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11-9-2006

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Nondipole Effects in Double Photoionization of He

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Abstract. Lowest-order nondipole effects are studied in double photoionization (DPI) of the He atom. *Ab initio* parametrizations of the quadrupole transition amplitude for DPI from the $^1S_0$-state are presented in terms of the exact two-electron reduced matrix elements. Parametrizations for the dipole-quadrupole triply differential cross section (TDCS) and doubly differential cross section (DDCS) are presented in terms of polarization-independent amplitudes for the case of an elliptically polarized photon. Expressions for the DDCS in terms of the reduced two-electron matrix elements are also given. A general analysis of retardation-induced asymmetries of the TDCS including the circular dichroism effect at equal energy sharing is presented. Our numerical results exhibit a nondipole forward-backward asymmetry in the TDCS for DPI of He at an excess energy of 450 eV that is in qualitative agreement with existing experimental data.

Keywords: double ionization, nondipole effects, circular dichroism, helium, quadrupole amplitude
PACS: 32.80.Fb

GENERAL RESULTS FOR THE DPI QUADRUPOLE TRANSITION AMPLITUDE

We consider double photoionization (DPI) from the $^1S_0$ state $|0\rangle$ to the final two-electron singlet state $|\mathbf{p}_1\mathbf{p}_2\rangle$, with asymptotic electron momenta $\mathbf{p}_1$ and $\mathbf{p}_2$, in the nonrelativistic domain of photon energies taking into account lowest-order retardation corrections. Because neither orbital nor spin-dependent parts of the magnetic dipole interaction contribute to the transition amplitude A for nonrelativistic photon energies [1], the spin dependence of the two-electron wave functions is suppressed in our analysis. The dipole-quadrupole TDCS for DPI is:

$$\frac{d^3\sigma}{d\varepsilon_1 d\Omega_1 d\Omega_2} \equiv \sigma = \mathscr{A}|A|^2, \tag{1}$$

where $\mathscr{A} = 4\pi^2\alpha p_1 p_2/\omega$ is a normalization factor, and $\alpha = 1/137.036$. Atomic units are used throughout this paper. The amplitude A involving E1 and E2 components has the form (where $\mathbf{e}$ is the photon polarization vector and $\mathbf{k}$ is the photon wavevector),

$$A = A_d + A_q = \langle \mathbf{p}_1\mathbf{p}_2|(\mathbf{e}\cdot\mathbf{D}) + (\{\hat{\mathbf{k}}\otimes\mathbf{e}\}_2\cdot Q_2)|0\rangle, \tag{2}$$

for both velocity (V) and length (L) gauges of the electron-photon interaction. $\{\mathbf{a}\otimes\mathbf{b}\}_{2m}$ is the irreducible tensor product of rank 2. In the V-gauge, $\mathbf{D} \equiv \mathbf{D}^{(V)} = -i(\nabla_1 + \nabla_2)$ and $Q_{2m} \equiv Q_{2m}^{(V)} = \alpha\omega(\{\mathbf{r}_1\otimes\nabla_1\}_{2m} + \{\mathbf{r}_2\otimes\nabla_2\}_{2m})$. In the L-gauge, $\mathbf{D} \equiv \mathbf{D}^{(L)} = i\omega(\mathbf{r}_1 + \mathbf{r}_2)$ and $Q_{2m} \equiv Q_{2m}^{(L)} = -(1/2)\alpha\omega^2(\{\mathbf{r}_1\otimes\mathbf{r}_1\}_{2m} + \{\mathbf{r}_2\otimes\mathbf{r}_2\}_{2m})$. (See Ref. [2] for details.)

