Two-photon single and double ionization of helium

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We describe an efficient technique for calculating cross sections for two-photon single and double ionization of an atom without the use of the final-state continuum wave function. We present results of an application to helium over the photon energy range 25–54 eV, and we compare our results to a representative sample of results obtained by others.

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I. INTRODUCTION

The subject of two-photon double ionization is currently drawing much interest. The availability of sources of intense extreme-ultraviolet (XUV) radiation has given birth to experiments [1,2] probing for the first time the complete breakup of a helium atom by two-photon absorption. Further experiments are in the planning stages. Meanwhile, a wide range of theoretical techniques has been brought to bear on the problem, but the results are quite disparate [3–11]. The purpose of this paper is to introduce yet another technique and to present results of calculations of the generalized cross section for two-photon single and double ionization of helium over the photon energy range 25–54 eV. One merit of our approach is that it is computationally undemanding; our results were obtained within a relatively short execution time using a standard desktop with 2Mb of memory.

Our approach avoids explicit use of the final-state continuum wave function in both the single- and double-ionization channels. We start from the observation that the inclusive rate for the breakup of a system, summed over all possible decay channels, is, aside from a factor $-2/\hbar$, just the imaginary part of the system's (quasi)energy. Thus the inclusive rate for the breakup of an atom by two-photon absorption is (up to a factor $-2/\hbar$) the imaginary part of the second-order term in the perturbative expansion of the atom's ac quasienergy in powers of the light intensity. However, at frequencies above the threshold for one-photon ionization, this term includes contributions from both one- and two-photon ionization. The contribution from one-photon ionization is due to the possibility that while an electron can escape after absorbing only one photon it may absorb and reemit, or emit and reabsorb, another photon as it moves outwards in the radiation field; see Fig. 1. In other words the electron can undergo a one-photon transition to a dressed continuum state, dressed by the radiation field.

In addition, at frequencies above the threshold for two-photon double ionization the second-order quasienergy includes contributions from both single and double ionization. Our goal is to separate these different contributions. To accomplish this we make an approximation: We neglect the dressing of the bound states of the He atom and the residual He^+ ion by the radiation field. The resulting error is relatively very small except at those frequencies very close to a transition frequency of the residual ion, at which there is a core-excited resonance [12]. At frequencies where the detuning from a core-excited resonance is less than, or comparable to, the dipole coupling energy of the relevant ionic states our approximation does break down; regardless, in this region a meaningful ionization rate can no longer be defined, a point we return to below.

In the next section we describe the details of the method. In Sec. III we present our results.

II. THEORY

A. Basics

We consider two-photon absorption by an atom composed of two electrons labeled 1 and 2, each of charge $-e$, which are bound to a nucleus with atomic number Z that is fixed in space. The electrons are separated by the dimensionless dis-

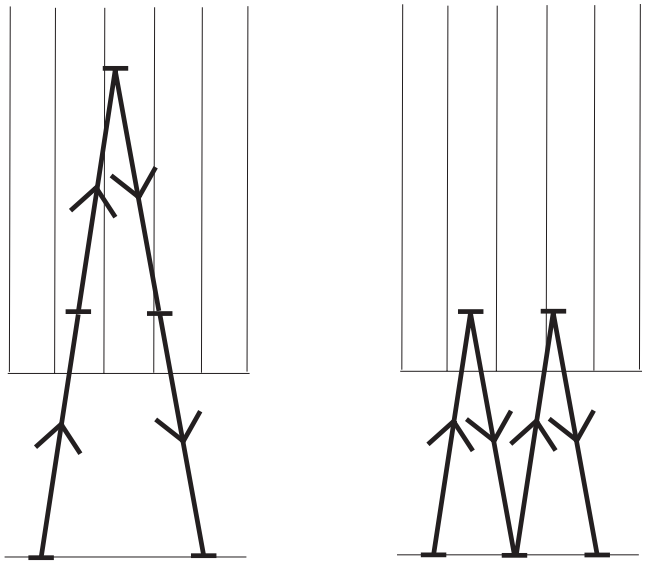

FIG. 1. Contributions to the imaginary part of the ac quasienergy, at second order in the intensity, for two-photon absorption at photon energies above the threshold for a one-photon transition to the continuum (the hatched region). The left diagram includes decay via both one- and two-photon transitions. Decay via a one-photon transition is accompanied by the absorption and reemission of a photon between two continuum states. The right diagram includes decay via a one-photon transition only; this transition is accompanied by the emission and reabsorption of a photon between a bound and continuum state.

tance $r_{12}=|\vec{r}_1-\vec{r}_1|$ where the dimensionless coordinates $\vec{r}_1$ and $\vec{r}_2$ locate the electrons relative to the nucleus. The Hamiltonian of the atom is $(e^2/a_0)H$ where a_0 is the Bohr radius and where

$$H = H_{aa} + \frac{1}{r_{12}}, \tag{1}$$

$$H_{aa} = h_a(1) + h_a(2), \tag{2}$$

$$h_a(j) = -\frac{1}{2}\nabla_j^2 - \frac{Z}{r_j} \tag{3}$$

with the subscript (aa) denoting the entrance channel of the atom. Photon absorption from a radiation field with frequency ω and electric field amplitude $\vec{F}_0$ (which is complex if the light is elliptically polarized) is mediated in the velocity gauge by the interaction $(e^2/a_0)V_+^{(v)}$ where

$$V_+^{(v)} = \frac{1}{2}\left(\frac{ea_0\vec{F}_0}{\hbar\omega}\right)\cdot(\vec{\nabla}_1 + \vec{\nabla}_2). \tag{4}$$

