

## LETTER TO THE EDITOR

**Two-electron interference in the helium (e, 2e) differential cross section at 64.6 eV**N J Bowring<sup>†</sup>, F H Read and A J Murray

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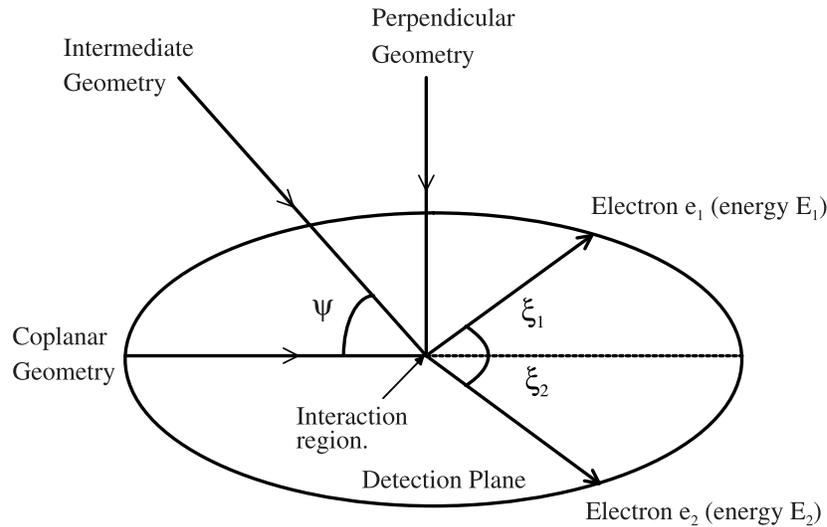
**Abstract.** The unexpected sharp dip which is known to exist in the helium (e, 2e) differential cross section at an incident electron energy of 64.6 eV, and for equal energies and angles of the two outgoing electrons, is known to be sensitive to three of the four available experimental parameters. We have now investigated the sensitivity of the dip to the fourth parameter, the asymmetry of the angles of the outgoing electrons, and have found that the dip exists over a wide range of asymmetric angles, but with an almost constant angle between the two electrons. A two-electron interference model is proposed to explain these results.

Understanding the mechanisms of atomic ionization by electron impact, particularly in the range of impact energies from a few eV above the ionization threshold energy to approximately 100 eV above, continues to present a considerable theoretical challenge (see, e.g., Botero and Macek 1992, Pan and Starace 1993, Jones and Madison 1994, Berakdar and Briggs 1994a, b, McCarthy and Weigold 1995, Bray *et al* 1997, Rasch *et al* 1997, Chen *et al* 1997). In this range, sometimes called the intermediate range of energies, the ionization mechanism involves all the complexities of exchange and capture, distortions in the incoming and outgoing channels, and both long- and short-range correlations, but the energy range is too far above the threshold to benefit from the simplifications offered by the Wannier model (Wannier 1953, Read 1985, Selles *et al* 1987). Angular correlation experiments, in which incident electrons ionize helium atoms and the resulting electrons are detected in coincidence provide particularly severe tests of the theoretical models.

Since the first experimental studies of this type (Ehrhardt *et al* 1969) attention has largely been confined to coplanar glancing collisions, but the testing and development of current theories (for a recent review, see McCarthy and Weigold (1995)) has been further challenged by including a wider range of scattering and ejected electron angles, as employed, for example, by Pochat *et al* (1983), Hawley-Jones *et al* (1992), Murray and Read (1992) and Röder *et al* (1996).

In the first thorough experimental exploration (Murray and Read 1993a, b) of electron impact ionization of helium in the intermediate energy region over a wide range of directions of the outgoing electrons and for several different incident energies, it was discovered that a deep minimum exists in the differential cross section (DCS) for a particular set of conditions. Figure 1 shows the type of coincidence geometry used in these studies and in the present study. For most of the experiments, including the present ones, the outgoing electrons are observed at the ‘doubly symmetric’ condition for which the scattering angles  $\xi_1$  and  $\xi_2$  of the two outgoing