CP811 *Ionization, Correlation, and Polarization in Atomic Collisions*, edited by A. Lahmam-Bennani and B. Lohmann
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In the electric dipole approximation (EDA), the parametrization of the EDA amplitude A_d in terms of scalar products of the vectors $\mathbf{e}$, $\hat{\mathbf{p}}_1$, and $\hat{\mathbf{p}}_2$ is well-known [3, 4]:

$$A_d = f_1(\mathbf{e}\cdot\hat{\mathbf{p}}_1) + f_2(\mathbf{e}\cdot\hat{\mathbf{p}}_2). \tag{3}$$

In this equation, $f_1 \equiv f(p_1,p_2,\cos\theta)$ and $f_2 \equiv f(p_2,p_1,\cos\theta)$ [where $\theta_{12} \equiv \theta$ is the mutual ejection angle, $\cos\theta = (\hat{\mathbf{p}}_1\cdot\hat{\mathbf{p}}_2)$] are defined by a single function,

$$f(p,p',\cos\theta) = \sum_{l=1}^{\infty}(-1)^{l+1}\left[\sum_{l'=l\pm1} D_{ll'}(p,p')\right]P_l^{(1)}(\cos\theta), \tag{4}$$

where $P_l^{(n)}(x)$ is the n-th derivative of the Legendre polynomial $P_l(x)$, $P_l^{(n)}(x) = (d^n/dx^n)P_l(x)$. The energy-dependent coefficient $D_{ll'}(p,p')$ is given by $D_{ll'}(p,p') = [(2l+1)(2l'+1)\max(l,l')]^{-1/2}d_{ll'}(p,p')$, where $d_{ll'}(p,p') \equiv \langle pp';(ll')1\|\mathbf{D}\|0\rangle$ is the reduced matrix element of the operator $\mathbf{D}$ between the ${}^1S_0$-state and the P-wave component of the final state $|\mathbf{pp}'\rangle$, with photoelectron angular momenta l and $l' = l\pm1$. We use an expansion of $|\mathbf{pp}'\rangle$ in terms of modified bipolar harmonics $\mathscr{C}_{LM}^{ll'}(\hat{\mathbf{p}},\hat{\mathbf{p}}')$,

$$|\mathbf{pp}'\rangle = \sum_{ll'LM}\mathscr{C}_{LM}^{ll'\,*}(\hat{\mathbf{p}},\hat{\mathbf{p}}')|pp';(ll')LM\rangle. \tag{5}$$

Similarly to the derivations of Eqs. (3) and (4) in Ref. [3], to derive a parametrization of the quadrupole amplitude A_q we use the expansion (5) and the reduction formulae [3] for the rank-2 bipolar harmonics. Thus, A_q may be presented as follows [2, 5]:

$$A_q = g_1(\mathbf{e}\cdot\hat{\mathbf{p}}_1)(\hat{\mathbf{p}}_1\cdot\hat{\mathbf{k}}) + g_2(\mathbf{e}\cdot\hat{\mathbf{p}}_2)(\hat{\mathbf{p}}_2\cdot\hat{\mathbf{k}}) + g_s\left[(\mathbf{e}\cdot\hat{\mathbf{p}}_1)(\hat{\mathbf{p}}_2\cdot\hat{\mathbf{k}}) + (\mathbf{e}\cdot\hat{\mathbf{p}}_2)(\hat{\mathbf{p}}_1\cdot\hat{\mathbf{k}})\right], \tag{6}$$

where the generally complex parameters $g_{1,2}$ and g_s depend on p_1, p_2, and θ. The parameter g_s is symmetric in p_1 and p_2, $g_s \equiv g_s(p_1,p_2,\cos\theta) = g_s(p_2,p_1,\cos\theta)$, while g_1 and g_2 are expressed in terms of a single function, $g(p,p',\cos\theta)$, with $g_1 \equiv g(p_1,p_2,\cos\theta)$ and $g_2 \equiv g(p_2,p_1,\cos\theta)$. The explicit forms of the functions $g(p,p',\cos\theta)$ and $g_s(p,p',\cos\theta)$ are as follows:

$$\begin{aligned} g_s(p,p',\cos\theta) &= \sum_{l=1}^{\infty}(-1)^{l+1}\Bigg[\sum_{l'=l\pm2} Q_{ll'}(p,p')P_{\frac{l+l'}{2}}^{(2)}(\cos\theta) \\ &\quad + \sqrt{6}Q_{ll}(p,p')\left(P_{l+1}^{(2)}(\cos\theta) - \frac{2l+3}{2}P_l^{(1)}(\cos\theta)\right)\Bigg], \\ g(p,p',\cos\theta) &= \sum_{l=2}^{\infty}(-1)^{l}\left[\sum_{l'=l\pm2} Q_{ll'}(p,p') + \sqrt{6}Q_{ll}(p,p')\right]P_l^{(2)}(\cos\theta), \end{aligned} \tag{7}$$