Photon emission is mediated in the velocity-gauge by the interaction $(e^2/a_0)V_-^{(v)}$ where $V_-^{(v)}$ differs from $V_+^{(v)}$ through complex conjugation of $\vec{F}_0$. Initially the atom is bound with energy $E_0(e^2/a_0)$ and its state is represented by $|\phi_0\rangle$. The response of the atom to n-photon absorption is represented by

$$|\phi_n^{(v)}\rangle = G(E_n)V_+^{(v)}|\phi_{n-1}^{(v)}\rangle, \tag{5}$$

where $|\phi_0^{(v)}\rangle=|\phi_0\rangle$, where E_n is the dimensionless energy of the atomic system after n-photon absorption, i.e.,

$$(E_n - E_0)(e^2/a_0) = n\hbar\omega \tag{6}$$

and where $G(E)$ is the resolvent

$$G(E) = (E-H)^{-1}. \tag{7}$$

We restrict E_1 to negative values, i.e., we assume the frequency lies below the threshold for one-photon double ionization. However, E_2 can be positive or negative.

If $E_2<0$ and if electron 2 is ejected while electron 1 remains bound (possibly in an excited state) the motion in the exit channel (ab) is governed by the Hamiltonian

$$H_{ab} = h_a(1) + h_b(2), \tag{8}$$

where $h_b(2)$ is the one-electron Hamiltonian

$$h_b(2) = -\frac{1}{2}\nabla_2^2 - \frac{Z-1}{r_2} - \frac{e^{-\alpha r_2}}{r_2} \tag{9}$$

with α real and positive (so the potential is Hermitian). The difference $(1-e^{-\alpha r_2})/r_2$ between $h_b(2)$ and $h_a(2)$ accounts for the partial screening of the nucleus at large distances. We take α to have the same value as the (inverse) length scale of the basis on which the states are expressed. The full Hamiltonian can be written as

$$H = H_{ab} + U_{ab}, \tag{10}$$

where U_{ab} is the final-state perturbation

$$U_{ab} = \frac{1}{r_{12}} - \left(\frac{1-e^{-\alpha r_2}}{r_2}\right), \tag{11}$$

which (for fixed r_1) falls off as an inverse square with increasing r_2 and (aside from the exceptional case $r_1=0$) is finite at $r_2=0$.

In the length gauge the interaction with the radiation field is $(e^2/a_0)V_+^{(l)}$, where

$$V_+^{(l)} = \frac{1}{2}\left(\frac{ea_0\vec{F}_0}{e^2/a_0}\right)\cdot(\vec{r}_1 + \vec{r}_2) \tag{12}$$

and the response of the atom to n-photon absorption is represented by

$$|\phi_n^{(l)}\rangle = G(E_n)V_+^{(l)}|\phi_{n-1}^{(l)}\rangle, \tag{13}$$

where $|\phi_0^{(l)}\rangle=|\phi_0\rangle$. Noting that

$$V_+^{(v)} = \left(\frac{e^2/a_0}{\hbar\omega}\right)[(E_n - H), V_+^{(l)}] \tag{14}$$

it is not difficult to show that

$$|\phi_n^{(v)}\rangle = \left(\frac{e^2/a_0}{\hbar\omega}\right)V_+^{(l)}|\phi_{n-1}^{(v)}\rangle - G(E_n)V_+^{(l)}|\phi_{n-1}^{(v)}\rangle - \left(\frac{e^2/a_0}{\hbar\omega}\right)G(E_n)V_+^{(l)}V_+^{(v)}|\phi_{n-2}^{(v)}\rangle. \tag{15}$$

In particular, the first-order response kets in the length and velocity gauges are related by

$$|\phi_1^{(v)}\rangle = \left(\frac{e^2/a_0}{\hbar\omega}\right)V_+^{(l)}|\phi_0\rangle - |\phi_1^{(l)}\rangle, \tag{16}$$

a result we use below.

B. Flux formulas

The second-order contribution to the inclusive rate for breakup is $-(2/\hbar)$ times the imaginary part of the second-order ac quasienergy. Let us denote this contribution by $(e^2/\hbar a_0)\Gamma_{\text{inc}}$, where Γ_{inc} is the dimensionless asymptotic flux. Referring to Fig. 1, both diagrams contribute to Γ_{inc} and their sum is

$$\Gamma_{\text{inc}} = -2\,\text{Im}[\langle\phi_0|V_-^{(v)}G(E_1)V_-^{(v)}G(E_2)V_+^{(v)}G(E_1)V_+^{(v)}|\phi_0\rangle + \langle\phi_0|V_-^{(v)}G(E_1)V_+^{(v)}G(E_0)(1-|\phi_0\rangle\langle\phi_0|) \times V_-^{(v)}G(E_1)V_+^{(v)}|\phi_0\rangle], \tag{17}$$

where the projection operator $(1-|\phi_0\rangle\langle\phi_0|)$ serves to eliminate the ground-state pole of $G(E)$. However, for our purpose it is more useful to express the inclusive flux directly as [12]

$$\Gamma_{\text{inc}} = i\langle\phi_2^{(v)}|(H-H^\dagger)|\phi_2^{(v)}\rangle. \tag{18}$$

Note that (use Green's theorem to convert the volume integral to an integral over the surface at infinity) only the asymptotic form of the response function $\langle\vec{r}_1\vec{r}_2|\,\phi_2^{(v)}\rangle$ contributes to the right side of Eq. (18). Using Eq. (5) in the second step below we can re-express Eq. (18) in a form more suitable for computation as

$$\Gamma_{\rm inc} = -2\ {\rm Im}\langle\phi_2^{(v)}|(E-H^\dagger)|\phi_2^{(v)}\rangle \tag{19}$$

$$= -2\ {\rm Im}\langle\phi_1^{(v)}|(V_+^{(v)})^\dagger|\phi_2^{(v)}\rangle. \tag{20}$$

In general, the right side of Eq. (20) includes contributions from both one- and two-photon ionization.

