Third-harmonic generation from a laser-induced autoionizing level

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A nonperturbative theory is presented for the gas polarization induced by two lasers. The first laser ionizes Li($2s$) by three-photon absorption, with the first two photons in resonance with the $3d$ level. A second laser stimulates recombination into the $3s$ level, thereby inducing an autoionizing-like structure in the continuum. It is found that the polarization (and hence possibly third-harmonic generation) is enhanced near the induced resonance.

I. INTRODUCTION

It has been suggested[1,2] that third-harmonic generation (THG) from an autoionizing level would be an important mechanism for the production of vacuum-ultraviolet radiation. This problem has recently been studied theoretically.[3] The theory of autoionization in the presence of a strong radiation field has received much attention recently,[4–6] and the theory has been extended to include both radiative decay[7] and a type of decay which affects only the off-diagonal elements of the density matrix.[8]

The theory of autoionizing-like resonances which are induced in the continuum by the presence of a second, strong radiation field has also appeared recently,[9,10] and a calculation[11] has been performed on atomic Li. It is the purpose of this paper to extend this theory[10,11] to THG.

Earlier studies of THG in gases include those of New, Ward, and Smith.[12] Recently work[13–15] has appeared on THG as a competing process on three-photon resonantly enhanced multiphoton ionization. Moreover, the first experimental results on the process described in the present paper have also appeared.[16]

II. THEORY

Electromagnetic (EM) wave generation in a gas is described by the equation

$$\nabla^2\mathbf{E}'(\mathbf{r},t)-\left(\frac{n'}{c}\right)^2\frac{\partial^2\mathbf{E}'(\mathbf{r},t)}{\partial t^2}=\frac{4\pi}{c^2}\frac{\partial^2\mathbf{P}(\mathbf{r},t)}{\partial t^2}\ , \tag{1}$$

where $\mathbf{E}'$ is the electric field of the generated wave, $\mathbf{P}$ is the gas polarization (dipole moment per unit volume) induced by an incident wave, and n' is the index of refraction for propagation of the generated wave at the appropriate frequency. $\mathbf{P}$ is

$$\mathbf{P}(\mathbf{r},t)=N\gamma(t)\hat{\rho}Ee^{i(\mathbf{k}\cdot\mathbf{r}-\omega t)}\ , \tag{2}$$

where N is the gas number density, $\gamma(t)$ is a generalized polarizability in cm^3/atom, and an "incident" EM wave with frequency ω and electric-field strength E propagates in the direction of $\mathbf{k}$ and is polarized in the direction of the unit vector $\hat{\rho}$. For example, Eq. (2) has been studied[13] for THG, where $\gamma=\frac{1}{4}\chi E^2$ (for an atom nonlinear susceptibility χ in cm^6/erg), $|\mathbf{k}|=3n\omega_p/c$, and $\omega=3\omega_p$ (for photon frequency ω_p and an index of refraction n for wave propagation at frequency ω_p, such that n' can now be identified as the index of refraction for wave propagation at $3\omega_p$). χ has been calculated for various atoms from third-order time-dependent perturbation theory.[12,17]

We consider the following resonant processes as a source of gas polarization in atomic Li:

$$\begin{array}{l} 2s\xrightarrow{2\epsilon_1}3d\xrightarrow{\epsilon_1}\epsilon p+\epsilon f\ ,\\ \qquad\quad\ \updownarrow \epsilon_2\\ \qquad\quad\ 3s\end{array} \tag{3}$$

where, on resonance $\epsilon_1=1.925$ eV for $\epsilon_{3d}-\epsilon_{2s}-2\epsilon_1=0$ and $\epsilon_2=2.432$ eV for $\epsilon_{3s}-\epsilon_{3d}-\epsilon_1+\epsilon_2=0$ (where we have used the atomic model discussed in Ref. 11, in which $\epsilon_{2s}=-5.342$ eV, $\epsilon_{3s}=-1.998$ eV, and $\epsilon_{3d}=-1.491$ eV). Note that laser 1 can ionize $3d$ but not $3s$ and that laser 2 can ionize both $3d$ and $3s$. The processes in Eq. (3) are described by solving the time-dependent Schrödinger equation. The exact wave function for a Li atom in the presence of two lasers is expanded in the $2s$, $3d$, ϵp, ϵf, and the set of np eigenstates of Li. As discussed in Ref. 11, the continuum and virtual states are eliminated by substitution in the equations for the probability amplitudes for the $2s$, $3d$, and $3s$ eigenstates. After some approximations[11,18] a set of coupled differential equations is obtained for these amplitudes. These are