where $Q_{ll'}(p,p') = [4(l+l'-2)!/(l+l'+3)!]^{1/2}\, q_{ll'}(p,p')$ and $q_{ll'}(p,p') \equiv \langle pp';(ll')2\|Q_2\|0\rangle$ is the reduced matrix element of the operator Q_{2m} between the initial ${}^1S_0$-state, $|0\rangle$, and the D-wave component of the two-electron continuum state $|\mathbf{pp}'\rangle$ with photoelectron angular momenta l and $l' = l, l\pm2$.

An alternative parametrization of the DPI amplitude A in terms of orthogonal vectors $\mathbf{p}_+ = (\hat{\mathbf{p}}_1 + \hat{\mathbf{p}}_2)/2$ and $\mathbf{p}_- = (\hat{\mathbf{p}}_1 - \hat{\mathbf{p}}_2)/2$ and the symmetrized ($f^{(\mathrm{g})} = f_1 + f_2$ and $g_\pm^{(\mathrm{g})} = g_1 + g_2 \pm 2g_s$) and antisymmerized ($f^{(\mathrm{u})} = f_1 - f_2$ and $g^{(\mathrm{u})} = g_1 - g_2$) amplitudes is rather obvious and can be easily obtained from Eqs. (3), (4), (6), and (7) [2]. The results in Eqs. (3), (4), (6) and (7) are general and do not depend upon the dynamical model used for calculation of the reduced matrix elements $d_{ll'}(p,p')$ and $q_{ll'}(p,p')$.

PARAMETRIZATIONS AND NUMERICAL ESTIMATES FOR THE DIPOLE-QUADRUPOLE TDCS

For the most general case of DPI by elliptically polarized light (described by the complex polarization vector $\mathbf{e}$ [$(\mathbf{e}\cdot\mathbf{e}^*) = 1$]), the TDCS in Eq. (1) (neglecting the small terms $\sim |A_q|^2$) has a model-independent parametrization similar to that for dipole DPI [3],

$$\sigma = \mathscr{A}\{c_1|\mathbf{e}\cdot\hat{\mathbf{p}}_1|^2 + c_2|\mathbf{e}\cdot\hat{\mathbf{p}}_2|^2 + \mathrm{Re}\,c_3\big[(1-\ell)\big((\hat{\mathbf{p}}_1\cdot\hat{\mathbf{p}}_2) - (\hat{\mathbf{k}}\cdot\hat{\mathbf{p}}_1)(\hat{\mathbf{k}}\cdot\hat{\mathbf{p}}_2)\big) + 2\ell(\hat{\boldsymbol{\varepsilon}}\cdot\hat{\mathbf{p}}_1)(\hat{\boldsymbol{\varepsilon}}\cdot\hat{\mathbf{p}}_2)\big] + \xi\,\mathrm{Im}\,c_3\,(\hat{\mathbf{k}}\cdot[\hat{\mathbf{p}}_1\times\hat{\mathbf{p}}_2])\}, \tag{8}$$

where, in contrast to the dipole case, the coefficients c_i depend upon $\mathbf{k}$:

$$\begin{aligned} c_1 &= |f_1|^2 + 2\mathrm{Re}\big[f_1 g_1^*(\hat{\mathbf{k}}\cdot\hat{\mathbf{p}}_1) + f_1 g_s^*(\hat{\mathbf{k}}\cdot\hat{\mathbf{p}}_2)\big],\\ c_2 &= |f_2|^2 + 2\mathrm{Re}\big[f_2 g_2^*(\hat{\mathbf{k}}\cdot\hat{\mathbf{p}}_2) + f_2 g_s^*(\hat{\mathbf{k}}\cdot\hat{\mathbf{p}}_1)\big],\\ c_3 &= f_1 f_2^* + (f_1 g_s^* + f_2^* g_1)(\hat{\mathbf{k}}\cdot\hat{\mathbf{p}}_1) + (f_2^* g_s + f_1 g_2^*)(\hat{\mathbf{k}}\cdot\hat{\mathbf{p}}_2). \end{aligned} \tag{9}$$