If $E_2>0$ two-photon double ionization occurs, and it contributes to $\Gamma_{\rm inc}$. In order to remove this contribution we introduce the projection operators $p_a(j;\mathcal{E})$, $j=1,2$, which project onto a subspace of the full two-electron space in which electron j resides in a localized state represented by a linear combination of those bound eigenstates of $h_a(j)$ with eigenvalues less than or equal to $\mathcal{E}$, where $\mathcal{E}<0$. Thus, if $|j;\mathcal{E}_a\rangle$ is an eigenket of $h_a(j)$ with eigenvalue $\mathcal{E}_a$,

$$p_a(1;\mathcal{E}) = \sum_{\mathcal{E}_a\le\mathcal{E}} |1;\mathcal{E}_a\rangle\langle 1;\mathcal{E}_a| \otimes \mathbf{1}_2, \tag{21}$$

where $\mathbf{1}_2$ is the identity operator acting on the space of electron 2. The symmetric projection operator

$$P_a(\mathcal{E}) \equiv p_a(1;\mathcal{E}) + p_a(2;\mathcal{E}) - p_a(1;\mathcal{E})p_a(2;\mathcal{E}) \tag{22}$$

projects onto a subspace of the full two-electron space in which at least one of the electrons is localized and has energy less than or equal to $\mathcal{E}<0$. Both $p_a(j;\mathcal{E})$ and $P_a(\mathcal{E})$ are Hermitian, and have the properties $p_a^2(j;\mathcal{E})=p_a(j;\mathcal{E})$ and $P_a^2(\mathcal{E})=P_a(\mathcal{E})$. Since the right side of Eq. (18) depends only on the asymptotic form of $\langle\vec{r}_1\vec{r}_2|\,\phi_2^{(v)}\rangle$, the rate $\Gamma_{\rm SI}(\mathcal{E}_{\rm max})$ for single ionization accompanied by excitation of the residual ion to all energy levels below $\mathcal{E}_{\rm max}$, where $\mathcal{E}_{\rm max}$ is negative and less than E_2, is given by making the replacement

$$|\phi_2^{(v)}\rangle \to P_a(\mathcal{E}_{\rm max})|\phi_2^{(v)}\rangle \tag{23}$$

in Eq. (18). Omitting $1/r_{12}$ from H (it cancels out) we have

$$\Gamma_{\rm SI}(\mathcal{E}_{\rm max}) = i\langle\phi_2^{(v)}|P_a(\mathcal{E}_{\rm max})(H_{aa}-H_{aa}^\dagger)P_a(\mathcal{E}_{\rm max})|\phi_2^{(v)}\rangle \tag{24}$$

$$= i\sum_{j=1}^{2}\langle\phi_2^{(v)}|p_a(j;\mathcal{E}_{\rm max})(H_{aa}-H_{aa}^\dagger)p_a(j;\mathcal{E}_{\rm max})|\phi_2^{(v)}\rangle \tag{25}$$

$$= 2i\langle\phi_2^{(v)}|p_a(1;\mathcal{E}_{\rm max})(H_{aa}-H_{aa}^\dagger)p_a(1;\mathcal{E}_{\rm max})|\phi_2^{(v)}\rangle \tag{26}$$

$$= 2i\langle\phi_2^{(v)}|p_a(1;\mathcal{E}_{\rm max})(H_{ab}-H_{ab}^\dagger)p_a(1;\mathcal{E}_{\rm max})|\phi_2^{(v)}\rangle. \tag{27}$$

We have just made an approximation: We have assumed that once the photoelectron escapes and is asymptotically far away, the residual ion, if it is bound, relaxes to an unperturbed state. This is not entirely correct, for within our formalism the residual ion remains in the radiation field and therefore its bound states are slightly perturbed by the field, and hence are not exactly eigenstates of the unperturbed Hamiltonian $h_a(1)$. In other words, we have neglected the virtual absorption and reemission, or emission and reabsorption, of a photon by the residual ion, which accompanies one-photon single ionization. As noted in the Introduction, this is a small correction except in the immediate vicinity of a core-excited resonance where the photon frequency differs from a transition frequency between two unperturbed states of the residual ion by less than the dipole coupling energy of these states. At these exceptional frequencies the virtual process becomes a real one, and the ion undergoes Rabi flopping so that it is no longer possible to define a meaningful ionization rate. Recall that we have not neglected the (generally important) real absorption and reemission, or emission and reabsorption, of a photon by the photoelectron as it escapes in the field after one-photon absorption.