$$i\begin{bmatrix}\frac{d}{dt}&0&0\\0&\frac{d}{dt}&0\\0&0&\frac{d}{dt}\end{bmatrix}\begin{bmatrix}a_{2s}\\a_{3d}\\a_{3s}\end{bmatrix}=\begin{bmatrix}S_{2s}&\frac{1}{2}\Omega_{2s,3d}e^{-i\Delta_2 t}&0\\ \frac{1}{2}\Omega_{3d,2s}e^{i\Delta_2 t}&S_{3d}-\frac{i}{2}\gamma_{3d}&\frac{1}{2}\Omega_{3d,3s}e^{-i\Delta_1 t}\\0&\frac{1}{2}\Omega_{3s,3d}e^{i\Delta_1 t}&S_{3s}-\frac{i}{2}\gamma_{3s}\end{bmatrix}\begin{bmatrix}a_{2s}\\a_{3d}\\a_{3s}\end{bmatrix}\ . \tag{4}$$

On resonance ($\Delta_2=\omega_{3d}-\omega_{2s}-2\omega_1=0$ and $\Delta_1=\omega_{3s}-\omega_{3d}-\omega_1+\omega_2=0$) and for linearly polarized light, the matrix elements of Eq. (4) are given in the Appendix.

Analysis shows that the gas polarization [Eq. (2)] for the processes given by Eq. (3) is

$$\mathbf{P}(\mathbf{r},t)=N[\gamma_1(t)\hat{\rho}_1E_1+\gamma_2(t)\hat{\rho}_2E_2]e^{i(3\mathbf{k}_1\cdot\mathbf{r}-3\omega_1t)}, \tag{5a}$$

$$E_j=\left[\frac{8\pi F_j\epsilon_j}{c}\right]^{1/2}, \tag{5b}$$

$$\gamma_1(t)=\frac{4a_0^3}{3\sqrt{5}}\left[\int_0^\infty dr\,u_{2s-1}(r)ru_{3d}(r)+k^{-1}\int_0^\infty dr\int_0^\infty dr'\,u_{2s}(r)rG_1(kr_>)F_1(kr_<)r'u_{3d}(r')\right.$$
$$\left.-ik^{-1}\int_0^\infty dr\,F_1(kr)ru_{2s}(r)\int_0^\infty dr\,F_1(kr)ru_{3d}(r)\right]b_{2s}^*(t)b_{3d}(t), \tag{5c}$$

$$\gamma_2(t)=\frac{2a_0^3}{3}\left[\int_0^\infty dr\,u_{2s-2}(r)ru_{3s}(r)+k^{-1}\int_0^\infty dr\int_0^\infty dr'u_{2s}(r)rG_1(kr_>)F_1(kr_<)r'u_{3s}(r')\right.$$
$$\left.-ik^{-1}\int_0^\infty dr\,F_1(kr)ru_{2s}(r)\int_0^\infty dr\,F_1(kr)ru_{3s}(r)\right]b_{2s}^*(t)b_{3s}(t), \tag{5d}$$

where b_{nl} are the slowly varying amplitudes defined by

$$b_{2s}=a_{2s}, \tag{6a}$$

$$b_{3d}=e^{-i\Delta_2t}a_{3d}, \tag{6b}$$

$$b_{3s}=e^{-i(\Delta_1+\Delta_2)t}a_{3s}. \tag{6c}$$

The contributions to $\gamma_1(t)$ and $\gamma_2(t)$ are given in Table I. The unperturbed radial orbitals (u_{nl}), first-order perturbed radial orbitals ($u_{nl\pm q}$, where $q=1,2$ refers to laser 1,2 for $+$ or $-$ frequency), and continuum radial orbitals are discussed in the Appendix.

