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Decomposition of experimentally determined atomic $(e,2e)$ ionization measurements

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A set of previously measured helium $(e,2e)$ coincidence ionization differential cross sections obtained over an exceptionally wide range of outgoing electron angles has been decomposed into tensorial angular components. A technique has been used which is familiar in nuclear physics for the analysis of cascade decay correlations but which does not appear to have been applied previously to atomic scattering or ionization processes. The amplitudes of the angular components, and their variation with energy, give information of a new type on the interplay between the incident and outgoing electrons.

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The derivation of angular momentum information from measured angular distributions of reaction products has a long and distinguished history, particularly in the general area of nuclear physics [1–4]. As far as we are aware, this type of analysis has not been attempted previously on the atomic $(e,2e)$ ionization process, although Klar and Fehr [5] have pointed out this possibility. The helium $(e,2e)$ experimental results, presented by Murray and Read [6] and obtained over an exceptionally wide range of angles, provide the first opportunity for such an analysis. The angular range has been achieved by measuring the differential scattering cross section over an unrestricted range of scattering geometries, from the coplanar to the perpendicular plane geometry.

In the experiments of Murray and Read [6], the incident electrons and target atoms are not polarized, the outgoing electron polarizations are not measured, and the target atom and final ion have zero angular momenta. The differential cross section for particular incident and outgoing energies can thus be decomposed into angular functions that depend only on the linear momentum vectors $\mathbf{k}_a$, $\mathbf{k}_b$, and $\mathbf{k}_0$ of the two outgoing electrons and the incident electron, respectively. These angular functions are necessarily scalars that do not depend on the choice of coordinate system. They are therefore [1–5] the scalar invariants

$$I_{l_a l_b l_0}(\hat{\mathbf{k}}_a,\hat{\mathbf{k}}_b,\hat{\mathbf{k}}_0) = \sum_{m_a m_b} \langle l_a m_a l_b m_b | l_0 m_0\rangle C_{l_a m_a}(\hat{\mathbf{k}}_a) C_{l_b m_b}(\hat{\mathbf{k}}_b) \times C^*_{l_0 m_0}(\hat{\mathbf{k}}_0) \ , \quad (1)$$

where $C_{lm}(\hat{\mathbf{k}}) = [4\pi/(2l+1)]^{1/2} Y_{lm}(\hat{\mathbf{k}})$, and where l_a, l_b, and l_0 are integers that label the members of the full set of functions $I_{l_a l_b l_0}$. Some examples of these scalar invariants (omitting normalizations for clarity) are

$$\begin{aligned} &I_{000} \sim \text{const}, \quad I_{101} \sim \hat{\mathbf{k}}_0\cdot\hat{\mathbf{k}}_a, \quad I_{110} \sim \hat{\mathbf{k}}_a\cdot\hat{\mathbf{k}}_b, \\ &I_{202} \sim 3(\hat{\mathbf{k}}_0\cdot\hat{\mathbf{k}}_a)^2-1, \quad I_{220} \sim 3(\hat{\mathbf{k}}_a\cdot\hat{\mathbf{k}}_b)^2-1, \\ &I_{211} \sim 3(\hat{\mathbf{k}}_0\cdot\hat{\mathbf{k}}_a)(\hat{\mathbf{k}}_a\cdot\hat{\mathbf{k}}_b)-(\hat{\mathbf{k}}_0\cdot\hat{\mathbf{k}}_b), \\ &I_{112} \sim 3(\hat{\mathbf{k}}_0\cdot\hat{\mathbf{k}}_a)(\hat{\mathbf{k}}_0\cdot\hat{\mathbf{k}}_b)-(\hat{\mathbf{k}}_a\cdot\hat{\mathbf{k}}_b). \end{aligned} \quad (2)$$

The decomposition of the $(e,2e)$ differential cross-section $\sigma(\mathbf{k}_a,\mathbf{k}_b,\mathbf{k}_0)$ is then given by

$$\sigma(\mathbf{k}_a,\mathbf{k}_b,\mathbf{k}_0) = \sum_{l_a l_b l_0} B_{l_a l_b l_0}(k_a,k_b,k_0) I_{l_a l_b l_0}(\hat{\mathbf{k}}_a,\hat{\mathbf{k}}_b,\hat{\mathbf{k}}_0), \quad (3)$$

where the amplitudes $B_{l_a l_b l_0}(k_a,k_b,k_0)$ depend on the magnitudes of the three momenta but not on their directions. These amplitudes provide in principle a complete parametrization of the measured cross section, allowing comparison with calculations and also allowing comparison between sets of experimental results obtained over different ranges and choices of scattering angles. They also provide a link [5] between coincidence and noncoincidence differential cross sections.

