

Laser probe of the atomic ionization continuum: Stimulated recombination into an excited state

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A two-laser theory is formulated for the photoionization of Li atoms. The first laser is tuned above the ionization threshold of Li(2s). The second laser probes the ionized target. Stimulated recombination into Li(*ns*) occurs near the Raman-type resonance point $\epsilon_{ns} - \epsilon_{2s} - \epsilon_1 + \epsilon_2 = 0$ (where $n > 2$ and ϵ_q is a photon energy). Near the resonance point the ionization probability is characterized by a line profile which resembles that for an autoionizing resonance.

I. INTRODUCTION

There has been recent theoretical work on the effect of a strong radiation field on autoionizing states embedded in the photoelectron continuum.¹⁻⁴ There has also been recent theoretical⁵ experimental⁶ work on radiation-field "induced resonances" in an otherwise structureless continuum. Previous theoretical work¹⁻⁵ involves the generalization, for strong fields, of the theory of Fano⁷ for atomic autoionization.

In this paper we study the second of these two processes. Our aims include: (1) some study on the connection between the two processes and (2) use of quantitative quantum-mechanical methods to calculate the atomic parameters required by the theory. The latter is intended as a guide to experimentalists in the choice of a target gas and of laser wavelengths and power levels.

Our methods involve solving the time-dependent Schrödinger equation for two bound levels lying below an ionization continuum (Fig. 1). The two bound levels have the same parity and thus are not coupled directly by the dipole component of the radiation field. The first level, Li(2s), is ionized by one laser. A second laser stimulates ejected-electron recombination into the second level, Li(3s), and then reionizes the atom by absorption from

this level. What emerges is a Raman-type $2s \rightarrow 3s$ excitation (followed by $3s \rightarrow 2s$ deexcitation), with the first of the two photons in each case resonant with the ejected-electron $\epsilon_c p$ continuum (where ϵ_c is the continuum-electron energy) and the second photon in each case stimulating a downward transition into 3s (2s). The probability for ionization is calculated as a function of time and shows a characteristic autoionizinglike structure near the Raman-type resonance point $\epsilon_{3s} - \epsilon_{2s} - \epsilon_1 + \epsilon_2 = 0$ (where ϵ_{ns} is the level orbital energy and ϵ_q is the photon energy).

II. THEORY

The time-dependent Schrödinger equation for the Li atom in the presence of two lasers is solved in the usual way by expanding the wave function in the set of eigenstates of the target. We retain the 2s and 3s bound states and the complete set of *p* states. Further, the laser photons are energetic enough that each *ns* state is coupled directly to the $\epsilon_c p$ set of continuum states by one or both lasers. The probability amplitudes for the *p* states are eliminated by substitution into the equations for the *ns*-state amplitudes. Retaining only matrix elements with a low-frequency time variation, we derive the effective set

$$\begin{aligned}
 i\dot{a}_{2s} = & \sum_{j,q} \langle \psi_{2s} | \Omega_q | \psi_j \rangle \langle \psi_j | \Omega_q | \psi_{2s} \rangle \left[\frac{e^{i(\omega_{2s} - \omega_j + \omega_q)t}}{i} \int_0^t dt' e^{i(\omega_j - \omega_{2s} - \omega_q)t'} a_{2s}(t') \right. \\
 & \left. + \frac{e^{i(\omega_{2s} - \omega_j - \omega_q)t}}{i} \int_0^t dt' e^{i(\omega_j - \omega_{2s} + \omega_q)t'} a_{2s}(t') \right] \\
 & + \sum_{\bar{j}} \langle \psi_{2s} | \Omega_1 | \psi_{\bar{j}} \rangle \langle \psi_{\bar{j}} | \Omega_2 | \psi_{3s} \rangle \left[\frac{e^{i(\omega_{2s} - \omega_{\bar{j}} + \omega_1)t}}{i} \int_0^t dt' e^{i(\omega_{\bar{j}} - \omega_{3s} - \omega_2)t'} a_{3s}(t') \right. \\
 & \left. + \frac{e^{i(\omega_{2s} - \omega_{\bar{j}} - \omega_2)t}}{i} \int_0^t dt' e^{i(\omega_{\bar{j}} - \omega_{3s} + \omega_1)t'} a_{3s}(t') \right], \tag{1a}
 \end{aligned}$$