In this work, the probability amplitudes [Eqs. (6)] are evaluated nonperturbatively by numerically solving Eq. (4); however, it is instructive to evaluate these amplitudes in the perturbative limit, in which case the gas polarization [Eq. (5a)] reduces to a familiar-looking expression in terms of the atomic nonlinear susceptibilities.[12] Thus

$$\mathbf{P}(\mathbf{r},t)=N[\chi_3\hat{\rho}_1E_1^3+\chi_5\hat{\rho}_2E_1^3E_2^2]e^{i(3\mathbf{k}_1\cdot\mathbf{r}-3\omega_1t)}, \tag{7}$$

for a third-order susceptibility χ_3 in $\mathrm{cm}^6/\mathrm{erg}$ and a fifth-order susceptibility χ_5 in $\mathrm{cm}^9/\mathrm{erg}^2$. In arriving at Eq. (7) we have used the following perturbative limits for the amplitudes:

$$b_{2s}\to1, \tag{8a}$$

$$b_{3d}\to\left[\frac{4a_0^3}{3\sqrt{5}\hbar\Delta_2}\int_0^\infty dr\,u_{3d}(r)ru_{2s+1}^{(1)}(r)\right]\frac{E_1^2}{4}, \tag{8b}$$

$$b_{3s}\to\left[\frac{16a_0^6}{45\hbar^2\Delta_1\Delta_2}\left[-k^{-1}\int_0^\infty dr\int_0^\infty dr'u_{3s}(r')G_1(kr_>)F_1(kr_<)r'u_{3d}(r')\right.\right.$$
$$\left.\left.-ik^{-1}\int_0^\infty dr\,F_1(kr)ru_{3s}(r)\int_0^\infty dr\,F_1(kr)ru_{3d}(r)\right]\int_0^\infty dr\,u_{3d}(r)ru_{2s+1}^{(1)}(r)\right]\frac{E_1^3E_2}{16}. \tag{8c}$$

The fifth-order term also depends on the factor $e^{i(3\mathbf{k}_1\cdot\mathbf{r}-3\omega_1t)}$ since the second-laser photon $\hbar\omega_2$ is emitted and reabsorbed in the transmission connecting the $3s$ and ϵp levels [Eq. (3)].

III. NUMERICAL RESULTS AND DISCUSSION

Although Eq. (4) can be solved exactly analytically, we choose to solve it numerically. Our results are shown in Figs. 1 and 2. The laser pulse is assumed to be square and of duration 5 ns. The intensities are 3.08 and 77.9 $\mathrm{MW\,cm^{-2}}$ for the first and second lasers, respectively. The ionization probability $P_I=1-|a_{2s}|^2-|a_{3d}|^2-|a_{3s}|^2$ is plotted at the bottom, and the magnitude of the induced dipole moment [magnitude of the terms in square brackets in Eq. (5a) with $\hat{\rho}_1=\hat{\rho}_2=1$], is plotted above it. In Fig. 1 these quantities are plotted versus Δ_2 for $\Delta_1=0.019\ \mathrm{cm}^{-1}$ and in Fig. 2 these quantities are plotted versus Δ_1 for $\Delta_2=0.01\ \mathrm{cm}^{-1}$. The dotted-dashed curve [see Eq. (5a)] is $|\gamma_1E_1|$, the dashed curve is

TABLE I. Contributions to the generalized polarizabilities given by Eqs. (5c) and (5d) (in units of $10^{-24}\ \mathrm{cm}^3$).

$\gamma_1(t)=(-6.439+7.200+1.363i)b_{2s}^*b_{3d}$
$\gamma_2(t)=(4.361-6.488-1.397i)b_{2s}^*b_{3s}$

