Double ionization of helium by highly-charged-ion impact analyzed within the frozen-correlation approximation

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We apply the frozen-correlation approximation (FCA) to analyze double ionization of helium by energetic highly charged ions. In this model the double ionization amplitude is represented in terms of single ionization amplitudes, which we evaluate within the continuum distorted wave-eikonal initial state (CDW-EIS) approach. Correlation effects are incorporated in the initial and final states, but are neglected during the time the collision process takes place. We implement the FCA using the Monte Carlo event generator technique, which allows us to generate theoretical event files and to compare theory and experiment using the same analysis tools. The comparison with previous theoretical results and with experimental data demonstrates, on the one hand, the validity of our earlier simple models to account for higher-order mechanisms, and, on the other hand, the robustness of the FCA.

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Although double ionization (DI) of atoms by charged particle impact has been studied extensively (for recent reviews see, e.g. [1,2]), our understanding of the underlying collision dynamics is still surprisingly incomplete. In an attempt to systematically analyze the collision dynamics it is common to discuss DI in terms of various reaction mechanisms, which are viewed as characteristic sequences of interactions within pairs of particles [3]. In one such mechanism, called the two-step-one (TS-1) projectile-electron interaction, electron-electron correlation plays an essential role in ejecting two target electrons to the continuum: here, the sequence consists of a projectile-electron interaction and an interaction between both electrons. A second mechanism, called two-step-two (TS-2) projectile-electron interaction, does not necessarily involve electron-electron correlation. Rather, the sequence consists of two independent interactions of the projectile with each electron. Both mechanisms have been investigated in a number of recent works (e.g. [4–7]), but on a quantitative level the question concerning their relative importance and thus of the role of correlation in DI remains, to a large extent, open.

A few years ago a novel analysis technique that proved to be very powerful in identifying the characteristic features of the various DI mechanisms was introduced. It is based on four-particle Dalitz (4-D) plots and was first applied to analyze kinematically complete data on electron ejection from both collision partners [8]. In a 4-D plot the data are shown in a tetrahedral coordinate system where each tetrahedron plane represents one of the collision fragments. The distance of a given data point to the four planes provides a set of the squares of the momentum changes of the four collision fragments normalized to the sum of the squares of all particles $\pi_i = p_i^2/\Sigma p_j^2$. This method offers two important advantages over conventional spectra analysis: First, since in a 4-D plot cross sections are shown in dependence of all four particles, it provides a comprehensive picture of the collision dynamics. Second, 4-D plots are proportional to multiple differential cross sections and yet the integral spectrum corresponds to the total cross section, i.e., a high level of detail is combined with comprehensiveness. In a series of joint experimental and theoretical studies on DI of helium by ion impact [5,9,10], we demonstrated that the various DI mechanisms lead to characteristic features in 4-D plots. Furthermore, a new DI channel, called TS-1-EL, which can be viewed as a hybrid between TS-1 and TS-2, could be identified in 4-D plots for fast proton impact [9].

For large perturbation parameter η (projectile charge to speed ratio, often called the Sommerfeld parameter), TS-2 is the dominant DI channel, but a theoretical treatment of this process is challenging because of its higher-order nature. Nevertheless, experimental 4-D plots for 158 MeV Au^{33+}+ He ($\eta = 5.8$) were amazingly well reproduced by a relatively simple simulation of the TS-2 mechanisms [10]. There, the DI cross sections were calculated in terms of a convolution of cross sections for single ionization of He with those for single ionization of He^+. However, from a formal point of view this approach could be criticized for a lack of rigor. For example, in a proper treatment single-ionization amplitudes, rather than cross sections, should be convoluted. Furthermore, for collision systems with intermediate η the TS-2 and TS-1-EL amplitudes may be of similar magnitude. In this case, the amplitudes for all channels have to be added coherently so that an approach which only calculates cross sections for TS-2 is questionable.

In this Brief Report we present a theoretical study of DI based on the frozen-correlation approximation (FCA) [11]. As is explained further below, this approach can be viewed as a more rigorous method to account for the TS-2 process than the simulation mentioned above. Earlier, it was applied to calculate total DI cross sections for proton and antiproton impact (for a discussion and complete list of references see [12]) and was used to examine the correlation function [13] and the two-electron cusp [14] in DI of helium by energetic ion impact. Here, we report theoretical 4-D plots calculated within the FCA model.