The parameters ℓ and ξ in Eq. (8) are the degrees of linear and circular polarization of an elliptically polarized photon; $\ell = \mathbf{e}^2 = \sqrt{1-\xi^2}$, $\xi \equiv i(\hat{\mathbf{k}}\cdot[\mathbf{e}\times\mathbf{e}^*])$, and the unit vector $\hat{\boldsymbol{\varepsilon}}$ is directed along the major axis of the polarization ellipse. The photon polarization dependence of the dipole-quadrupole TDCS is thus determined by *four* real $\hat{\mathbf{k}}$-dependent parameters, c_1, c_2, $\mathrm{Re}\,c_3$, and $\mathrm{Im}\,c_3$. These parameters may be determined from four measurements with different polarizations of the photon beam, e.g., two experiments with linearly polarized photons and two experiments with circularly polarized photons. For circularly polarized photons ($\ell = 0$, $\xi = \pm 1$) Eq. (8) simplifies,

$$\sigma = \frac{\mathscr{A}}{2}\Big(c_1[\hat{\mathbf{k}}\times\hat{\mathbf{p}}_1]^2 + c_2[\hat{\mathbf{k}}\times\hat{\mathbf{p}}_2]^2 + \mathrm{Re}\,c_3\,\big([\hat{\mathbf{k}}\times\hat{\mathbf{p}}_1]\cdot[\hat{\mathbf{k}}\times\hat{\mathbf{p}}_2]\big) + 2\xi\,\mathrm{Im}\,c_3\,(\hat{\mathbf{k}}\cdot[\hat{\mathbf{p}}_1\times\hat{\mathbf{p}}_2])\Big). \tag{10}$$

The TDCSs in Eqs. (8) and (10) both contain a term that is proportional to ξ, which is responsible for the circular dichroism (CD) effect. This effect is usually characterized by the absolute CD parameter, $\Delta_{cd} \equiv \sigma(\xi = +1) - \sigma(\xi = -1)$:

$$\Delta_{cd} = 2\mathscr{A}\,\mathrm{Im}\,c_3\,(\hat{\mathbf{k}}\cdot[\hat{\mathbf{p}}_1\times\hat{\mathbf{p}}_2]). \tag{11}$$

The term Δ_{cd} involves both the dipole-dipole and dipole-quadrupole contributions,

$$\Delta_{cd} = \Delta_{cd}^{(dip)} + \Delta_{cd}^{(quadr)}, \quad \text{where } \Delta_{cd}^{(dip)} = 2\mathscr{A}\,\mathrm{Im}(f_1 f_2^*)\,(\hat{\mathbf{k}}\cdot[\hat{\mathbf{p}}_1\times\hat{\mathbf{p}}_2]), \text{ and}$$
$$\Delta_{cd}^{(quadr)} = 2\mathscr{A}\,\mathrm{Im}[(f_1 g_s^* + f_2^* g_1)(\hat{\mathbf{k}}\cdot\hat{\mathbf{p}}_1) - (f_2 g_s^* + f_1^* g_2)(\hat{\mathbf{k}}\cdot\hat{\mathbf{p}}_2)]\,(\hat{\mathbf{k}}\cdot[\hat{\mathbf{p}}_1\times\hat{\mathbf{p}}_2]). \tag{12}$$

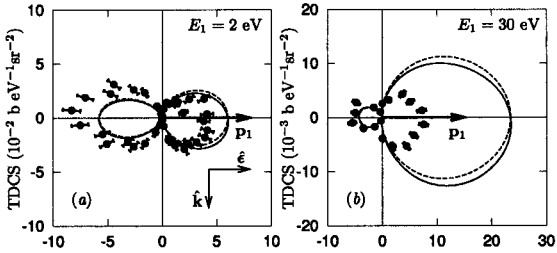

FIGURE 1. Comparison of our LOPT results for the TDCS at an excess energy of 450 eV for the case of linear polarization with the normalized experimental data of Ref. [10]. The directions of the photon wavevector $\hat{\mathbf{k}}$ and polarization $\hat{\boldsymbol{\varepsilon}}$ are as shown in (*a*); the electron having energy E_1 is ejected along ε. Full curves: dipole-quadrupole results; dashed curves: EDA results.

