# Double photoionization of helium using many-body perturbation theory

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(Received 8 March 1993)

Cross sections for double photoionization of He are calculated using the lowest-order many-body perturbation theory. There are three amplitudes contributing in the present calculation. They represent the three mechanisms for double ionization, namely, two-step-1, shake off, and ground-state correlation. It is explicitly shown that the cross section for each of these mechanisms depends strongly on the adopted form of the dipole interaction as indicated by Dalgarno and Sadeghpour [Phys. Rev. A **46**, 3591 (1992)]. Our final results obtained by the sum of three amplitudes do not depend on the choice of the dipole formula at photon energies above 1 keV. The ratios of the cross sections for double ionization to single ionization are in excellent agreement with recent experimental results at energies 2–12 keV.

PACS number(s): 32.80.Fb, 31.20.−d

## I. INTRODUCTION

The double excitation and ionization of atomic targets with two electrons are the simplest and the most fundamental processes in which the dynamics of the electron-electron correlation interaction may be studied. At high energies, the interaction occurs so quickly that the important processes are limited to relatively simple ones. The dynamics of electron correlation in two electron atoms is easier to understand in interactions with photons than with charged particles. Recent experiments [1, 2] using synchrotron radiation now provide data on the ratio of double to single photoionization of helium, against which current theoretical understanding [3–10] may be examined.

In 1991, Ishihara, Hino, and McGuire [3] published results of calculations using the many-body perturbation theory (MBPT), which predicted a ratio of double to single ionization of about 1.6% at the photon energy of 2.8 keV shortly before the experimental result of 1.6 ± 0.3% was obtained [1]. This result was in excellent agreement with earlier predictions by Dalgarno and Ewart [11], Åberg [7], and Byron and Joachain [8], who also obtained this ratio in between 1.6% and 1.7%. However, such good agreement among the theorists seemed fortuitous because their theories appeared to be quite different. In particular, Ishihara, Hino, and McGuire found that one of the largest amplitudes came from the two-step-1 (TS1) term corresponding to the contribution of one electron, after an interaction with a photon, knocking the other electron out of an atom in a second step of the collision. Dalgarno and Ewart had ignored this term, and so had Åberg and also Byron and Joachain. On the other hand, Carter and Kelly [5] had emphasized the importance of this TS1 term as had Samson [9], who used only the TS1 term in his model of double photoionization. Moreover, Végh and Burgdörfer [10] showed that TS1 gave the largest contribution in the case of double ionization by a fast charged particle. Amusia [12] gave a mathematical argument indicating that TS1 was negligible for high-energy photons, and he further suggested that the result might depend on whether the length, velocity, or acceleration form was used. So, while the theorists agreed that the ratio at high energies was about 1.6%, there was disagreement as to the dominant process by which double photoionization was caused.

Recently, Dalgarno and Sadeghpour [4] have suggested a resolution to this dilemma, namely that TS1 is negligible in the acceleration form and may be large in the length form. Specifically, they gave two general mathematical arguments at the limit of high photon energy $\omega$ that while the TS1 term is non-negligible in the length form, it drops off in the acceleration form as $\frac{1}{\omega}$ relative to the leading terms.

In this paper, we present the lowest-order MBPT calculation of the double photoionization of He in the energy range 0.2–12 keV. The transition amplitude consists of three terms which correspond to the three mechanisms called in the literature as the two-step-1, the shake off (SO), and the ground-state correlation (GSC). Cross sections for individual mechanisms are also calculated separately.

Cross-section formulas are given in Sec. II in the length, velocity, and acceleration form of the dipole operator. Numerical results are shown in Sec. III. Three forms of the cross sections using the total transition am-

 

plitude agree very well with each other at high energies. Agreement with experiments is excellent at high energies for the ratio of double- to single-ionization cross sections. On the other hand, cross sections for individual mechanisms differ in the three forms of the dipole operator. Detailed discussions on this differences are given. Also given in this section is the calculated energy distribution of the ejected electrons in the double-ionization process. Conclusions are given in Sec. IV.

