Calculation of Circular Dichroism in Helium Double Photoionization

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We apply the convergent close-coupling (CCC) method to the calculation of the circular dichroism in helium double photoionization for a photon energy of 99 eV. Comparison is made with the measurements of Mergel *et al.* [Phys. Rev. Lett. **80**, 5301 (1998)], and substantial discrepancy is found. This is the first example where the CCC theory yields such disagreement with experiment for either $(\gamma, 2e)$ or $(e, 2e)$ angular differential cross sections. Nevertheless, we argue that there are good reasons to believe in the accuracy of the theoretical predictions. [S0031-9007(98)07687-X]

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In recent times there has been much progress in the field of double photoionization of helium, both experimentally and theoretically. On the experimental side, the ratio of total double to single photoionization has been determined to a high accuracy over a broad energy range [1–3], which is in good agreement with close-coupling-based theories [4–6]. The next challenge is to study the double photoionization angular distributions. Extensive experimental effort has already been expended in this direction. Using linearly polarized light Schwarzkopf *et al.* [7] first reported measurements of double photoionization of helium where the two outgoing electrons are detected in coincidence. Subsequently, the technique of cold target recoil ion momentum spectroscopy (COLTRIMS) was applied to also perform such fully differential measurements [8–10]. Following the prediction of Berakdar and Klar [11] that left- and right-hand circular polarized light should yield different angular distributions for double photoionization (unlike single photoionization) Viefhaus *et al.* [12] confirmed this experimentally. Most recently Mergel *et al.* [13] applied the COLTRIMS technique using circularly polarized light and presented the most detailed measurements of circular dichroism (CD). For a single photon energy of 99 eV, leading to an excess energy $E = 20$ eV, they presented both the absolute cross sections $\sigma_{\pm}$ and the resultant CD $= (\sigma_+ - \sigma_-)/(\sigma_+ + \sigma_-)$ where $\pm$ correspond to the positive or negative helicity.

On the theoretical side, calculations involve the evaluation of the dipole matrix element for an initial ground state of helium and a final state governing two-electron escape. Since this matrix element may be evaluated in the three gauges known as length (L), velocity (V), and acceleration (A) it is vital that the results do not depend on the choice of gauge. Unfortunately, it is not uncommon for calculations to differ substantially depending on the gauge [14]. This endangers any claim of a theory to yield predictive results. For example, the 3C theory presented by Mergel *et al.* [13] shows substantial shape variation and an order of magnitude difference between the L- and V-gauge generated cross sections. Thus, the discrepancy between the calculations and experiment could clearly be attributed solely to theory.

The fundamental strength of the 3C description of the final two-electron state is that it has correct boundary conditions for infinite separation of the electrons and the residual ion. It is analytical and is a product of two Coulomb waves and three Coulomb phases. Its weakness is that it ignores the interaction region, relying instead primarily on the strength of the Coulomb repulsion in the asymptotic regions. Nevertheless, it has been particularly successful in yielding qualitative agreement with experiment, with its simplicity allowing for a ready analysis of the underlying physics of ionization processes.

By contrast, the convergent close-coupling (CCC) method is a much more computationally intensive approach. For the final state it attempts to solve the Schrödinger equation for the system of interest relying on the close-coupling (CC) expansion of the total wave function. Recent applications of the method to problems involving helium with low-energy two-electron escape reproduced very detailed $(e, 2e)$ [15] and $(\gamma, 2e)$ [10,16] differential cross sections. Using a highly correlated Hylleraas-type ground state the CCC photoionization results are essentially gauge independent, yield correct integrated cross sections, and so have a hope of being predictive [6,16].

