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Distinction between sequential and direct ionization in two-photon double ionization of helium

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This paper aims to shed some light on the role of the direct, or nonsequential, ionization channel in the regime in which the sequential channel is open in two-photon double ionization (TPDI) of helium. In this regime the sequential channel dominates any direct contribution unless the laser pulse is of very short duration, in which case their distinction is hard to draw. Based on both a simple model and full solutions of the time-dependent Schrödinger equation, we aim to provide evidence of direct double ionization by identifying a term proportional to the pulse duration in the double ionization yield. Indeed, such a term is identified in the energy-differential yield. When it comes to the total double ionization probability, however, it turns out that the net first-order contribution is negative. The nature of the negative first-order contribution is discussed, and we argue that it is of correlated origin.

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I. INTRODUCTION

The vast volume of publications dealing with the process of double ionization of helium by the absorption of two photons clearly demonstrates the interest in this subject [1–24]. Despite tremendous effort, both theoretically and experimentally, certain issues related to the process remain unsettled. One of these issues is the role of the *direct* or *nonsequential* ionization mechanism in the regime where sequential ionization dominates.

Of course, monochromatic sources with photon energies below the binding energy of the He^+ ion at 54.4 eV can only induce double ionization in a nonsequential way. Above 54.4 eV, however, the sequential mechanism dominates. Still, we do expect a certain direct contribution also above threshold. Such a contribution is indeed seen in several works. In Refs. [25–27] nonsequential ionization was revealed by studying the momentum distribution of the photo electrons, whereas Horner and co-workers suggested using nuclear recoil to identify a direct contribution [28]. Evidence of electron correlation has also been seen in the angular photo electron distribution in the sequential regime [29,30].

In order to fully distinguish the two double ionization mechanisms, we would need a quantitative criterion. In Ref. [25] it is stated that "*The momentum-space picture nicely distinguishes this process* [direct double ionization] *from the less interesting sequential double-ionization of He, an uncorrelated process which can be accounted for entirely with one-electron models*." In literature there seems to be some consensus on this way of distinguishing between the direct and sequential process; the latter may be understood within an independent particle picture, whereas the interaction between the electrons is crucial for the former process to come about. Thus, for the sequential process, energy conservation should apply to each electron separately, while it only applies to the two-particle system as a whole in the direct case. In Ref. [27] it is pointed out that this distinction is blurred in the case of finite pulse lengths, i.e., nonmonochromatic light sources, and consequently the authors refrain from using the terms "sequential" and "direct" or "nonsequential" and instead use the terms "uncorrelated" and "correlated," respectively. However, several works (see, e.g., Refs. [9,31]) suggest that under proper conditions, the probability for any direct process to take place should scale linearly with the pulse duration, whereas a sequential one should scale quadratically. The proper conditions are that ionization takes place slowly and that the pulse duration is long enough so that the laser light may be treated as near monochromatic.

In Ref. [32] Ishikawa and Midorikawa studied the process of above-threshold double ionization of the helium atom. They identified an "anomalous component" in the singly differential photoelectron distribution, which previously had been associated with direct double ionization. They were, however, able to find reasonable agreement with predictions based on a process coined "sequential ionization during core relaxation" (SIDCR), thus concluding that the anomalous contribution was consistent with a sequential process. SIDCR is understood as a sequential process in which the second electron has not yet had time to relax into the ground state of the He^+ residual ion and thus experiences an effectively lower ionization potential. However, as such a process involves at least *some* correlation, one may question whether this mechanism is a purely sequential one. Finally, the authors demonstrated that the total double ionization probability did not seem to have any term linear in pulse duration for the photon energy at hand ($\hbar\omega = 91.45$ eV).

Simple models based on a semi-independent particle picture have proven able to give quite precise predictions of the TPDI process both below and above the sequential threshold [7,13,14,32–34]. Recently, a similar model was applied in a study of direct TPDI of H_2 [35]. The quantitative agreement with full calculations, both in terms of total cross sections and cross sections differential in energy, is quite remarkable given the simplicity of the models. In the present work, we will make use of such a model to identify a term linear in pulse duration

above the sequential threshold. We will then discuss whether this terms can be attributed to the nonsequential ionization channel. The findings are confirmed by full solutions of the time-dependent Schrödinger equation (TDSE) for the problem.