## II. FORMULATION

The cross sections for the double ionization of helium following absorption of a photon is given by

$$\sigma^{2+} = \frac{4\pi^2}{c}\frac{1}{\omega}\int_0^{\varepsilon_{\max}} |M_{fi}|^2 d\varepsilon_2, \tag{1}$$

where $M_{fi}$ stands for the dipole-transition matrix element and $c$ is the speed of light. Atomic units are used unless otherwise stated. Here the radial part of the continuum wave function is energy normalized, i.e., $\int \psi_{\varepsilon_1}\psi^*_{\varepsilon_2} dr = \delta(\varepsilon_1 - \varepsilon_2)$. In Eq. (1), the energies of the two ejected electrons satisfy the energy-conservation relation

$$\varepsilon_1 + \varepsilon_2 = \omega + E_0 \tag{2}$$

with $E_0$ being the ground-state energy of a helium atom, $\varepsilon_1$ and $\varepsilon_2$ being the energies of the two ejected electrons, and the upper bound of the $\varepsilon_2$ integration being $\varepsilon_{\max} = \omega + E_0$.

We shall consider three forms for the dipole interaction which are known to give the same cross sections. Choosing the direction of polarization of a photon to be the $z$ axis, the three forms of the dipole operator are

$$d^L = z \qquad \text{(length form)}, \tag{3}$$

$$d^V = \frac{d}{dz} \qquad \text{(velocity form)}, \tag{4}$$

$$d^A = \frac{2z}{r^3} \qquad \text{(acceleration form)}, \tag{5}$$

where $z$ and $r$ are the $z$ coordinate and the radial distance of an electron with respect to the helium nucleus (whose nuclear charge $Z = 2$ appears in the acceleration form). The cross sections of Eq. (1) in these three forms are calculated according to

$$M_{fi} = -\omega D^L, \quad D^V, \quad \text{or} \quad -\frac{1}{\omega}D^A, \tag{6}$$

where $D^a$ ($a = L, V$, or $A$) is the matrix element of the dipole operator $d_1^a + d_2^a$ for the two electron target. These three forms of $M_{fi}$ are identical, provided that the atomic wave functions are exact.

In MBPT [13], the transition amplitude $T^a$ is expanded in powers of the electron-electron correlation interaction $v$. For the basis set $\{\phi_n\}$ of the expansion we choose the usual $V^{N-1}$ potential defined by

$$h\,\phi_n(\mathbf{r}) = \varepsilon_n\,\phi_n(\mathbf{r}), \tag{7}$$

where

$$h = -\frac{1}{2}\nabla^2 + V^{N-1}(\mathbf{r}) \tag{8}$$

with

$$V^{N-1}(\mathbf{r}) = -\frac{2}{r} + \int \frac{|\phi_{1s}(\mathbf{r}')|^2}{|\mathbf{r}-\mathbf{r}'|}d\mathbf{r}'. \tag{9}$$

Equation (7) is the Hartree-Fock equation for the $1s$ state $\phi_{1s}$ for which we use the results of Ref. [14]. An electron in an excited orbital $\phi_n$ ($n \neq 1s$) moves in the static potential of the $1s$ electron.

There are three diagrams contributing to double photoionization in the lowest-order MBPT as shown in Fig. 1. In these diagrams, it is understood that the hole-hole interactions are incorporated to all orders to give the Hartree-Fock energy

$$E_0 = 2\varepsilon_{1s} - \langle 1s1s|v|1s1s\rangle \tag{10}$$

for the ground-state energy $E_0$ of helium instead of the sum of the $1s$ orbital energies $2\varepsilon_{1s}$.

The diagram labeled as TS1 is the so-called two-step-1 amplitude that corresponds to the collision process in which one electron is ejected from the ground state by an incident photon and then interacts with the other electron on the way out of the collision region. The SO diagram corresponds to the shake off where one electron is ejected by the photon and the wave function of the re-