We can put $\Gamma_{\rm SI}(\mathcal{E}_{\rm max})$ in a form more suitable for computation by introducing the resolvent

$$G_{ab}(E) = \frac{1}{E-H_{ab}}. \tag{28}$$

If $E<0$ we have

$$G(E) = G_{ab}(E) + G_{ab}(E)U_{ab}G(E). \tag{29}$$

Combining Eqs. (5) and (29) gives

$$p_a(1;\mathcal{E}_{\rm max})|\phi_2^{(v)}\rangle = p_a(1;\mathcal{E}_{\rm max})G_{ab}(E_2)[1+U_{ab}G(E_2)]V_+^{(v)}|\phi_1^{(v)}\rangle \tag{30}$$

$$= p_a(1;\mathcal{E}_{\rm max})G_{ab}(E_2)[V_+^{(v)}|\phi_1^{(v)}\rangle + U_{ab}|\phi_2^{(v)}\rangle]. \tag{31}$$

As noted earlier if r_1 remains finite U_{ab} vanishes as $1/r_2^2$ for $r_2\to\infty$. However, at energies above the complete breakup threshold both electrons escape, so U_{ab} has a long-range Coulomb tail, and therefore Eq. (29) is invalid when $E>0$. Nevertheless, Eq. (31) is valid even if $E_2>0$ since $\mathcal{E}_{\rm max}<0$ and $p_a(1;\mathcal{E}_{\rm max})|\phi_2^{(v)}\rangle$ excludes the double-ionization channel. Noting that $p_a(1;\mathcal{E}_{\rm max})$ commutes with H_{ab}, it follows from Eqs. (27) and (31) that

$$\Gamma_{\rm SI}(\mathcal{E}_{\rm max}) = -4\ {\rm Im}\langle\phi_2^{(v)}|p_a(1,\mathcal{E}_{\rm max})(E_2-H_{ab}^\dagger)p_a(1,\mathcal{E}_{\rm max})|\phi_2^{(v)}\rangle \tag{32}$$

$$= -4\ {\rm Im}[\langle\phi_1^{(v)}|(V_+^{(v)})^\dagger p_a(1,\mathcal{E}_{\rm max})|\phi_2^{(v)}\rangle + \langle\phi_2^{(v)}|U_{ab}p_a(1,\mathcal{E}_{\rm max})|\phi_2^{(v)}\rangle]. \tag{33}$$

In general, the right side of Eq. (33) includes contributions from both one- and two-photon single ionization.

C. Two-photon single and double ionization

Since $E_1<0$ the difference

$$\Gamma_{\rm DI}^{(2)} = \Gamma_{\rm inc} - \Gamma_{\rm SI}(0) \tag{34}$$

gives the rate for two-photon double ionization. To obtain the rate for two-photon single ionization we must eliminate the contribution to $\Gamma_{\rm SI}(\mathcal{E}_{\rm max})$ from one-photon single ionization. We begin this task by isolating that part of the modified second-order response ket $p_a(1;\mathcal{E}_{\rm max})|\phi_2^{(v)}\rangle$ which accounts for one-photon ionization.

Using Eq. (31), noting that $p_a(1;\mathcal{E}_{\max})$ commutes with $G_{ab}(E)$, and inserting $[p_a(1;E_1)+1-p_a(1;E_1)]$ between $V_+^{(v)}$ and $|\phi_1^{(v)}\rangle$, we have

$$p_a(1;\mathcal{E}_{\max})|\phi_2^{(v)}\rangle = G_{ab}(E_2)(|\tilde{\psi}_{\text{unloc}}\rangle + |\tilde{\psi}_{\text{loc}}\rangle), \tag{35}$$

where

$$|\tilde{\psi}_{\text{unloc}}\rangle = p_a(1;\mathcal{E}_{\max})V_+^{(v)}p_a(1;E_1)|\phi_1^{(v)}\rangle, \tag{36}$$

$$|\tilde{\psi}_{\text{loc}}\rangle = p_a(1;\mathcal{E}_{\max})\{V_+^{(v)}[1-p_a(1;E_1)]|\phi_1^{(v)}\rangle + U_{ab}|\phi_2^{(v)}\rangle\}. \tag{37}$$

At frequencies above the threshold for one-photon ionization $|\phi_1^{(v)}\rangle$ contains open subchannels in which one of the electrons can escape. Although $p_a(1;\mathcal{E}_{\max})$ acts to suppress the escape of electron 1, electron 2 can escape and therefore $|\tilde{\psi}_{\text{unloc}}\rangle$ represents an unlocalized state and is nonnormalizable. Indeed, if $|\mathcal{E}_a\mathcal{E}_b\rangle \equiv |1;\mathcal{E}_a\rangle \otimes |2;\mathcal{E}_b\rangle$ is an eigenket of H_{ab}, the matrix element $\langle \mathcal{E}_a\mathcal{E}_b|\tilde{\psi}_{\text{unloc}}\rangle$ is proportional to $\delta(\mathcal{E}_b - E_1 + \mathcal{E}_a)$. This delta function expresses energy conservation for the process in which the atom absorbs one photon and electron 2 escapes with energy $\mathcal{E}_b$ leaving behind electron 1 which is bound in the residual ion with energy $\mathcal{E}_a < E_1$. On the other hand, $|\tilde{\psi}_{\text{loc}}\rangle$ represents a localized state and is normalizable due to the joint action of $p_a(1;\mathcal{E}_{\max})$ (which suppresses the escape of electron 1) and either $[1-p_a(1;E_1)]$ (which suppresses the escape of electron 2) or U_{ab} (which suppresses the escape of both electrons).