We start from the FCA transition amplitude for DI at a given impact-parameter vector **b** [13],

$$a^{2e}_{i\to\mathbf{k}_1,\mathbf{k}_2}(\mathbf{b}) = \frac{\varphi(k_{12})}{\sqrt{2}} \sum_{j_1 j_2} C_{j_1 j_2}\big[a^{1e}_{j_1\to\mathbf{k}_1}(\mathbf{b})a^{1e}_{j_2\to\mathbf{k}_2}(\mathbf{b}) + a^{1e}_{j_1\to\mathbf{k}_2}(\mathbf{b})a^{1e}_{j_2\to\mathbf{k}_1}(\mathbf{b})\big], \quad (1)$$

where each $a^{1e}_{j_i\to\mathbf{k}_i}$ denotes a single-ionization amplitude obtained from an effective one-electron calculation. In the present work the $a^{1e}_{j_i\to\mathbf{k}_i}$ are calculated in the continuum distorted wave-eikonal initial state (CDW-EIS) approximation [15]. The coefficients $C_{j_1 j_2}$ arise due to a configuration interaction (CI) ansatz for the initial ground state of the helium atom, and the factor $\varphi(k_{12})$ due to a simplified treatment of final-state correlations in the two-electron continuum. As in previous works we use the CI wave function of Silverman *et al.* [16] and account for electron-electron correlation effects in the continuum retroactively (see, e.g. [5] for details), i.e., we set $\varphi(k_{12}) = 1$.

If we would neglect initial-state correlation, Eq. (1) would reduce to a (symmetrized) product of one-electron amplitudes, each of which describes the ionization of one electron due to interactions with the projectile. This would correspond to an uncorrelated TS-2 amplitude. With initial- and final-state correlations included, the FCA amplitude becomes more intricate, but maintains a TS-2 like character.

The first step in obtaining theoretical 4-D plots is to calculate fully differential cross sections (FDCS's) [17]. FCDS's are proportional to the square of the Fourier transform (FT) of an impact-parameter-dependent amplitude $a_{ik}(\mathbf{b})$ [18], i.e.,

$$R_{ik}(\mathbf{q}_T) = \int d\mathbf{b}\, \exp(i\mathbf{b}\cdot\mathbf{q}_T)a_{ik}(\mathbf{b}), \quad (2)$$

where $\mathbf{q}_T$ is the transverse momentum transfer ($\mathbf{q}_T \cdot \mathbf{v}_P = 0$, $\mathbf{v}_P$ being the velocity of the incoming projectile). For the FCA approach we need to calculate the FT of Eq. (1), i.e.,

$$R^{2e}_{i\to\mathbf{k}_1,\mathbf{k}_2}(\mathbf{q}_T) = \int d\mathbf{b}\, \exp(i\mathbf{b}\cdot\mathbf{q}_T)a^{2e}_{i\to\mathbf{k}_1,\mathbf{k}_2}(\mathbf{b}) = \frac{1}{\sqrt{2}}\sum_{j_1 j_2} C_{j_1 j_2} \times \left[\int d\mathbf{b}\, \exp(i\mathbf{b}\cdot\mathbf{q}_T)a^{1e}_{j_1\to\mathbf{k}_1}(\mathbf{b})a^{1e}_{j_2\to\mathbf{k}_2}(\mathbf{b}) + \int d\mathbf{b}\, \exp(i\mathbf{b}\cdot\mathbf{q}_T)a^{1e}_{j_1\to\mathbf{k}_2}(\mathbf{b})a^{1e}_{j_2\to\mathbf{k}_1}(\mathbf{b})\right]. \quad (3)$$

Each term in Eq. (3) is the FT of a product of functions in a two-dimensional space, and consequently we can apply the convolution theorem: $\mathcal{F}[f\cdot g] = \mathcal{F}[f] * \mathcal{F}[g]$, where $\mathcal{F}$ is the FT and $*$ denotes the convolution of two functions defined by $f(t) * g(t) = \int_{-\infty}^{\infty} f(\tau)g(t-\tau)d\tau$ for one-dimensional (well-behaved) functions. The convolution can be extended to multidimensional functions, i.e., $f(\mathbf{r}) * g(\mathbf{r}) = \int_{-\infty}^{\infty} f(\mathbf{r}')g(\mathbf{r}-\mathbf{r}')d\mathbf{r}'$ and consequently we can write Eq. (3) as

$$R^{2e}_{i\to\mathbf{k}_1,\mathbf{k}_2}(\mathbf{q}_T) = \frac{1}{\sqrt{2}}\sum_{j_1 j_2} C_{j_1 j_2}\big[R^{1e}_{j_1\to\mathbf{k}_1}(\mathbf{q}_T) * R^{1e}_{j_2\to\mathbf{k}_2}(\mathbf{q}_T) + R^{1e}_{j_1\to\mathbf{k}_2}(\mathbf{q}_T) * R^{1e}_{j_2\to\mathbf{k}_1}(\mathbf{q}_T)\big]. \quad (4)$$

