

## Effect of laser radiation on the mobility of electrons in gases

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Numerical solutions of Boltzmann's equation are obtained for the distribution of electrons in gaseous Ar in the presence of a dc electric field and a laser field. The calculations are performed at field strengths below those either of electrical or optical gas breakdown in order to study the effect of the radiation on dc-field-induced electron mobilities. The isotropic component of the electron distribution function oscillates with energy as a result of the discrete nature of free-free absorption and stimulated emission. Similar structure has been observed in previous studies at laser-field strengths close to optical breakdown with no dc field present. The radiative processes cause the mobility to increase by roughly the same amount as that obtained by use of the classical model of microwave electron heating, or by removing the laser and increasing the temperature to a thermal energy close to one laser photon. Thus, the energy oscillations in the distribution are not significantly probed by the  $e, \text{Ar}$  mobility. These findings suggest the use of molecular gases, having vibrationally inelastic-threshold energies in the region of oscillations, to find mobilities more sensitive to the discrete nature of the radiative processes.

### INTRODUCTION

Theoretical results<sup>1</sup> have been available for some time for the isotropic component of the distribution function ( $f_0$ ) for electrons in a gas in the presence of an intense laser beam. These results are presented in the study of ionization growth and gas breakdown. Other studies, both experimental<sup>2,3</sup> and theoretical,<sup>4</sup> have been concerned with gas breakdown in the presence of an intense laser beam and a strong dc-electric field. The previous theoretical results<sup>1</sup> for the laser beam (and of the present study) reveal oscillatory structure in  $f_0$  as a function of the electron energy  $\epsilon$ . Recent work<sup>5</sup> has appeared on dc-field electron mobilities through molecular gases which reveals a critical dependence of the drift velocity on such details of the electron-molecule scattering as the relative energy position of a Ramsauer minimum in the elastic channel and the first vibrationally inelastic channel and the first vibrationally inelastic threshold. Such a drift velocity would be expected to be sensitive to an oscillatory  $f_0$ , if the experiments<sup>6</sup> were repeated in the presence of a laser field, for example.

It is the purpose of the present work to examine the effect of laser-induced structure in  $f_0$  on the electron drift velocity at field strengths for both the laser and the dc field that are substantially below optical or electrical gas breakdown voltages. It is hoped that such studies as these might provide theoretical evidence that the laser can be used to influence  $f_0$  such that the mobility may be affected in predictable and interesting ways. Calculations are performed for electrons in Ar. The mobility is increased by electron absorption of radiation, but it appears not to be sensitive to the energy oscillations in  $f_0$ . This is perhaps not sur-

prising for  $e, \text{Ar}$ . Only elastic scattering occurs in the energy range studied, and the drift velocity depends on an integration over energy which tends to smooth any oscillatory structure present in the distribution. However, the  $e, \text{Ar}$  elastic cross section is well known both experimentally<sup>7</sup> and theoretically<sup>8</sup> and, more importantly, the free-free absorption cross section is reliably known theoretically.<sup>9,10</sup> Thus, this choice provides a good test of theoretical methods used to evaluate the free-free absorption cross section (Appendix A) and to solve the Boltzmann equation (Appendix B).

### THEORY

Swarm experiments<sup>7</sup> measure a drift velocity  $W$  of electrons in the direction opposite to the direction of an applied dc-electric field of strength  $E$ .  $W$  is proportional to an integral over electron energy,  $\epsilon$  erg, containing  $f_1^{(\text{dc})}(\epsilon)$ , the strength of the dipolar component of the electron distribution function. For electrons in Ar (for which the only low-energy loss mechanism is gas-atom recoil),<sup>11</sup>

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

where  $Q_m$  is the  $e, \text{Ar}$  momentum-transfer cross section and  $f_0^{(\text{dc})}(\epsilon)$  is the isotropic component of the electron distribution function, which is Maxwellian when  $E/N = 0$ . The mobility  $\mu$  is defined as the ratio of  $W$  [see Eq. (14)] to  $E$ . Results for  $W$ , rather than  $\mu$ , are presented in Table I in order to facilitate comparison with dc-field calculations<sup>7</sup> as a function of  $E/N$ . The theory of ac-field gas discharges<sup>11</sup> has been available for some time. Under steady-state conditions, a relaxation time  $\tau_m = [N\nu Q_m(\epsilon)]^{-1}$  is defined, where  $\nu$  is the instantaneous electron velocity. In an ac field, at optical frequencies  $\omega$  for which  $\omega\tau_m \gg 1$ , Eq. (1) is

TABLE I. Drift velocities for combined dc- and laser-fields at  $E/N = 10^{-21}$  V cm<sup>2</sup>.

