Theory of laser-induced electron transport through molecular gases

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Conditions are found for the growth of a current in an electron swarm in gaseous media in the presence of intense, monochromatic radiation. For transport to occur by free-free absorption, it is necessary for the medium to have left- or right-handed chirality (as in a sample of chiral molecules), such that the radiative-dipole rate of absorption by the swarm is unequal for electron velocities $\vec{v}$ and $-\vec{v}$ relative to the photon direction.

I. INTRODUCTION

In a previous paper,[1] a study was made of the effect of free-free absorption and stimulated emission of intense, monochromatic radiation on the transport of electrons through gaseous Ar. Swarm experiments measure drift velocity W (ratio of a drift current to the electron number density) in the presence of a uniform dc-electric field of strength E,

$$W=\frac{1}{3}\left(\frac{2}{m}\right)^{1/2}\int_0^\infty d\epsilon\,\epsilon f_1(\epsilon)\Big/\int_0^\infty d\epsilon\,\epsilon^{1/2}f_0(\epsilon), \quad (1a)$$

$$f_1(\epsilon)=-\left(\frac{E}{N}\right)\frac{e}{Q_m(\epsilon)}\,\frac{df_0(\epsilon)}{d\epsilon}, \quad (1b)$$

where $f_0(\epsilon)$ [see Eq. (3) of Ref. 1, for example] and $f_1(\epsilon)$ are the monopolar and dipolar components of the electron distribution function; ϵ, e, and m are the electron energy, charge, and mass; N is the neutral number density, and $Q_m(\epsilon)$ is the electron-scattering momentum-transfer cross section. The distribution function has been expanded,[2]

$$f(\epsilon,\hat{\upsilon})\simeq f_0(\epsilon)+f_1(\epsilon)\cos\theta, \quad (2)$$

where θ is the angle between the electron and field directions along $\hat{\upsilon}$ and $\vec{E}$, respectively. The components $f_0(\epsilon)$ and $f_1(\epsilon)$ are coupled by the $\vec{E}$ field, and this coupling is responsible for the growth of $f_1(\epsilon)$. Recent work[3] has shown that Eq. (2) may not be a converged series under certain circumstances; thus care should be exercised in calculations. In Ref. 1 it was argued that free-free radiative processes affect $f_0(\epsilon)$ strongly and affect $f_1(\epsilon)$ through its coupling to the derivative of $f_0(\epsilon)$ [Eq. (1b)]. For the laser pulse characteristics considered, however, the component of $f_1(\epsilon)$ contributed by the radiation field is negligible. Thus W is significant only if the $\vec{E}$ field is present.

In the present paper we examine physical conditions under which the radiation field can couple $f_0(\epsilon)$ and $f_1(\epsilon)$. In this case the $\vec{E}$ field can be quenched, and a drift velocity will be supported by the radiation alone. We refer to a drift current whose physical origin is a dipolar $\cos\theta$ component[4] in the free-free absorption and stimulated-emission rates. These rates occur in driver terms in the Boltzmann equation [see Eqs. (8) of Ref. 1 for the uncoupled equation for $f_0(\epsilon)$], and $\cos\theta$ components in the rates cause $f_0(\epsilon)$ and $f_1(\epsilon)$ to be coupled. This effect is different from free-space acceleration produced by radiation-field gradients[5] and from the coupling of the time-dependent components $f_0(\epsilon,t)$ and $f_1(\epsilon, t)$ by an ac-electric field oscillating as $\vec{E}\cos\omega t$ studied by Holstein[2] and found[1] to be negligible for the pulse widths considered. Thus to our knowledge the coupling of $f_0(\epsilon)$ and $f_1(\epsilon)$ and, as a consequence, the growth of a drift current induced by photoabsorption rates which are not invariant under space inversion[4] ($\vec{\upsilon}\rightarrow-\vec{\upsilon}$) has not been previously studied.