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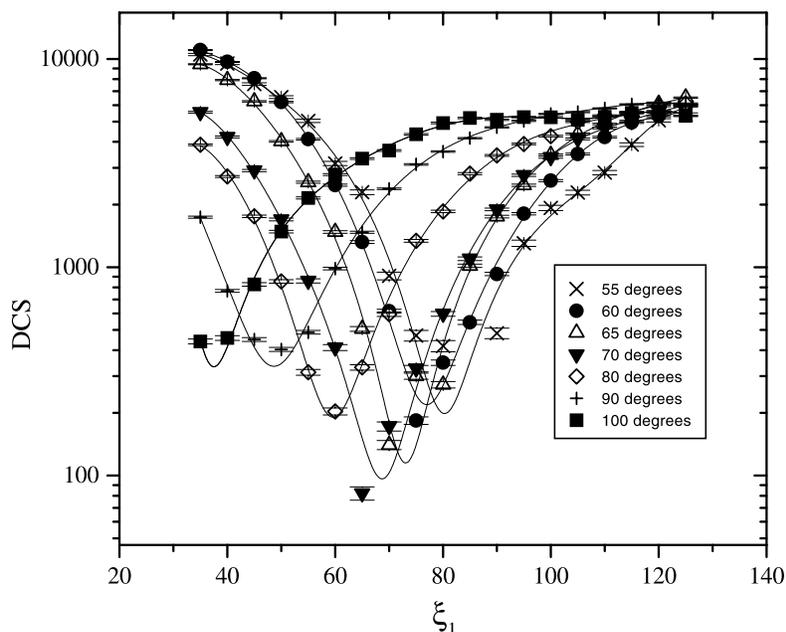
**Figure 1.** The (e, 2e) coincidence geometry used in the present experiments. The gun angle  $\psi$  is the angle between the direction of the incident electron beam and the detection plane. The scattering angles  $\xi_1$  and  $\xi_2$  in the detection plane are also shown.

electrons with respect to the incoming electron beam are the same (but with the electrons on opposite sides of the incoming direction) and the energies  $E_1$  and  $E_2$  of the electrons are also the same. At each incident electron energy  $E_i$  the gun angle  $\psi$  between the direction of the incident beam and the detection plane can be varied continuously from the coplanar geometry ( $\psi = 0^\circ$ ) through to the perpendicular plane ( $\psi = 90^\circ$ ) geometry.

The conditions found (Murray and Read 1993a, b) for the deep minimum are  $E_i = 64.6$  eV (i.e. 40 eV above the ionization threshold),  $E_1 = E_2$ ,  $\xi_1 = \xi_2 = 70^\circ$  and  $\psi = 67.5^\circ$ . The magnitude of the differential cross section at the minimum was measured to be approximately  $3 \times 10^{-5}$  au (before any allowance is made for the finite angular resolution of the detectors), which is 100 times smaller than the value at the local maximum at larger values of  $\xi$ . When the angular response function is deconvolved from the measured differential cross section it is found that the minimum is somewhere in the range from zero to approximately  $10^{-6}$  au.

Shallower minima have been seen in (e, 2e) differential cross sections for a variety of other conditions, and a sharp dip has been observed previously in the coplanar symmetric geometry ( $\psi = 0$ ,  $E_1 = E_2$ ) at an incident energy of 500 eV and a scattering angle  $\xi$  of approximately  $90^\circ$  (Rösel *et al* 1991). This dip, and that of the present study, are sufficiently sharp and deep to indicate the presence of some form of destructive interference involving only two major collision mechanisms or destructive interference within one major mechanism. This is surprising and intriguing in view of the complexity usually assumed for the (e, 2e) process, particularly at the low energy considered in the present study.

It was observed in the earlier studies (Murray and Read 1993a) that the magnitude of the minimum differential cross section is sensitive to changes in  $\psi$ , increasing to  $5 \times 10^{-4}$  au when  $\psi$  is decreased to  $45^\circ$ , but is less sensitive to changes in  $E_i$ , increasing to approximately  $5 \times 10^{-5}$  au when  $E_i$  is reduced to 54.6 eV. In a later study (Murray and Read 1993b) the sensitivity to the asymmetry in the partitioning of the excess energy between  $E_1$  and  $E_2$  was explored, and a strong sensitivity was observed, the magnitude of the minimum differential cross section increasing to  $3 \times 10^{-4}$  au when  $E_1$  and  $E_2$  were caused to be different by 10 eV.