According to the above discussion, we will make the *ansatz* that the total double ionization probability may be written as a sum of a contribution proportional to the pulse duration T and another proportional to its square, i.e.,

$$P_{\mathrm{DI}} = \left(\frac{I_0}{\hbar\omega}\right)^2 \left(\alpha\, T_{\mathrm{eff},2} + \frac{1}{2}\beta\, T_{\mathrm{eff},1}^2\right), \tag{1}$$

where I_0 is the amplitude of the field at peak intensity, and the effective pulse duration for an n-photon ionization process is given by [36]

$$T_{\mathrm{eff},n} = \int_0^T \left(\frac{I(t)}{I_0}\right)^n dt. \tag{2}$$

Here, $\hbar\omega$ is the photon energy, and α and β are the proportionality factors. The *ansatz* (1) is written this way in order to identify α and β in a manner which is independent of intensity and pulse shape. As mentioned, for such a picture to make sense, the pulse duration T must be large so that the laser is near monochromatic, and the intensity must be low enough so that the population in the initial state is not significantly altered at the time scale of ionization [31]. In our calculations we have made sure that these criteria are met—both in the model, which allows for arbitrarily weak fields, and in the full solutions of the TDSE.

In the next section, we will present the model in some detail. The technique for solving the full TDSE for the problem at hand is also outlined. In Sec. III we will present differential and total double ionization probabilities—both based on the model and full solutions. The nature of its dependence on pulse duration is analyzed in order to identify the ω-dependent parameters α and β in Eq. (1). The conclusions are drawn in Sec. IV. Atomic units are used unless stated otherwise.

II. THEORY

A. The model

Although various versions of the simple model on which we will base our analysis are described elsewhere (see Refs. [7,13,14,32,33]), we will present it here for completeness. We will follow the train of thought of Ref. [13]. Our assumption is that the two electrons, which we will take to be distinguishable for now, may be seen as an outer electron (a) seeing the effective potential of the nucleus combined with the inner electron, i.e., a *single active electron* (SAE) potential, and an inner electron (b) which only sees the potential of the bare nucleus, i.e., the potential of the He^+ ion. Our initial state is assumed to be the product state $|\varepsilon_a^0,\varepsilon_b^0\rangle = |\varepsilon_a^0\rangle|\varepsilon_b^0\rangle$, i.e., the respective ground states, and our final state is $|\varepsilon_a,\varepsilon_b\rangle = |\varepsilon_a\rangle|\varepsilon_b\rangle$, which corresponds to both electrons being in their respective continua with energies ε_a and ε_b, respectively. The ionization potential of electron a is $I_a = 0.904$ a.u. $= 24.5$ eV, the ionization potential of electron b is $I_b = 2$ a.u. $= 54.4$ eV, and the total double ionization potential is their sum, $I = I_a + I_b$. We insist that the electrons are ionized in the order: first a, then b. Note that this restriction in fact correlates the electrons; the underlying Hamiltonian is *not* merely the sum of an SAE and a He^+ part. Now, second-order perturbation theory in the length gauge using the dipole approximation and the rotating wave approximation provides [14,32]

$$\langle\varepsilon_a,\varepsilon_b|\Psi(T)\rangle = \frac{E_0^2}{4}\langle\varepsilon_a|x|\varepsilon_a^0\rangle\langle\varepsilon_b|x|\varepsilon_b^0\rangle \times \int_0^T dt\, f(t)e^{ibt}\int_0^t dt'\, f(t')e^{iat'}, \tag{3}$$

where

$$a \equiv \varepsilon_a + I_a - \hbar\omega, \tag{4}$$

and correspondingly for b. Here E_0 is the maximum electric field strength and $f(t)$ is the envelope of the laser field. For the simplest case of a rectangularly shaped laser pulse, the amplitude reads

$$\langle\varepsilon_a,\varepsilon_b|\Psi(T)\rangle = -\frac{E_0^2}{4}\langle\varepsilon_a|x|\varepsilon_a^0\rangle\langle\varepsilon_b|x|\varepsilon_b^0\rangle \times \frac{1}{a}\left(\frac{e^{i(a+b)T}-1}{a+b} - \frac{e^{ibT}-1}{b}\right). \tag{5}$$

The squared absolute value of this quantity gives the doubly differential double ionization probability $\partial^2 P_{\mathrm{DI}}/\partial\varepsilon_a\partial\varepsilon_b$, which for photon energies above 54.4 eV, i.e., in the sequential regime, features a pronounced peak centered at the point $\varepsilon_a + I_a - \hbar\omega = \varepsilon_b + I_b - \hbar\omega = 0$ in the energy plane. Clearly, this peak corresponds to energy conservation for each electron separately, consistent with a sequential process. Finally we impose exchange symmetry on the amplitude [Eq. (3)]:

$$\langle\varepsilon_a,\varepsilon_b|\Psi(T)\rangle \to \frac{1}{\sqrt{2}}[\langle\varepsilon_a,\varepsilon_b|\Psi(T)\rangle + \langle\varepsilon_b,\varepsilon_a|\Psi(T)\rangle]. \tag{6}$$

Consequently, the energy differential double ionization probability will feature two peaks rather than just one. In the so-called direct regime, $I/2 < \hbar\omega < I_b$, these peaks, which become increasingly sharp with increasing T, lie outside the first quadrant of the energy plane, and the last term in the parentheses of Eq. (5) may be disregarded as $T \to \infty$.