II. THEORY

The radiative driving terms in the Boltzmann equation (BE) depend on the rates in s^{-1} for free-free absorption and stimulated emission. One-photon rates are assumed to be dominant.[6] These rates depend on the directions of incidence along $\vec{\upsilon}'$ (from the solid angle Ω') and scattering along $\vec{\upsilon}$ (into the solid angle Ω) in a laboratory frame (Fig. 1). The BE driver contains appropriate combinations of these rates (which express electron density growth and loss at ϵ), integrated over the solid angle of incidence Ω'. The density-growth ("in-scattering") terms depend on the distribution function [Eq. (2)] at $\vec{\upsilon}'$. Thus the general form of the radiative collisional (rc) driver in the BE equation is

$$\left(\frac{\partial f}{\partial t}\right)_{\rm rc}=\sum_{j=1}^{2}\int d\Omega'[R_j^{(i)}(\epsilon_j,\epsilon,\hat{\upsilon},\hat{\upsilon}')f(\epsilon_j,\hat{\upsilon}')-R_j^{(0)}(\epsilon,\epsilon_j,\hat{\upsilon},\hat{\upsilon}')f(\epsilon,\hat{\upsilon})], \quad (3)$$

where $R_1^{(i)}$, $R_1^{(0)}=R_E$, R_A for $\epsilon_1=\epsilon+\hbar\omega$ and $R_2^{(i)}$, $R_2^{(0)}=R_A$, R_E for $\epsilon_2=\epsilon-\hbar\omega$, where $\hbar\omega$ is the photon energy and R_A, R_E are the absorption, stimulated-emission rates[1] in s^{-1}. When the distribution function f of Eq. (3) is expanded as in Eq. (2) and only the first term retained, the result is the driver studied in Ref. 1 (and references therein) for the effect of radiation on f_0. Note that the absorption and stimulated emission rates in Eq. (3) depend on both $\hat{\upsilon}'$ *and* $\hat{\upsilon}$, as shown in Fig. 1. Both incident and scattered electron directions relative to the photon direction along $\hat{k}_p$ are needed to define a free-free rate. Ordinary field-free collisional terms in the BE equation[2] depend on $\hat{\upsilon}\cdot\hat{\upsilon}'$ only (Fig. 1). In the present work we are concerned with f_0 and f_1 [Eq. (2)] and thus with terms in the rates of Eq. (3) which are *nonisotropic* in $\hat{\upsilon}$. Specifically we are concerned with finding conditions for which these rates contain a $\cos\theta$ (dipolar) term [see Eq. (2) and Fig. 1, where now the photon beam and not the $\vec{E}$ field is considered to be along z]. This dipolar term in the free-free rates would couple opposite-parity components of the distribution function (the leading members of which are f_0 and f_1 presumably). Thus, depending on the sign of the dipolar term, an electron drift would be generated in the direction of, or opposite to, the photon direction (Fig. 1). In other words, the free-free dipolar term is intended to replace the dipolar $\vec{E}$-field driver $-a\partial f/\partial \upsilon_z$ (where a is the electron acceleration) in ordinary transport.

In Eq. (3) the two $j=1$ terms account, respectively, for density growth at ϵ by stimulated emission from $\epsilon+\hbar\omega$ to ϵ and density loss at ϵ by absorption from ϵ to $\epsilon+\omega$. The two $j=2$ terms account, respectively, for density growth at ϵ by absorption from $\epsilon-\hbar\omega$ to ϵ and density loss by stimulated emission from ϵ to $\epsilon-\hbar\omega$. The two rates are related by the detailed-balance relationship

$$R_E(\epsilon,\epsilon',\hat{\upsilon},\hat{\upsilon}') = R_A(\epsilon',\epsilon,\hat{\upsilon},\hat{\upsilon}')(1-\hbar\omega/\epsilon)^{1/2}, \quad (4a)$$

$$\epsilon = \epsilon' + \hbar\omega . \quad (4b)$$

The uncoupled equation for $f_0(\epsilon)$ and the rate R_A integrated over Ω and Ω' have received detailed study.[1]

Analysis shows that

$$\int d\Omega' R_A = 4\pi N F[b_0(\epsilon',\epsilon) + b_2(\epsilon',\epsilon)P_2(\cos\theta)], \quad (5)$$

for nonchiral scatterers, where F is the flux due to an assumed constant-amplitude radiation field.[7] For example, $4\pi NFb_0(\epsilon',\epsilon)$ is given by Eq. (11a) or Eq. (12) of Ref. 1. This form emphasizes that free-free rates, integrated over all incident-electron directions, have the same angular dependence for electrons emerging from the field of the scatterer as bound-free rates have for electrons ejected from a nonoriented absorber. In Eq. (5) and what follows, analysis is limited to terms in Eq. (3) for which only the f_0 component of f is retained. Our interest is in terms coupling f_0 to f_s for $s>0$. The second term in Eq. (5) couples f_2 to f_0. Below and in the Appendix, the term is derived which couples f_1 to f_0. [In calculations, however, all components f_s would be retained in Eq. (3) consistent with terms retained in expansions[3] for f.]