In this Letter we compare CCC results with the experiment and theory of Mergel *et al.* [13]. The fully resolved (energy integrated) cross section of atomic double photoionization is [16]

$$\sigma_M(\boldsymbol{k}_1, \boldsymbol{k}_2) = C \left| \sum_{l_1 l_2} (-i)^{l_1 + l_2} B_{1M}^{l_1 l_2}(\hat{k}_1, \hat{k}_2) \times e^{i[\delta_{l_1}(E_1) + \delta_{l_2}(E_2)]} D_{l_1 l_2}(E_1, E_2) \right|^2, \quad (1)$$

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where $E = E_1 + E_2$ is the total excess energy, B is a bipolar harmonic [17], and C is the double photoionization constant which depends on the normalization of the continuum wave functions and the gauge of the electromagnetic operator. The index M indicates polarization of light and is set to 0 for linearly polarized light and to ± 1 for circularly polarized light depending on the helicity. Hence, $\sigma_{M=\pm 1}$ is identified with $\sigma_{\pm}$. Note that for $M = 0$ the z axis is chosen along the polarization axis of the photon, whereas for $M = \pm 1$ it is directed along the momentum of the photon.

Expression (1) complies with the general formalism of circular dichroism in double photoionization developed previously by several authors, Berakdar *et al.* [18], for example. For the initial helium ^1S ground state, using the Clebsch-Gordan expansion for the product of two bipolar harmonics [17] we may write Eq. (1) as

$$\sigma_M(\boldsymbol{k}_1, \boldsymbol{k}_2) = C \sum_{J L_1 L_2} C^{J0}_{1M,1-M} \times B^{L_1 L_2}_{J0}(\hat{k}_1, \hat{k}_2) \gamma_{L_1 L_2}(E_1, E_2), \quad (2)$$

where γ is a complicated function of angular momentum coefficients, phases, and the D matrix elements, but has no M dependence. The only dependence on M comes simply from the Clebsch-Gordan coefficient. The summations over $J = 0, 1, 2$, L_1, and L_2 are independent of M. In the case of the CD term $\sigma_+ - \sigma_-$ only the $J = 1$ and $L_1 = L_2$ terms survive, leading to the CD expression identical to Eq. (15) of Berakdar *et al.* [18]. We note that, in contrast to the fully resolved cross section (2), the CD is a much less sensitive parameter requiring only diagonal γ_{LL} terms in the numerator.

The quantity $D_{l_1 l_2}(E_1, E_2)$ in Eq. (1) is the reduced dipole matrix element which is stripped of its angular dependence. To calculate this matrix element we employ the CCC method by expanding the final two-electron continuum state using N square-integrable (L^2) states, with the double ionization processes being identified with excitation of the positive-energy pseudostates. These states, $\phi^{N_l}_{nl}$ with energy $\epsilon^{N_l}_{nl}$, are obtained by diagonalizing the He^+ Hamiltonian in a Laguerre basis of size N_l. For each target-space orbital angular momentum $l \leq l_{\max}$ the Laguerre exponential falloff parameter is varied until one of the energies is E_2. Upon completion of the N-state ($N = \sum_l N_l$) CCC calculation we obtain matrix elements $D^N_{l_1 l_2}(E_1, n_2)$, which correspond to photoionization with a true continuum electron of energy E_1 and orbital angular momentum l_1, together with excitation of the ionic electron to state denoted by $n_2 l_2$ with energy E_2.

We write the N-state approximation

$$e^{i[\delta_{l_1} + \delta_{l_2}]} D^N_{l_1 l_2}(E_1, E_2) = e^{i[\delta_{l_1}(Z=1) + \delta_{l_2}(Z=2)]} \times D^N_{l_1 l_2}(E_1, n_2) \langle l_2 E_2 \parallel n_2 l_2 \rangle, \quad (3)$$

where the phases are for the given Coulombic charges Z, and the overlap is between a $Z = 2$ Coulomb radial orbital of angular momentum l_2 and energy E_2 and the $n_2 l_2$ pseudostate of the same energy. The formalism relies on the completeness of the Laguerre basis to ensure the full solution of the problem in the limit of infinite N. In practice, we increase N until convergence in the angular distributions is obtained. In the present case this is found by taking $l_{\max} = 4$ with $N_l = 17 - l$ making a total of 75 states.

