Comment on "Partial Photoionization Cross Sections and Angular Distributions for Double Excitation of Helium up to the $N = 13$ Threshold"

In a recent Letter [1], Czasch *et al.* report on measurements of partial cross sections and asymmetry parameters for ionization of He with excitation to well defined levels $n = 8$ to 13 of the residual ion in the energy region just below the double ionization (DI) threshold. The main interest of the study, beyond the technical challenge it represents, is that it brings new elements to discuss the possible relationship between the dynamics that takes place, respectively, above and below the DI threshold. These elements consist of measured asymmetry parameters and a related model. The authors first show that their measured threshold asymmetry parameter decreases steadily with increasing n, which is consistent with the usual expectation that it should tend towards -1 for n infinite, thus matching the expected value of the threshold asymmetry parameter for double ionization [2]. The authors then refer to Wannier's ideas [3] to propose, as an explanation of their observations, the dynamical model which motivates this comment. I discuss two aspects of this below.

Considering a helium atom in the Wannier configuration $r_1 = r_2$ and $\theta_{12} = \pi$, the authors state "If the (Wannier) configuration is oriented perpendicular to the polarization vector, the electric field of the photon can drive the system along the stable direction of the saddle." This is illustrated by the top part of Fig. 3(c) of Ref. [1] where the motions of the nucleus and the two electrons are indicated by arrows of opposite directions but both parallel to the polarization vector. This statement and the associated figure are incorrect. To show it, let us suppose, with the authors, that, at some intermediate stage of the dynamics, the particles are aligned in the Wannier configuration, and that this configuration is perpendicular to the polarization vector. This may certainly happen. However, the authors assume that the subsequent evolution will consist, at least for some time, of a mere motion of the particles along the photon field in a direction determined by their charge as shown in Fig. 3(c). This idea is incorrect because (i) it neglects the nonzero linear and angular momenta the particles have at the time considered, and (ii) because it neglects the Coulomb interactions inside the system despite the fact that they are orders of magnitude larger than the perturbative coupling of the system to the photon field.

The other part of the author's argument rests implicitly on the following two ideas: (i) if the system stays on the Wannier ridge $r_1 = r_2$, strong correlations develop, which is true; (ii) if strong correlations develop, the system will end with $\beta = -1$, which, despite current wisdom, is false. This can be easily understood by considering double ionization of $^3S^e$ helium, a process which, although as correlated as double ionization of $^1S^e$ helium, leads to $\beta = +2$ as early noted by Greene [4]. More recently, a series of numerical experiments devoted to the dynamics of He at 100 meV above the DI have been performed [5], leading to a comprehensive study of the relation between the value of the asymmetry parameter and the degree of correlation in the system [6]. This study is based on an analysis of the ionization excitation cross sections and asymmetry parameters in terms of parabolic partial waves in the photoelectron frame, which can be denoted $(n_1 n_2 |m|)$ following the parabolic convention, or (nKT) following the group theoretical convention currently in use for doubly excited states. It shows, in particular, that the mean value of $\cos\theta_{12}$ tends towards -1 as n tends towards infinity not only within the $(n, T = 1)$ subspace for which $\beta = -1$, but also within the $(n, T = 0)$ subspace for which $\beta = +2$. This provides a definite refutation of the common belief outlined just above in (ii), which appears as an essential element of the author's argument.

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