Inherent symmetries in the $(e,2e)$ process give rise to restrictions on the allowed combinations of l_a, l_b, and l_0. The first restriction arises from the parity of the angular functions, which follows from the parity of the spherical harmonics, and is given by

$$I_{l_a l_b l_0}(-\hat{\mathbf{k}}_a,-\hat{\mathbf{k}}_b,-\hat{\mathbf{k}}_0) = (-1)^{l_a+l_b+l_0} I_{l_a l_b l_0}(\hat{\mathbf{k}}_a,\hat{\mathbf{k}}_b,\hat{\mathbf{k}}_0). \quad (4)$$

Since the differential cross section is unaffected by an inversion of the coordinate system, odd values of $(l_a + l_b + l_0)$ must therefore be excluded from the summation in Eq. (3). A further, general restriction is that l_a, l_b, and l_0 must satisfy the triangulation condition.

Another restriction can arise from the exchange symmetry:

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$$I_{l_b l_a l_0}(\hat{\mathbf{k}}_b, \hat{\mathbf{k}}_a, \hat{\mathbf{k}}_0) = (-1)^{l_a+l_b+l_0} I_{l_a l_b l_0}(\hat{\mathbf{k}}_a, \hat{\mathbf{k}}_b, \hat{\mathbf{k}}_0). \quad (5)$$

The differential cross section is unaffected by the exchange of $\mathbf{k}_a$ and $\mathbf{k}_b$, which therefore implies that the amplitudes B have the exchange symmetry

$$B_{l_b l_a l_0}(k_b, k_a, k_0) = B_{l_a l_b l_0}(k_a, k_b, k_0). \quad (6)$$

In the experiments of Murray and Read [6] the two outgoing electrons have the same energy, giving

$$B_{l_b l_a l_0}(k_a, k_b, k_0) = B_{l_a l_b l_0}(k_a, k_b, k_0). \quad (7)$$

The summation in Eq. (3) can therefore be restricted to $l_b \geq l_a$ when analyzing experiments for which $E_a = E_b$. In summary, the three conditions considered so far are

$$l_a + l_b + l_0 = \text{even}, \quad \Delta(l_a, l_b, l_0) \neq 0, \quad l_b \geq l_a. \quad (8)$$

An additional restriction on the allowed combinations of l_a, l_b, and l_0 arises from the symmetry of the angular configuration used by Murray and Read [6]. Because of this, the angular functions for this experiment are functions of only two independent mutual angles ψ and ξ [6]. The full angular set $I_{l_a l_b l_0}(\hat{\mathbf{k}}_a, \hat{\mathbf{k}}_b, \hat{\mathbf{k}}_0)$ is a function of three independent mutual angles, which can be taken as θ_a, θ_b, and $(\phi_a - \phi_b)$, where $\mathbf{k}_0$ defines the z axis. The functions $I_{l_a l_b l_0}(\xi, \psi)$ appropriate to the present data are in this sense a subset of the complete functions $I_{l_a l_b l_0}(\theta_a, \theta_b, \phi_a - \phi_b)$. Although the functions of the complete set are linearly independent (see below), the functions of the subset are not, so that we can write

$$I_{l_a l_b l_0}(\xi, \psi) = \sum_{l'_a l'_b l'_0} c_{l'_a l'_b l'_0} I_{l'_a l'_b l'_0}(\xi, \psi). \quad (9)$$

The coefficients $c_{l'_a l'_b l'_0}$ are found to be nonzero if the following conditions are satisfied:

$$l'_0 = l_0, l_0 - 2, \ldots, |l_a - l_b| \quad \text{and} \quad l'_a + l'_b = l_a + l_b, l_a + l_b - 2, \ldots, l'_0. \quad (10)$$