The close-coupling boundary conditions have the true continuum electron always being shielded by the L^2 one, irrespective of their energies. This is physical when the energy of the L^2 electron is substantially smaller than that of the continuum one, but not when the energies are the other way around. This leads to a highly asymmetric energy distribution within the ionization channels, the so-called singly differential cross section (SDCS). In fact, Bray [19] argued that the SDCS should converge to a step function (nonzero for $0 \leq E_2 \leq E/2$ and zero for $E/2 < E_2 \leq E$) in the limit of infinite N. At sufficiently low energies, where the SDCS at $E/2$ is substantial, the finite calculations yield small, but nonzero cross sections for $E_2 > E/2$ and have unphysical oscillations for $0 \leq E_2 \leq E/2$, though the integral yields a stable (independent of N) total (double) ionization cross section. Furthermore, nonzero amplitudes for $E_2 > E/2$ imply two independent estimates of the direct and exchange pairs corresponding to the same ionization process. This requires an incoherent combination of the two pairs of coherently combined direct and exchange amplitudes. The latter is particularly substantial for the equal energy-sharing kinematical region and is necessary to yield excellent angular distributions [15,20], though the absolute values are affected by the unphysical oscillations in the SDCS. This may be remedied if the true SDCS happens to be known [19]. For example, in the near threshold region the true SDCS is approximately flat and so may be obtained directly from the stable integrated SDCS. Subsequently, the CCC results may be rescaled to ensure that the flat SDCS is obtained after integration over all the angular variables of the fully differential cross sections. This semiempirical procedure does not affect the angular distributions or their relative magnitudes. If the CCC theory yields correctly all angular distributions with correct relative magnitudes then the rescaling by the single constant (a function of E_2) ensures correct absolute magnitudes also.

For asymmetric energy sharing, of interest in the present case, the close-coupling formalism seems ideal. The fast $Z = 1$ Coulomb outgoing wave is shielded from the $Z = 2$ ion by the slow $Z = 2$ Coulomb wave. In the case of $(e, 2e)$ equivalent formulation works very well [21,22]. There are no conceptual problems due to an incoherent summation of amplitudes as the second pair of direct and exchange amplitudes, corresponding to excitation of the state with energy E_1, is usually too small.

In Fig. 1 we present the CCC calculations of double photoionization of helium at $E = 20$ eV excess energy