For molecular targets it is necessary to average the rates over all molecular orientations[8] (see the Appendix). It is well known[9,10] that the form given by Eq. (5) still obtains and in fact is a statement for nonoriented molecules of Yang's theorem,[11] originally stated for radiative-dipole angular distributions for nonoriented nuclei. In a previous paper[4] it was shown that nonoriented chiral molecules provide an exception to this theorem by introducing a term linear in the cosine of the ejection angle into the angular distribution. In the present paper, it is shown (see the Appendix) that molecular chirality causes this same linear cosine dependence to occur in free-free radiative-dipole rates which have been integrated over incident-electron directions. This rate has the form

$$\int d\Omega' R_A = 4\pi NF[b_0(\epsilon',\epsilon) \pm b_1(\epsilon',\epsilon)P_1(\cos\theta) + b_2(\epsilon',\epsilon)P_2(\cos\theta)], \quad (6)$$

where the parity-violating component $b_1(\epsilon',\epsilon)$ exists only if the photon is left (+) or right (−) circularly polarized[4] (Fig. 1). On physical grounds it is not surprising that Ω'-integrated free-free rates have the same dependence on $P_L(\cos\theta)$ as bound-free rates in which the initial state is spatially unoriented.[4,9-11]

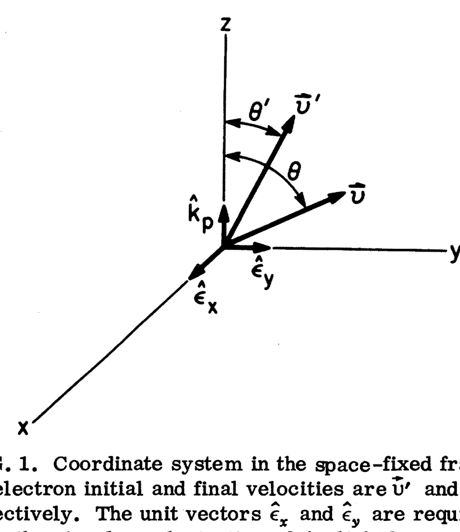

FIG. 1. Coordinate system in the space-fixed frame. The electron initial and final velocities are $\hat{\upsilon}'$ and $\hat{\upsilon}$, respectively. The unit vectors $\hat{\epsilon}_x$ and $\hat{\epsilon}_y$ are required to define the circular polarization of the light beam, and the unit vector $\hat{k}_p$ is in the direction of the light beam.

In the absence of the $\vec{E}$ field, which causes $f_1(\epsilon)$ to grow by coupling it to $f_0(\epsilon)$, the only mechanism for the growth of $f_s(\epsilon)$ for $s>0$ is its coupling to $f_0(\epsilon)$ according to Eq. (3). This coupling is determined by the series in $P_L(\cos\theta)$ given in the most general form by Eq. (6). Thus we note (quite generally for radiative-dipole processes) that gaseous media separate into two categories, nonchiral, for which $f_0(\epsilon)$ and $f_2(\epsilon)$ are coupled, and chiral, for which, in addition to the coupling of $f_0(\epsilon)$ and $f_2(\epsilon)$, $f_0(\epsilon)$ and $f_1(\epsilon)$ are coupled. In the first category, in fact, all $\Delta s=\pm 2$ (same parity) $f_s(\epsilon)$ components are coupled, and in the second category, all $\Delta s=\pm 2$ and $\Delta s=\pm 1$ (all parity) $f_s(\epsilon)$ components are coupled. Note, however, that the growth of electron density in any $s>0$ $f_s(\epsilon)$ component originates with its coupling to $f_0(\epsilon)$. Thus in the first category only even-parity components exist, while in the second category even- and odd-parity components exist. A drift current [see Eqs. (1)] is generated only for indefinite-parity electron distributions (as in the second category); thus, to leading order in the electron-radiation field interaction, this requires the existence of the parity-violating term in the Ω'-integrated absorption rate.

