

Deep interference minima in experimental ionization differential cross sections

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A deep, narrow interference minimum that exists for symmetric noncoplanar ($e,2e$) ionization experiments, first discovered in helium, has now been found in neon. In helium, the $1s$ electron is ionized, whereas in neon, the minimum is observed ionizing the inner $2s$ electron. The depth of the measured minima indicates that a simple, underlying coherent process is occurring in the reaction.

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Fundamental to the understanding of all physical processes on an atomic scale is a theoretical framework based upon quantum mechanics and wave-particle duality. The success of the quantum theory has been proven time and again, and has led to many of the technological and scientific advances that evolved during the latter part of the 20th century. This theory provides the framework for all successful models of atomic, molecular, and nuclear reactions including elastic scattering, excitation, ionization, and fragmentation.

Under certain conditions the wave nature of the particles manifests itself directly, allowing characteristics of this nature to be measured. The most usual manifestations are diffraction and interference, where both the amplitude and phase of the wave front play a key role. Diffraction of electrons at a boundary is routinely observed in electron microscopy, and can be directly related to the de Broglie wavelength determined from the electron momentum.

Wave effects such as interference minima are less frequently observed, since a coherent summation of both the amplitudes and phases of contributing wave vectors relating to the process under study is necessary to produce a minimum. One striking example where quantum-mechanical interference is seen is in elastic scattering of electrons from atoms [1–3]. In this case, a number of minima are observed in the elastic differential cross section as a function of scattering angle. This is a direct result of the complex scattering amplitudes relating to the incident and scattered electrons adding coherently to produce minima in the cross section. A further manifestation of interference is seen in electron-impact ionization through a resonance target state. In this case, a Fano profile may be observed in the scattered electron cross section [4,5]. This is a result of an interference between two possible pathways to the continuum, either directly or via the resonance state.

Observations of strong interference effects in electron-impact experiments become fewer as the complexity of the reaction increases, since the probability of occurrence of these minima decreases. This is a consequence of the stringent requirement that both the amplitude and phase of the scattering amplitudes contributing to the reaction must coherently add to be very small to produce a sharp minimum in the measured cross section. For inelastic electron-scattering experiments involving target excitation, the incident channel involves an electron and a neutral target, as does the final channel following excitation. It is necessary to include the complex process of resonant target excitation in the model,

together with any subsequent decay of the target following excitation. These additional terms reduce the likelihood that sharp minima will be observed, although evidence of such effects has been found [6].

For electron impact ($e,2e$) ionization experiments, the reaction is substantially more complex. In the incident channel, an electron and neutral target are again involved, while in the final channel following ionization two electrons and an ion are involved. Coulombic interplay between all the charged particles leads to postcollisional interactions between the outgoing electrons, the remaining bound-state electrons, and the ionic core. At low incident energies short- and long-range correlations, ingoing and outgoing channel wave-front distortions, postcollisional interactions, and electron exchange processes all play significant roles in the reaction. The processes leading to ionization in this energy regime are therefore varied and complex.

The most common geometry chosen for these ($e,2e$) experiments is coplanar geometry, where the incident, scattered, and ejected electrons all occupy the same plane. In these experiments, the complexity of the reaction is reflected in the differential cross sections that are measured, where maxima and minima are observed due to the summation of many different contributing partial waves [7–9]. For noncoplanar geometries, the interactions that lead to ionization are further complicated, since multiple-scattering processes must be involved. Again, broad maxima and minima are generally observed due to the complex interaction of many partial waves [10–13].

It is for these reasons that the results presented in this paper are so remarkable and unexpected. Deep and narrow interference minima are observed in the ionization differential cross section from both helium and neon in noncoplanar geometry at low incident energies. Under these conditions the reaction is expected to be complex, yet the results indicate that a simpler, coherent reaction is occurring. The helium results that have been presented previously are for ionization from the outer $1s$ shell [12]. The results presented here for neon are from the inner $2s$ shell, and so ionization is further complicated by the influence of the outer closed shell of $2p^6$ electrons on the ionized $2s$ electron as it emerges from the reaction. In both helium and neon, the interference minima are found to be narrow and deep, indicating that the scattering amplitudes describing all contributions to the reaction coherently sum to be close to zero. For this to occur in one target seems unlikely, but is conceivable. For two quite

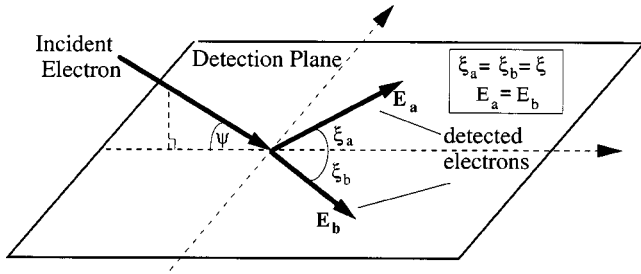


FIG. 1. The experimental geometry that produces deep and narrow interference minima in the ionization differential cross section.

different targets, one from a valence shell and the other from an inner shell, to show such effects indicates a more fundamental underlying process is occurring. The nature of this process remains an open question at this time.