For example,

$$I_{222} = \sqrt{2/7}\left\{\frac{3\sqrt{21}}{5} I_{132} + \frac{4}{5} I_{022} + \sqrt{5} I_{220} + \frac{9\sqrt{3}}{10} I_{110} - \frac{1}{10} I_{000}\right\}. \quad (11)$$

We have established these dependencies algebraically for values of l_a, l_b, and l_0 up to 9 and have confirmed them numerically at selected angles. The effect of the linear dependences for the present analysis is that some combinations of l_a, l_b, and l_0 become redundant, the ordering of the l_a, l_b, and l_0 determining the particular combinations that have to be omitted. We have chosen to scan l_0 from 0 to a maximum preset value, and at each value of l_0 have scanned all the possible combinations of l_a and l_b in order of increasing l_a and l_b up to their maximum preset values, using the conditions given by (8) above. The redundant combinations then become those for which the term $(l_a + l_b)$ has already appeared in the list, for the given value of l_0.

The reduction in the number of combinations of l_a, l_b, and l_0 due to the energy and angular symmetries is by approximately a factor of 2 in the present analysis and is very helpful because it allows a larger range of values to be explored when fitting to the experimental data.

For given incident and outgoing electron energies the amplitudes can be extracted by fitting Eq. (3) to the measured differential cross sections. Taking the z axis along the direction $\hat{\mathbf{k}}_0$, the angular functions have the form

$$I_{l_a l_b l_0}(\theta_a, \theta_b, \phi) = \sum_m (-1)^m \langle l_a m l_b - m | l_0 0\rangle \left[\frac{(l_a - |m|)!}{(l_a + |m|)!}\frac{(l_b - |m|)!}{(l_b + |m|)!}\right]^{1/2} P_{l_a}^{|m|}(\theta_a) P_{l_b}^{|m|}(\theta_b) e^{im\phi}, \quad (12)$$

where $\phi = \phi_a - \phi_b$. Note that this expression differs from that given in [1], by the factor $(-1)^{l_0+l_b}(2l_0+1)^{-1/2}$. In the present symmetrical configuration, $\theta_a = \theta_b$ and the angles θ_a and ϕ are related to the gun angles ψ and the detection plane angle ξ by [6]

$$\cos\theta_a = \cos\xi \, \cos\psi, \quad \cot\frac{\phi}{2} = \cot\xi \, \sin\psi. \quad (13)$$

In principle the full set of amplitudes can be obtained by making use of the orthogonality property of the angular functions,

$$\int I_{l_a l_b l_0}(\theta_a, \theta_b, \phi) I_{l'_a l'_b l'_0}(\theta_a, \theta_b, \phi) \sin\theta_a \sin\theta_b d\theta_a d\theta_b d\phi = \frac{8\pi}{(2l_a+1)(2l_b+1)} \delta_{l_a l'_a} \delta_{l_b l'_b} \delta_{l_0 l'_0}. \quad (14)$$

Note that this expression also differs by a factor from that given by [1]. The present experiment does not, however, cover the whole range of values of θ_a, θ_b, and ϕ. We have therefore used a least-squares fitting method to analyze the experimental results. The quantity that we have minimized is

$$\chi^2 = \sum_i w_i\{\sigma_{\rm calc}(\mathbf{k}_a, \mathbf{k}_b, \mathbf{k}_0) - \sigma_{\rm expt}(\mathbf{k}_a, \mathbf{k}_b, \mathbf{k}_0)\}^2, \quad (15)$$

where $\sigma_{\rm calc}$ is given by Eq. (3), $\sigma_{\rm expt}$ is the measured differential cross section, and the summation is over the experimental points i. The weighting factors w_i are based on the statistical weights of the experimental data points.

Three conditions are incorporated in the minimization procedure, all of which are concerned with the calculated cross sections at the end points $\xi = 0°$ and $180°$ for each value of the gun angle ψ, and in the gaps that are present in the data near these end points. The first condition is that $\sigma_{\rm calc}$ should be zero at the end points, because here the two outgoing electrons coincide in direction. The second condition is that $\sigma_{\rm calc}$ should not be allowed to become negative anywhere, which would be unphysical. The third is that the calculated cross section should have not more than one point of inflection in each gap, to ensure a monotonic behavior, without any peaks or shoulders. Details of the computer programs used for the analysis will be given in a future publication.