$$\Omega_q = (2\pi F_q \alpha \omega_q)^{1/2} \hat{\rho}_q \cdot \vec{r}, \tag{1b}$$

and similarly for \dot{a}_{3s} , interchanging 2s and 3s and interchanging the photon labels. The q th laser has flux F_q $\text{cm}^{-2}\text{s}^{-1}$, frequency ω_q , and polarization direction $\hat{\rho}_q$; α is the fine structure constant, \vec{r} is the electron position, and the summation over j runs over the complete set of p states. In the cases where $\hbar\omega_q$ exceeds the ionization energy of either ns state (i.e., where $\omega_{ns} + \omega_q > 0$), use of scattered-wave boundary conditions on the intermediate set requires that we integrate over continuum-electron momenta $\hbar\vec{k}$, where $\omega_j = \hbar k^2 / (2m)$. The time integrations in Eq. (1a) are done by parts. For example, for $q=1$, one of the diagonal terms is given by

$$(2\pi)^{-3} \int d\vec{k} \langle \psi_{2s} | \Omega_1 | \psi_{\vec{k}} \rangle \langle \psi_{\vec{k}} | \Omega_1 | \psi_{2s} \rangle \frac{e^{i[\omega_{2s} - (\hbar k^2)/(2m) + \omega_1]t}}{i} \int_0^t dt' e^{i[(\hbar k^2)/(2m) - \omega_{2s} - \omega_1]t'} a_{2s}(t')$$

$$= (2\pi)^{-3} \int d\vec{k} \langle \psi_{2s} | \Omega_1 | \psi_{\vec{k}} \rangle \langle \psi_{\vec{k}} | \Omega_1 | \psi_{2s} \rangle \left[\omega_{2s} - \frac{\hbar k^2}{2m} + \omega_1 \right]^{-1}$$

$$\times \left[a_{2s}(t) - e^{i[\omega_{2s} - (\hbar k^2)/(2m) + \omega_1]t} \left[1 + \int_0^t dt' e^{i[(\hbar k^2)/(2m) - \omega_{2s} - \omega_1]t'} \dot{a}_{2s}(t') \right] \right], \quad (2)$$

where $\psi_{\vec{k}}$ is the continuum wave function, and similarly for all of the terms on the right-hand sides (rhs) of Eq. (1a) and of the equation for \dot{a}_{3s} . We obtain an approximate set by retaining only the first terms on the right-hand sides of the equations for \dot{a}_{2s} and \dot{a}_{3s} . The terms which are dropped contain factors of the form $\exp[i(\hbar/2m)(k_{nq}^2 - k^2)t]$ [where $(\hbar k_{nq}^2)/(2m) = \omega_{ns} + \omega_q$] or $\exp[i(\omega_{\pm nq} - \omega_j)t]$ (where $\omega_{\pm nq} = \omega_{ns} \pm \omega_q$). Rapid oscillation of these factors is expected to produce strong cancellation in the integrations over k (for a fixed t where resonance occurs at $k = k_{nq}$) or over t (for fixed $\omega_{\pm nq} - \omega_j$ where no resonance occurs for any $\omega_{\pm nq}$ or ω_j , i.e., $\omega_j \neq \omega_{\pm nq}$). These terms are thus expected to be much smaller than the terms which are retained. However, since the continuum contributions are brought into resonance at $k = k_{nq}$, the validity of dropping these terms is examined quantitatively below.

Integration of the retained continuum contributions over \vec{k} gives the Green's function for an effective e, Li^+ potential,

$$G_{nq}^{(+)} = \lim_{\eta \rightarrow 0} (2\pi)^{-3} \int d\vec{k} \frac{\psi_{\vec{k}}(\vec{r}) \psi_{\vec{k}}^*(\vec{r}')}{(k_{nq}^2 - k^2) + i\eta}$$

$$= -\frac{(4\pi)^{-1}}{krr'} \sum_l [G_l(k_{nq}r_>) F_l(k_{nq}r_<)]$$

$$+ iF_l(k_{nq}r) F_l(k_{nq}r') P_l(\hat{r} \cdot \hat{r}'), \quad (3)$$

where for $n=2, q=1$ and for $n=3, q=1$ and 2 (Fig. 1), outgoing boundary conditions are used for $G_{nq}^{(+)}$, and G_l and F_l are, respectively, the irregular and regular radial partial waves for the ejected electron. All other $ns \rightarrow p$