This leads to the following double ionization probability doubly differential in electron energies:

$$\frac{\partial^2 P_{\mathrm{DI}}}{\partial\varepsilon_a\partial\varepsilon_b} = \frac{E_0^4}{16}\frac{1-\cos[(a+b)T]}{(a+b)^2}|f(\varepsilon_a,\varepsilon_b) + f(\varepsilon_b,\varepsilon_a)|^2, \tag{7}$$

with

$$f(\varepsilon_a,\varepsilon_b) \equiv \frac{\langle\varepsilon_a|x|\varepsilon_a^0\rangle\langle\varepsilon_b|x|\varepsilon_b^0\rangle}{\varepsilon_a + I_a - \omega}. \tag{8}$$

Equation (7) integrates to a total TPDI probability proportional to the pulse duration T. The distribution narrows towards the line given by $\varepsilon_a + \varepsilon_b = 2\hbar\omega - I \equiv \varepsilon_{\mathrm{tot}}$, i.e., total energy conservation, as the laser becomes near monochromatic. The corresponding singly differential cross section reads

$$\frac{\partial\sigma_{\mathrm{dir}}}{\partial\varepsilon_{\mathrm{r}}} = \frac{\sqrt{2}\varepsilon_{\mathrm{tot}}\hbar^3\omega^2}{4\pi}|g(\varepsilon_a,\varepsilon_b) + g(\varepsilon_b,\varepsilon_a)|^2|_{\varepsilon_a+\varepsilon_b=\varepsilon_{\mathrm{tot}}}, \tag{9}$$

where the energy sharing $\varepsilon_r \equiv \varepsilon_b/\varepsilon_{\rm tot}$, and

$$g(\varepsilon_a,\varepsilon_b) \equiv \frac{\sqrt{\sigma_{\rm SAE}(\varepsilon_a + I_a)\,\sigma_{\rm He^+}(\varepsilon_b + I_b)}}{\sqrt{(\varepsilon_a + I_a)(\varepsilon_b + I_b)}\,(\varepsilon_a + I_a - \hbar\omega)}. \quad (10)$$

We have here substituted the matrix elements in Eq. (8) with one-photon single ionization cross sections [37],

$$\sigma_{\rm SAE}(\hbar\omega) = 4\pi^2\alpha\hbar\omega\left|\langle\varepsilon|x\left|\varepsilon_a^0\right\rangle\right|^2\Big|_{\hbar\omega=\varepsilon+I_a}, \quad (11)$$

$$\sigma_{\rm He^+}(\hbar\omega) = 4\pi^2\alpha\hbar\omega\left|\langle\varepsilon|x\left|\varepsilon_b^0\right\rangle\right|^2\Big|_{\hbar\omega=\varepsilon+I_b}, \quad (12)$$

where α here is the fine structure constant. The fact that only the absolute square enters on the right-hand side in Eqs. (11) and (12) causes no problem as we may safely assume that the radial wave functions involved are real valued. Accurate analytical fits to both the SAE and He^+ cross sections are given in Ref. [38].

As has already been demonstrated in previous works [7,13,34], the simple model Eq. (9) is able to reproduce both the total and differential TPDI cross sections in the direct regime to a remarkable degree of accuracy.

Above the sequential threshold the "sequential peaks" will dominate the energy distribution of the photoelectrons. The direct ionization channel is still open, however, and it should, at least in principle, contribute to the double ionization probability. As the first term in Eq. (5) defined the cross section in the direct regime, it may seem reasonable to assume that the very same amplitude provides the direct contribution above the threshold as well, and that the second term is simply the amplitude for the sequential component. This does not lead to any meaningful, however, as the absolute square of each of these terms alone would provide divergent "probability distributions."