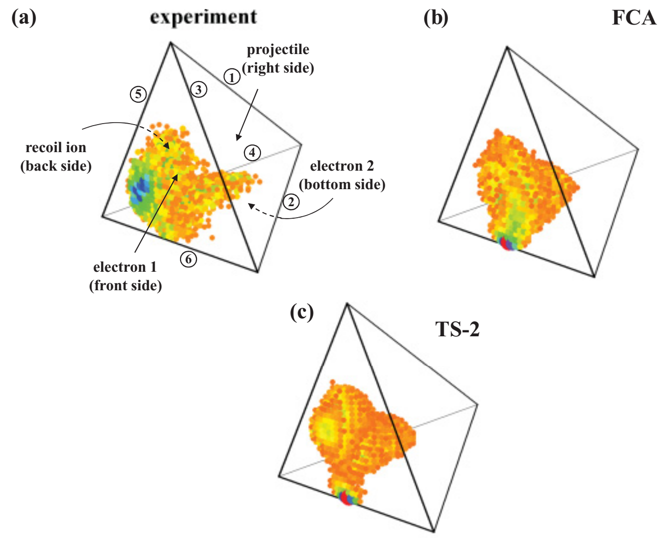

FIG. 1. (Color online) 4-D plots for three-dimensional momenta for double ionization of helium bombarded by 158 MeV Au^{33+} projectiles. (a) Experimental data [10], (b) FCA theory, and (c) TS-2 simulation [10] (see text for details). We have included numbers in the lines intersecting the tetrahedral planes and indicative labels (for further details see [10]).

Based on the calculated FDCS a theoretical event file is generated using the Monte Carlo event generator (MCEG) technique described by Dürr *et al.* [19]. The event file contains the momentum components of all collision fragments for a large number (typically about a million) of DI events such that the occurrence frequency of specific configurations of momentum components reflects the calculated FDCS. These events are then sorted into 4-D spectra in exactly the same manner as in the experiment. The computation of Eq. (4) involves in each FDCS calculation the evaluation of a two-dimensional integration that we have performed using Gaussian quadratures. This additional step increases the computational time substantially, but using the parallel programming technique presented in [17] it is possible to generate the event files needed in a reasonable time.

In Fig. 1 an experimental 4-D plot for DI of helium by 158 MeV Au^{33+} impact is shown in comparison with the TS-2 simulation of Ref. [10] and with our present FCA calculation. The front and bottom planes of the tetrahedron represent the ejected electrons, the back plane the target nucleus, and the right plane the projectile. A detailed interpretation of the experimental 4-D plots was provided by Fischer *et al.* [10] and will not be repeated here. Rather, we focus on a comparison of the new FCA calculation with the experimental data and the TS-2 simulation. First we note that the TS-2 and FCA results are very similar. Given that both models are designed to describe the same underlying DI process, but the computer codes for both models were developed independently, this gives us confidence in the numerical implementation. Furthermore, this similarity suggests that the convolution of the cross sections for single ionization of He and He^{+} represents a reasonably accurate simulation of TS-2. On the other hand, the agreement of both models with experiment is reasonable, but not very good. Interestingly, the

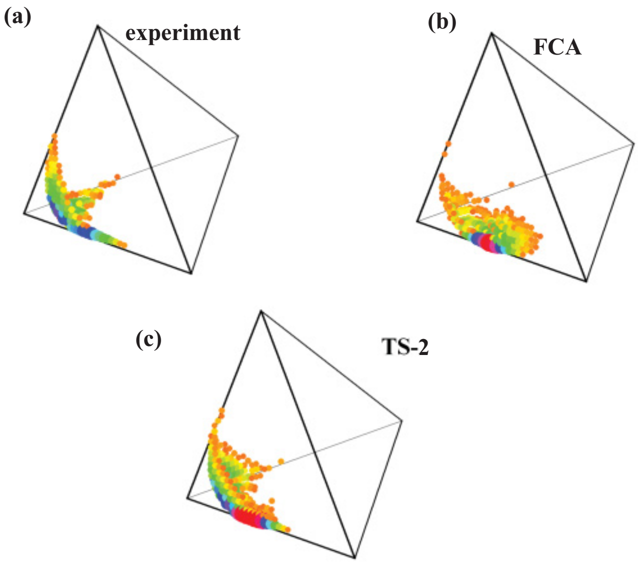

FIG. 2. (Color online) Same as Fig. 1, but the 4-D plots are now generated using only the momentum components in the transverse direction.