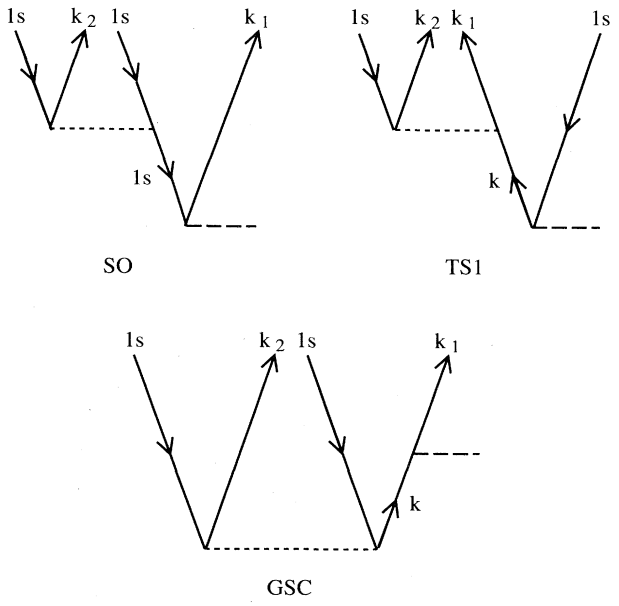


FIG. 1. The lowest-order MBPT diagrams for double photoionization of helium. SO, TS1, and GSC correspond to shake off, two-step-1 and ground-state correlation as explained in the text. The electron-photon dipole interaction $d$ is represented by a dashed line, while the electron-electron interaction $v$ is shown by a dotted line. The angular momenta of the final continuum state $(k_1, k_2)$ for TS1 and GSC are either $(s,p)$ or $(p,d)$ and exchange diagrams are included. For SO, only $(s,p)$ contributes.

maining electron later readjusts to the change in nuclear screening. The combination of TS1 and SO is referred to conventionally as the final-state correlation since both correspond to the electron-electron interactions occurring after the interaction with the photon [5]. The GSC diagram corresponds to the ground-state correlation where the effect of the interaction between the electrons in the ground state is taken into account. Transition amplitudes in the $a$ form of the dipole operator ($a = L, V$, or $A$) for these diagrams are expressed as

$$T^a(\mathrm{TS1}) = \sum_k \frac{\langle k_1 k_2|v|k\ 1s\rangle\langle k|d^a|1s\rangle}{\varepsilon_{1s} - \varepsilon_k + \omega + i\eta}, \tag{11}$$

$$T^a(\mathrm{SO}) = -\frac{\langle k_2 1s|v|1s1s\rangle\langle k_1|d^a|1s\rangle}{\varepsilon_{1s} - \varepsilon_{k_1} + \omega}, \tag{12}$$

$$T^a(\mathrm{GSC}) = \sum_k \frac{\langle k_1|d^a|k\rangle\langle kk_2|v|1s1s\rangle}{E_0 - \varepsilon_k - \varepsilon_{k_2}}, \tag{13}$$

where it is understood that the summation with respect to $k$ is taken over all excited (including continuum) states $\phi_k$ ($k \neq 1s$) and $\eta \to 0^+$.

Cross-section formulas in the length, velocity, and acceleration form are obtained by substituting

$$M_{fi} = -\omega T^L, \quad T^V, \quad -\frac{1}{\omega}T^A, \tag{14}$$

respectively, into Eq. (1), where

$$T^a = T^a(\mathrm{TS1}) + T^a(\mathrm{SO}) + T^a(\mathrm{GSC}) \tag{15}$$

is the transition amplitude in the lowest-order MBPT. If our lowest-order approximation is good enough, these three forms of cross sections must be close to each other.

The cross section for each diagram is also obtained similarly using the amplitudes given in Eqs. (11)–(13). However, they depend on the form of the dipole operator because those amplitudes correspond to the dipole matrix elements calculated using a part of the wave functions for the initial and final states expanded in the perturbation series.

## III. RESULTS AND DISCUSSION

We have evaluated cross sections for double photoionization of helium using the lowest-order MBPT in the length ($L$), velocity ($V$), and acceleration ($A$) form of the dipole operator. Figure 2 shows the cross section ratio of double to single ionization in the $L$ form. Curves TS1, SO, and GSC are the ratios where the double-ionization cross sections are calculated using the corresponding amplitudes Eqs. (11), (12), and (13), respectively. The curve marked "Total" is the cross-section ratio obtained using the coherent sum Eq. (15) of three amplitudes for double ionization. In the $L$ form, TS1 and GSC appear to be the largest and the magnitudes are greater by roughly one order than "Total" at high energies. Therefore, it is apparent that the TS1 and GSC amplitudes interfere *destructively* in the $L$ form [3]. Though TS1 is large at high photon energy $\omega$, it does not seem to become constant contrary to the suggestion by Dalgarno and Sadeghpour [4].