It may be shown that, on the long time scale, integrating the doubly differential double ionization probability, as provided by Eq. (3), over an energy region which includes a peak, will be of leading order T^2 and that any region consistent with energy conservation but excluding the peaks yields a contribution of leading order T. However, the rather unphysical scenario of an instantaneous turn on of the laser pulse is problematic for finite pulses. Instead we resort to a pulse with a sine shaped envelope,

$$E(t) = E_0 f(t)\sin(\omega t),$$
$$\text{where} \qquad f(t) = \begin{cases} \sin\left(\frac{\pi}{T}t\right), & 0 < t < T \\ 0, & \text{otherwise} \end{cases}, \quad (13)$$

aiming to distinguish between T and T^2 contributions in a semianalytical manner. While, e.g., a sine squared or a Gaussian shape would be more realistic, this is a convenient choice as it facilitates the analysis. Solving the TDSE, both within the model and *ab initio* frameworks, it is found that the conclusions drawn are independent on the particular choice of pulse.

The time integral in Eq. (3) can now be expressed as

$$\mathcal{T}(\varepsilon_a,\varepsilon_b,\omega,T) = \frac{e^{i(a+b)T}-1}{4}\left[\frac{1}{(a+\pi/T)(a+b+2\pi/T)} + \frac{1}{(a-\pi/T)(a+b-2\pi/T)} - \frac{2a}{(a+b)(a^2-\pi^2/T^2)}\right] + (e^{ibT}+1)\,\frac{\pi^2/T^2}{(a^2-\pi^2/T^2)(b^2-\pi^2/T^2)}, \quad (14)$$

which leads to the following doubly differential double ionization probability:

$$\frac{\partial^2 P}{\partial\varepsilon_a\partial\varepsilon_b} = \frac{E_0^4}{32}\left|\langle\varepsilon_a|x\left|\varepsilon_a^0\right\rangle\langle\varepsilon_b|x\left|\varepsilon_b^0\right\rangle\mathcal{T}(\varepsilon_a,\varepsilon_b,\omega,T) + \langle\varepsilon_b|x\left|\varepsilon_a^0\right\rangle\langle\varepsilon_a|x\left|\varepsilon_b^0\right\rangle\mathcal{T}(\varepsilon_b,\varepsilon_a,\omega,T)\right|^2. \quad (15)$$

If we now consider an even simpler model in which we take the a and b electrons to be completely independent particles, i.e., we assume a Hamiltonian of the form,

$$H = H_{\rm SAE} + H_{\rm He^+}, \quad (16)$$

the problem decouples. Then, the ionization dynamics may be treated independently for the two electrons by first-order perturbation theory. For a sine shaped envelope, we have

$$\frac{\partial P_a}{\partial\varepsilon_a} = \frac{E_0^2}{2}\pi^2\left|\langle\varepsilon_a|x\left|\varepsilon_a^0\right\rangle\right|^2 T^2\,\frac{1+\cos(aT)}{((aT)^2-\pi^2)^2}, \quad (17)$$

and correspondingly for the b electron. The double differential double ionization probability will be their (symmetrized) product—times a global factor of $1/2$ according to the time ordering. By applying the common peaking approximation and extending the lower limit of the energy integrals to minus infinity, we arrive at a total double ionization probability directly proportional to T^2. Moreover, the proportionality factor β in Eq. (1) is in this picture simply identified as the product of the corresponding photoionization cross sections [9],

$$\beta = \sigma_{\rm SAE}(\hbar\omega)\sigma_{\rm He^+}(\hbar\omega). \quad (18)$$

B. The full solution

In order to verify the model results, the time-dependent Schrödinger equation is solved numerically for the fully correlated two-electron problem and presented for comparison. The TDSE in the case of atomic helium interacting with a classical electromagnetic field reads (velocity gauge)

$$i\frac{\partial}{\partial t}\Psi(\mathbf{r}_1,\mathbf{r}_2,t) = \left[\sum_{i=1,2}\left(-\frac{\nabla_{\mathbf{r}_i}^2}{2} - \frac{2}{|\mathbf{r}_i|} + \mathbf{A}(t)\cdot\mathbf{p}_i\right) + \frac{1}{|\mathbf{r}_1-\mathbf{r}_2|}\right]\Psi(\mathbf{r}_1,\mathbf{r}_2,t), \quad (19)$$

where the electric dipole approximation has been assumed. The applied propagation scheme is based on an expansion

in B-spline functions for the radial directions and coupled spherical harmonics in the angular coordinates,

$$\Psi(\mathbf{r}_1,\mathbf{r}_2,t) = \sum_{i,j,k} c_{i,j,k} \frac{B_i(r_1)}{r_1} \frac{B_j(r_2)}{r_2} \mathcal{Y}_k(\Omega_1,\Omega_2), \qquad (20)$$