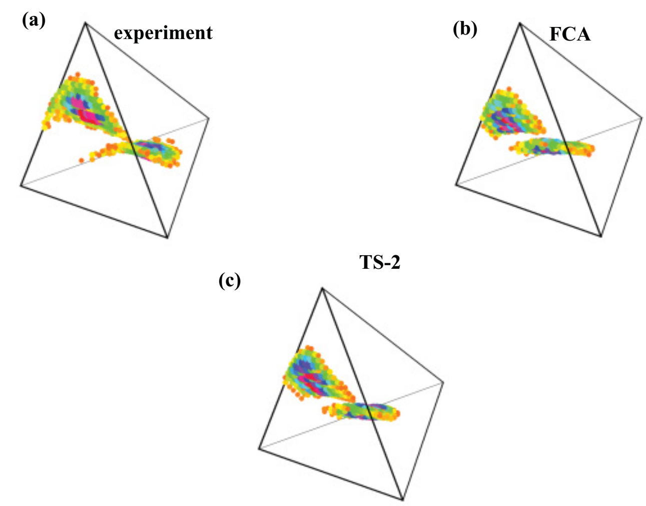

FIG. 3. (Color online) Same as Fig. 1, but the 4-D plots are now generated using only the momentum components in the longitudinal direction.

conceptually simpler TS-2 simulation fares slightly better in that it rudimentarily reproduces (at a slightly higher location) the most intense contribution in the experimental data near the lower left corner of the tetrahedron. Both models severely overestimate the momentum exchange between the heavy particles, which leads to a pronounced peak structure at the intersection lines between the two electron planes (line 6).

4-D plots can also be generated for specific components of the momentum change vectors of the collision fragments. In Fig. 2 such plots are shown for the components in the direction of the transverse (i.e., perpendicular to the projectile beam axis) projectile momentum change. Once again, the plots for the TS-2 simulation and the FCA calculation are quite similar. Here too the momentum exchange between the heavy particles is strongly overestimated by both models. However, otherwise the experimental data are nicely reproduced at least by the TS-2 simulation, which once again achieves somewhat better agreement than the FCA calculation.

In Fig. 3, 4-D plots are shown for the longitudinal (i.e., parallel to the projectile beam axis) momentum change components. In this direction the interaction between the heavy particles does not lead to any momentum exchange because there is no inelasticity involved so that the collision is symmetric with respect to the distance of closest approach. Therefore, for small scattering angles, which are always realized for fast heavy-ion impact, the longitudinal force components in the incoming and outgoing parts of the collision cancel each other almost exactly in the integral over time. As a result, the feature which causes the largest discrepancies between theory and experiment in the transverse direction is not present in the longitudinal direction. Here, the TS-2 simulation and the FCA calculation yield nearly identical results. Furthermore, rather good agreement with the experimental data is obtained.

In summary, we have presented 4-D plots for double ionization of helium by highly-charged-ion impact calculated within the frozen-correlation approximation. In this model electron-electron correlations are contained in the initial and final states but are neglected in the collision dynamics, thereby accounting only for TS-2 like contributions to DI. The FCA calculations closely resemble the results of a simulation of TS-2 in terms of a convolution of two single ionization events, and both models are in reasonable overall agreement with experimental data. This supports the commonly held assumption that for a collision system with such large perturbation parameters, DI is dominated by TS-2. Furthermore, the similarity between the FCA calculation and the TS-2 simulation suggests that initial-state correlation, which is not accounted for in the simulation, is insignificant when TS-2 is dominant.

As an outlook we plan to extend the present work by developing a code based on the *convergent* frozen-correlation approximation [20]. This approach is an extension of the FCA and includes TS-1 like contributions to DI. It should be well suited to treat DI for collision systems with intermediate perturbation parameters, where the TS-1 and TS-2 amplitudes are expected to be of similar magnitude.

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