Hence $G_{ab}(E_2)|\tilde{\psi}_{\text{unloc}}\rangle$ describes both one- and two-photon ionization while $G_{ab}(E_2)|\tilde{\psi}_{\text{loc}}\rangle$ describes only two-photon ionization. It is useful to look at $G_{ab}|\tilde{\psi}_{\text{unloc}}\rangle$ in position space:

$$\langle \vec{r}_1\vec{r}_2|G_{ab}(E_2)|\tilde{\psi}_{\text{unloc}}\rangle = \sum_{\mathcal{E}_a \le \mathcal{E}_{\max}} \left(\sum_{\mathcal{E}_b \le 0} + \int_0^\infty \rho(\mathcal{E}_b)d\mathcal{E}_b \right) \times \langle \vec{r}_1\vec{r}_2|\mathcal{E}_a\mathcal{E}_b\rangle \frac{\langle \mathcal{E}_a\mathcal{E}_b|\tilde{\psi}_{\text{unloc}}\rangle}{E_2 - \mathcal{E}_a - \mathcal{E}_b}, \tag{38}$$

where $\rho(\mathcal{E}_b)$ is the density of continuum states of electron 2. When $|\vec{r}_2|$ is large $\langle \vec{r}_1\vec{r}_1|\mathcal{E}_a\mathcal{E}_b\rangle$ oscillates very rapidly as $\mathcal{E}_b$ varies, and so the main contribution to the integral over $\mathcal{E}_b$ comes from the region where the integrand is singular. Since $\langle \mathcal{E}_a\mathcal{E}_b|\tilde{\psi}_{\text{unloc}}\rangle \propto \delta(\mathcal{E}_b - E_1 + \mathcal{E}_a)$ the response function $\langle \vec{r}_1\vec{r}_2|G_{ab}(E_2)|\tilde{\psi}_{\text{unloc}}\rangle$ contains, at asymptotically large distances, a wave $e^{ik^{(1)}(\mathcal{E}_b)r_2}$ which represents electron 2 moving radially outwards with momentum $k^{(1)}(\mathcal{E}_b) = \sqrt{2(E_1 - \mathcal{E}_a)}$ after the atom has absorbed one photon, leaving behind a residual ion with internal energy $\mathcal{E}_a < E_1$. The integrand is also singular at $\mathcal{E}_b = E_2 - \mathcal{E}_a$, where the denominator is proportional to $\delta(\mathcal{E}_b - E_2 + \mathcal{E}_a)$. This delta function expresses energy conservation for the process in which the atom absorbs two photons and electron 2 escapes with energy $\mathcal{E}_b$ leaving behind a residual ion with internal energy $\mathcal{E}_a < \mathcal{E}_{\max}$ where $\mathcal{E}_{\max}$ is negative and less than E_2. Hence at asymptotically large distances $\langle \vec{r}_1\vec{r}_2|G_{ab}(E_2)|\tilde{\psi}_{\text{unloc}}\rangle$ also contains a wave $e^{ik^{(2)}(\mathcal{E}_b)r_2}$ which represents electron 2 moving radially outwards with momentum $k^{(2)}(\mathcal{E}_b) = \sqrt{2(E_2 - \mathcal{E}_a)}$.

Evidently we must eliminate all asymptotic waves of the form $e^{ik^{(1)}(\mathcal{E}_b)r_2}$ from $\langle \vec{r}_1\vec{r}_2|G_{ab}(E_2)|\tilde{\psi}_{\text{unloc}}\rangle$. Since $k^{(1)}(\mathcal{E}_b)$ depends on the value of $\mathcal{E}_a$, it is expedient to resolve $|\tilde{\psi}_{\text{unloc}}\rangle$ into components, each of which depends on a specific value of the energy $\mathcal{E}_a$ of the (unperturbed) residual ion at asymptotically large distances. Thus we write

$$|\tilde{\psi}_{\text{unloc}}\rangle = \sum_{\mathcal{E}_a \le E_1} |\tilde{\chi}_{\text{unloc}}(\mathcal{E}_a)\rangle, \tag{39}$$

where $\langle \vec{r}_1\vec{r}_2|\tilde{\chi}_{\text{unloc}}(\mathcal{E}_a)\rangle$ is proportional to a specific wave $e^{ik^{(1)}(\mathcal{E}_b)r_2}$ at asymptotically large r_2. Noting that $h_b(2)$ commutes with $G_{ab}(E)$ we remove this wave from $G_{ab}(E_2)|\tilde{\chi}_{\text{unloc}}(\mathcal{E}_a)\rangle$ by applying $[E_1 - \mathcal{E}_a - h_b(2)]$ to $G_{ab}(E_2)|\tilde{\chi}_{\text{unloc}}(\mathcal{E}_a)\rangle$. However, $G_{ab}(E_2)|\tilde{\chi}_{\text{unloc}}(\mathcal{E}_a)\rangle$ also contains the waves $e^{ik^{(2)}(\mathcal{E}_b)r_2}$ and, as a result of applying $[E_1 - \mathcal{E}_a - h_b(2)]$, their amplitudes are erroneously enhanced by a factor $[E_1 - E_2 - \mathcal{E}_a + \mathcal{E}_a']$ where here $\mathcal{E}_a'$ is a possible energy of the residual ion after two-photon ionization. We correct for this by dividing by $[E_1 - E_2 - \mathcal{E}_a + h_a(1)]$. Thus, we introduce the operator