1. $I = 0$ , $T = 300$ K [Eqs. (3) and (7a)], $W = 1.5987 \times 10^3$ cm s <sup>-1</sup> (see Ref. 5).
2. $I \cong 0.19$ MW cm <sup>-2</sup> , $T = 300$ K, using $E_{ss}^{(dc)}(\epsilon)$ -modified Eq. (8a), $W = 3.9468 \times 10^3$ cm s <sup>-1</sup> .
3. $I \cong 0.19$ MW cm <sup>-2</sup> , $T = 300$ K, using Eqs. (3) and (7a) with $E_{ss}^{(ac)}$ [Eq. (7b)] added to $k_B T$ , $W = 3.3713 \times 10^3$ cm s <sup>-1</sup> .
4. $I = 0$ , $T = 1200$ K [Eqs. (3) and (7a)], $W = 3.5292 \times 10^3$ cm s <sup>-1</sup> .

replaced by

$$f_1^{(ac)}(\epsilon, t) = -E_{rms} \frac{eV}{\omega} \sin \omega t \frac{df_0^{(ac)}(\epsilon)}{d\epsilon}, \quad (2)$$

where  $E_{rms}$  is the ac-field root-mean-square field strength. Thus,  $E[NQ_m(\epsilon)]^{-1} = E\nu\tau_m$  of Eq. (1) is replaced by  $E_{rms}\nu\omega^{-1}\sin\omega t$ . If the field strengths and  $f_0^{(j)}(\epsilon)$  derivatives are assumed to have comparable magnitudes, then the ratio  $f_1^{(ac)}/f_1^{(dc)} \sim (\omega\tau_m)^{-1}\sin\omega t$ . Thus the ac  $f_1$  is damped as  $(\omega\tau_m)^{-1} \ll 1$ , and when it is averaged with respect to a pulse width characteristic of laser irradiation, rapid oscillations in  $\sin\omega t$  produce a result which combines with the damping factor to give a minuscule result for pulse widths ( $>ns$ ) and power levels ( $<10^9$  W cm<sup>-2</sup>) at which such experiments would be performed. Thus, the effect of the laser field on the  $f_1$ -component of the distribution function, *except* through its effect on the derivative of the  $f_0$ -component, can be ignored.

The equation for  $f_0^{(j)}(\epsilon)$ , according to the theory of Ref. 11, has the simple form

$$[k_B T + E_{ss}^{(j)}(\epsilon)] \frac{df_0^{(j)}(\epsilon)}{d\epsilon} = -f_0^{(j)}(\epsilon). \quad (3)$$

The solution is

$$f_0^{(j)}(\epsilon) = A \exp\left(-\int_0^\epsilon \frac{d\epsilon'}{k_B T + E_{ss}^{(j)}(\epsilon')}\right), \quad (4)$$

where  $A$  is the normalization constant determined from

$$\int_0^\infty d\epsilon \epsilon^{1/2} f_0^{(j)}(\epsilon) = 1, \quad (5)$$

and  $k_B T$  is the thermal energy.  $E_{ss}^{(j)}(\epsilon)$  is an energy of the electrons in an ac or dc field under steady-state conditions, that is,

$$\epsilon_{ss}^{(j)}(\epsilon) = \frac{1}{3} \frac{M}{m} \frac{(E_j e)^2}{m_e} \tau_j^2, \quad (6)$$

where  $m_e$  is the electron mass,  $M$  is the gas-atom mass,  $\tau_j$  is a "relaxation" time appropriate for a steady-state ac- or dc-field Boltzmann equation [Eq. (3)], and  $E_j$  is the field strength in either case. Explicitly,

$$E_{ss}^{(dc)}(\epsilon) = \frac{1}{6} \frac{M}{m_e} \left(\frac{E}{N}\right)^2 \frac{e^2}{Q_m^2(\epsilon)\epsilon}, \quad (7a)$$

$$E_{ss}^{(ac)} = \frac{1}{3} \frac{M}{m_e} \frac{(E_{rms} e)^2}{m_e \omega^2}, \quad (7b)$$

where  $\tau_{dc} = [N\nu Q_m(\epsilon)]^{-1}$  or  $\tau_{ac} = \omega^{-1}$ , respectively, have been used. These cases are well known. Equations (3) and (7a) were used in Ref. 7 to fit dc-field drift velocity data to determine  $Q_m(\epsilon)$ . Equations (3) and (7b) are appropriate in the limit  $\epsilon \gg \hbar\omega$  (where  $\hbar\omega$  is the photon energy) and have been used to describe gas breakdown.<sup>12</sup> They derive from the Boltzmann equation for finite photon energies<sup>13,14</sup>

