Nuclear Recoil Cross Sections from Time-dependent Studies of Two-Photon Double Ionization of Helium

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We examine the sensitivity of nuclear recoil cross sections produced by two-photon double ionization of helium to the underlying triple differential cross sections (TDCS) used in their computation. We show that this sensitivity is greatest in the energy region just below the threshold for sequential double ionization. Accurate TDCS, extracted from non-perturbative solutions of the time-dependent Schrödinger equation, are used here in new computations of the nuclear recoil cross section.

PACS numbers: 32.80.Fb, 32.80.Rm

Two-photon double ionization of helium, a subject of intense theoretical interest in recent years, has now become the target of new experiments with high harmonic and free-electron laser sources. Of particular interest is the energy region just below the threshold for sequential double ionization at 54.4 eV, where theoretical studies have predicted marked changes in both total and single differential cross sections due to an effect we have termed “virtual sequential ionization”, where the signature of sequential ionization can be seen below the energetic threshold for that process.

The photoelectron angular distributions produced in two-photon non-sequential and sequential ionization are markedly different, the former characterized by largely back-to-back electron ejection while the latter is typically characterized by the uncorrelated product of dipole distributions ($\cos^2\theta$) for each electron along the direction of photon polarization. These distributions in turn leave clear signatures in the nuclear recoil distributions, even when they are integrated over all possible energy sharings of the ejected electrons, and are therefore likely to be **the first** differential quantities associated with two-photon double ionization that will be measured, since they do not require detection of the photoemitted electrons.

We define the nuclear recoil cross section in terms of the TDCS for two-photon double ionization,

$$\frac{d\sigma}{d^3\mathbf{Q}dE_1} = \int d\Omega_1 \int d\Omega_2 \frac{d\sigma^{2\omega}}{dE_1 d\Omega_1 d\Omega_2} \delta^3(\mathbf{Q} + \mathbf{k}_1 + \mathbf{k}_2) \quad (1)$$

where $\mathbf{Q}$ is the recoil momentum of the nucleus due to the ejection of two electrons of momenta $\mathbf{k}_1$ and $\mathbf{k}_2$,

$$\mathbf{Q} = -(\mathbf{k}_1 + \mathbf{k}_2) \quad (2)$$

Ω_1 and Ω_2 define the directions of ejection of the photoelectrons and E_1 is the energy of one ejected electron at a given photon frequency. The delta function picks out

$\mathbf{k}_1$ and $\mathbf{k}_2$ that satisfy Eq. (2) with

$$\begin{aligned} |\mathbf{k}_1| &= k_1 \\ |\mathbf{k}_2| &= k_2. \end{aligned} \quad (3)$$

Numerical implementation of the integral in Eq. (1) is outlined in ref.??.

Our earlier study of nuclear recoil used TDCS extracted from solutions of the time-independent coupled Lewis-Dalgarno equations that describe the absorption of two photons of frequency ω by an atom initially in state Φ_0 in second order perturbation theory,

$$\begin{aligned} (E_0 + \hbar\omega - H)\Psi_1^{sc}(\mathbf{r}_1, \mathbf{r}_2) &= \mu\Phi_0 \\ (E_0 + 2\hbar\omega - H)\Psi_2^{sc}(\mathbf{r}_1, \mathbf{r}_2) &= \mu\Psi_1^{sc}. \end{aligned} \quad (4)$$

Outgoing wave boundary conditions were imposed by solving Eq. (4) under exterior complex scaling (ECS). As we have recently shown, TDCS for two-photon double ionization, for sufficiently weak field intensities, can also be obtained from solutions of the time-dependent Schrödinger equation using second-order time-dependent perturbation theory. Ideally, nuclear recoil cross sections obtained from the two approaches would be equivalent. However, numerical difficulties complicate the calculations near the sequential double ionization threshold.

For real photon energies above the first ionization potential of the atom, Ψ_1^{sc} will behave as non-normalizable, undamped scattered wave. This complicates the solution of the equation for Ψ_2^{sc} since Ψ_1^{sc} appears in the driving term. To circumvent this problem, we added a small positive imaginary part to ω in the equation for Ψ_1^{sc} , which causes it to fall off exponentially, and then numerically extrapolated the results to real ω to obtain physical TDCS. The polynomial fits used to extrapolate the individual partial-wave amplitudes to real energies evidently become less reliable as we move to within several eV of the sequential threshold, where the total cross section in lowest-order perturbation theory formally diverges. This

becomes evident when comparing total and single differential cross sections computed using extrapolated time-independent methods and more recent time-dependent methods, which do not require any extrapolation. Differences in the cross sections computed using these two approaches systematically increase in the energy range between 50 and 54 eV, where non-sequential and virtual sequential processes, which evidently have very different analytic behavior, compete strongly. We have therefore recomputed the nuclear recoil cross sections using TDCS extracted from a time-dependent treatment.