Within the EDA, the CD effect is described by the term $\Delta_{cd}^{(dip)}$ [3, 6], which vanishes at equal energy sharing (because for $p_1 = p_2$ one has $f_1 = f_2$). However, the quadrupole term, $\Delta_{cd}^{(quadr)}$, produces a non-zero CD effect even at equal energy sharing. The dichroic term $\mathrm{Im}\, c_3(\hat{\mathbf{k}} \cdot [\hat{\mathbf{p}}_1 \times \hat{\mathbf{p}}_2])$ in Eqs. (8) and (10) results also in an unusual feature of the TDCS, a different symmetry of the TDCS with respect to two transformations: (i) $\mathbf{k} \to -\mathbf{k}$ (i.e., the inversion of the photon beam direction) and (ii) $(\theta_1, \theta_2) \to (\pi - \theta_1, \pi - \theta_2)$ (i.e., the reflection of the photoelectron pair in the polarization plane) [2].

The expression for the TDCS in terms of the symmetrized amplitudes has a form identical to those in Eqs. (8)-(10) provided the following substitutions are made: $\{\hat{\mathbf{p}}_1, \hat{\mathbf{p}}_2, f_1, f_2, g_1, g_2, g_s\} \to \{\mathbf{p}_+, \mathbf{p}_-, f^{(\mathrm{g})}, f^{(\mathrm{u})}, g_+^{(\mathrm{g})}, g_-^{(\mathrm{g})}, g^{(\mathrm{u})}\}$. This parametrization leads to a simpler form of the TDCS for equal energy sharing [7]:

$$\sigma^{(eq)} = \mathscr{A}\Big\{ \Big[|f^{(\mathrm{g})}|^2 + 2\mathrm{Re}\{f^{(\mathrm{g})*} g_+^{(\mathrm{g})}\}(\hat{\mathbf{k}} \cdot \mathbf{p}_+) \Big] |\mathbf{e} \cdot \mathbf{p}_+|^2 + \mathrm{Re}\{f^{(\mathrm{g})*} g_-^{(\mathrm{g})}\}(\hat{\mathbf{k}} \cdot \mathbf{p}_-)$$
$$\times \Big[2l(\hat{\boldsymbol{\varepsilon}} \cdot \mathbf{p}_+)(\hat{\boldsymbol{\varepsilon}} \cdot \mathbf{p}_-) + (l-1)(\hat{\mathbf{k}} \cdot \mathbf{p}_+)(\hat{\mathbf{k}} \cdot \mathbf{p}_-) \Big] \Big\} + (\xi/2)\Delta_{cd}^{(eq)},$$
$$\Delta_{cd}^{(eq)} \equiv \Delta_{cd}^{(quadr)}|_{p_1=p_2} = 2\mathscr{A}\, \mathrm{Im}\{f^{(\mathrm{g})*} g_-^{(\mathrm{g})}\}\, (\hat{\mathbf{k}} \cdot [\mathbf{p}_- \times \mathbf{p}_+])\, (\hat{\mathbf{k}} \cdot \mathbf{p}_-). \tag{13}$$

Numerical estimates of nondipole effects in TDCSs calculated within lowest-order perturbation theory (LOPT) in the interelectron interaction [8, 9] have been reported in Refs. [2, 5, 7] for excess energies ranging from tens to hundreds of eV. In Fig. 1 we present an example of such predictions for an excess energy of 450 eV that are compared to the experimental data of Ref. [10]. Despite the deviation of our LOPT predictions within EDA from the experimental data, one sees that both the experimental data and our nondipole results exhibit a noticeable forward-backward asymmetry as compared to our EDA results: the angular distributions of the fast electron are shifted along the direction of the vector $\mathbf{k}$, especially in the angular ranges $0 < \theta_2 < \pi/2$ and $3\pi/2 < \theta_2 < 2\pi$. The relative magnitudes of such an asymmetry at particular angles θ_2, measured by the quantity $R = [\sigma(2\pi - \theta_2) - \sigma(\theta_2)]/\sigma(2\pi - \theta_2)$, appear to be in reasonable agreement with the experimental data. Indeed, for $\theta_1 = 0.174$ (in radians), one has the theoretical value $R^{th} = 0.051$ vs. the experimentally measured value $R^{exp} = 0.049$; for $\theta_1 = 0.516$,