where k is an index which enumerates all allowed combinations of the angular quantum numbers l_1, l_2, L, and M. Expanding the TDSE in Eq. (20) yields a system of first-order ordinary differential equations which is solved numerically with the Cayley-Hamilton propagation scheme,

$$\left(\mathbf{S} + \frac{i\Delta t}{2}\mathbf{H}(t+\Delta t)\right)\mathbf{c}(t+\Delta t) = \left(\mathbf{S} - \frac{i\Delta t}{2}\mathbf{H}(t)\right)\mathbf{c}(t), \qquad (21)$$

where $\mathbf{S}$ is the basis overlap matrix (due to the nonorthogonal B splines), $\mathbf{H}$ is the Hamiltonian matrix, and Δt is the propagation time step. The propagation starts from the initial vector $\mathbf{c}(t=0)$ which corresponds to the helium ground-state wave function. As the Hamiltonian matrix is too large for direct diagonalization, $\mathbf{c}(t=0)$ is obtained by applying the iterative implicitly restarted Arnoldi method. Equation (21) is then solved repeatedly until the laser-atom interaction is concluded at $t=T$ and we label the wave function corresponding to the last set of coefficients $\Psi_f(\mathbf{r}_1,\mathbf{r}_2)$. The details of our numerical scheme are presented in previous works and readers are referred to Ref. [12] for more elaboration on the methodology.

From the final wave function, the energy-differential probability distribution is obtained by projecting Ψ_f onto products of $Z=2$ one-electron Coulomb waves [9,12],

$$\frac{\partial^2 P}{\partial\varepsilon_1\partial\varepsilon_2} = \sum_{l_1,l_2,L,M} \left|\left\langle \psi^{Z=2}_{\varepsilon_1,l_1}(\mathbf{r}_1)\right| \times \left\langle \psi^{Z=2}_{\varepsilon_2,l_2}(\mathbf{r}_2)\middle|\Psi_f^{l_1,l_2,L,M}(\mathbf{r}_1,\mathbf{r}_2)\right\rangle\right|^2, \qquad (22)$$

with ε_1 and ε_2 being the continuum energy of electrons 1 and 2, respectively. Equation (22) constitutes an approximation in the sense that electron-electron repulsion is neglected in the product state spanning the two Coulomb waves. The error associated with this approach can be controlled by a technique known as postpropagation where the final wave function is exposed to additional field-free propagation. The idea is simply to leave the system to evolve freely in time after the end of the laser pulse in order to let the ionized electrons drift apart due to the electron-electron repulsion. This causes the mean distance between the ionized electrons to increase rapidly, thus reducing the importance of the electron-electron interaction. It is usually found that the error associated with such an approach becomes small after only a few cycles of postpropagation [9,17].

III. RESULTS AND DISCUSSION

Isolating the linear and quadratic terms in the double ionization probability necessitates a well-resolved sampling of the total double ionization probability. In terms of the fully correlated time-dependent Schrödinger equation for the two electrons, several independent simulations with as long pulse duration as possible were executed. The TDSE was solved in a basis with 410 B-spline functions distributed in a radial box extending up to 500 a.u. in each radial direction, including all angular channels up to $l_1 = l_2 = l_{\max} = 6$. This gives rise to a ground-state energy of -2.9035 a.u., which is in close agreement with the benchmark value -2.9037 a.u. [39]. The calculated total, as well as energy-differential, double ionization probabilities were found to be well converged with respect to the numerical representation, but it should be pointed out that larger $l_{\max}$ might be necessary if fully converged angular-differential properties were to be obtained [40].

Before addressing the issue of a possible first-order contribution to the total double ionization yield above the sequential threshold, we will demonstrate the predictive power of the simple model, Eq. (15), by comparing it with the full solution to the TDSE, Eq. (22). Although similar results for energy-differential double ionization probabilities are presented elsewhere [14], we include them here for completeness.

Figure 1 depicts the singly differential double ionization probability for the photon energy $\hbar\omega = 2.1$ a.u. $= 57.1$ eV.