In the time-dependent treatment, we solve, on a real grid, the time-dependent Schrödinger equation for an atom that is subjected to a radiation pulse that starts at $t = 0$ and ends at $t = T$. We treat the laser-atom interaction, V_L , in the dipole approximation. In the length gauge, it is given in terms of the dipole operator, $\boldsymbol{\mu} = -e \sum_i \mathbf{r}_i$ and the electric field $\mathbf{E}(t)$ by $V_L = \mathbf{E}(t) \cdot \boldsymbol{\mu}$. We express $\mathbf{E}(t)$ as

$$\mathbf{E}(t) = \begin{cases} E_0 F_\omega(t) \hat{\mathbf{e}} & t \in [0, T] \\ 0 & \text{elsewhere} \end{cases} \quad (5)$$

where E_0 is the maximum field strength, and $\hat{\mathbf{e}}$ is the polarization vector. We choose a sine squared envelope for the time dependence of the pulse, $F_\omega(t)$,

$$F_\omega(t) = \sin^2\left(\frac{\pi}{T}t\right) \sin(\omega t). \quad (6)$$

At time T , the pulse terminates and the wavepacket $\Psi(t)$ describing the system continues to evolve under the influence of the field-free atomic Hamiltonian H_0 and the outgoing electrons continue to interact with each other and the nucleus. As we have previously shown, we can effectively propagate the wavepacket to infinite time by defining a scattered wave corresponding to a specific final total energy E as the Fourier transform of the time propagated packet:

$$\Psi_{sc} \equiv -i \lim_{\gamma \rightarrow 0} \int_0^\infty dt e^{i(E+i\gamma-H_0)t} \Psi(T). \quad (7)$$

We note that Ψ_{sc} , from which all the physical information will be extracted, satisfies the time-independent driven equation

$$(E - H_0)\Psi_{sc} = \Psi(T), \quad (8)$$

with pure outgoing boundary conditions. It is in solving the driven equation for Ψ_{sc} that the ECS transformation on the electronic coordinates is introduced.

Amplitudes for double ionization corresponding to final state momenta $\mathbf{k}_1$ and $\mathbf{k}_2$ can be constructed from the scattered wave Ψ_{sc} . The derivation, which will not be repeated here, is based on formally expanding the wavepacket at the end of the pulse in a complete set of bound, single and double continuum states of the atom,

and using the asymptotic form of the full Green's function $(E+i\gamma-H_0)^{-1}$ to identify the coefficients which are the ionization amplitudes. A surface integral involving Ψ_{sc} and atomic Coulomb functions extracts the double ionization amplitude:

$$C(\mathbf{k}_1, \mathbf{k}_2) = \frac{1}{2} \int [\phi_{\mathbf{k}_1}^{-*}(\mathbf{r}_1) \phi_{\mathbf{k}_2}^{-*} \nabla \Psi_{sc}(\mathbf{r}_1, \mathbf{r}_2) - \Psi_{sc}(\mathbf{r}_1, \mathbf{r}_2) \nabla (\phi_{\mathbf{k}_1}^{-*}(\mathbf{r}_1) \phi_{\mathbf{k}_2}^{-*}(\mathbf{r}_2))] \cdot d\mathbf{S}, \quad (9)$$

where the two-electron gradient is $\nabla = (\nabla_1, \nabla_2)$ and the testing functions $\phi_{\mathbf{k}}^{-}$ are momentum-normalized Coulomb functions with a nuclear charge $Z = 2$. The quantities $C(\mathbf{k}_1, \mathbf{k}_2)$ are amplitudes specific to a particular radiation pulse. They can be computed over the range of energies within the bandwidth of that pulse. If the field intensities are such that two-photon double ionization can be treated with second-order time-dependent perturbation theory, we can use $C(\mathbf{k}_1, \mathbf{k}_2)$ to construct double ionization cross sections for energies within that bandwidth from the solution of the time-dependent Schrödinger equation for a single pulse. The TDCS for two-photon double ionization is then given by,