Figure 1 shows the experimental geometry that is adopted in these experiments. The deep, narrow interference minima are found to occur only in a noncoplanar geometry ($\psi \neq 0$) and require that the scattered and ejected electrons emerge from the reaction with equal energy and with equal angles with respect to the projection of the incident electron momentum onto the detection plane ($\xi_a = \xi_b = \xi$). A high degree of symmetry appears to be necessary in the final channel for these effects to be observed [14].

Since the electrons emerge from the reaction in a plane that does not contain the incident electron momentum, multiple scattering must occur. A number of different scattering mechanisms are possible. One possibility is that the incident electron initially scatters from the target into the detection plane followed by a binary collision between incident and ejected electrons leading to ionization. A second possibility is that the incident electron suffers a binary collision with a bound electron followed by both electrons scattering from the ionic core into the detection plane. Other higher-order processes that lead to symmetric ionization into the detection plane are also possible.

Figure 2 shows the deep and narrow interference minimum previously discovered in helium, together with results for coplanar geometry ($\psi = 0^\circ$) and the perpendicular plane ($\psi = 90^\circ$) that are presented for comparison [12]. In this case the incident electron energy was 64.6 eV and the scattered and ejected electrons were each detected with an energy of 20 eV. The interference minimum is seen at an incident beam angle $\psi = 67.5^\circ$ to the detection plane and the minimum appears at a value near $\xi = 70^\circ$. The analyzers had detection angles of $\pm 3^\circ$ and the incident electron pencil angle was $\pm 2^\circ$. This apparatus angular profile tends to fill out the minimum, and so the true minimum is deeper and narrower than measured. The actual depth was estimated by deconvolving a Gaussian angular profile of $\pm 4.5^\circ$ from the results as shown. From this analysis, the minimum is estimated to be very close to zero, indicating almost complete cancellation of all contributing scattering amplitudes.

This remarkable result prompted a number of different theoretical groups to attempt to calculate this minimum. The first calculations of Berekdar and Briggs [15] emulated the minimum by considering scattering amplitudes for the incident, ejected, and bound spectator electron that remained

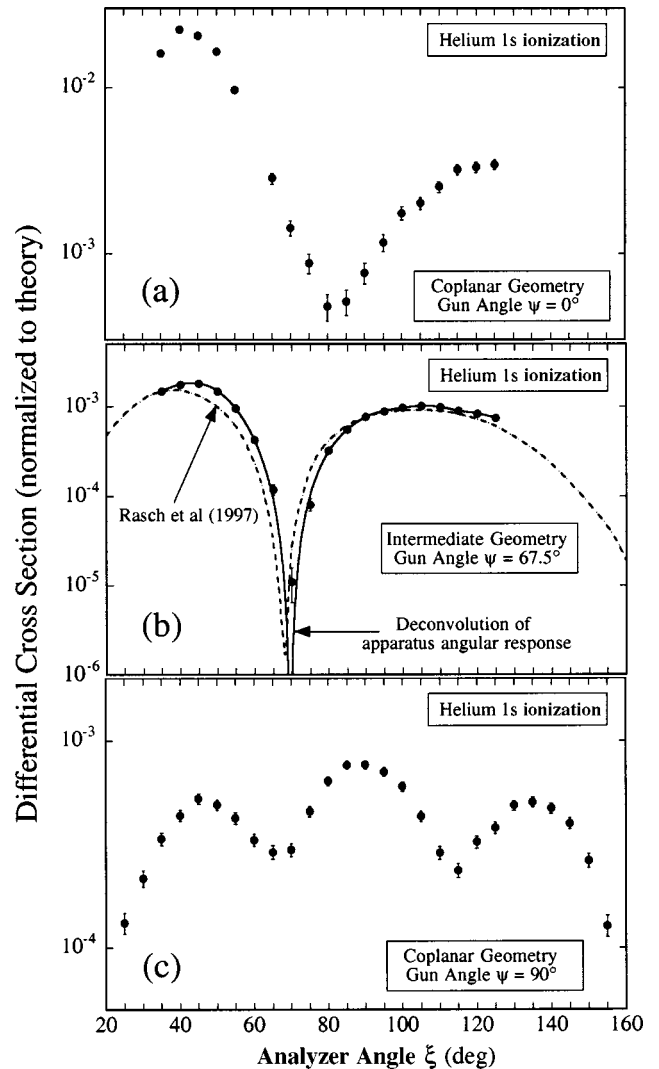


FIG. 2. The $(e,2e)$ ionization cross section for helium at 64.6 eV incident energy for (a) $\psi = 0^\circ$, (b) $\psi = 67.5^\circ$, and (c) $\psi = 90^\circ$, showing the deep, narrow minimum at $\psi = 67.5^\circ$ for $\xi \sim 70^\circ$. The result of deconvolving the estimated apparatus angular response is shown together with the calculations of Rasch *et al.* [17].