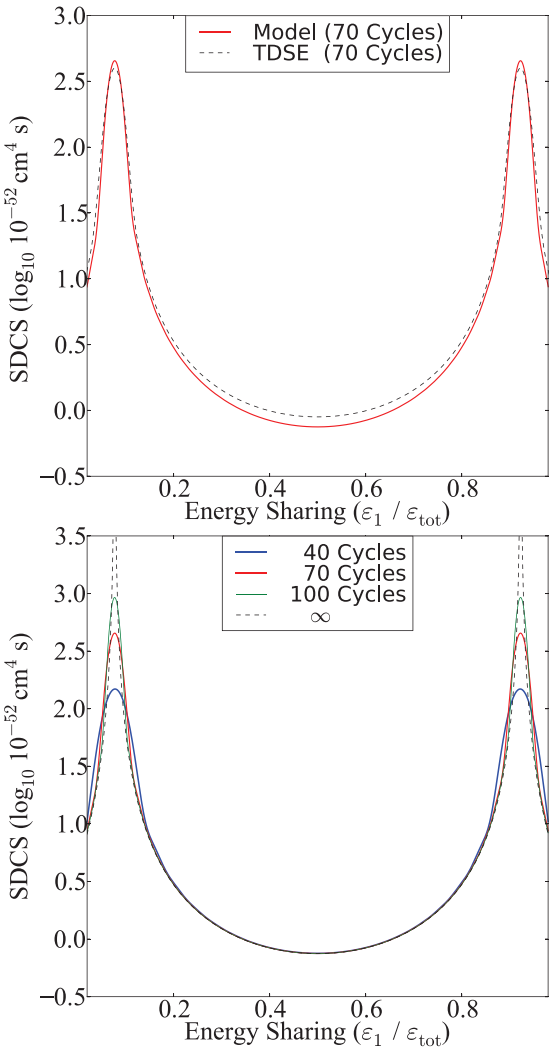

FIG. 1. (Color online) Singly differential "cross sections" (SDCS) for the photon energy 57.1 eV. The upper panel depicts a comparison between the full solution of the time-dependent Schrödinger equation (dashed curve) and the model prediction (full curve), for a pulse duration corresponding to 70 optical cycles (about 5 fs). The lower panel shows the model result for various pulse durations—including the limit of an infinitely long pulse.

The double ionization probability is given in terms of the generalized cross section for two-photon direct double ionization, which is related to the TPDI probability by

$$\sigma = \left(\frac{\hbar\omega}{I_0}\right)^2 \frac{P_{\rm DI}}{T_{\rm eff,2}}, \tag{23}$$

c.f. Eqs. (1) and (2). Although technically this "cross section" is not a meaningful measure above the threshold, we still find it a convenient quantity here. One reason for this is that it is independent of intensity. Another reason is that it allows us to compare the numbers both below and above threshold on an equal footing.

The upper panel in Fig. 1 shows a direct comparison between the full solution of the TDSE for the TPDI process and the corresponding result obtained by the model. The agreement with the simple model is in fact very good, both in terms of shape and total yield. The lower panel shows the model predictions for various pulse durations—including the limit of an infinitely long pulse; c.f. Eq. (9). Moreover, we observe that in the regions away from the peaks, i.e., when the two ionized electrons share the available excess energy more or less equally, the singly differential double ionization "cross section" quickly becomes T independent as the distance to the peaks (in terms of energy sharing) exceeds the bandwidth of the pulse, in agreement with the findings in Refs. [14,26]. This implies the contribution of a component to the double ionization yield proportional to T.

As already mentioned in the theory section, within a purely independent particle picture the double ionization yield becomes proportional to the square of the pulse duration, and the proportionality constant β is given by Eq. (18). Figure 2 shows the difference between the total double ionization yield as obtained by the model Eq. (15), which includes both first- and second-order contributions, and the corresponding probability obtained from an independent particle model, Eq. (18), for the photon energy 57.1 eV. The figure depicts the former subtracted from the latter. As it turns out, the β term from Eq. (18) coincides with the term proportional to T^2 in the model Eq. (15). Indeed, the difference is seen to become proportional to the pulse duration for increasing T, consistent with the *ansatz* (1). Moreover, and surprisingly, the slope, i.e., α in Eq. (1), turns out to be negative! The value of the parameter α at the current photon energy equals -0.46×10^{-52} cm^4 s, which was determined by using a linear least squares fit to the data in Fig. 2.