$$\left(k_B T \frac{df_0(\epsilon)}{d\epsilon} + \frac{D_+(\epsilon) + D_-(\epsilon)}{(2m_e/M)N\nu Q_m(\epsilon)\epsilon^{3/2}}\right) = -f_0(\epsilon), \quad (8a)$$

$$D_+(\epsilon) = \int_0^\epsilon d\epsilon' \epsilon'^{1/2} [R_E(\epsilon' + \hbar\omega) f_0(\epsilon' + \hbar\omega) - R_A(\epsilon' + \hbar\omega) f_0(\epsilon')], \quad (8b)$$

$$D_-(\epsilon) = \int_0^\epsilon d\epsilon' \epsilon'^{1/2} [R_A(\epsilon') f_0(\epsilon' - \hbar\omega) - R_E(\epsilon') f_0(\epsilon')]. \quad (8c)$$

$R_A$  and  $R_E$  are the free-free absorption and stimulated-emission rates, respectively, in s<sup>-1</sup> per electron. By detailed balance,

$$R_E(\epsilon') = R_A(\epsilon')(1 - \hbar\omega/\epsilon')^{1/2}. \quad (9)$$

Using Eq. (9) in Eqs. (8b) and (8c) and the condition  $\epsilon' \gg \hbar\omega$ , expansion<sup>13,14</sup> of  $\epsilon'^{1/2} R_A(\epsilon' + \hbar\omega)$  and  $f_0(\epsilon' \pm \hbar\omega)$  in a Taylor series about  $\epsilon'$  results in cancellation of terms to order  $(\hbar\omega)^2$ . For  $\epsilon' \gg \hbar\omega$ , the absorption rate has a simple relationship to  $Q_m(\epsilon)$ ,

$$R_A(\epsilon') \approx \frac{2(E_{rms} e)^2 N\nu Q_m(\epsilon') \epsilon'}{3m_e \omega^2 (\hbar\omega)^2}. \quad (10)$$

Use of Eq. (10) and the expanded forms of  $D_+$  and  $D_-$  gives the limiting result, Eqs. (3) and (7b).

As stated above, the interest in the present work is the region  $\epsilon' \approx \hbar\omega$ . Here, a form of  $R_A$  other than Eq. (10) must be used. Provided the laser intensity is small enough<sup>15</sup> so that  $R_A$  can be calculated from first-order perturbation theory, it is (in s<sup>-1</sup>),<sup>10</sup>

$$R_A = (4\pi)^{-1} \int d\Omega_i d\Omega(2\pi)^{-1} \left( \frac{F\alpha a_0^2}{E_p} \frac{m_e}{\hbar} N\nu |\langle \psi_k^{(-)}(\vec{r}) | \hat{p} \cdot (-i\vec{\nabla}) | \psi_k^{(+)}(\vec{r}) \rangle|^2 \right), \quad (11a)$$

where  $F = 10^7 I / (\hbar \omega)$  is the photon flux in  $\text{cm}^{-2} \text{s}^{-1}$  for an intensity  $I$  in  $\text{W cm}^{-2}$ ,  $\alpha$  is the fine-structure constant,  $E_p$  in the photon energy is in atomic units, and the surface integrals are over all directions of incidence along  $\vec{k}_i$  and scattering along  $\vec{k}$ , where  $k^2 = k_i^2 + 2E_p$ . The intensity is related to  $E_{\text{rms}}$  according to

$$E_{\text{rms}} = \left( \frac{4\pi 10^7 I}{c} \right)^{1/2}, \quad (11b)$$

where  $c$  is the velocity of light. Equation (11a) can be rewritten as

$$R_A = FN \sigma_A(\epsilon), \quad (12)$$

in terms of an absorption "cross section"  $\sigma_A(\epsilon)$  in  $\text{cm}^5$ .

### RESULTS AND DISCUSSION

Values of  $\sigma_A(\epsilon)$  for  $e, \text{Ar}$  have been presented<sup>10</sup> for photon energies of 0.62 and 6.2 eV. In the present study, however,  $\sigma_A(\epsilon)$  is required for a photon energy of 0.117 eV. Use of an approximate form (Appendix A) of the radiative dipole matrix element of Eq. (11a) is investigated.