**Figure 2.** The He (e, 2e) differential cross section as a function of  $\xi_1$  for a series of values of  $\xi_2$ , and for  $E_i = 64.6$  eV,  $E_1 = E_2 = 20$  eV and  $\psi = 67.5^\circ$ . The normalization is arbitrary but constant. The curves are least-squares fits to ninth-order polynomials.

In the present study, the sensitivity of the dip to the fourth experimental parameter, the asymmetry of the angles  $\xi_1$  and  $\xi_2$  of the outgoing electrons, is explored.

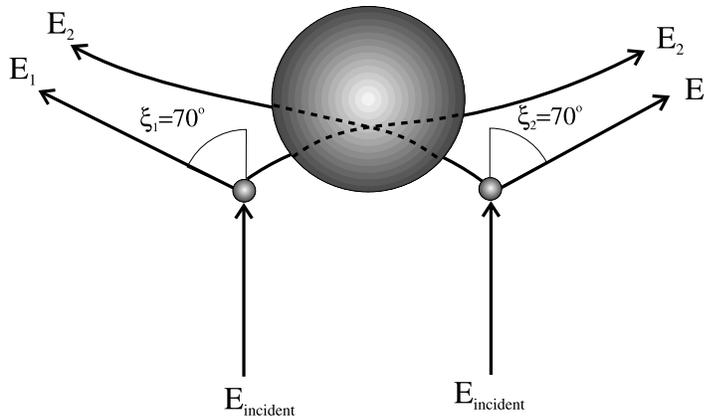
The spectrometer is fully computer controlled and computer optimized and is essentially that described previously (Murray *et al* 1992, Murray and Read 1993a). An unselected electron gun produces a beam of energy width 600 meV, which is focused onto the interaction region with a zero beam angle and a pencil angle of approximately  $2^\circ$ . The electrons scattered and ejected from the ionization process are collected by two hemispherical deflection analysers, which have energy resolutions of 500 meV and acceptance half angles of approximately  $3^\circ$ .

The experimental results are shown in figure 2. Three parameters are kept constant at the values previously found for the dip ( $E_i = 64.6$  eV,  $E_1 = E_2$ ,  $\psi = 67.5^\circ$ ), while  $\xi_1$  is scanned for a series of values of  $\xi_2$ . The data sets at each value of  $\xi_2$  are normalized relative to each other by using the fact that the differential cross section is unchanged by an exchange of the angles  $\xi_1$  and  $\xi_2$ . There are enough exchange pairs  $(\xi_1, \xi_2)$ ,  $(\xi_2, \xi_1)$  present in the data to enable this to be done for the data sets at all values of  $\xi_2$ . No attempt has been made to give an absolute normalization to the differential cross sections. Finally, the curves shown in figure 2 are empirical fits of a ninth-order polynomial function to each set of raw data, to find the positions of the minima for each value of  $\xi_2$ .

The scatter of the data points at the minima of the curves is caused by the extreme difficulty in measuring a cross section which is nearly zero. However, one trend which is clear is that, for each  $5^\circ$  increase in  $\xi_2$ , the value of  $\xi_1$  at the position of the minimum decreases by approximately  $5^\circ$ . In other words, the relative angle  $\xi_1 + \xi_2$  between the directions of the two electrons at the minimum remains approximately constant. It lies in the narrow range  $136$ – $140^\circ$  as  $\xi_2$  is varied from  $55$ – $100^\circ$ .

The fitted curves in the figure also indicate that the lowest differential cross section occurs when  $\xi_1$  and  $\xi_2$  are the same, at  $70^\circ$ . It therefore seems, from this and the earlier study (Murray and Read 1993b), that the differential cross section has its lowest value at the ‘doubly symmetric’ ridge  $E_1 = E_2$  and  $\xi_1 = \xi_2$ . Finally, we see from the figure that the dip broadens as  $\xi_1$  and  $\xi_2$  move away from the symmetric condition.