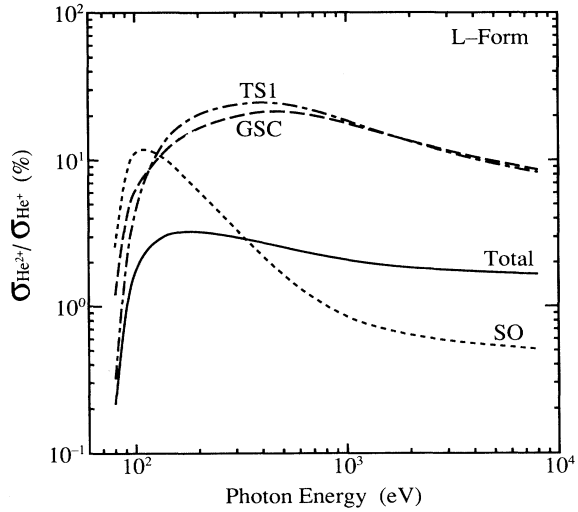


FIG. 2. Cross-section ratios of double to single ionization calculated using the dipole operator in the length form. TS1, SO, and GSC are the results for corresponding amplitudes Eqs. (11), (12), and (13), respectively. "Total" is our final results for the sum Eq. (15) of three amplitudes.

The ratios in the $V$ form are shown in Fig. 3. At higher $\omega$ the TS1 contribution falls off rapidly and both SO and GSC interfere *constructively* to result in "Total." The ratios in the $A$ form are shown in Fig. 4. Here all three amplitudes seem to interfere *constructively*, and TS1 falls off at high $\omega$. The $\omega$ dependence of this decline is reasonably close to $\frac{1}{\omega}$ given by the analysis of Dalgarno and Sadeghpour [4].

Thus it has been demonstrated that the individual MBPT cross sections indeed depend upon the form of the dipole operator, as we have mentioned in Sec. II. At the high-$\omega$ limit, TS1 is dominant in the $L$ form but neg-

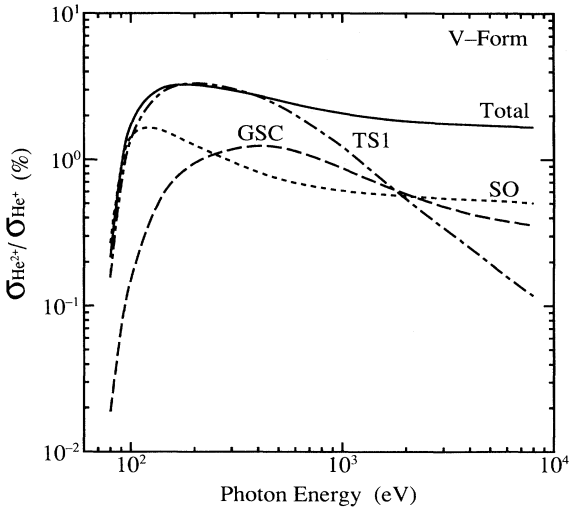


FIG. 3. Same as Fig. 2 in the velocity form.

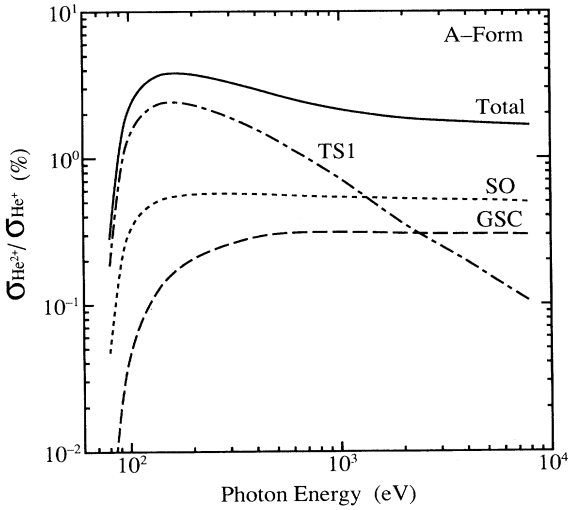


FIG. 4. Same as Fig. 2 in the acceleration form.