In the TDSE calculations the value of α is extracted in a slightly different manner. Instead of identifying the second-order term by analytical means, we extrapolate the quantity $P_{\rm DI}/T^2$ to infinity and subtract the corresponding second-order term from $P_{\rm DI}(T)$. The result is seen in Fig. 3, where the upper panel shows $P_{\rm DI}/T^2$ versus inverse time in black and the dashed red line (extrapolation for $T \to \infty$) is used to identify the term proportional to T^2. This contribution is subtracted from the total double-ionization probability and

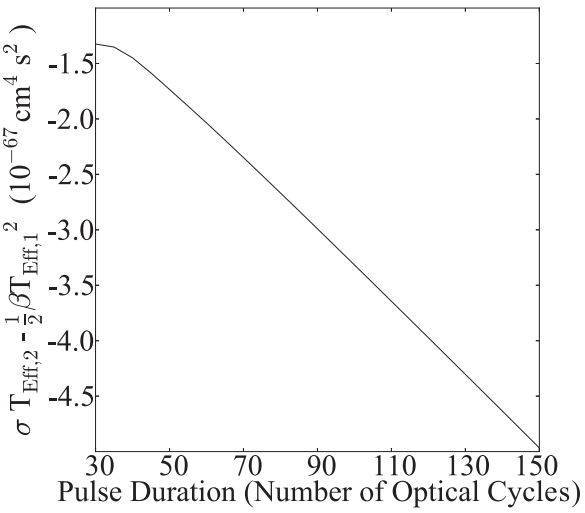

FIG. 2. Difference between the total double ionization probability given by the model Eq. (15) and the corresponding probability obtained from a fully independent particle model. The latter is proportional to the square of the pulse duration with the proportionality constant given by Eq. (18). The photon energy $\hbar\omega = 2.1$ a.u. $=$ 57.1 eV.

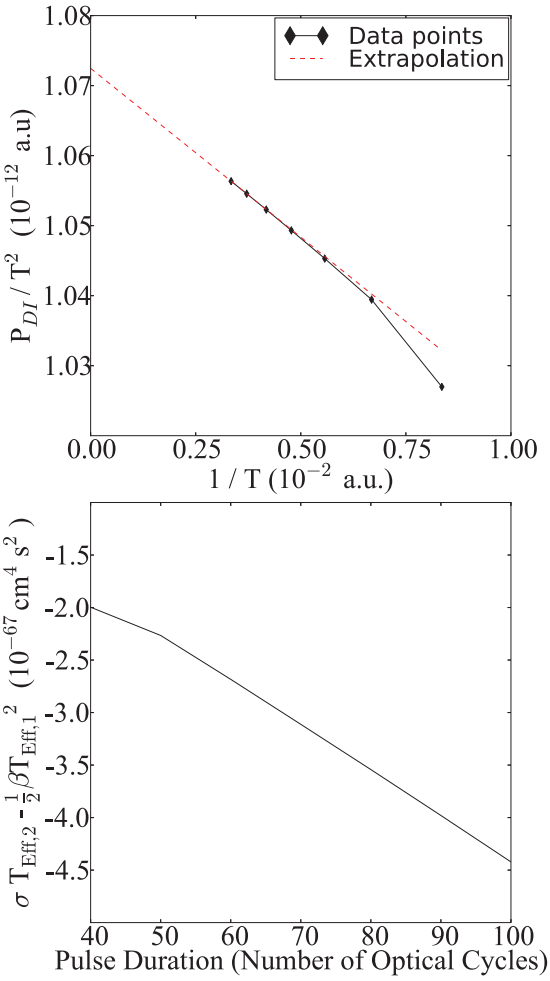

FIG. 3. (Color online) This figure, which is analogous to Fig. 2, is obtained from the full solutions of the TDSE. The β parameter, c.f. Eq. (1), is estimated by extrapolating $P_{\rm DI}/T^2$ to infinity (upper panel). The difference between the total double ionization yield and the corresponding quadratic term, βT^2, with β obtained as above, is displayed in the lower panel.

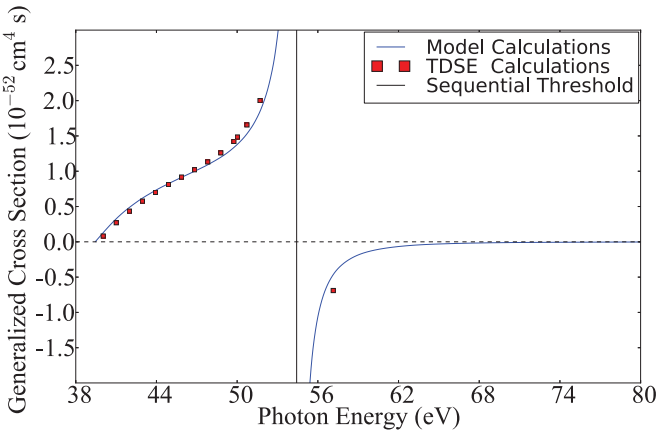

FIG. 4. (Color online) The first-order contribution to the total probability of double ionization, as obtained from the model—both under and over the sequential threshold. As in Fig. 1, the results have been given in units corresponding to a generalized cross section. The red points are results obtained from the full solutions of the TDSE. The TDSE results below threshold are taken from Ref. [12].

shown in the lower panel. Again we find that this difference becomes proportional to T for sufficiently long pulses, and, as in the case of the model, the *ansatz* (1) is seen to be adequate. More importantly, the full calculations confirm that the value of α is negative! Here, the extracted value for α at the photon energy 57.1 eV is $-0.65(\pm 0.05) \times 10^{-52}$ cm^4 s, which is somewhat lower than the value obtained within the model framework.