III. MODEL CALCULATION

We adopt a simplified model which may be useful to obtain an order of magnitude estimate of the drift velocity due to the parity-violating (pv) term in Eq. (6). The photon energy is assumed to be large (1.17 eV for an Nd^{+3} glass laser) compared with a characteristic vibrational spacing but small compared with a characteristic electronic spacing in the molecule, such that there is no photoabsorption by the scatterer. The average electron energy is also assumed to be between the two extremes[8] of molecular vibrational and electronic excitation, but it is assumed to be large compared with the photon energy ($\epsilon \gg \hbar\omega$). The latter criterion makes it possible[1] to simplify Eq. (3) by expanding the rates and distribution functions in the Taylor series about ϵ. On using the leading term of Eq. (4) there is cancellation of terms to order $(\hbar\omega)$.[2] It is further assumed that Eq. (2) is valid and that the equations for $f_0(\epsilon)$ and $f_1(\epsilon)$, which are coupled by the pv term in Eq. (6), can be solved using an $f_0(\epsilon)$ from the uncoupled equation. Use of approximate forms of Eq. (3) and of $4\pi NFb_0(\epsilon',\epsilon)$ [Eq. (6)], described below, leads to an uncoupled equation for $f_0(\epsilon)$ whose solution is available in analytic form [Eqs. (4) and (7b) of Ref. 1 and below]. Although not valid when $\epsilon\simeq\hbar\omega$, this solution is shown (Fig. 4 of Ref. 1) to be a qualitative average of the oscillatory $f_0(\epsilon)$ obtained from the numerical solution of Eq. (8a), Ref. 1; thus when $\epsilon\gg\hbar\omega$, it is expected to be quite reliable. The pv term coupling $f_1(\epsilon)$ and $f_0(\epsilon)$ is then evaluated using this approximate $f_0(\epsilon)$.

The truncation of the Legendre-polynomial series for f after two terms [Eq. (2)] is likely to be valid when the electron scattering is predominantly elastic.[3] The neglect of vibrationally inelastic scattering places severe limitations on the validity of the model, first because these cross sections can be appreciable at energies well beyond the energies at which they reach maximum values, and second because the BE is not local in the energy.

The steady-state[2,3] equation for $f_0(\epsilon)$ is

$$k_BT\,\frac{df_0(\epsilon)}{d\epsilon}+D_L=-f_0(\epsilon), \tag{7}$$

where D_L (below) is the laser driver. Vibrationally inelastic scattering contributes terms[12] of the form

$$D_V=\left(2\,\frac{m}{M}\,\epsilon^2Q_m(\epsilon)\right)^{-1}\int_{\epsilon}^{\epsilon+\Delta\epsilon}d\epsilon'\,\epsilon'Q_h(\epsilon')f_0(\epsilon'), \tag{8}$$

where $Q_h(\epsilon)$ is an inelastic cross section for the vibrational transition $\Delta\epsilon$ and M is the molecular mass. If $Q_h(\epsilon)$ and $f_0(\epsilon)$ are reasonably smooth in the interval from ϵ to $\epsilon+\Delta\epsilon$, then, for $\epsilon\gg\Delta\epsilon$,

$$D_V\simeq\frac{\Delta\epsilon}{\epsilon}\,\frac{Q_h(\epsilon)f_0(\epsilon)}{(m/M)Q_m(\epsilon)}\,. \tag{9}$$

For $\epsilon\gg\Delta\epsilon$, it is expected that $(m/M)Q_m(\epsilon)\simeq Q_h(\epsilon)$, i.e., that the rates of translational and vibrational energy loss are comparable. At $\epsilon\simeq\Delta\epsilon$ the latter is likely to be dominant. Thus for D_V to be dropped $\Delta\epsilon/\epsilon\ll D_L$. If a Ramsauer minimum occurs in $Q_m(\epsilon)$, the present model would break down,[3] however, such minima are not likely to occur at the energies considered here.

The expanded form[1] of the laser driver is

$$D_L\simeq\left(2\frac{m}{M}N\upsilon Q_m(\epsilon)\epsilon\right)^{-1}4\pi NFb_0(\epsilon)\,(\hbar\,\omega)^2\,\frac{df_0(\epsilon)}{d\epsilon}\,. \tag{10}$$

Note that D_L contains the ratio of an absorption rate $NF4\pi b_0(\epsilon)$ to the translational energy loss rate $(m/M)\,N\upsilon Q_m(\epsilon)$. For $\epsilon\gg\hbar\omega$, such that $\epsilon'\simeq\epsilon$ [Eq. (4b)], the absorption rate of Eq. (6) has been expressed[1] in terms of $Q_m(\epsilon)$,

$$4\pi NFb_0(\epsilon',\epsilon)\simeq\frac{4}{3}\,\epsilon_f\,\frac{N\upsilon Q_m(\epsilon)\epsilon}{(\hbar\,\omega)^2}=4\pi NFb_0(\epsilon)\,, \tag{11a}$$

$$\epsilon_f=\frac{1}{4}\,\frac{(Ee)^2}{m\,\omega^2}\,, \tag{11b}$$

where ϵ_f is just the classical mean energy of an electron oscillating in a force $Ee\cos\omega t$. Note that D_L reduces to $\epsilon_L\, df_0(\epsilon)/d\epsilon$, such that a constant laser "heating" energy $\epsilon_L = (2M/3m)\epsilon_f$ can be added to the thermal energy k_BT in Eq. (7).