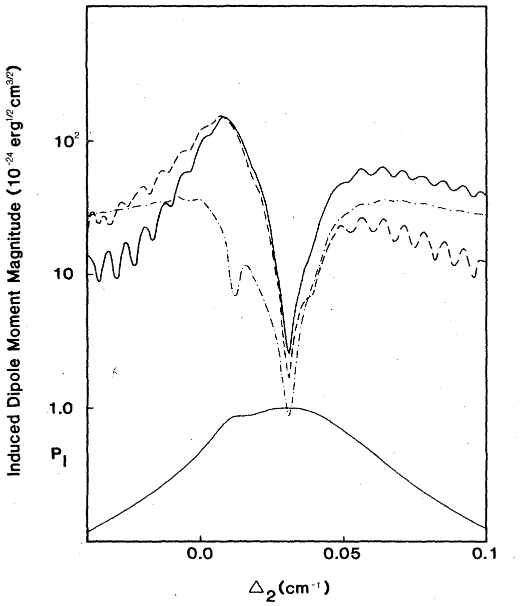

FIG. 1. Ionization and induced-dipole moment vs $\Delta_2=\omega_{3D}-\omega_{2S}-2\omega_1$ for $\Delta_1=\omega_{3S}-\omega_{3D}-\omega_1+\omega_2=0.019$ cm^{-1}; $I_1=3.08$ MW cm^{-2} and $I_2=77.9$ MW cm^{-2}.

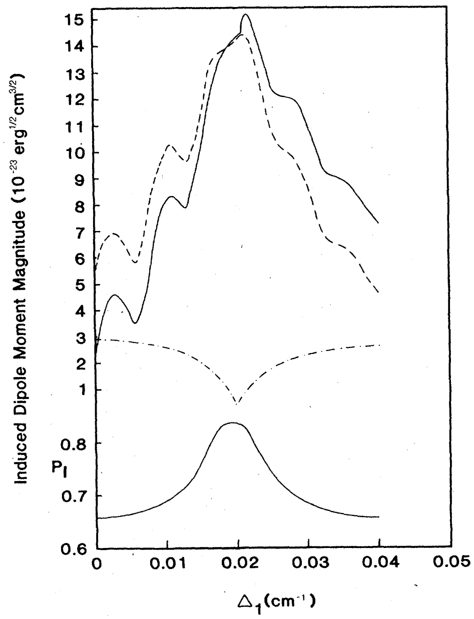

FIG. 2. Ionization and induced-dipole moment vs $\Delta_1=\omega_{3S}-\omega_{3D}-\omega_1+\omega_2$ for $\Delta_2=\omega_{3D}-\omega_{2S}-2\omega_1=0.01$ cm^{-1}; $I_1=3.08$ MW cm^{-2} and $I_2=77.9$ MW cm^{-2}.

$|\gamma_2E_2|$, and the solid curve is $|\gamma_1E_1+\gamma_2E_2|$. Other calculations show that when $|\Delta_1|$ is large, the induced-dipole moment magnitude reduces to $|\gamma_1E_1|$.

For $\Delta_1=0.019$ cm^{-1}, the squared magnitude of the induced dipole moment is enhanced by the growth of the γ_2E_2 contribution (Fig. 1 at $\Delta_2\simeq0.01$ cm^{-1}) near the point where the bump occurs in the ionization curve due to the "induced autoionizing resonance" by the second laser.[10,11] Then, near unit ionization probability, both contributions to the induced-dipole moment pass through a minimum. This is due simply to the smallness of the bound-state amplitudes at this point [Eqs. (5c) and (5d)]. Figure 2 shows a blow-up of the third-harmonic maximum at $\Delta_2\simeq0.01$ cm^{-1}. Now the plot versus Δ_1 clearly shows the induced autoionizing resonance peak in the ionization curve and the rapid growth of $|\gamma_2E_2|$. The structure in $|\gamma_1E_1+\gamma_2E_2|$ results from the interference of the two contributions γ_1E_1 and γ_2E_2. Note that the ionization curve here does not show the characteristic "Fano asymmetry" of previous calculations.[10,11] However, these were calculations of one-photon (rather than three-photon) ionization rates.

These results imply that THG can possibly be enhanced by the growth of the induced-dipole moment near the induced resonance. However, this question is a complicated one, and it requires the solution of Eq. (1) for wave propagation through the medium for an answer. This leads to the important problem of phase mismatch due to the refractive-index difference of the generated wave (at $3\omega_1$) and the incident wave (at ω_1). Phase mismatch is further complicated in the case of resonant absorption by population growth in excited states, a situation which has been studied recently by Puell *et al.*[20] for a saturated bound-bound transition. In our case the bound-to-continuum transition is nearly saturated so that the effect of this saturation on the phase mismatch should be carefully examined before any conclusions can be made on THG enhancement.