$$M \cong \frac{2\pi}{E_p} \alpha_0^2 \vec{\rho} \cdot [\vec{k} f_i(k_i, \hat{k} \cdot \hat{k}_i) - \vec{k}_i f(k, \hat{k} \cdot \hat{k}_i)], \quad (13)$$

where  $f_i, f$  are the elastic-scattering amplitudes at  $k_i^2, k^2$  Ry. Using the  $e, \text{Ar}$  phase shifts of Pindzola and Kelly,<sup>8</sup> we test the accuracy of this approximation against the exact dipole results of Ref. 10 (Figs. 1–3), allowing for an estimated 10% error inherent in the numerical procedures used in Ref. 10. For small  $k_i$  the agreement is good. A disagreement (see Appendix A for a discussion) has set in by  $k_i = 0.6 \alpha_0^{-1}$  for the polarization-potential<sup>10</sup> (PP) results and by  $k_i = 0.8 \alpha_0^{-1}$  for the optical-potential<sup>8</sup> (OP) and Hartree-Fock-potential<sup>8</sup> (HF) results. The radial dipole matrix elements were calculated using (PP) and HF radial waves; thus, strictly speaking, there is no dipole result against which to compare the OP results of Fig. 1. The OP and PP calculations differ significantly above  $k_i = 0.75 \alpha_0^{-1}$  in the  $d$ -wave phase shifts,<sup>10</sup> and this is the origin of the OP and PP disagreement at higher  $k_i$  in Fig. 1. The higher  $k_i$  disagreement of results derived from Eq. (13) with the dipole results is of some interest since it is believed (see Ref. 19 and references therein, for example) that Eq. (13) (which has been derived before by Low<sup>16</sup>) is accurate when  $k_i^2 \gg E_p$ .

Figures 4–5 show  $f_0$ , normalized according to Eq. (5) from the numerical solution of Eq. (8a) (described in Appendix B) at different laser intensities  $I$ . Figure 4 shows the thermal distribution ( $I = 0$ ) and the laser-deformed distribution ( $I \cong 0.075$

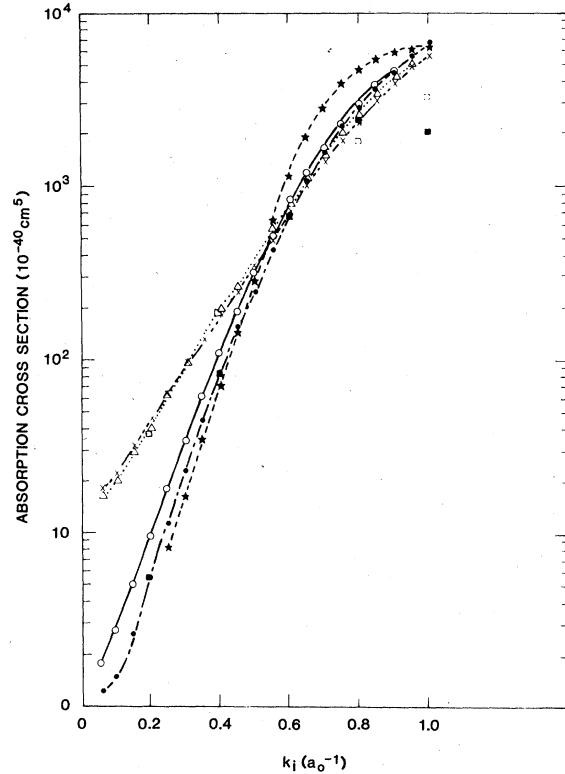


FIG. 1. Absorption cross section vs initial electron momentum  $k_i$  for 0.62-eV photons. Solid circles:  $\sigma_A$  evaluated using  $k = k_i$  in  $f$ , Eq. (13), and optical potential (OP) phase shifts (Ref. 8); Open circles:  $\sigma_A$  evaluated using  $k_i = k$  in  $f_i$  and OP phase shifts;  $\times$ :  $\sigma_A$  evaluated using  $k = k_i$  in  $f$  and Hartree-Fock (HF) phase shifts (Ref. 8); triangles:  $\sigma_A$  evaluated using  $k_i = k$  in  $f_i$  and HF phase shifts; stars:  $\sigma_A$  evaluated using  $k = k_i$  in  $f$  and polarization potential (PP) phase shifts (Ref. 10); closed squares: radiative-dipole results of Ref. 10, PP theory; open squares: radiative-dipole results of Ref. 10, HF theory.

MW  $\text{cm}^{-2}$ ). Also shown is the Holstein<sup>11</sup> ac-field or "laser-heating" solution  $f_0^{(\text{ac})}(\epsilon)$  from Eqs. (3) and (7b). Although valid for  $\epsilon \gg \hbar\omega$ , it is seen to be a crude average of the oscillatory  $f_0(\epsilon)$  in the range of  $\epsilon$  comparable to  $\hbar\omega$ . Figure 5 shows results for the lowest and highest intensities used. It would be difficult to use the present numerical algorithm (Appendix A) to go beyond  $I \cong 0.19$  MW  $\text{cm}^{-2}$  since  $\epsilon_{\text{max}}$  of Eq. (8a) (0.65 eV) would have to be larger, requiring a larger number of grid points ( $> 130$ ) to achieve the same resolving power for the continuous function  $f_0(\epsilon)$  as that currently obtained.