Two attempts to account for the dip theoretically are those of Berakdar and Briggs (1994a, b) and Rasch *et al* (1997). Both sets of calculations reproduce the essential properties of the dip for the doubly symmetric condition  $E_1 = E_2$  and  $\xi_1 = \xi_2$ . Berakdar and Briggs (1994a, b) considered the three amplitudes  $T_1$ ,  $T_2$  and  $T_3$  which represent the interactions between the incident electron and the three particles in the target atom, i.e. the target electron which is ionized, the nucleus and the spectator electron, respectively. The final state wavefunction used by Berakdar and Briggs takes account of all the post-collision interactions. They found that all three amplitudes must be included to reproduce the dip, and that a condition exists at which the real and imaginary parts of the summed amplitude become zero simultaneously, thus giving the dip. An interpretation or explanation of why the real and imaginary parts become zero simultaneously is not available at present. Berakdar and Briggs also explored the dependence of the parameters of the dip on the incident energy, again only for the doubly symmetric condition. Rasch *et al* (1997) used a distorted-wave Born approximation and found that distorted waves are needed in both the incident and final channels in order to reproduce the dip. They used a Gamow-type factor to allow partially for the post-collision interactions in the final channel. The dip appears as an interference effect between the incident and final channel distorted waves. Rasch *et al* considered only the scattering conditions that give the dip experimentally and, therefore, no information is available at present on how well this model will reproduce the differential cross section at other similar scattering conditions. More recently, Khajuria and Tripathi (1998) have looked at the dependence of the dip on the energy asymmetry, using a distorted-wave approximation. The sensitivity of the dip to the asymmetry in  $\xi_{1,2}$  has not yet been explored theoretically.



**Figure 3.** Schematic diagram showing two semiclassical routes for electrons observed in coincidence at  $E_i = 64.6$  eV,  $E_1 = E_2 = 20$  eV and  $\psi = 67.5^\circ$ . The plane of the diagram is the detection plane. An incident electron that is initially at  $67.5^\circ$  to this plane undergoes a binary collision in one of two localized volumes. One of the electrons resulting from the collision lies in the detection plane, while the other is deflected by the field of the ion (indicated by the larger sphere) to follow the broken path, which brings it into the detection plane. Destructive interference occurs when the difference in the path lengths between both routes is  $\lambda/2$ .

It was suggested by Murray *et al* (1996) that the existence of the sharp dip may be explained in terms of a destructive interference between two ‘routes’ within a single dominant scattering mechanism. This type of interference is illustrated in figure 3.

Of paramount importance for this explanation is the nature of the single dominant mechanism. Two mechanisms are effectively excluded for the present scattering angles and energies. Firstly, the simplest mechanism of a single-stage binary collision is improbable, because the present out-of-plane scattering angles can be produced only if the target electron has a large initial momentum. We calculate that the target electron would need to be moving in a direction almost perpendicular to the detector plane, with an initial kinetic energy of approximately 55 eV, which corresponds to a momentum of 2.0 au. Since the momentum profile of the bound electrons peaks at 0 au and falls by 50% at approximately 0.6 au (McCarthy and Weigold 1995) this process is unlikely to contribute significantly. A second mechanism which is improbable is the two-stage process in which the incident electron is scattered in the field of the ion core and then undergoes a binary collision with a target electron. This is markedly less probable than the two-stage process in which the incident electron has a binary collision with a target electron followed by the scattering of one of the outgoing electrons in the field of the ion, because the Rutherford scattering cross section (Landau and Lifshitz 1959) for an incident electron of energy 64.6 eV is an order of magnitude smaller than for an electron of energy 20 eV. It should also be noted that the symmetric conditions for  $E_{1,2}$  and  $\xi_{1,2}$  eliminate the triplet state contributions (Mazeau *et al* 1986).

The dominant mechanism for the present conditions therefore seems to be a binary collision followed by scattering of one of the outgoing electrons in the field of the ion. This would not necessarily be true for other scattering conditions, in particular for higher impact energies, near-threshold energies, ‘glancing’ collisions and in-plane scattering.