with the ion. They argued that the coherent addition of all three complex scattering amplitudes must add to zero to produce the observed dip, but when the resulting parameters are applied at other incident angles the theoretical results failed to match experimental data. Clearly any calculation that attempts to explain the interference minimum must also predict the experimental data for all other geometries. This provides a considerable challenge to any theory.

Khajuria and Tripathi [16] used a distorted-wave Born approximation (DWBA), but when the parameters are adjusted to produce the interference minimum, this theory again does not predict the cross section for other scattering geometries. Rasch *et al.* [17] have also used a DWBA theory together with a modified Gamow factor representing final-state electron correlations to reproduce the interference minimum. Results of their calculation at other scattering geometries are not available, although the position of the minimum is reasonably well reproduced with an angle of ξ

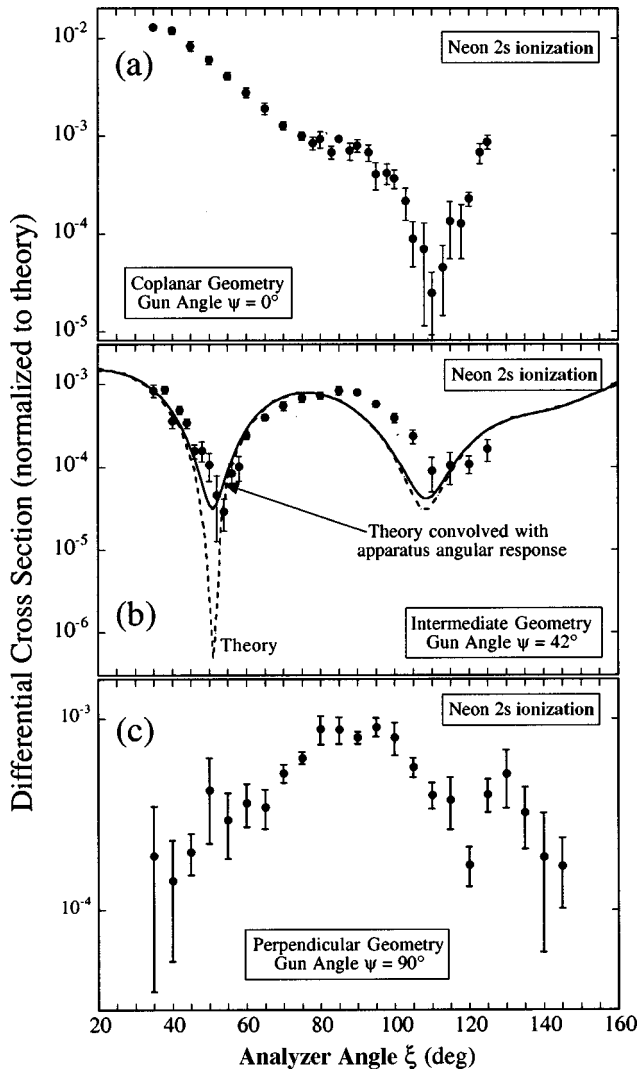


FIG. 3. The $(e,2e)$ ionization cross section from the neon 2s inner shell for (a) $\psi=0^\circ$, (b) $\psi=42^\circ$, and (c) $\psi=90^\circ$. The theoretical prediction for $\psi=42^\circ$ is shown together with convolution of this calculation with the angular response of the apparatus.

$=68^\circ$ being close to experimental observation, as shown in Fig. 2. The depth of the predicted minimum does not match the deconvolved estimate. It remains to be seen if this theory can reproduce the results at other angles as is required.

Rasch *et al.* [17] extend their theory to look for similar effects in different targets, and predicted deep and narrow interference minima from s -shell ionization in neon, argon, and lithium. Specifically, they predict such a minimum for the ionization of the 2s shell of neon at an incident electron energy of 110.5 eV for an incident angle $\psi=42^\circ$.

These predictions prompted the experiments that are detailed here to be conducted, since the incident and detected electron energies were within the range accessible to the apparatus at Manchester. Further, the overall magnitude of the ionization cross section was predicted to be similar to that obtained for helium, with the minimum occurring at a detection angle $\xi=51^\circ$.