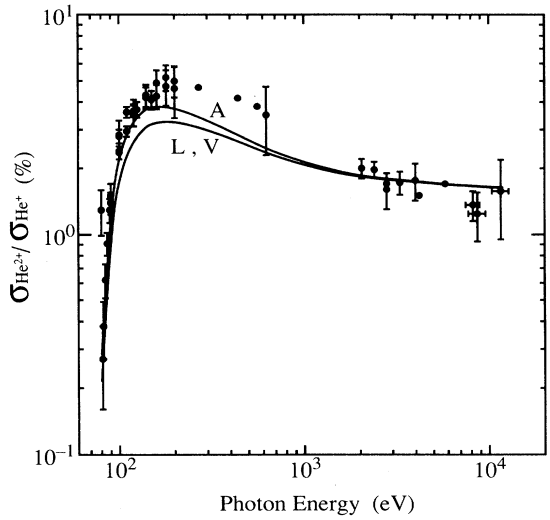


FIG. 5. Comparison of the cross-section ratios for double to single photoionization. Curves $L$, $V$, and $A$ are the length, velocity, and acceleration calculations, respectively. Experimental data are taken from Refs. [1, 2, 16–22]. Note that $L$ and $V$ are the same as shown by Eq. (24), but $A$ differs a little, in agreement with Eq. (27).

ligible in the $V$ and $A$ forms. SO's in all forms of the dipole operator converge to 0.5% at the asymptotic high $\omega$.

In Table I [15], we tabulate the cross sections for the single and "Total" double ionizations and the associated ratios in each form. The single-ionization cross section varies as $\omega^{-\frac{7}{2}}$. Figure 5 compares the ratios of the calculated cross sections for the double to single ionization taken from Table I and the existing experimental data [1, 2, 16–22]. As we have mentioned in Sec. II, agreement of all three formulas is a necessary condition for the validity of the approximation. We show below that our lowest-order results in the $L$ and $V$ form are identical at all values of $\omega$. Small differences seen in Table I are due to round-off errors. There are some differences between the $A$ form and $V$ (or $L$) form that are negligible at high energies. Differences are less than 1% at energies above 2 keV. Our results are in excellent agreement with the recent observations in the keV region [1, 2, 22]. We note that our ratio of double to single ionization is not quite independent of $\omega$ at the highest energy of our calculation.

It was recently pointed out by Samson, Greene, and Bartlett [23] that Compton scattering might affect the measured cross-section ratio of the double to single ionization at photon energies higher than 6 keV. They claim that the contribution of the Compton scattering dominates at the higest energy for which experimental data are available. The satisfactory agreement of the measured value at this energy with the present results and other theoretical predictions, in which no account is taken for the contribution of the Compton scattering, may imply that the ratio of the double- to single-ionization cross section due to the Compton scattering

TABLE I. Cross sections of the single photoionization $\sigma^+$ and the double photoionizations $\sigma^{2+}$ of helium, and the ratios of double to single photoionization vs the incident photon energy $\omega$. $L$, $V$, and $A$ stand for the length, velocity, and acceleration form, respectively. The numbers in square brackets represent powers of 10.

| $\omega$ (keV) | $\sigma^+$ (cm$^2$) | $\sigma^{2+}$ (cm$^2$) | | | Ratios (%) | | |
|---|---|---|---|---|---|---|---|
| | | $L$ | $V$ | $A$ | $L$ | $V$ | $A$ |
| 0.2 | 4.95[-20] | 1.66[-21] | 1.66[-21] | 1.90[-21] | 3.35 | 3.35 | 3.85 |
| 0.4 | 6.48[-21] | 1.79[-22] | 1.79[-22] | 1.92[-22] | 2.77 | 2.77 | 2.97 |
| 0.6 | 1.88[-21] | 4.48[-23] | 4.48[-23] | 4.69[-23] | 2.39 | 2.39 | 2.50 |
| 0.8 | 7.64[-22] | 1.68[-23] | 1.68[-23] | 1.74[-23] | 2.19 | 2.20 | 2.28 |
| 1.0 | 3.77[-22] | 7.82[-24] | 7.82[-24] | 8.06[-24] | 2.08 | 2.08 | 2.14 |
| 2.0 | 3.99[-23] | 7.23[-25] | 7.24[-25] | 7.31[-25] | 1.81 | 1.82 | 1.83 |
| 4.0 | 4.03[-24] | 6.98[-26] | 7.02[-26] | 7.04[-26] | 1.73 | 1.74 | 1.75 |
| 6.0 | 1.03[-24] | 1.74[-26] | 1.75[-26] | 1.75[-26] | 1.68 | 1.69 | 1.69 |
| 8.0 | 3.92[-25] | 6.53[-27] | 6.56[-27] | 6.53[-27] | 1.66 | 1.67 | 1.66 |
| 10.0 | 1.85[-25] | 3.02[-27] | 3.04[-27] | 3.03[-27] | 1.64 | 1.65 | 1.64 |
| 12.0 | 9.96[-26] | 1.62[-27] | 1.63[-27] | 1.62[-27] | 1.63 | 1.64 | 1.63 |