$$O(\mathcal{E}_a) \equiv \left(\frac{E_1 - \mathcal{E}_a - h_b(2)}{E_1 - E_2 - \mathcal{E}_a + h_a(1)} \right) p_a(1;\mathcal{E}_{\max}) \tag{40}$$

$$= -g_a(1;E_2 - E_1 + \mathcal{E}_a)[E_1 - \mathcal{E}_a - h_b(2)]p_a(1;\mathcal{E}_{\max}), \tag{41}$$

where $g_\beta(j;E)$ is the one-particle resolvent

$$g_\beta(j;E) = 1/[E - h_\beta(j)]. \tag{42}$$

Defining

$$|\tilde{\zeta}_{\text{loc}}\rangle \equiv \sum_{\mathcal{E}_a \le E_1} O(\mathcal{E}_a)|\tilde{\chi}_{\text{unloc}}(\mathcal{E}_a)\rangle, \tag{43}$$

we delete the contribution from one-photon single ionization to $\Gamma_{\text{SI}}(\mathcal{E}_{\max})$, and thereby obtain $\Gamma_{\text{SI}}^{(2)}(\mathcal{E}_{\max})$, the two-photon single ionization rate, by the replacement

$$p_a(1,\mathcal{E}_{\max})|\phi_2^{(v)}\rangle \to G_{ab}(E_2)[|\tilde{\zeta}_{\text{loc}}\rangle + |\tilde{\psi}_{\text{loc}}\rangle] \tag{44}$$

in Eq. (27).

If $\mathcal{E}_a' - \mathcal{E}_a = E_2 - E_1$, where $\mathcal{E}_a'$ and $\mathcal{E}_a$ are any two bound-state energies of the residual ion, $g_a(1;E_2 - E_1 + \mathcal{E}_a)$ has a pole. This pole is the signature a core-excited resonance. However, due to the interaction between the electrons, the corresponding pole in the complete scattering matrix is shifted slightly, and displaced off the real energy axis into the complex energy plane. Therefore we can add a tiny imaginary part to E_1 without incurring any physical consequence; we do so to improve numerical stability. We do not alter E_0 and E_2.

It remains to identify the components $|\tilde{\chi}_{\text{unloc}}(\mathcal{E}_a)\rangle$ in the decomposition of $|\tilde{\psi}_{\text{unloc}}\rangle$. This task exposes a difficulty: When $[E_1 - \mathcal{E}_a - h_b(2)]$ acts on $|\tilde{\chi}_{\text{unloc}}(\mathcal{E}_a)\rangle$ the resulting function in position space is prohibitively singular at $r_2 = 0$. To

remedy this we modify $|\tilde{\chi}_{\rm unloc}(\mathcal{E}_a)\rangle$, thereby changing $|\tilde{\zeta}_{\rm loc}\rangle$ and $|\tilde{\psi}_{\rm loc}\rangle$ on the right side of Eq. (44). We first use Eq. (16) to rewrite Eq. (36) as

$$|\tilde{\psi}_{\rm unloc}\rangle = (e^2/a_0)(1/\hbar\omega)p_a(1;\mathcal{E}_{\rm max})V_+^{(v)}p_a(1;E_1)V_+^{(1)}|\phi_0\rangle - p_a(1;\mathcal{E}_{\rm max})V_+^{(v)}p_a(1;E_1)|\phi_1^{(1)}\rangle. \quad (45)$$

The first term on the right side of Eq. (45) is normalizable, and so we subtract it from $|\tilde{\psi}_{\rm unloc}\rangle$ and add it to $|\tilde{\psi}_{\rm loc}\rangle$, i.e., we replace $|\tilde{\psi}_{\rm unloc}\rangle$ and $|\tilde{\psi}_{\rm loc}\rangle$, respectively, by

$$|\psi_{\rm unloc}\rangle = -p_a(1;\mathcal{E}_{\rm max})V_+^{(v)}p_a(1;E_1)|\phi_1^{(1)}\rangle \quad (46)$$

and

$$|\psi_{\rm loc}\rangle = |\tilde{\psi}_{\rm loc}\rangle + (e^2/a_0)(1/\hbar\omega)p_a(1;E_2)V_+^{(v)}p_a(1;E_1)V_+^{(1)}|\phi_0\rangle = p_a(1;\mathcal{E}_{\rm max})\{V_+^{(v)}[p_a(1;E_1)-1]|\phi_1^{(v)}\rangle + U_{ab}|\phi_2^{(v)}\rangle\}, \quad (47)$$

$$+ (e^2/a_0)(1/\hbar\omega)p_a(1;\mathcal{E}_{\rm max})V_+^{(v)}V_+^{(1)}|\phi_0\rangle, \quad (48)$$

where we used

$$[1-p_a(1;E_1)]|\phi_1^{(v)}\rangle + \frac{e^2}{a_0\hbar\omega}p_a(1;E_1)V_+^{(1)}|\phi_0\rangle = \frac{e^2}{a_0\hbar\omega}V_+^{(1)}|\phi_0\rangle + [p_a(1;E_1)-1]|\phi_1^{(1)}\rangle. \quad (49)$$