We have also performed the  $I \cong 0.19$  MW  $\text{cm}^{-2}$  calculation in the presence of a dc field and have calculated  $W$  from

$$W = \frac{1}{3} \left( \frac{2}{m_e} \right)^{1/2} \int_0^\infty d\epsilon \epsilon f_1(\epsilon), \quad (14)$$

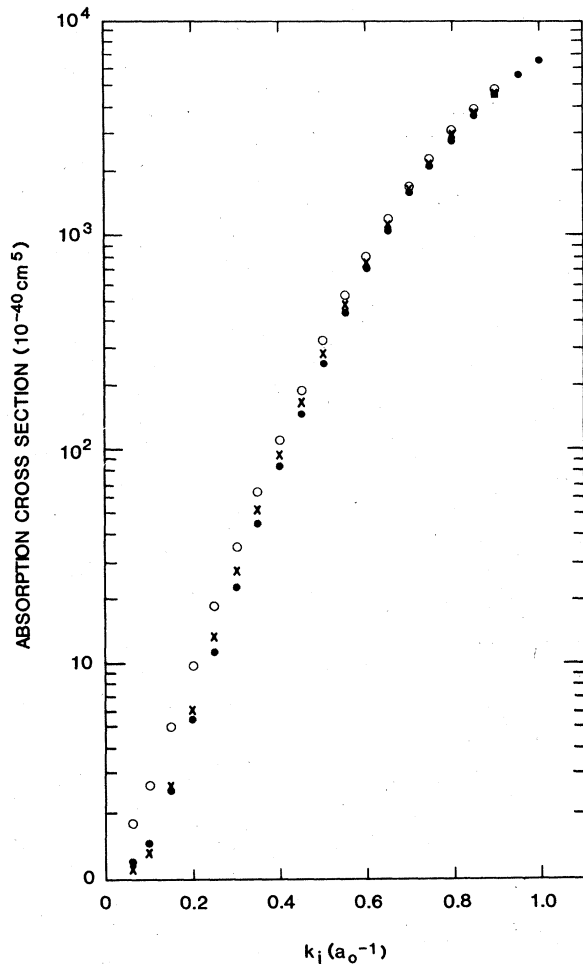


FIG. 2. Solid and open circles same as in Fig. 1;  $\times$ :  $\sigma_A$  evaluated using Eq. (13) as written and OP phase shifts.

where  $f_1$  is given by Eq. (1) with  $f_0^{(dc)}(\epsilon)$  replaced by  $f_0(\epsilon)$  from Eq. (8a), which is modified by adding  $E_{ss}^{(dc)}(\epsilon)$  [Eq. (7a)] to  $k_B T$ . The solution of Eqs. (3) and (7a) at  $E/N = 10^{-21}$  V cm<sup>2</sup>, the lowest strength of Ref. 7, was repeated using the numerical methods described in the Appendix A (and by direct integration of the differential equation as a further check). At this,  $E/N f_0^{(dc)}(\epsilon)$  is nearly Maxwellian; thus,  $f_0(\epsilon)$  from  $E_{ss}^{(dc)}(\epsilon)$ -modified Eq. (8a) differs slightly from the  $I \cong 0.19$  MW cm<sup>-2</sup>  $f_0(\epsilon)$  of Fig. 5 from unmodified Eq. (8a), since  $k_B T \gg E_{ss}^{(dc)}(\epsilon)$  for most of  $\epsilon$ . Values of  $W$  are shown in Table I. Also shown is a  $W$  calculated by using  $f_0$  from Eqs. (3) and (7a), modified by adding  $E_{ss}^{(ac)}$  to  $k_B T$  at  $T = 300$  K and a  $W$  calculated by using  $f_0^{(dc)}$  from Eqs. (3) and (7a) at  $T = 1200$  K. This temperature for a dc-field calculation (no laser present) is chosen to correspond to a thermal energy close to 0.12 eV (one CO<sub>2</sub> photon energy). The similarity