It is instructive at this stage to continue the discussion in semiclassical terms (whilst acknowledging that the electron wavelengths are not very much smaller than the relevant distances; for example,  $\lambda = 0.04$  nm for a 20 eV electron). We can then say that the undeflected outgoing electron emerges from the binary collision in approximately the direction of one of the detectors, while the other outgoing electron needs to be deflected by approximately  $75^\circ$  in order for it to be brought into the detection plane and to be received by the other detector. Post-collision interactions between the final particles will also clearly play a role.

In this model, the site of the initial binary collision has to be such that the undeflected electron can emerge with little further interaction, while the deflected electron needs to have a long path length in the field of the ion to give it the required deflection angle. The site is therefore localized to a small volume. This therefore implies that the range of initial angular momenta which are involved in the interaction is small, which, in quantum-mechanical terms, is consistent with a single dominant angular momentum.

As illustrated in figure 3 there are two possible sites, and so two ‘routes’, by which the two detectors can receive the two outgoing electrons. Quantum mechanical interference is therefore possible between the wavefronts which propagate to the two detectors. Note that this interference is not that of the conventional ‘two-slit’ type since, here, two particles emerge from each region (or ‘slit’). For destructive interference, the phase differences between the two routes should be  $\pi$ , which is equivalent to a path difference of  $\lambda/2$  if the kinetic energy is approximately constant. The required path difference is therefore 0.14 nm (2.6 au) for a 20 eV electron (ignoring the increase in kinetic energy due to the potential energy in the field of the ion). This implies that the mean distance of the trajectory of the deflected electron from the centre of the ion is approximately 0.1 nm (2 au) in order to give the required deflection of  $75^\circ$ . This is also consistent with the mean distance required to give a Rutherford deflection of  $75^\circ$ .

The semiclassical model is therefore internally consistent. It also provides a qualitative explanation of the sensitivities of the dip to the various experimental parameters. Firstly, the required ranges of incident energy and gun angle (Murray and Read 1993a) are clearly related to the need to exclude other contributions to the ionization process. Secondly, the strong sensitivity to an asymmetry in the energies of the two final electrons (Murray and Read 1993b) follows from the requirement that the two routes should have the same path lengths. Finally, the near constancy found in the present study for the relative angle  $\xi_1 + \xi_2$  follows from the fact that the relative path lengths change by only a small amount if all the outgoing trajectories are rotated in the same direction by the same small angle.

Refinements and improvements of this model are clearly possible—for example to allow a small change in direction of the incident electron before the binary collision and to take account of the post-collision interactions—but these are not warranted at this stage, since our intention is only to provide a qualitative suggestion for the collision mechanism that causes the sharp dip. This type of semiclassical explanation can, of course, only be a crude representation of the correct ionization process, but we believe that it embodies the essential physics of the process under the present scattering conditions.

The semiclassical model allows some predictions to be made about the conditions for which sharp dips might occur for other target atoms. Focusing attention on the heavier rare-gas atoms, their ground state ‘radii’ are larger than that of helium. For example, krypton has a ‘radius’ of 1.99 au (Cowan 1981) as opposed to 0.93 au for helium. The mean distance  $\bar{R}$  of the orbit of the deflected electron from the centre would therefore increase by a factor of approximately 2 for krypton, while the Rutherford deflection angle  $\theta_d$ , caused by the ion core, would decrease by approximately the same factor. The path length difference, which is approximately  $\bar{R}\theta_d$ , would, therefore, not be very sensitive to the radius of the atom, and so the energies  $E_1$  and  $E_2$  would continue to be about 20 eV. The smaller value of  $\theta_d$  would imply that the detection plane is less far removed from the planes of the initial binary collisions which, in turn, implies that the gun angle  $\psi$  would be smaller. The angles  $\xi_1$  and  $\xi_2$  would be less affected. The reduction in  $\psi$  would bring the scattering conditions nearer to those of a one-stage binary-collision mechanism, which might therefore contribute significantly, causing the dip to have the form of a less pronounced local reduction in the cross section. We would, however, still expect this dip to be strongly sensitive to any asymmetry in  $E_1$  and  $E_2$ , and for the relative angle  $\xi_1 + \xi_2$  still to be approximately constant. We intend to carry out experiments to test these predictions. We shall also look for the dips found by Rasch *et al* (1997) in their calculated (e, 2e) cross sections for Ne and Ar.

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