The equation coupling $f_1(\epsilon)$ to $f_0(\epsilon)$ is

$$f_1(\epsilon) = -[N\upsilon Q_m(\epsilon)]^{-1} 4\pi NF(\hbar\omega)^2 \frac{db_1(\epsilon)}{d\epsilon}\frac{df_0(\epsilon)}{d\epsilon}, \tag{12}$$

where the expanded form of Eq. (3) has been used. Based on Eqs. (11) we use the *ansatz*,

$$4\pi NFb_1(\epsilon',\epsilon) \simeq \frac{4}{3}\epsilon_f \frac{N\upsilon Q_1(\epsilon)\epsilon}{(\hbar\omega)^2} = 4\pi NFb_1(\epsilon), \tag{13}$$

where $Q_1(\epsilon)$ is a cross section whose existence depends on the chirality of the scatterer (see the Appendix). For the relationship between the free-free radiative-dipole matrix element [Eq. (A1), see the Appendix] and the elastic-scattering amplitude, on which Eqs. (11) and (13) are based, see Eq. (13) and Appendix 1 of Ref. 1.

If we assume that $Q_1(\epsilon)$ is reasonably smooth with energy, then the derivative of $b_1(\epsilon)$ depends on $\frac{3}{2}\epsilon^{1/2}$. At power levels such that the thermal energy can be ignored in Eq. (7),

$$f_1(\epsilon) \simeq 3\frac{m}{M}\frac{Q_1(\epsilon)}{Q_m(\epsilon)} e^{-\epsilon/\epsilon_L}. \tag{14}$$

From Eq. (1a),

$$W \simeq \frac{2}{\sqrt{\pi}}\upsilon_L \frac{m}{M}\langle Q_1/Q_m\rangle, \tag{15a}$$

$$\upsilon_L = (2\epsilon_L/m)^{1/2}, \tag{15b}$$

where the brackets on the cross-section ratio indicate a mean value of $Q_1(\epsilon)/Q_m(\epsilon)$ [Eq. (14)] used in the integrand belonging to the numerator of Eq. (1a). If the energy dependence of $Q_1(\epsilon)$ and $Q_m(\epsilon)$ is roughly the same in the region in which $\exp(-\epsilon/\epsilon_L)$ is appreciable, such that their ratio is nearly constant, Eq. (15a) may be useful for an order of magnitude estimate of W. Finally, expressing υ_L in terms of laser intensity I in $\mathrm{W\,cm^{-2}}$ and using numerical values for m, e, and c (velocity of light),

$$W \simeq 3.145\times10^{16}\langle Q_1/Q_m\rangle\,\omega^{-1}\left(\frac{m}{M}I\right)^{1/2}\ \mathrm{cm\,s^{-1}}. \tag{16}$$

The dependence of W on $(m/M)^{1/2}$ arises from the weighting of ϵ_f in ϵ_L by M/m, causing $f_0(\epsilon)$ to be diffuse in the energy as a result of the slow rate of translational energy loss $(m/M)N\upsilon Q_m(\epsilon)$. The derivative of $f_0(\epsilon)$ with ϵ thus depends on m/M and υ_L [Eq. (15b)] depends on $(M/m)^{1/2}$, causing W to depend on $(m/M)^{1/2}$.

IV. CONCLUSIONS

For 1.17-eV photons, Eq. (16) is

$$W \simeq 17.68\langle Q_1/Q_m\rangle\left(\frac{m}{M}I\right)^{1/2}. \tag{17}$$