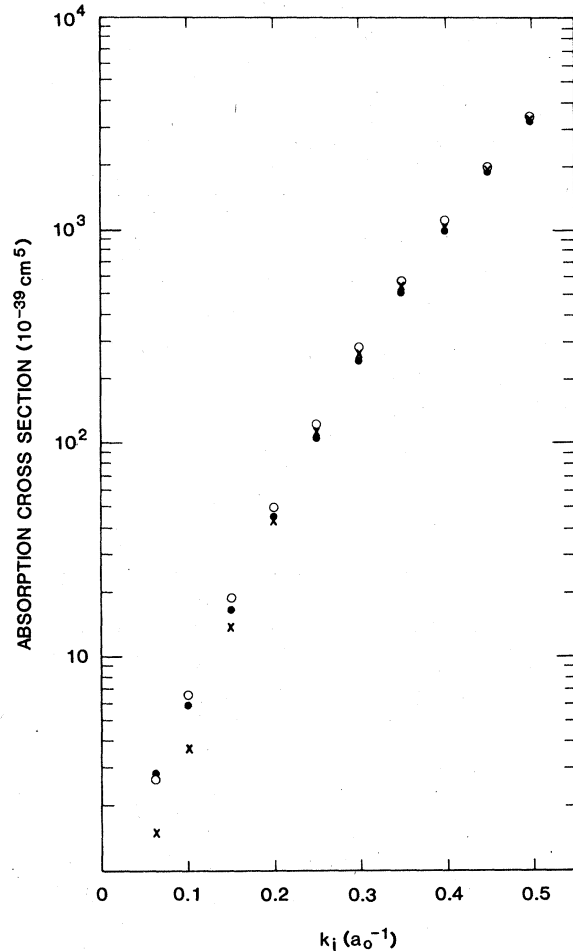


FIG. 3. Absorption cross section for 0.117-eV photons. Solid and open circles same as in Fig. 1;  $\times$  same as in Fig. 2.

of this elevated-temperature mobility to the 300 K laser-influenced mobilities emphasizes the laser-induced "heating" nature of the problem, as opposed to aspects ( $f_0$  vs electron energy, for example) which depend on the absorption and stimulated emission of radiation quanta. Thus  $W$  is shown to be fairly insensitive to laser-induced multiple-absorption structure in  $f_0$ . In fact, a temperature of 1200 K in Eqs. (3) and (7a) or a laser "temperature" of  $E_{ss}^{(ac)}/k_B$  added to  $T$  (for  $T = 300$  K) in Eqs. (3) and (7a) gives a  $W$  comparable to that obtained using  $f_0$  from  $E_{ss}^{(dc)}(\epsilon)$ -modified Eq. (8a). The use of "laser-heating" may be important, however, if elevated temperatures are experimentally impractical. The structure in  $f_0$  (Figs. 4-5) suggests that target gases having inelastic thresholds in this range of  $\epsilon$  may have  $W$ 's which differ more strongly as a function of laser power.

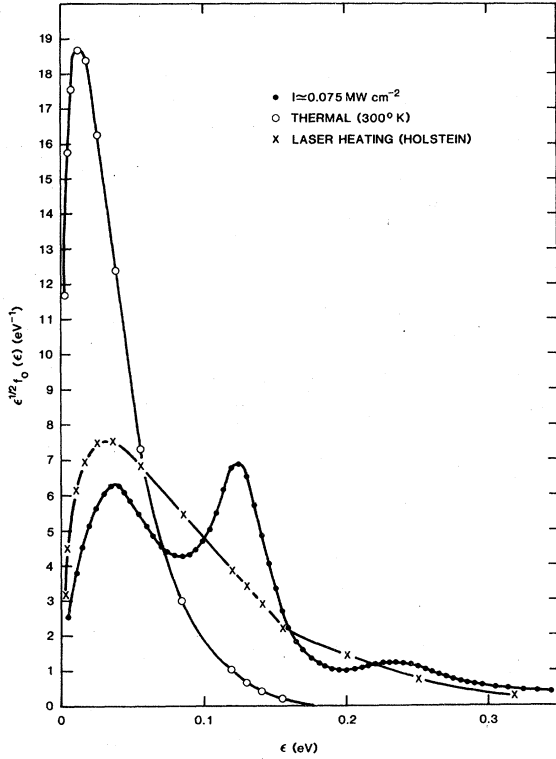


FIG. 4. Solid circles:  $f_0$  from Eq. (8a); open circles:  $f_0$  from Eq. (8a),  $I=0$ ; x:  $f_0$  from Eqs. (3) and (7b).