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APPENDIX

The matrix elements of Eq. (4) (in s^{-1}) are presented in this appendix. We have

$$S_{2s}=\frac{4\pi}{3}\alpha a_0^2F_1E_1\left[\int_0^\infty dr\,u_{2s}(r)ru_{2s+1}^{(1)}(r)+\int_0^\infty dr\,u_{2s}(r)ru_{2s-1}^{(1)}(r)\right]$$
$$+\frac{4\pi}{3}\alpha a_0^2F_2E_2\left[\int_0^\infty dr\,u_{2s}(r)ru_{2s+2}^{(1)}(r)+\int_0^\infty dr\,u_{2s}(r)ru_{2s-2}^{(1)}(r)\right], \tag{A1}$$

$$\tfrac{1}{2}\Omega_{3d,2s}=\tfrac{1}{2}\Omega_{2s,3d}=\frac{8\pi}{3\sqrt{5}}\alpha a_0^2F_1E_1\int_0^\infty dr\, u_{3d}(r)ru_{2s+1}^{(1)}(r)\ , \tag{A2}$$

$$\begin{aligned}S_{3d}=4\pi\alpha a_0^2F_1E_1\Big[&-\tfrac{4}{15}k^{-1}\int_0^\infty dr\int_0^\infty dr' u_{3d}(r)rG_1(kr_>)F_1(kr_<)r'u_{3d}(r')\\&-\tfrac{9}{35}k^{-1}\int_0^\infty dr\int_0^\infty dr' u_{3d}(r)rG_3(kr_>)F_3(kr_<)r'u_{3d}(r')\\&+\tfrac{4}{15}\int_0^\infty dr\, u_{3d}(r)ru_{3d-1}^{(1)}(r)+\tfrac{9}{35}\int_0^\infty dr\, u_{3d}(r)ru_{3d-1}^{(3)}(r)\Big]\\+4\pi\alpha a_0^2F_2E_2\Big[&-\tfrac{4}{15}k'^{-1}\int_0^\infty dr\int_0^\infty dr' u_{3d}(r)rG_1(k'r_>)F_1(k'r_<)r'u_{3d}(r')\\&-\tfrac{9}{35}k'^{-1}\int_0^\infty dr\int_0^\infty dr' u_{3d}(r)rG_3(k'r_>)F_3k'r_<)r'u_{3d}(r')\\&+\tfrac{4}{15}\int_0^\infty dr\, u_{3d}(r)ru_{3d-2}^{(1)}(r)+\tfrac{9}{35}\int_0^\infty dr\, u_{3d}(r)ru_{3d-2}^{(3)}(r)\Big]\ ,\end{aligned} \tag{A3}$$

$$\begin{aligned}\gamma_{3d}=8\pi\alpha a_0^2F_1E_1&\left[\tfrac{4}{15}k^{-1}\left[\int_0^\infty dr\,F_1(kr)ru_{3d}(r)\right]^2+\tfrac{9}{35}k^{-1}\left[\int_0^\infty dr\,F_3(kr)ru_{3d}(r)\right]^2\right]\\+8\pi\alpha a_0^2F_2E_2&\left[\tfrac{4}{15}k'^{-1}\left[\int_0^\infty dr\,F_1(k'r)ru_{3d}(r)\right]^2+\tfrac{9}{35}k'^{-1}\left[\int_0^\infty dr\,F_3(k'r)ru_{3d}(r)\right]^2\right]\ ,\end{aligned} \tag{A4}$$

$$\begin{aligned}\tfrac{1}{2}\Omega_{3s,3d}=\tfrac{1}{2}\Omega_{3d,3s}=&\frac{8\pi}{3\sqrt{5}}\alpha a_0^2(F_1E_1F_2E_2)^{1/2}\\&\times\Big[-k^{-1}\int_0^\infty dr\int_0^\infty dr' u_{3s}(r)G_1(kr_>)F_1(kr_<)r'u_{3d}(r)\\&+\int_0^\infty dr\, u_{3s}(r)ru_{3d-2}^{(1)}-ik^{-1}\int_0^\infty dr\,F_1(kr)ru_{3s}(r)\int_0^\infty dr\,F_1(kr)ru_{3d}(r)\Big]\ ,\end{aligned} \tag{A5}$$