takes a similar value to the photoionization. In principle, electrons ejected by the Compton scattering can be distinguished by measuring the energy distribution or observing the Compton scattered photons coincidentally. More detailed measurement in which the component of the Compton scattering is separated is desirable as well as theoretical study of double-ionization mechanism induced by the Compton scattering.

As we have seen that the double-ionization cross sections corresponding to each diagram depend generally upon the form of the dipole operator even at high-energy limit. Let us examine those differences explicitly in the following.

Since $V^{N-1}(\mathbf{r})$ is chosen to be a local potential, we have

$$d^V = [d^L, h]. \tag{16}$$

Using this relation in Eqs. (11)–(13), we obtain

$$T^V(\mathrm{TS1}) + \omega T^L(\mathrm{TS1}) = \sum_k \langle k_1 k_2|v|k1s\rangle\langle k|d^L|1s\rangle. \tag{17}$$

Similarly, we obtain the differences for other amplitudes,

$$T^V(\mathrm{SO}) + \omega T^L(\mathrm{SO}) = -\langle k_2 1s|v|1s1s\rangle\langle k_1|d^L|1s\rangle \tag{18}$$

and

$$T^V(\mathrm{GSC}) + \omega T^L(\mathrm{GSC}) = -\sum_k \langle k_1|d^L|k\rangle\langle k k_2|v|1s1s\rangle. \tag{19}$$

These differences, in particular in TS1 and GSC, are not small as we have seen in Figs. 2 and 3. In SO, the difference vanishes at high-$\omega$ limit because the right-hand side of Eq. (18) vanishes in the limit $k_1 \to \infty$ or $k_2 \to \infty$.

Provided that our lowest-order perturbation is a good approximation, the right-hand sides of Eqs. (17)–(19) when summed should cancel with each other to give a small difference in the "Total" cross section. In our present case, exact cancellation occurs as we show below. We begin by rewriting the right-hand side of Eq. (19) using the closure relation

$$\sum_{k\ (\neq 1s)} |k\rangle\langle k| = 1 - |1s\rangle\langle 1s| \tag{20}$$

to obtain

$$-\sum_k \langle k_1|d^L|k\rangle\langle k k_2|v|1s1s\rangle = -\langle k_1|d^L v_{k_2 1s}(\mathbf{r})|1s\rangle + \langle k_1|d^L|1s\rangle\langle 1s k_2|v|1s1s\rangle, \tag{21}$$

where

$$v_{k_2 1s}(\mathbf{r}) = \int \frac{\phi^*_{k_2}(\mathbf{r}')\phi_{1s}(\mathbf{r}')}{|\mathbf{r}-\mathbf{r}'|} d\mathbf{r}'. \tag{22}$$

The second term of Eq. (21) cancels the right-hand side of Eq. (18). The first term of Eq. (21) also cancels the right-hand side of Eq. (17) because

$$\sum_k \langle k_1 k_2|v|k1s\rangle\langle k|d^L|1s\rangle = \langle k_1|v_{k_2 1s} d^L|1s\rangle - \langle k_1 k_2|v|k1s\rangle\langle 1s|d^L|1s\rangle = \langle k_1|d^L v_{k_2 1s}|1s\rangle, \tag{23}$$

where use has been made of $[v_{k_2 1s}, d^L] = 0$ and $\langle 1s|d^L|1s\rangle = 0$. Hence all the *extra* terms appearing in Eqs. (17)–(19) sum to zero and we obtain

$$T^V(\mathrm{TS1}) + T^V(\mathrm{SO}) + T^V(\mathrm{GSC}) = -\omega\left\{T^L(\mathrm{TS1}) + T^L(\mathrm{SO}) + T^L(\mathrm{GSC})\right\}. \tag{24}$$

Thus there is no difference between the $L$ and $V$ form in our double-ionization cross sections.