From Eqs. (13) and (29) we have

$$p_a(1;E_1)|\phi_1^{(1)}\rangle = p_a(1;E_1)G_{ab}(E_1)(V_+^{(1)}|\phi_0\rangle + U_{ab}|\phi_1^{(1)}\rangle) \quad (50)$$

and hence

$$p_a(1;E_1)|\phi_1^{(1)}\rangle = \sum_{\mathcal{E}_a\le E_1}|\xi_{\rm unloc}(\mathcal{E}_a)\rangle, \quad (51)$$

where

$$|\xi_{\rm unloc}(\mathcal{E}_a)\rangle = |1;\mathcal{E}_a\rangle\langle 1;\mathcal{E}_a|\otimes g_b(2;E_1+i0-\mathcal{E}_a)(V_+^{(1)}|\phi_0\rangle + U_{ab}|\phi_1^{(1)}\rangle). \quad (52)$$

It follows that

$$|\psi_{\rm unloc}\rangle = \sum_{\mathcal{E}_a\le E_1}|\chi_{\rm unloc}(\mathcal{E}_a)\rangle, \quad (53)$$

where

$$|\chi_{\rm unloc}(\mathcal{E}_a)\rangle = -p_a(1;\mathcal{E}_{\rm max})V_+^{(v)}|\xi_{\rm unloc}(\mathcal{E}_a)\rangle. \quad (54)$$

In place of $|\tilde{\zeta}_{\rm loc}\rangle$ we define

$$|\zeta_{\rm loc}\rangle \equiv \sum_{\mathcal{E}_a\le E_1}O(\mathcal{E}_a)|\chi_{\rm unloc}(\mathcal{E}_a)\rangle. \quad (55)$$

The presence of $V_+^{(1)}$ rather than $V_+^{(v)}$ on the right side of Eq. (52) ensures that $O(\mathcal{E}_a)|\chi_{\rm unloc}(\mathcal{E}_a)\rangle$ is nonsingular, or at least acceptably singular, in position space. We have

$$O(\mathcal{E}_a)|\chi_{\rm unloc}(\mathcal{E}_a)\rangle = p_a(1;E_2)g_a(1;E_2-E_1+\mathcal{E}_a)\{[V_+^{(v)},h_b(2)]\times|\xi_{\rm unloc}(\mathcal{E}_a)\rangle + V_+^{(v)}(|1;\mathcal{E}_a\rangle\langle 1;\mathcal{E}_a|\otimes 1_2)\times(V_+^{(1)}|\phi_0\rangle + U_{ab}|\phi_1^{(1)}\rangle)\}. \quad (56)$$

We delete the contribution of one-photon single ionization to $\Gamma_{\rm SI}(\mathcal{E}_{\rm max})$ by the replacement [instead of Eq. (44)]

$$p_a(1,\mathcal{E}_{\rm max})|\phi_2^{(v)}\rangle \to G_{ab}(E_2)[|\zeta_{\rm loc}\rangle + |\psi_{\rm loc}\rangle] \quad (57)$$

in Eq. (27). Note that $p_a(1;E_2)$ and $O(\mathcal{E}_a)$ commute with $G_{ab}(E_2)$, and with each other, so

$$\Gamma_{\rm SI}^{(2)}(\mathcal{E}_{\rm max}) = 2i\langle\zeta_{\rm loc}|G_{ab}^\dagger(E_2)(H_{ab}-H_{ab}^\dagger)G_{ab}(E_2)|\zeta_{\rm loc}\rangle + 2i\langle\psi_{\rm loc}|G_{ab}^\dagger(E_2)(H_{ab}-H_{ab}^\dagger)G_{ab}(E_2)|\psi_{\rm loc}\rangle - 4\,{\rm Im}\langle\psi_{\rm loc}|G_{ab}^\dagger(E_2)(H_{ab}-H_{ab}^\dagger)G_{ab}(E_2)|\zeta_{\rm loc}\rangle \quad (58)$$

$$= -4\,{\rm Im}\{\langle\zeta_{\rm loc}|G_{ab}(E_2)|\zeta_{\rm loc}\rangle + \langle\psi_{\rm loc}|G_{ab}(E_2)|\psi_{\rm loc}\rangle + \langle\psi_{\rm loc}|[G_{ab}(E_2)-G_{ab}^\dagger(E_2)]|\zeta_{\rm loc}\rangle\}. \quad (59)$$

III. APPLICATION

We have applied the preceding formalism, in particular Eqs. (34) and (59), to two-photon ionization of helium. We expressed the response functions on a complex Sturmian basis, composed of functions $r_1^{l_1}r_2^{l_2}L_{m-l_1-1}^{2l_1+1}(-2i\kappa r_1)L_{m-l_2-1}^{2l_2+1}\times(-2i\kappa r_2)e^{i\kappa(r_1+r_2)}$ with κ chosen to encompass outgoing-wave and exponentially damped-wave behavior, i.e., ${\rm Re}(\kappa)>0$ and ${\rm Im}(\kappa)>0$. We included values of l_1 and l_2 in the range $0\le l_1,l_2\le 3$ subject to angular momentum coupling rules and identical particle symmetry. For the reason noted in Sec. II C, at photon energies above 40 eV we added a tiny imaginary part 0.14 eV to E_1.