For reasonably strong power levels $(m/M)I \simeq 10^6$ $\mathrm{W\,cm^{-2}}$; thus,

$$W \simeq 1.768\times10^4\langle Q_1/Q_m\rangle. \tag{18}$$

A reasonably accurate estimate of the pv cross section Q_1 is essential to proceed further in estimating W. If the pv to momentum-transfer cross section ratio turned out to be as large as $\frac{1}{10}$, then W would be about 1.8×10^3 $\mathrm{cm\,s^{-1}}$ according to the model. (According to the ω^{-1} dependence of Eq. (16), however, lower intensities are required for the same W if the photon frequency is lower.) It is interesting to compare this estimate with a dc-field drift velocity[13] of about 1.6×10^3 $\mathrm{cm\,s^{-1}}$ in the e, Ar system at $E/N = 10^{-2}$ $\mathrm{V\,cm^2}$. For $N = 10^{18}$ $\mathrm{cm^{-3}}$, $E = 10^{-3}$ $\mathrm{V\,cm^{-1}}$. On the other hand, the E field corresponding to an intensity of 10^{10} $\mathrm{W\,cm^{-2}}$ is about 2.75×10^6 $\mathrm{V\,cm^{-1}}$. Thus radiation-supported transport by the pv mechanism is very inefficient when compared with conventional electrode transport. However, the use of a laser for transport or the pv mechanism may suggest interesting applications. That a chiral gaseous medium should make radiation-supported tranport possible through the violation of a symmetry theorem (space invariance of radiative-dipole angular distributions for nonoriented targets) would appear to be an interesting phenomenon in itself and worthy of further study.

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APPENDIX: FREE-FREE ABSORPTION RATE ANGULAR DEPENDENCE

To first order in perturbation theory[6] the free-free absorption rate is defined[14] in the dipole length form

$$R_A(\epsilon',\epsilon,\hat{\upsilon}) = NF\int d\Omega'(2\pi)^{-1}[\alpha a_0^5 E_p k|\langle\psi^{(-)}(\vec{r},\vec{k})|\hat{p}\cdot\vec{r}|\psi^{(+)}(\vec{r},\vec{k}')\rangle|^2], \tag{A1}$$

where in atomic units E_p is the photon energy and $\vec{k}'$ and $\vec{k}$ are the initial and final electron momenta. The unit vector $\hat{\rho}$ is in the direction of polarization of the photon. Use of partial wave expansions[4,8] in the matrix element leads to the form,

$$R_A(\epsilon',\epsilon,\hat{\upsilon}) = 4\pi NF \sum_{L=0,1}^{2} b_L(\epsilon',\epsilon)P_L(\cos\theta), \tag{A2a}$$

$$b_L(\epsilon',\epsilon) = 8\pi\alpha a_0^5 E_p k(-1)^{m_p} \sum (-1)^{m+m_1}(2\ell+1)^{1/2}(2\lambda+1)^{1/2}(2L+1)$$
$$\times\begin{pmatrix}1 & 1 & L\\ -m_p & m_p & 0\end{pmatrix}\begin{pmatrix}1 & 1 & L\\ m_1 & -\mu_1 & -(m-\mu)\end{pmatrix}\begin{pmatrix}\ell & \lambda & L\\ 0 & 0 & 0\end{pmatrix}$$
$$\times\begin{pmatrix}\ell & \lambda & L\\ m & -\mu & -(m-\mu)\end{pmatrix}(a_{\ell'\lambda 1m'\mu\mu_1})^* a_{\ell'\ell 1m'mm_1}, \tag{A2b}$$

$$a_{\ell'\ell 1m'mm_1} = \left(\frac{4\pi}{3}\right)^{1/2} \int d\vec{r}\,\psi_{\ell m}^{(-)*}(\vec{r},k) r Y_{1m_1}(\theta_B,\phi_B)\psi_{\ell'm'}^{(+)}(\vec{r},k'), \tag{A2c}$$

where θ_B, ϕ_B are the angles of the electron position $\vec{r}$ in a molecule-fixed frame.[4,8,15] Free-free radial-dipole matrix elements [analogous to those defined in Eq. (A2c)] have recently been calculated for e, Ar by Pindzola and Kelly.[14]

The argument for the existence of the $L=1$ component in Eq. (A2a) is the same as that of Ref. 4 for photoionization of nonoriented chiral molecules. First, the photon must be left- or right-circularly polarized ($m_p = \pm 1$). Second the final-state partial waves l and λ must be of opposite parity. Third, the electric-dipole amplitudes of Eq. (A2b) must be unequal for positive and negative values of final-state waves azimuthal quantum numbers m and μ. Unless the latter criterion is met, the $L=1$ branches of the rate will cancel by the rule

$$\begin{pmatrix}1 & 1 & L\\ m_1 & -\mu_1 & -(m-\mu)\end{pmatrix} = (-1)^L\begin{pmatrix}1 & 1 & L\\ -m_1 & \mu_1 & (m-\mu)\end{pmatrix}, \tag{A3}$$

in the summations over all m and μ.

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