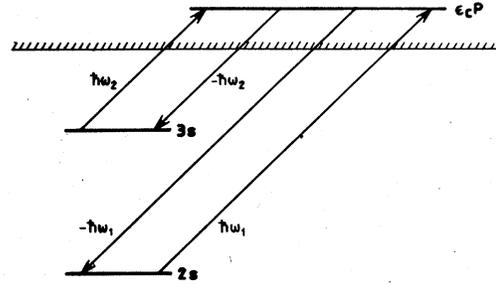


FIG. 1. Level scheme showing the continuum contributions for $k = k_{21} = k_{32}$.

transitions are virtual [i.e., $(\omega_{ns} \pm \omega_q) < 0$], and we have evaluated the summations over j implicitly by the differential equations technique. That is, the equations (in a.u.) from first-order perturbation theory,

$$[\nabla^2 - 2V_{SE}(r) + 2(E_{ns} \pm E_q)] \psi_{ns \pm q} = \hat{\rho}_q \cdot \vec{r} \psi_{ns}, \quad (4)$$

are solved (where E_{ns} and E_q are, respectively, the ns orbital and photon energies in a.u.). The radial continuum waves of Eq. (3) are calculated using a local potential for e, Li^+ described in a previous paper.⁸ The unperturbed orbitals ψ_{ns} we obtained as the second and third eigenvectors in a Ritz variational calculation using the nonlocal e, Li^+ static-exchange potential V_{SE} and ten S -symmetry Slater-type basis members. This calculation was checked by comparison with a previous calculation⁹ using the same potential. The radial parts of the perturbed orbitals in Eq. (4) were calculated by expanding $\psi_{ns \pm q}$ in ten P -symmetry Slater-type basis members and use of matrix inversion. This calculation was checked by using ψ_{2s+2} to calculate the two-photon ionization cross section for Li and comparison with a previous calculation.¹⁰

Thus we arrive at an approximate but exactly soluble set of equations for the bound-state probability amplitudes

$$i \begin{bmatrix} \frac{d}{dt} & 0 \\ 0 & \frac{d}{dt} \end{bmatrix} \begin{bmatrix} a_{2s} \\ a_{3s} \end{bmatrix} = \begin{bmatrix} S_{2s} - \frac{i}{2} \gamma_{2s} & \frac{1}{2} \Omega_{2s3s} e^{-i\Delta t} \\ \frac{1}{2} \Omega_{3s2s} e^{i\Delta t} & S_{3s} - \frac{i}{2} \gamma_{3s} \end{bmatrix} \begin{bmatrix} a_{2s} \\ a_{3s} \end{bmatrix}. \quad (5)$$

Ω_{ij} is an effective Rabi frequency for the indirect 2s-3s coupling

$$\begin{aligned} \frac{1}{2}\Omega_{2s3s} = & \frac{4}{3}\pi\alpha a_0^2(F_1E_1F_2E_2)^{1/2} \left[-k_{32}^{-1} \int_0^\infty dr \int_0^\infty dr' u_{2s}(r)rG_1(k_{32}r_>)F_1(k_{32}r_<)r'u_{3s}(r') \right. \\ & + \int_0^\infty dr u_{2s}(r)ru_{3s-1}(r) - ik_{32}^{-1} \int_0^\infty dr F_1(k_{32}r)ru_{2s}(r) \\ & \left. \times \int_0^\infty dr F_1(k_{32}r)ru_{3s}(r) \right] = S_0 + \frac{i}{2}\gamma_0 \end{aligned} \quad (6)$$

and similarly for $\frac{1}{2}\Omega_{3s2s}$ (replacing k_{32} by k_{21} , u_{3s-1} by u_{2s-2} , and interchanging u_{2s} and u_{3s}). $\Omega_{2s3s} = \Omega_{3s2s}$ on resonance ($\Delta = \omega_{3s} - \omega_{2s} - \omega_1 + \omega_2 = 0$); for the narrow range of detuning shown in Figs. 3 and 4, $\Omega_{2s3s} \cong \Omega_{3s2s} = \Omega$ and $k_{32} \cong k_{21} = k$.