$$\begin{aligned}S_{3s}=&\frac{4\pi}{3}\alpha a_0^2F_1E_1\left[\int_0^\infty dr\, u_{3s}(r)ru_{3s+1}^{(1)}+\int_0^\infty dr\, u_{3s}(r)ru_{3s-1}^{(1)}(r)\right]\\&+\frac{4\pi}{3}\alpha a_0^2F_2E_2\left[-k^{-1}\int_0^\infty dr\int_0^\infty dr' u_{3s}(r)rG_1(kr_>)F_1(kr_<)r'u_{3s}(r')+\int_0^\infty dr\, u_{3s}(r)ru_{3s-2}^{(1)}(r)\right]\ ,\end{aligned} \tag{A6}$$

$$\gamma_{3s}=\frac{8\pi}{3}\alpha a_0^2F_2E_2k^{-1}\left[\int_0^\infty dr\,F_1(kr)ru_{3s}(r)\right]^2\ , \tag{A7}$$

$$\frac{k^2}{2}=E_{3d}+E_1=E_{3s}+E_2\ , \tag{A8}$$

$$\frac{k'^2}{2}=E_{3d}+E_2\ , \tag{A9}$$

where α is the fine-structure constant, a_0 the Bohr radius, F_j the jth laser (with photon energy E_j in a.u.) flux in $\mathrm{cm^{-2}\,s^{-1}}$, u_{nl} are the unperturbed radial orbitals having energies E_{nl} in a.u., and F_l (G_l) are the regular (irregular) partial waves of the photoelectron. These values are used throughout although Eq. (4) is solved for various values of Δ_1 and Δ_2 (Figs. 1 and 2) about zero detuning, valid since the matrix elements are slowly varying with energy.

The perturbed orbitals $u_{nl,\pm q}$ (where q refers to laser 1 or laser 2) are calculated from the equation of first-order perturbation theory (in a.u.),

$$\left[\frac{d^2}{dr^2}-\frac{|l\pm1|(|l\pm1|+1)}{r^2}-2V_{\mathrm{SE}}(r)+2(E_{nl}\pm E_q)\right]u_{nl,\pm q}^{(|l\pm1|)}(r)=r^2u_{nl}(r)\ , \tag{A10}$$

where V_{SE} is the nonlocal static-exchange potential.[11]

The unperturbed and perturbed bound orbitals are calculated[11] in the static-exchange approximation for e,Li^+, and the continuum partial waves are calculated[11,19] in a local approximation to the static-exchange approximation for e,Li^+. The numerical values for each matrix of Eq. (5) are given in Table II. Note that terms involving $u_{2s+1}^{(1)}$

TABLE II. Matrix elements of Eq. (4) [see Eqs. (5)]. The coefficients of laser flux are in units of $10^{-17}\ \mathrm{cm}^2$. The terms in Eqs. (5) are grouped consecutively below (where the $l=1$ and $l=3$ contributions to S_{2d} and γ_{3d} have been combined).

$S_{2s}=(16.601-0.377)F_1+(2.973-0.421)F_2$
$\tfrac{1}{2}\Omega_{3d,2s}=16.6384F_1$
$S_{3d}=(10.335+11.487)F_1+(9.276-9.858)F_2$
$\gamma_{3d}=2.365F_1+0.632F_2$
$\tfrac{1}{2}\Omega_{3s,3d}=(-4.150+6.005+i0.569)(F_1F_2)^{1/2}$
$S_{3s}=(5.733-5.315)F_1+(5.472-4.666)F_2$
$\gamma_{3s}=0.656F_2$

are large since the laser-1 photon (1.925 eV) is almost in tune with the $2s,2p$ transition (1.841 eV in our atomic model). Despite this fact, the $u_{2s+1}^{(1)}$ perturbed orbital was found to be numerically stable against increasing the number of basis orbitals in the variational method[11] used to calculate it. See Ref. 11 for further discussion on the calculation of matrix elements of this form.

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