In Fig. 2 we show our results for the two-photon single-ionization generalized cross section over a range of photon energies beginning just above the threshold for one-photon single ionization of the ground-state He atom, and ending just below the threshold for one-photon ionization of the residual ground-state He^+ ion. This cross section was summed over all states of the He^+ ion. The results were obtained using 30 radial basis functions per electron. The numerous fluctuations in the cross section are signatures of resonances. At photon energies below 40.8 eV these resonances are due to one- or two-photon transitions to autoionizing states of He. The broad and high peak centered at 40.8 eV is due to the lowest core-excited resonance, where the photon frequency coincides with the $1s$-$2p$ transition frequency of He^+. The next core-excited resonance is centered at 48.4 eV, and infinitely many core-excited resonances accumulate at 54.4 eV, the threshold for one-photon ionization of ground-state He^+. Similar core-excited resonances were found [12] in a study of two-photon detachment of H^-. The computed heights of the core-excited resonance peaks are limited not only by the interaction between the electrons but also by our addition of an artificial imaginary energy of 0.14 eV to E_1. The inclusion of this artificial energy does not

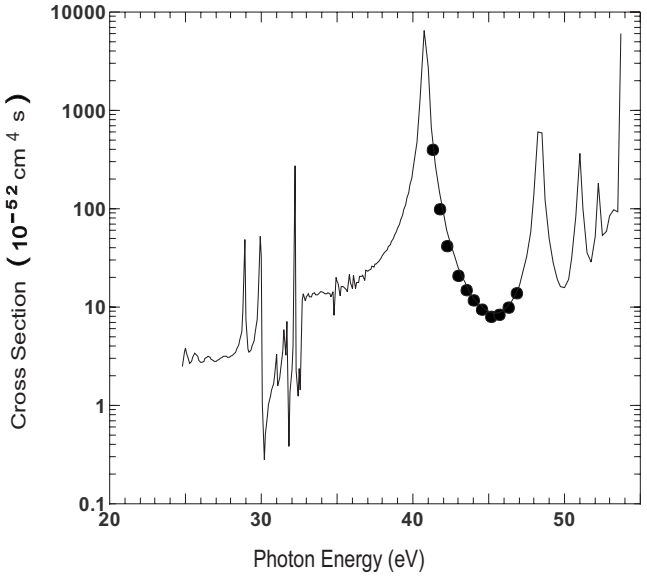

FIG. 2. Generalized cross section for two-photon single ionization of He summed over all states of the residual He^+ ion. The solid circles are the results of Feng and van der Hart [8].

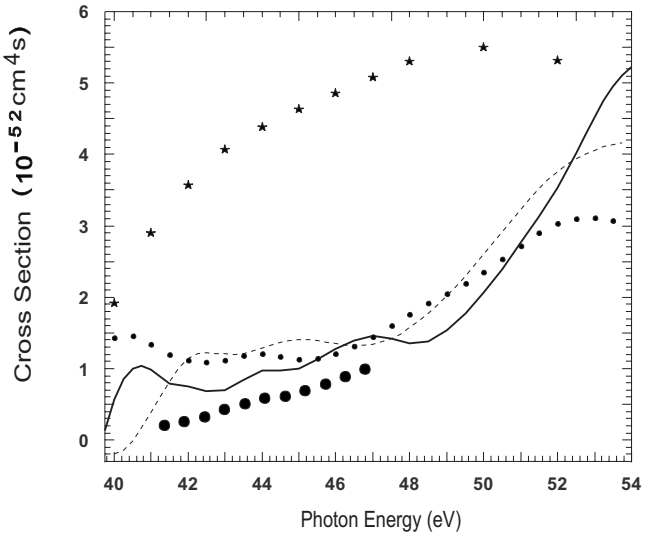

FIG. 3. Generalized cross section for two-photon double ionization of He. The solid circles are the results of Feng and van der Hart [8], the stars are the results of Foumouo *et al.* [4]. The remaining results are from the present work: Dots, 21 radial basis functions per electron; broken line, 30 radial basis functions per electron; solid line, 40 radial basis functions per electron.

affect the widths of the lowest few core-excited resonance peaks, which are much broader than 0.14 eV. At photon energies very close to the maxima of these peaks the dressing by the radiation field of the bound states of He^+, which we have omitted, becomes important, and a rate can no longer be defined, so the exact heights of the core-excited resonance peaks are of little interest. We also show in Fig. 2 the data of Feng and van der Hart [8] (solid circles) which cover the region between the first and second core-excited resonances. The agreement with our results is good.

In Fig. 3 we show our results for the two-photon double-ionization generalized cross section over a range of photon energies beginning just above the threshold 39.5 eV for two-photon double ionization and ending just below the threshold 54.4 eV for sequential double ionization [11]. We performed calculations with three different basis sets, containing different numbers of radial basis functions per electron; the first set (dots) contained 21 basis functions, the second set (broken line) contained 30, and the third set (solid line) contained 40 (which is about as many basis functions as can be accommodated on a 2Mb desktop). The three sets of results are in fair agreement far from the two thresholds but clearly diverge from each other as either threshold is approached. In fact, the true cross section should vanish at the lower threshold, but the 21×21 cross section rises near this threshold, while the 30×30 cross section reaches zero about 1 eV above this threshold. The numerical stability of our results (for double ionization) appears to be impaired by the presence of the core-excited resonances. Evidently a 40×40 basis is insufficient to achieve convergence; nevertheless, it yields results which may warrant some confidence, except near the threshold for sequential ionization, where an infinite number of core-excited resonances accumulates and perturbation theory is of questionable validity. We also show in Fig. 3 a representative sample of the results of others, specifically Feng and van der Hart [8] (solid circles) and Foumouo *et al.* (stars). The results of Nikolopoulos and Lambropoulos [3] (not shown) lie somewhat above those of Foumouo *et al.* while the results of Laulan and Bachau [9] (also not shown) lie between our results and those of Feng and van der Hart. In summary, two-photon double ionization of helium remains an open problem.

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