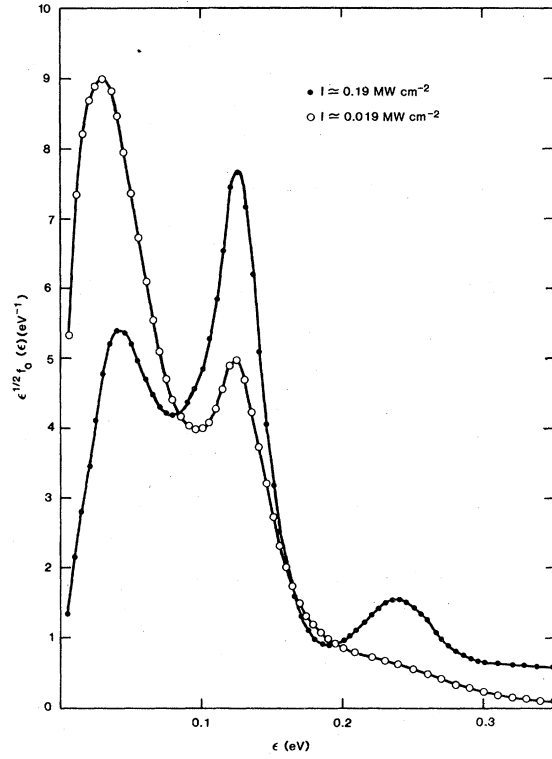


FIG. 5. Two  $f_0$ 's from Eq. (8a).

#### APPENDIX A: EVALUATION OF THE FREE-FREE RADIATIVE-DIPOLE MATRIX ELEMENT

$\sigma_A$  of Eq. (12) has been calculated by partial-wave expansion of the scattering wave functions in the radiative-dipole matrix element of Eq. (11a), with analytic evaluation of the angular parts and numerical evaluation of the radial parts.<sup>10</sup> Although it is estimated<sup>10</sup> that there is up to 10% error due to the use of numerical procedures designed to force the radial integrals to be absolutely convergent, the use of the dipole matrix element should be considered the most accurate means of calculating free-free absorption cross sections, provided the first-order theory is valid.<sup>15</sup> In this appendix, therefore, we examine the validity of the approximate form given by Eq. (13) against the results of Ref. 10 as a standard.

To study the matrix element, it is convenient to substitute

$$\psi_{\vec{k}_j}^{(\pm)}(\vec{r}) = e^{i\vec{k}_j \cdot \vec{r}} + \lim_{\gamma \rightarrow 0} (2\pi)^{-3} \int d\vec{k}' \int d\vec{r}' \frac{e^{i\vec{k}' \cdot (\vec{r}-\vec{r}')}}{[(k_j^2 - k'^2) \pm i\gamma]} U(\vec{r}') \psi_{\vec{k}_j}^{(\pm)}(\vec{r}') \quad (\text{A1})$$

for the initial ( $\vec{k}_j = \vec{k}_i$ ) and final ( $\vec{k}_j = \vec{k}_f$ ) states, where  $V = \hbar^2/2m_e U$  is the  $e$ , target potential. Operation with  $-i\vec{\nabla}$ , integration over  $\vec{r}$ , integration over  $\vec{k}$ -space variables, wherever analytic, taking the limits, and identification of the off-shell scattering amplitudes present give the identity for the matrix element,

$$M = (2\pi)^3 \hat{p} \cdot \vec{k}_i \delta(\vec{k}_i - \vec{k}_f) + \frac{2\pi a_0^2}{E_p} \hat{p} \cdot [\vec{k}_f f_i(\vec{k}_f \cdot \vec{k}_i) - \vec{k}_i f_f(\vec{k}_i \cdot \vec{k}_f)] + \frac{2}{\pi} \int d\vec{k}' (\hat{p} \cdot \vec{k}') f_i(\vec{k}' \cdot \vec{k}_i) f_f(\vec{k}' \cdot \vec{k}_f) \left[ \frac{1}{(k_i^2 - k'^2)(k_f^2 - k'^2)} - i\pi \left( \frac{\delta(k_i^2 - k'^2)}{k_f^2 - k'^2} + \frac{\delta(k_f^2 - k'^2)}{k_i^2 - k'^2} \right) \right]. \quad (\text{A2})$$

The off-shell scattering amplitudes are defined for any momentum  $\vec{k}$ ,

$$f_j(\vec{k} \cdot \vec{k}_j) = -(4\pi)^{-1} \langle e^{i\vec{k} \cdot \vec{r}} | U(\vec{r}) | \psi_{\vec{k}_j}^{(\pm)}(\vec{r}) \rangle. \quad (\text{A3})$$