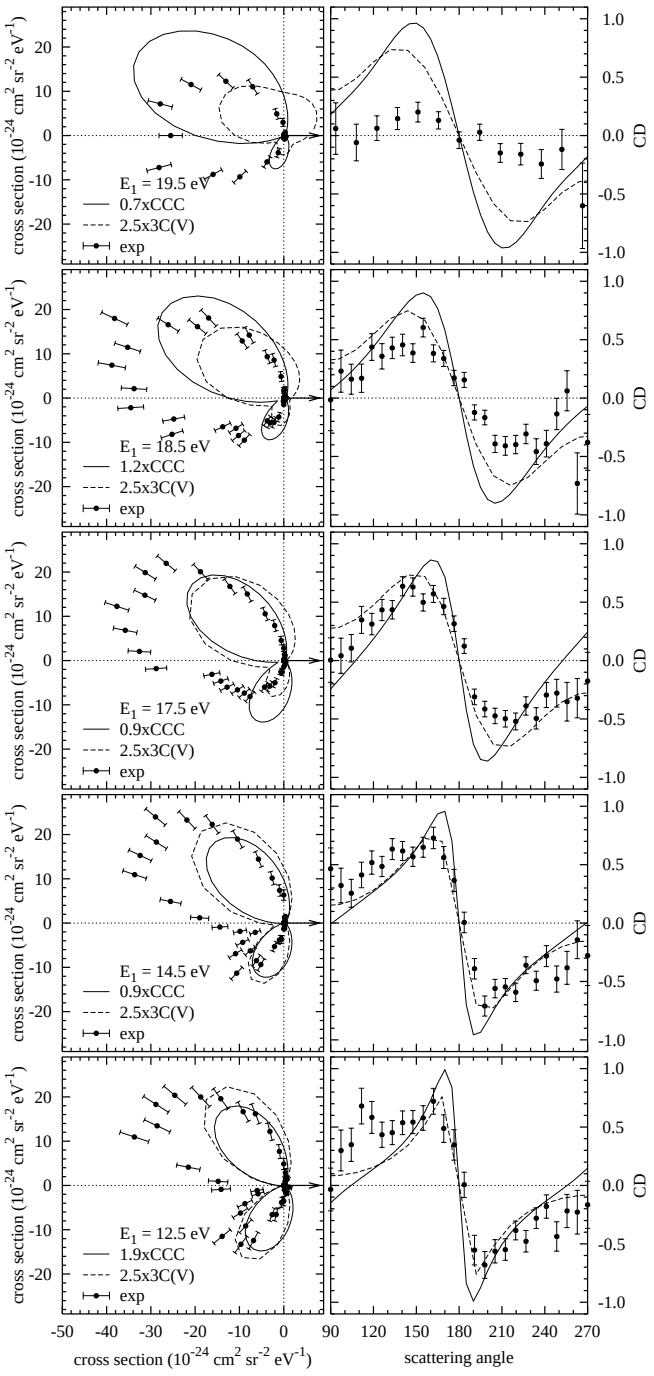

FIG. 1. Fivefold differential cross sections σ_+ and CD $= (\sigma_+ - \sigma_-)/(\sigma_+ + \sigma_-)$ at excess energy $E = 20$ eV, with electron of energy E_1 detected at $\theta_1 = 0°$. Note that σ_- are a mirror image of σ_+ about the horizontal axis. See text for the description of the CCC theory. Experiment and the 3C theory are from Ref. [13].

using spherically polarized light, and compare with the experiment and the velocity-gauge 3C theory of Mergel *et al.* [13]. Though the 3C(V) and 3C(L) results vary in shape and an order of magnitude in absolute value [13], the CCC theory would be barely distinguishable in the three gauges, and so only the L form is plotted. It yields correct values for single photoionization with excitation and total double ionization cross section (9.3×10^{-21} cm^2) [6]. As argued in Ref. [10], the experimental work of Wehlitz *et al.* [23] and the theoretical work of Pont and Shakeshaft [24] suggest that the SDCS at this energy may be assumed to be flat, and so we obtain the value of 9.3×10^{-22} cm^2/eV. This determines the CCC scaling factors given in Fig. 1. These are near unity for all but the least asymmetric energy-sharing case. The single scaling factor of 2.5 for the 3C(V) theory was taken for best visual fit to the CCC theory.

We see discrepancy between the CCC theory and experiment for both the magnitude and angular distribution of the σ_+ cross section. Though the two theories disagree in angular distributions for the more asymmetric energy-sharing cases, agreement improves dramatically as symmetric energy sharing is approached. The agreement for the experimental CD is much improved, as expected from the analysis of (2). However, it is overshadowed by the discrepancy in the absolute cross sections.

The angular agreement of the two theories is the first minor indication that the CCC theory may be correct. A much more major indication of this is obtained by comparison with the linear polarization data of Bräuning *et al.* [10], which is not only taken at the same total energy, but gives up to four different fixed electron angles θ_1 for a number of fixed electron energies E_1. In Fig. 2 we present the case of $E_1 = 3$ and 17 eV. All of the presented eight CCC(3.0) curves, calculated as σ_0 from (2), arise using the nine matrix elements obtained from the 75-state CCC calculation where $E_2 = 3$ eV for the $n_2 = 9 - l_2$ and $l_2 \leq 4$ pseudostate. This provides for a much more thorough test of the theory at a given E_1 than given in Fig. 1. The agreement between experiment and the CCC theory is excellent. Though the E_1 given by Bräuning *et al.* differ from the E_1 of Mergel *et al.* by 0.5 eV, the CCC(2.5) curves arising from the nine matrix elements used for $E_1 = 12.5$ eV in Fig. 1 also yield excellent agreement with the experiment of Bräuning *et al.* and the CCC(3.0) calculations. This suggests that at least the CCC $E_1 = 12.5$ eV results given in Fig. 1 are also correct.