Figure 3 shows the experimental results. The energy of

the incident electron was 110.5 eV and the scattered and ejected electrons were detected with equal energy of 31 eV. Measurements were taken for incident electron angles $\psi=0^\circ$ (coplanar geometry), $\psi=42^\circ$, and $\psi=90^\circ$ (the perpendicular plane geometry). By measuring the cross section at these angles, a more stringent test of theory can be conducted, as noted above for helium.

The results for coplanar geometry ($\psi=0^\circ$) show that the cross section varies over a very large range of nearly 1000: 1 from forward scattering at $\xi=35^\circ$ through to a minimum in the cross section at $\xi=110^\circ$. A point of inflection is seen around $\xi=80^\circ$. For all results described here, the cross section must be zero at $\xi=0^\circ$ and 180° due to postcollisional interactions between outgoing electrons of equal energy. Hence, the cross section must decrease for $\xi<35^\circ$ and $\xi>125^\circ$ where measurements cannot be conducted due to apparatus constraints [18].

These coplanar results are similar to the measurements of Rioual *et al.* [9] who performed coplanar symmetric ionization experiments from the 2s shell at 126.9 eV incident energy, although the minimum seen here at $\xi=110^\circ$ is deeper. Rioual *et al.* [9] indicate that the effects of polarization of the target are important to explain the shape of this cross section. It remains to be seen whether these calculations can reproduce the coplanar results presented here, with the added constraint that they must also reproduce the noncoplanar results of Figs. 3(b) and 3(c).

The data for the perpendicular plane ($\psi=90^\circ$) show that the cross section is a maximum at $\xi=90^\circ$ in this geometry. A three-peak structure is observed with peaks at $\xi=50^\circ$ and 130° , although the relative uncertainties in the data make it difficult to ascertain further details. This structure is similar to the perpendicular plane results previously seen in helium, as shown in Fig. 2 [10].

At an incident electron angle $\psi=42^\circ$ an interference minimum is observed in the cross section at $\xi\sim 55^\circ$. A second smaller minimum is also observed at $\xi=115^\circ$, in contrast to that predicted at $\xi=105^\circ$. The experimental and theoretical results have been normalized at $\xi=35^\circ$ where the measurement is most accurately defined.

The theoretical cross section reproduced from [17] is shown together with the result of convolving this calculation with the estimated experimental angular profile. The depth of the predicted narrow minimum at $\xi=51^\circ$ is much closer to observation after this convolution is performed. The minimum at $\xi=105^\circ$ does not change significantly as it is broader, and the angle of this minimum also does not change appreciably.

A further prediction of [17] is that the cross sections for helium at $\psi=67.5^\circ$ and neon at $\psi=42^\circ$ are of similar magnitude, with a ratio of 1.75: 1 at $\xi=35^\circ$. This contrasts with experimental observation, where it was found that measurements from the 2s inner shell of neon were far more difficult than for helium. To estimate this difference, experiments were performed at ($\xi=35^\circ$, $\psi=67.5^\circ$) on helium and at ($\xi=35^\circ$, $\xi=42^\circ$) on neon using conditions set as closely equal as possible. Under these conditions the rate of coincidence counts from helium was found to be 16.2 ± 2.0 times higher

than from neon, a factor of 9.2 ± 1.1 times greater than predicted by theory.

Although the calculations of [17] predict the observed minima (apart from an overall scaling factor as noted above), the complexity of the calculation does not elucidate the nature of the underlying physical process that leads to such strong interference effects. These sophisticated calculations generate distorted waves for the incident and outgoing electrons, orthogonalize these waves by projecting onto the atomic basis set, include postcollisional interactions of the outgoing electrons using a modified Gamow factor, do not include polarization of the target atom, and yet still produce deep interference minima.

It is not clear why such deep and narrow minima are seen for such a complex reaction. The semiclassical model of Bowring *et al.* [19] assumes that in the region of the dip the dominant mechanism is a binary collision followed by scattering of one of the outgoing electrons in the field of the ion. This model also accounts for the requirement that the angles and energies of the outgoing electrons be equal. It is certainly not clear, however, why this particular mechanism should be so dominant.

To try to understand these interactions further, experiments are being set up at Manchester to study noncoplanar ionization reactions using excited calcium atoms prepared in a well-defined P state using laser radiation. Rasch *et al.* [17] predict an absence of sharp interference minima when ionizing a p electron due to incoherent contributions from participating substates. By contrast, in these experiments, laser preparation of the atoms produces a coherent initial state that can be manipulated by controlling the power, polarization, and direction of the laser radiation [20]. Deep and narrow minima in the ionization cross section, from these coherently prepared excited atoms, are then expected. The effect of the initial target state on these minima can then be ascertained by varying the laser parameters and, hence, the initial target substate coherences and amplitudes. A deeper understanding of the underlying physical processes producing these minima should then be obtained.

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