$R^{th} = 0.143$ vs. $R^{exp} = 0.117$; for $\theta_1 = 0.868$, $R^{th} = 0.212$ vs. $R^{exp} = 0.181$; and for $\theta_1 = 1.216$, $R^{th} = 0.229$ vs. $R^{exp} = 0.398$.

DIPOLE-QUADRUPOLE DDCS

The DDCS for DPI from an initial ^1S state can be written in a form that is identical to that of the angle-differential cross section for single photoionization,

$$\frac{d^2\sigma}{d\Omega_{\mathbf{p}_1} dE_1} = \frac{\sigma_0}{4\pi}\Big\{1 + \beta P_2(|\mathbf{e}\cdot\hat{\mathbf{p}}_1|) + \big[\delta + \gamma|\mathbf{e}\cdot\hat{\mathbf{p}}_1|^2\big](\hat{\mathbf{k}}\cdot\hat{\mathbf{p}}_1)\Big\}, \tag{14}$$

where, however, the singly-differential cross section (SDCS) σ_0, the dipole asymmetry parameter β, and the nondipole asymmetry parameters γ and δ depend upon both the photon frequency ω *and* the energy sharing (i.e., upon the energy of one of the photoelectrons). For linear polarization ($\mathbf{e} = \mathbf{e}^* \equiv \hat{\varepsilon}$), $P_2(|\mathbf{e}\cdot\hat{\mathbf{p}}|) = P_2(\cos\alpha)$ is a Legendre polynomial, where α is the angle between the vectors $\hat{\mathbf{p}}$ and $\hat{\varepsilon}$; for circular polarization, $P_2(|\mathbf{e}\cdot\hat{\mathbf{p}}|) = (-1/2)P_2(\cos\phi)$, where ϕ is the angle between the vectors $\hat{\mathbf{p}}$ and $\hat{\mathbf{k}}$.

We have derived *ab initio* representations for σ_0, β, γ, and δ at two different levels of detail. The first one is in terms of integrals of the polarization-invariant amplitudes,

$$\sigma_0 = \mathscr{A}\frac{8\pi^2}{3}\int_{-1}^{1}\big[|f_1|^2 + |f_2|^2 + 2\mathrm{Re}(f_1 f_2^*)x\big]dx,$$

$$\beta = \mathscr{A}\frac{(4\pi)^2}{3\sigma_0}\int_{-1}^{1}\big[|f_1|^2 + |f_2|^2 P_2(x) + 2\mathrm{Re}(f_1 f_2^*)x\big]dx = 2 - \mathscr{A}\frac{8\pi^2}{\sigma_0}\int_{-1}^{1}|f_2|^2(1-x^2)dx,$$

$$\gamma = \mathscr{A}\frac{(4\pi)^2}{\sigma_0}\int_{-1}^{1}\mathrm{Re}\left\{f_1^*\big[g_1 + g_2P_2(x) + 2g_s x\big] + f_2^*\big[g_1 x + g_2 P_3(x) + 2g_s P_2(x)\big]\right\}dx,$$

$$\delta = \mathscr{A}\frac{8\pi^2}{\sigma_0}\int_{-1}^{1}\mathrm{Re}\big[f_2^*(g_s + g_2 x)\big](1-x^2)dx, \tag{15}$$

where the amplitudes $f_{1,2}(x)$, $g_{1,2}(x)$, and $g_s(x)$ depend upon $x \equiv \cos\theta$. The second parametrization is in terms of an infinite sum over the reduced matrix elements,

