectra of scattered electron intensities as a function of incident energy $e(V_a + W)$ for different values of final energy eW. The ionization threshold is marked I. The magnitude of the spectra have been arbitrarily normalized to give best visual fit, and the target pressure is $\approx 1\times 10^{-4}$ torr.

signal sizes, their magnitudes have been arbitrarily changed for best visual fit. The instrumental resolution is seen to be virtually independent of W. For Fig. 3 we have utilized the low-pressure data mentioned above, and to avoid confusion, the individual data points have been replaced by smooth curves. This figure clearly demonstrates that correlation effects are important, and hence the Wannier formulation is valid, for excess energies up to about 2.0 eV.

Whereas at first sight this may appear to be a surprisingly large energy, our conclusions are complemented by the results of Cvejanovic and Read.[2] They find that the Wannier threshold law,[1] in which correlation effects are *implied*,[3] does, in fact, hold extremely well to energies of 1.7 eV above the ionization threshold for scattered energies of less than 0.02 eV. Preliminary measurements indicate that similar effects occur in electron-argon collisions, though in this case we have not yet determined the range of excess energies over which correlation effects are important. Though many theoretical models of threshold ionization have been proposed, the energy range of validity of these models has usually remained unspecified. It is hoped that the present work will provide further impetus on this aspect of electron-impact ionization theory.

Note added in proof. Since submission of this manuscript, we have learned (R. J. Celotta, private communication) that N. Swanson and R. J. Celotta, National Bureau of Standards, Washington, have obtained spectra similar to those of Fig. 2, for excess electron energies between about 0.2 and 2 eV using a double electrostatic hemispherical analyzer apparatus. Consistent results were not obtainable below about 0.2 eV because of their rapidly falling apparatus transmission function with decreasing electron energy.

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