Equation (13) (with  $\vec{k}$  identified with  $\vec{k}_f$ ) emerges

from Eq. (A2) if the first and fourth terms are dropped and if the off-shell amplitudes in the second term are replaced by the on-shell amplitudes  $f_i(k_i, \hat{k}_f \cdot \hat{k}_i)$  and  $f_f(k_f, \hat{k}_f \cdot \hat{k}_i)$ , respectively. To order  $E_p^0$  [when the on-shell amplitudes of Eq. (13)

are expanded about the average energy  $(k_i^2 + k_f^2)/2$  Ry], Eq. (13) is equivalent to the well-known form derived by Low.<sup>16</sup> The on-shell replacement requires that  $E_p/k_i^2 \ll 1$  and  $E_p/k_f^2 \ll 1$ , as stated in Ref. 16. Since these validity indicate that the on-shell approximation should improve with increasing electron energy, the reason for the on-set of the disagreement with the dipole results of Ref. 10 at the higher  $k_i$  values (Fig. 1) would appear to be the result of dropping the fourth term (since the first term is zero for  $\vec{k}_i \neq \vec{k}_f$ ). Scattering amplitudes which are very much off shell should be small. For finite photon energies the two off-shell amplitudes under the  $d\vec{k}'$  integral sign can never be on shell at the same  $k'$ . This indicates that the fourth term may be small. However, questions of its magnitude relative to the second term can really be answered only after all contributing terms in Eq. (A2) have been calculated using the off-shell definition of the amplitudes [Eqs. (A3)].

#### APPENDIX B: NUMERICAL METHODS USED TO SOLVE EQ. (8a)

The presence of  $f_0(\epsilon' \pm \hbar\omega)$  prevents the use of direct integration methods to solve Eq. (8a). The integral terms in  $D_+$  and  $D_-$  are eliminated by converting to a second-order differential equation for  $f_0$  and then converting this equation to a  $130 \times 130$  homogeneous algebraic set using finite-difference formulas for the first and second derivatives [Eqs. (25.3.5)  $p=0$  and (25.3.23), respectively, from Ref. 17]. Following Riley and Matzen,<sup>18</sup> this set was diagonalized, and the eigenvalues and selected eigenvectors were inspected. Small relative values of the eigenvalues were enforced by repeating the closest to the diagonal and diagonal elements of the last three rows of the matrix. First, the thermal solution was obtained and the

accuracy was determined to be satisfactory by comparison with the analytic normalized Boltzmann factor  $A \exp(-\epsilon/k_B T)$ . Since a second-order equation is solved, a linear combination of the eigenvectors belonging to the eigenvalues having the lowest absolute values is, in principle, possible of finding the physical solution. For the thermal case it turned out that two degenerate zero eigenvalues occurred; however, the Eq. (5) normalized eigenvector which compared to excellent accuracy with  $A \exp(-\epsilon/k_B T)$  was that belonging to the second-lowest, real positive eigenvalue. With the laser field on, the physically acceptable eigenvectors [whose Eq. (5) normalized versions are plotted in Figs. 4-5] also belonged to the second lowest, real positive eigenvalues. These second-lowest eigenvalues were a factor of  $10^{-3}$  to  $10^{-4}$  smaller than the largest absolute values in the eigenvalue list. In both the thermal and laser cases the rejected eigenvectors had at least one node and did not decay at  $\epsilon_{\max}$ .

The normalized eigenvector for the highest power (Fig. 5) was insensitive to increasing the number of grid points from 100 to 130. The peak heights and valley depths were insensitive to about 1% to increasing  $\epsilon_{\max}$  from 0.60 to 0.65 eV. The function  $f_0$  is less accurate near  $\epsilon_{\max}$ . For example,  $f_0$  shows about a 10% change at 0.5 eV when  $\epsilon_{\max}$  is increased from 0.60 to 0.65 eV.  $f_0$  is small at these  $\epsilon$ 's, however, and greater relative error is to be expected. At  $\epsilon \cong 0.60$  eV,  $f_0$  is about 1% of the first peak height; thus an  $\epsilon_{\max}$  of 0.65 eV is considered satisfactory.

#### ACKNOWLEDGMENTS

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$$\rho = (8\pi 10^7 I \alpha / \hbar^3)^{1/2} (m_e a_0^3 / E_p^2) \hat{\rho} \cdot (\vec{k} - \vec{k}_i)$$

(see text for definitions). For backward scattering of 1 Ry electrons, when  $\hat{\rho} \cdot (\vec{k}_f - \vec{k}_i) \approx 2$  for  $\hat{\rho} \parallel \vec{k}_i$ ,  $\rho_{\max} \approx 1.07 \times 10^{-8} I^{1/2} E_p^2$ . For CO<sub>2</sub> photons  $\rho_{\max} \approx 10^{-3} \times (I/3)^{1/2}$ . The first-order dipole approximation can be expected to be accurate for  $\rho_{\max} < 1$ .

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