$$\sigma_0 = (4\pi)^2\frac{\mathscr{A}}{3}\sum_{l_2=0}^{\infty}\sum_{l_1=l_2\pm1}\frac{|d_{l_1 l_2}|^2}{(2l_1+1)(2l_2+1)},$$

$$\beta = \frac{(4\pi)^2}{\sigma_0}\mathscr{A}\sqrt{\frac{2}{3}}\sum_{l_2=0}^{\infty}\sum_{l_1=l_2\pm1}\sum_{l_1'=l_2\pm1}\frac{(-1)^{l_2}}{2l_2+1}C^{20}_{l_1 0 l_1' 0}\begin{Bmatrix} 1 & l_1' & l_2 \\ l_1 & 1 & 2 \end{Bmatrix} d_{l_1 l_2} d^*_{l_1' l_2},$$

$$\gamma = -\frac{(4\pi)^2}{\sigma_0}\sqrt{10}\mathscr{A}\sum_{l_2=2}^{\infty}\sum_{l_1=l_2\pm1}\sum_{l_1'=l_2,l_2\pm2}\frac{(-1)^{l_2}}{2l_2+1}C^{30}_{l_1 0 l_1' 0}\begin{Bmatrix} 2 & l_1' & l_2 \\ l_1 & 1 & 3 \end{Bmatrix}\mathrm{Re}(d_{l_1 l_2} q^*_{l_1' l_2}),$$

$$\delta = \frac{(4\pi)^2}{\sigma_0}\frac{\mathscr{A}}{\sqrt{5}}\sum_{l_2=2}^{\infty}\sum_{l_1=l_2\pm1}\sum_{l_1'=l_2,l_2\pm2}\frac{(-1)^{l_2}}{2l_2+1}\mathrm{Re}(d_{l_1l_2}q^*_{l_1'l_2})$$
$$\times\left(\sqrt{3}C^{10}_{l_10l_10}\left\{\begin{array}{ccc} 2 & l_1' & l_2 \\ l_1 & 1 & 1\end{array}\right\}+\sqrt{2}C^{30}_{l_10l_10}\left\{\begin{array}{ccc} 2 & l_1' & l_2 \\ l_1 & 1 & 3\end{array}\right\}\right). \tag{16}$$

The dipole-quadrupole terms, which are of the order of ω/c, do not appear in the SDCS given by the parameter σ_0 in Eqs. (15) and (16), i.e., the lowest-order nondipole corrections that contribute to the SDCS are the quadrupole-quadrupole and dipole-octupole terms, which are of the order $(\omega/c)^2$ and are thus not accounted for here. Also, the parameter δ is generally non-zero. This is in contrast to SPI, for which δ^{SPI} vanishes for ionization from atomic s subshells [11].

We have calculated the SDCS, σ_0, and the asymmetry parameters β, γ, and δ by the LOPT approach for an excess energy of 450 eV for two energy sharings used in the experiment of Ref. [10]. For the angular distribution of the electron having energy $E_1 = 448$ eV, we find that $\sigma_0 = 3.06$ b/eV, $\beta = 1.93$, $\gamma = 0.51$, and $\delta = 0.0043$, as compared to the CCC results $\sigma_0 = 2.54$ b/eV, $\beta = 1.92$ [10]. For the angular distribution of the electron having energy $E_1 = 420$ eV, we find that $\sigma_0 = 1.19$ b/eV, $\beta = 1.83$, $\gamma = 0.46$, and $\delta = 0.011$, as compared to the CCC results, $\sigma_0 = 0.73$ b/eV, $\beta = 1.78$ [10].

Comparisons of the LOPT predictions for dipole-quadrupole TDCSs, as well as for the parameters γ and δ, with those calculated by a more elaborate approach would be of great interest and are now in progress [12].

ACKNOWLEDGMENTS

This work was supported in part by the U.S. Department of Energy, Office of Science, Division of Chemical Sciences, Geosciences, and Biosciences, under grant DE-FG03-96ER14646, by RFBR Grant 04-02-16350, and by the joint Grant VZ-010-0 of the CRDF and the RF Ministry of Education and Sciences (NLM and AVM).

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