$$\frac{d\sigma^{2\omega}}{dE_1 d\Omega_1 d\Omega_2} = \frac{8\pi^3 (\Delta E_{fi}/2)^2 m^2 \hbar k_1 k_2}{c^2 |E_0|^4} \frac{|C(\mathbf{k}_1, \mathbf{k}_2)|^2}{|\mathfrak{F}(E_f, E_i, \omega, T)|^2}, \quad (10)$$

where the energy shape function $\mathfrak{F}$, whose explicit form is given in ref.???, depends on the pulse length T , the frequency ω and the difference between the initial-state target energy E_i and the final energy $E_f = k_1^2/2 + k_2^2/2$. While Eq. (10) only becomes exact as $T \rightarrow \infty$, the long time limit, as we have shown, is reached quickly, ie., computed 2-photon cross sections become T -independent, when no intermediate resonances are involved. In the present application, we are working near the sequential ionization threshold, above which the 2-photon cross section formally diverges in the long time limit. This divergence will of course be smoothed out for any finite length pulse, but the apparent cross sections calculated using Eq. (10) will then depend on pulse length as we approach the sequential threshold if the pulse bandwidth is large enough to sample the above-threshold region significantly.

Figure 1 shows SDCS and TDCS extracted from our time dependent calculations. For the results we are reporting here, Ψ_{sc} was obtained from an expansion in partial waves that included terms up to $\ell = 7$ for each electron. It is useful to compare these results with a simple analytical model that ignores correlation in the initial, intermediate and final states, but nevertheless gives a qualitatively correct description of sequential ionization. That model, whose details are derived elsewhere [?],

FIG. 1: Two-photon double ionization cross sections at (left to right) 48, 50 and 52 eV. Top row: SDCS from 1fs, 2fs and model calculations. Middle row: TDCS from 2fs calculations as a function of Θ_2 and energy sharing with the first electron aligned with the photon polarization. Bottom row: TDCS from model calculations.

gives the following expression for the TDCS,

$$\frac{d\sigma^{\text{seq}}}{dE_1 d\Omega_1 d\Omega_2} \approx \frac{\hbar}{4\pi} \left(\frac{3}{4\pi} \right)^2 \cos^2(\theta_1) \cos^2(\theta_2) \left[\frac{\sqrt{\sigma^{\text{He}^+}(E_2)\sigma^{\text{He}}(E_1)}}{E_0 + \hbar\omega - \epsilon_{1s} - E_1} + \frac{\sqrt{\sigma^{\text{He}^+}(E_1)\sigma^{\text{He}}(E_2)}}{E_0 + \hbar\omega - \epsilon_{1s} - E_2} \right]^2, \quad (11)$$

where σ^{He} and σ^{He^+} are the photoionization cross sections of helium and He^+ , respectively, and ϵ_{1s} is the energy of He^+ . When integrated over Ω_1 and Ω_2 , Eq.(11) gives an SDCS proportional to the square of the term in square brackets.

As Fig. 1 shows, the signature of sequential ionization is clearly manifested in the SDCS by the appearance of wings in the cross section that become increasingly sharp and peaked about extreme unequal energy sharing as the

threshold for sequential ionization is approached. This behavior is seen in both the *ab initio* and model results. The TDCS, however, tell a somewhat different story. Sequential ionization is characterized by the uncorrelated product of $\cos^2\theta$ angular distributions for each electron, as seen in the model TDCS results shown in Fig. 1, where the probability for back-to-back and parallel ejection of the two electrons are equal. The *ab initio* results, on the other hand, show that while the energetics favor extreme unequal energy sharing near the sequential threshold, the propensity for back-to-back ejection dominates, even at 52 eV photon energy.

This work was performed under the auspices of the US Department of Energy by the Los Alamos National Laboratory and the University of California Lawrence Berkeley National Laboratory under Contract DE-AC02-05CH11231 and was supported by the U.S. DOE Office

FIG. 2: Nuclear recoil cross sections at 48, 50 and 52 eV. Top row: 1fs pulse; middle row: 2fs pulse; bottom row: model calculation.

of Basic Energy Sciences, Division of Chemical Sciences. Foundation (Grant No. PHY-0604628).
CWM acknowledges support from the National Science