Comment on ‘’ Separation and Identification of Dominant Mechanisms in Double Photoionization’’

In a recent Letter, Schneider *et al* [1] calculated the ratios R of double to single photoionization of He by a single photon in terms of two contributing mechanisms: a knockout (KO) process and a shakeoff (SO) process. They used a classical-trajectory Monte Carlo phase space method to evaluate the *probability* of the KO process and the sudden approximation to evaluate the SO process. The important conclusion of this work is that double photoionization of He can be understood in terms of simple *incoherent* sum of the *probabilities* from these two mechanisms. They showed that the ratios R, as well as the singly differential cross sections, thus obtained are in good agreement with the most recent experimental results [2,3]. Thus Schneider *et al* offered a surprisingly simple interpretation of the mechanism of double ionization of He at all photon energies. However, such simplicity did not emerge from available elaborate *ab initio* calculations [4], nor from many-body perturbation calculations which showed that contributions from the different mechanisms should be added coherently [5].

In this Comment, we point out that the claim of Schneider *et al* is premature since their conclusion is based on the approximate evaluations of KO and SO probabilities. Specifically, they assumed that the first electron was created at $\vec{r}_1=0$, an approximation which is valid only in the limit of infinite photon energy. They also used a hydrogenic wavefunction with effective charge Z=1.6875 for the second electron to calculate the KO probability and another effective charge Z=1.49 to calculate the SO probability. The authors did not address how sensitive are their results with respect to the choice of the effective charges, but it is well known that SO probability can be calculated only with accurate correlated ground state wavefunctions [6]. In their model, the SO was evaluated without the consideration of electron correlation since they used a single effective charge for any escape velocity of the first electron.

We have recently [7] evaluated the SO probabilities vs the velocity of the first escape electron without the approximations employed by Schneider *et al.* Our starting point was completely identical to the expressions in their paper, Eqs. 6-9. Using accurate ground state wavefunction we obtained SO probabilities vs the escape velocity of the first electron. We compare our results with the SO probabilities of Schneider *et al* [1] in Fig.1, together with the experimental R [2]. Note that our SO probabilities are very different from those given by Schneider *et al*. Our SO results are already larger than the experimental ratios R. Thus if the KO contribution is to be added, it cannot be added incoherently. In our SO calculation, the dependence of R with respect to the escape velocity is a consequence of ground state correlation which was completely absent in the model of Schneider *et al*. Based on this discrepancy alone we doubt the conclusion of Schneider *et al* that double photoionization of He can be separated in terms of a simple incoherent sum of KO and SO processes at all photon energies. Such an important claim can be believed only after the properly defined KO and SO probabilities are evaluated accurately without the additional approximations in [1].

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Figure Caption: Double to Single photoabsorption cross section ratio R of He. The dotted lines are from experimental data [2] at low energies and from accurate calculations at high energies [4]. The ratio R_{SO} calculated from shakeoff theory : solid line, from [6] ; dashed lines, from [1].

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