Figure 4 shows the first-order contribution (α) to the total double ionization probability versus photon energy, both below and above the sequential threshold at $I_b = 54.4$ eV. As can be seen from the figure, the model results compare very well with the full solutions of the TDSE for photon energies below the threshold energy. Above the threshold, on the other hand, the agreement is not quantitative as already noted. It does, however, support the main finding, namely that there is indeed a first-order term present also above the threshold—and that it is negative, as indicated in Figs. 2 and 3. The reason why only one single data point, i.e., at $\hbar\omega = 57.1$ eV, is included from the full TDSE calculations is twofold: First, the calculations are computationally very demanding in terms of computational time, and second, for higher values of $\hbar\omega$, the process becomes somewhat more complex by the fact that both singly and doubly excited states are involved in the TPDI process, thus obstructing a direct comparison with the model, which does not include the effect of excited states.

The fact that the net first-order component to the total TPDI yield turns out to be negative may seem odd in light of the observation in the lower panel of Fig. 1, where the contribution proportional to T is seen to be positive in the regions away from the "sequential peaks." Thus, the negative first-order contribution must stem from the region of these peaks.

As apparent from Fig. 4, the first-order contribution to the total double ionization yield seems to diverge as the photon energy approaches the sequential threshold energy—from both sides. As the photon energy increases far into the sequential regime, however, it vanishes—in agreement with what was found in Ref. [32]. Nevertheless, the nonsequential ionization dynamics cannot be disregarded altogether for higher photon energies. Inspecting the energy differential double ionization probabilities at higher photon energies, a positive contribution proportional to T is seen away from the peaks—much like what is demonstrated in the lower panel of Fig. 1 and in Refs. [14,26]. Consequently, there should be an equally large negative component manifested within the peaks, where distinguishing between the sequential and direct channel is problematic [41]. This contribution causes $\alpha(\omega)$ to approach zero for increasing photon energies.

Now, what is the origin of this negative first-order contribution; does it arise due to the presence of the direct channel? In the absence of a clear-cut, quantitative definition of what is meant by sequential double ionization, this question is hard to answer. If we, e.g., identify the sequential contribution to the total yield to be the term proportional to T^2, exclusively, any lower order term must be due to the direct channel—by definition. This is not a very interesting "conclusion," however. More interestingly, we could try and ascribe the negative first-order term at the peaks to destructive interference between the sequential and direct channels at the peaks. In order to investigate this candidate further, the simple, incoherent *ansatz* (1) must be replaced by one in which the amplitudes are considered. Such an *ansatz* would suggest that a possible interference term should scale as $T^{3/2}$, not as T. In arriving at this, we have assumed that the direct part gives rise to a finite value of the "cross section" at the peaks. If this were not the case, the notion of a separate direct contribution could hardly be meaningful. Thus, we are led to dismiss destructive interference as the origin of the negative first-order term. Moreover, we do not find any evidence for any $T^{3/2}$ term, neither in the model nor the full solution of the TDSE.

Instead one may ask whether the negative first-order contribution may be considered a correction to the leading sequential contribution. This may seem plausible given that the negative contribution is only supported in the vicinity of the peaks. However, if we again consider the independent particle model, Eq. (16), in which correlation is fully excluded, the model only supports corrections of order less than T^1. This indicates that the negative first-order contribution, be it sequential or direct, is indeed of correlated origin. Whether the effect is sequential or direct remains a matter of definition; in order to answer this question, a more precise distinction between the two processes is called for.

IV. CONCLUSIONS

We investigated the impact of the direct channel in the two-photon double ionization dynamics of helium above the sequential threshold at 54.4 eV, where the sequential ionization process dominates. We confirmed that there indeed is a direct contribution in the form of a part proportional to the pulse length in the double ionization yield differential in energy sharing. When it comes to the total double ionization probability, the net first-order contribution turns out to be negative. The negative contribution is not due to interference between the two ionization channels. However, it does seem to be induced by electron correlation.

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