For the $A$ form, we first note the relation

$$[d^V, h] = d^A + \frac{d}{dz} v_{1s1s} \equiv \tilde{d}^A, \tag{25}$$

where $v_{1s1s}$ is the static potential appearing in Eq. (9). Using this relation, we obtain expressions similar to Eqs. (17)–(19) above except that $d^L$ is replaced by $d^V$ and $d^V$ by $\tilde{d}^A$. However, since $d^V = \frac{d}{dz}$ does not commute with $v_{k_2 1s}$, we obtain

$$T^V(\mathrm{TS1}) + T^V(\mathrm{SO}) + T^V(\mathrm{GSC}) = -\frac{1}{\omega}\{\tilde{T}^A(\mathrm{TS1}) + \tilde{T}^A(\mathrm{SO}) + \tilde{T}^A(\mathrm{GSC}) - \langle k_1|[v_{k_2 1s}, d^V]|1s\rangle\}, \tag{26}$$

where

$$\tilde{T}^A = T^A + \Delta T^A \tag{27}$$

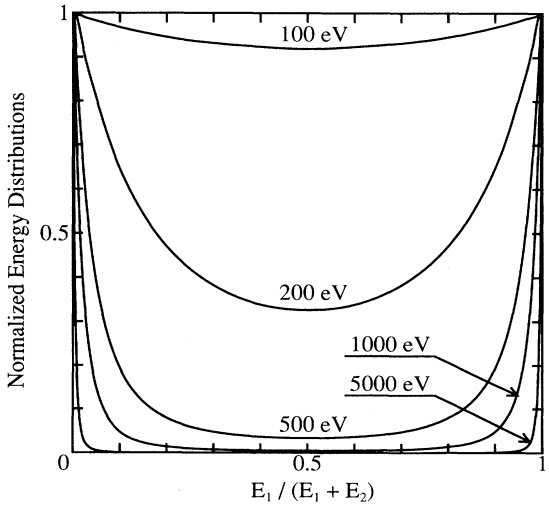


FIG. 6. The normalized energy distributions of ejected electrons for the double photoionization at photon energies of 100, 200, 500, 1000, and 5000 eV. $E_1$ and $E_2$ are the energies of two ejected electrons.

with $\Delta T^A$ obtained using $\frac{d}{dz}v_{1s1s}$ in place of $d^A$ in the defining equation of $T^A$. Thus we see that the difference between our cross sections in the $V$ and $A$ form is due in part to the noncommutativity of $v_{k_2 1s}$ and $d^V$, and due in part to the difference between $d^A$ and $\tilde{d}^A$.

The normalized energy distribution of the ejected electrons obtained from the sum Eq. (15) of three amplitudes are shown in Fig. 6. It justifies the common argument that at high energies one of the ejected electrons carries away most of the photon energy and the other electron leaves the target relatively slowly. The energy distribution calculated for each of the three mechanisms, however, depends upon the dipole formula and the above argument is not always valid for any one of these terms.

## IV. CONCLUSIONS

We have carried out calculations of the double photoionization of He at high energies using many-body perturbation theory in the lowest order. Double-ionization cross sections in the length and velocity forms of the dipole operator are identical because we have used a local potential to define the basis set of expansion. The cross section in the acceleration formula agrees well with other formulas at energies above 1 keV. Calculated ratios of the cross sections for double ionization to single ionization also agree very well with experiments at these energies. Three diagrams contribute to the lowest-order calculation of double ionization of He. They are called in the literature as the two-step-1, the shake off, and the ground-state-correlation mechanisms. However, the cross section for each of these individual mechanisms depends strongly on the form of the dipole operator as indicated by Dalgarno and Sadeghpour [4]. Therefore, they have no consistent meaning as mechanisms of the double ionization by photoabsorption unless we define the form of the dipole interaction.

## ACKNOWLEDGMENTS

We gratefully acknowledge discussion with M. Amusia, A. Dalgarno, M. Pindzola, and H. Sadeghpour. The present numerical computations were supported by the Institute of Physical and Chemical Research (RIKEN). This work was also supported in part by the Japanese Ministry of Education and in part by the Division of Chemical Sciences, Office of Basic Energy Science, Office of Energy Research, U.S. Department of Energy.

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