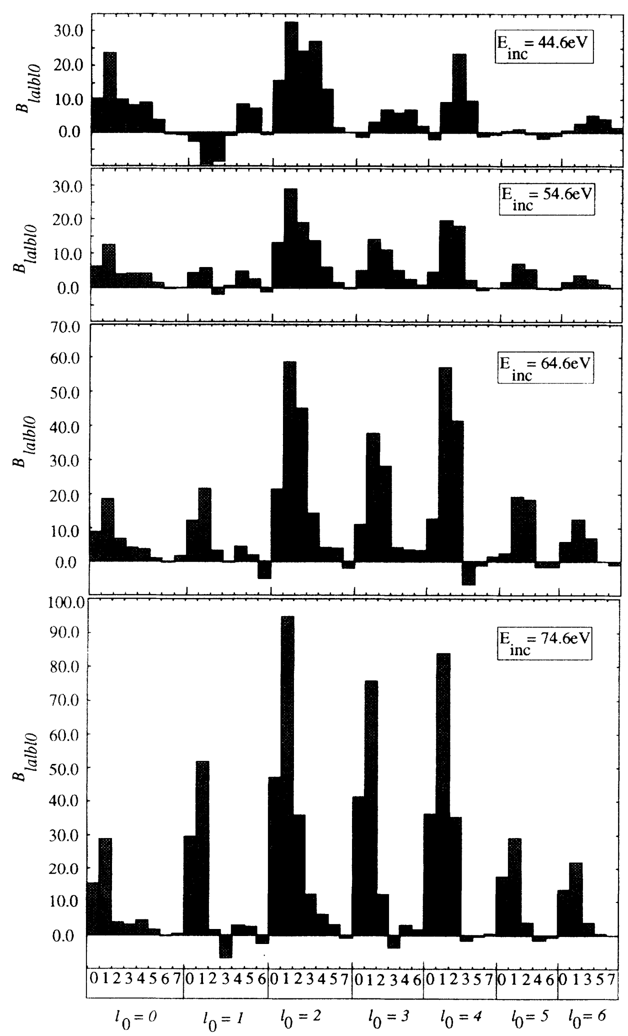

FIG. 1. The calculated fitting amplitudes $B_{l_a l_b l_0}(k_a, k_b, k_0)$ for the helium $(e, 2e)$ differential cross section symmetric in energy and scattering angle. The amplitudes are grouped into equal l_0 values, the associated value of l_a being shown immediately above these. The l_b component can be deduced from the condition $l_a + l_b = l_0, l_0 + 2, \ldots$.

The four incident energies used by Murray and Read [6] are 44.6, 54.6, 64.6, and 74.6 eV, with corresponding outgoing electron energies of 10, 15, 20, and 25 eV. Several different sets of values of $\max(l_a, l_b)$ and $\max(l_0)$ have been explored. The way in which the quality of the fits depends on these maximum values will be described in a future publication. We present here the results obtained by applying the set that has $\max(l_a, l_b) = 7$ and $\max(l_0) = 6$ to the data at the four incident energies.

The values of the amplitudes $B_{l_a l_b l_0}(k_a, k_b, k_0)$ obtained from these fits are shown in Fig. 1, grouped by the values of l_0. Within each group the values of l_a are indicated. The corresponding values of l_b can be deduced from conditions given above, and are such that within each l_0 group the sum $l_a + l_b$ takes the values $l_0, l_0 + 2, l_0 + 4, \ldots$.

At all four incident energies the largest amplitude occurs for $l_a = 1$, $l_b = 3$, and $l_0 = 2$. This amplitude incorporates that for $l_a = 2$, $l_b = 2$, and $l_0 = 2$, with which it is linearly related, as given by Eq. (11).

The higher values of l_0 feature more prominently at the higher values of $E_{\rm inc}$. A result of much greater significance is that the lowest values of $E_{\rm inc}$ give the highest values of l_a and l_b. For example, when $l_0 = 2$ the combination $(l_a, l_b) = (4, 6)$ contributes significantly at $E_{\rm inc} = 44.6$ eV, where the two outgoing electrons have the energy 10 eV, but less significantly at 74.6 eV, where the outgoing energy is 25 eV. The lowest of the values of $E_{\rm inc}$ studied here therefore gives rise to the highest complexity of combinations of $\mathbf{k}_a$, $\mathbf{k}_b$, and $\mathbf{k}_0$ [see Eq. (2)] in the tensorial angular functions I.