To summarize, the following are the reasons why we believe that the CCC $\sigma_{M=\pm1}$ results presented in Fig. 1 are correct. The CCC results are much the same in all three gauges of the dipole operator. The theory yields correct absolute integrated cross sections. The close-coupling approximation is ideal for the asymmetric energy sharing of the outgoing electrons. Consistency is found with the 3C calculation as equal energy sharing is approached. Absolute agreement with the closely related and much more detailed experiment of $\sigma_{M=0}$ is obtained. Equation (2) shows simple dependence on M, while the CCC calculations yield M-independent quantities γ.

Should the present theoretical results prove to be incorrect, for other than implementation reasons, then

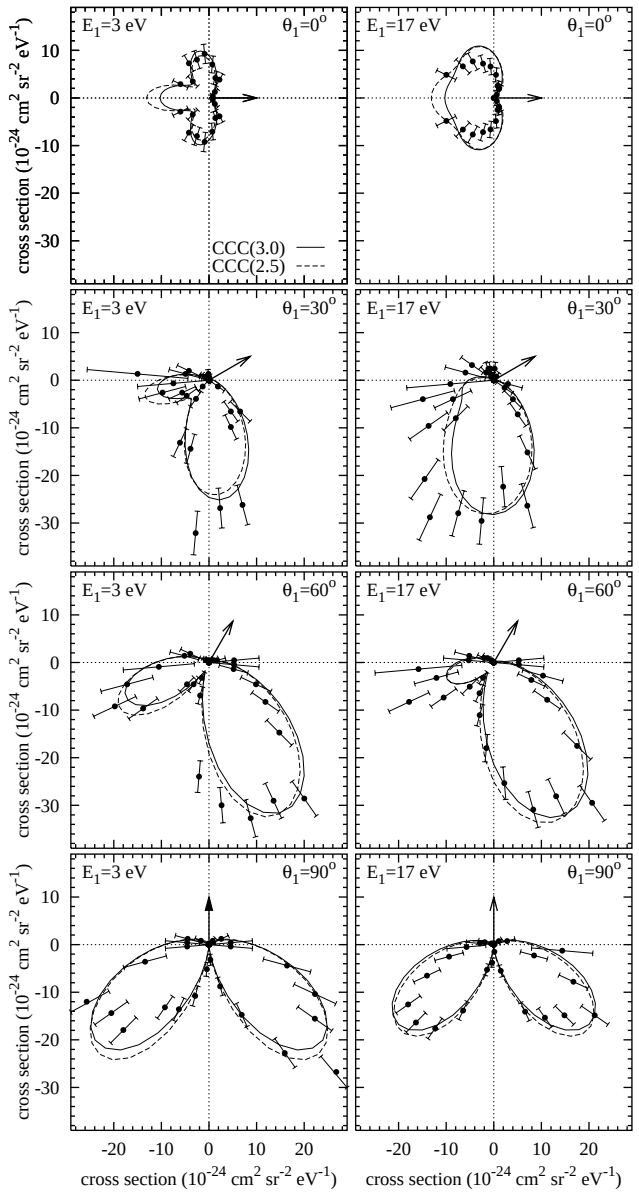

FIG. 2. Fivefold differential cross sections for double photoionization of helium using linearly polarized light at excess energy of 20 eV with an electron of energy E_1 being detected at θ_1. The CCC(3.0) lines and the experiment are from Ref. [10], with the "3.0" indicating that the curves were generated from a CCC calculation which had a 3 eV pseudostate for each l. The CCC(2.5) curves are generated from the same matrix elements used to generate the CCC result for the $E_1 = 17.5$ eV part of Fig. 1, where the CCC calculation had 2.5 eV pseudostates.

the CCC theory is invalidated as a predictive theory of scattering processes leading to two-electron escape. This would not be a problem of convergence, but rather a fundamental flaw in the CCC formalism.

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