It is well known [7–10] that at incident energies within 2 eV of the ionization threshold the interaction between the outgoing electrons leads to substantial changes in their energies, directions, and angular momenta, but the present analysis gives a clear indication that interactions involving all three momenta have profound effects up to 20 eV or more above threshold.

Experiments are at present in progress in our laboratory to measure differential cross sections for which the two detected electrons have energies and also scattering angles that are different from each other. Analysis of these will provide a complete set of amplitudes $B_{l_a l_b l_0}$, which will in turn enable a comparison to be made for the first time with measured noncoincidence double-differential and total cross sections.

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[1] L. C. Biedenharn, *Nuclear Spectroscopy, Part B*, edited by F. Ajzenberg-Selove (Academic, New York, 1960), p. 732.

[2] J. M. Blatt and L. C. Biedenharn, Rev. Mod. Phys. **24**, 258 (1952); **25**, 729 (1953).

[3] D. M. Brink and G. R. Satchler, *Angular Momentum* (Clarendon, Oxford, 1979).

[4] L. C. Biedenharn and J. D. Louck, *Angular Momentum In Quantum Physics* (Addison-Wesley, Reading, MA, 1981).

[5] H. Klar and M. Fehr, Z. Phys. D **23**, 295 (1992).

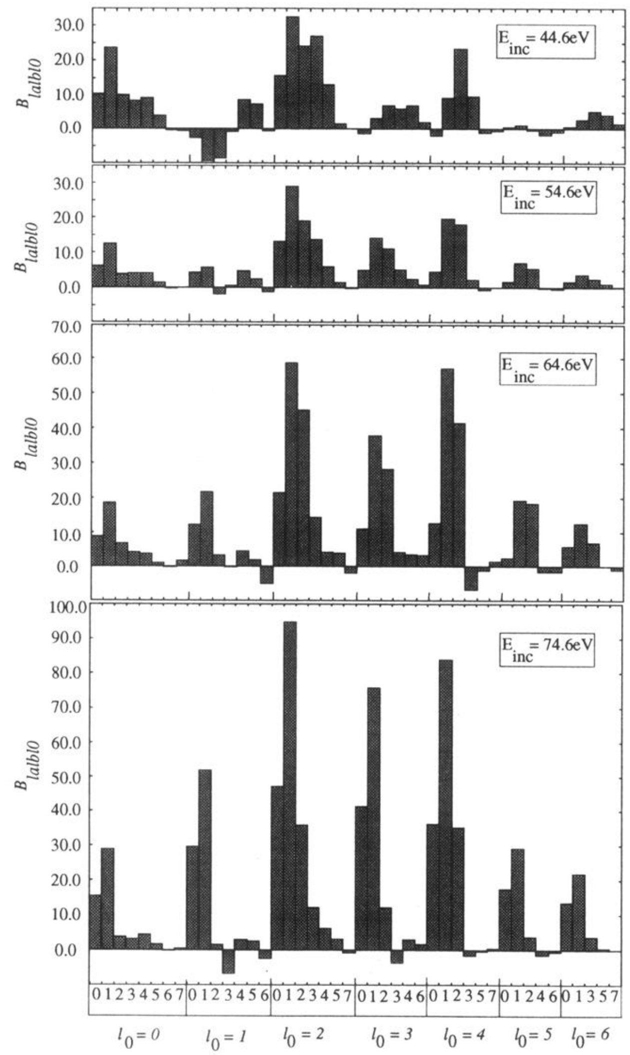
[6] A. J. Murray and F. H. Read, Phys. Rev. Lett. **69**, 2912 (1992).

[7] F. W. Byron and C. J. Joachain, Phys. Rep **179**, 221 (1989).

[8] G. H. Wannier, Phys. Rev. **90**, 817 (1953).

[9] F. H. Read, *Electron Impact Ionization*, edited by T. D. Märk and G. H. Dunn (Springer-Verlag, Vienna, 1985), p. 42.

[10] S. Jones, D. H. Madison, and M. K. Srivastava, J. Phys. B **25**, 1899 (1992).



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