The diagonal terms in Eq. (5) are proportional to the frequency-dependent polarizabilities of the ns states. The real part is the ac Stark shift of the level,

$$\begin{aligned} S_{2s} = & \frac{4\pi}{3}\alpha a_0^2 F_1 E_1 \left[-k^{-1} \int_0^\infty dr \int_0^\infty dr' u_{2s}(r)rG_1(kr_>)F_1(kr_<)r'u_{2s}(r') + \int_0^\infty dr u_{2s}(r)ru_{2s-1}(r) \right] \\ & + \frac{4\pi}{3}\alpha a_0^2 F_2 E_2 \left[\int_0^\infty dr u_{2s}(r)ru_{2s+2} + \int_0^\infty dr u_{2s}(r)ru_{2s-2} \right], \end{aligned} \quad (7a)$$

$$\begin{aligned} S_{3s} = & \frac{4\pi}{3}\alpha a_0^2 F_1 E_1 \left[-k'^{-1} \int_0^\infty dr \int_0^\infty dr' u_{3s}(r)rG_1(k'r_>)F_1(k'r_<)r'u_{3s}(r') + \int_0^\infty dr u_{3s}(r)ru_{3s-1}(r) \right] \\ & + \frac{4\pi}{3}\alpha a_0^2 F_2 E_2 \left[-k^{-1} \int_0^\infty dr \int_0^\infty dr' u_{3s}(r)rG_1(kr_>)F_1(kr_<)r'u_{3s}(r') + \int_0^\infty dr u_{3s}(r)ru_{3s-2}(r) \right], \end{aligned} \quad (7b)$$

$$\frac{k'^2}{2} = E_{3s} + E_1, \quad (7c)$$

and the imaginary part is one half the level photoionization rate,

$$\gamma_{2s} = \frac{8\pi}{3}\alpha a_0^2 F_1 E_1 k^{-1} \left[\int_0^\infty dr F_1(kr)ru_{2s}(r) \right]^2, \quad (8a)$$

$$\begin{aligned} \gamma_{3s} = & \frac{8\pi}{3}\alpha a_0^2 F_1 E_1 k'^{-1} \left[\int_0^\infty dr F_1(k'r)ru_{3s}(r) \right]^2 \\ & + \frac{8\pi}{3}\alpha a_0^2 F_2 E_2 k^{-1} \left[\int_0^\infty dr F_1(kr)ru_{3s}(r) \right]^2. \end{aligned} \quad (8b)$$

The justification for the approximate treatment of the bound-continuum coupling in the original set of equations [Eq. (1)] has been discussed in the literature.¹¹ From the theory of optical double resonance for three bound levels,¹²⁻¹⁴ however, we know that equations describing the time evolution of three bound-state probability amplitudes cannot be reduced to an effective 2×2 set analogous to Eq. (5), except when the second laser is a *weak* probe. This suggests that terms on the rhs of Eq. (2) cannot be dropped, for example, when a bound level (i.e., an autoionizing resonance¹⁻⁴) is embedded in the continuum. The terms which have been dropped in arriving at Eq. (5) have the form

$$R(t) = \frac{2}{\pi} \int_0^\infty dq q^2 \frac{\psi_1(qr)\psi_1(qr')}{(k_0^2 - q^2)} e^{i(k^2 - q^2)t/2} f(q, t). \quad (9)$$

The function $f(q, t)$ is unknown since it depends on the amplitudes. This suggests an iteration procedure, where $f(q, t)$ is 0 or 1 [see Eq. (2)] in the zeroth iteration. The retained terms behave as $R(0)$ [for $f(q, 0) = 1$] for all times. Thus it is appropriate to compare results for $R(0)$ and $R(t)$ [for $f(q, t) = 1$ in the zeroth iteration]. The approximations leading to Eq. (5) will be justified (at least in

the zeroth iteration) if $|R(t)| \ll |R(0)|$. Figure 2 shows results for $|R(t)|$ versus potential-well width a , where for purposes of this analysis the p waves ψ_1 are calculated in a square well of strength $V_0 = 3.53$ a.u. for $k_0 = 0.3a_0^{-1}$, $r = 0.5a_0$, and $r' = 1a_0$. This potential is

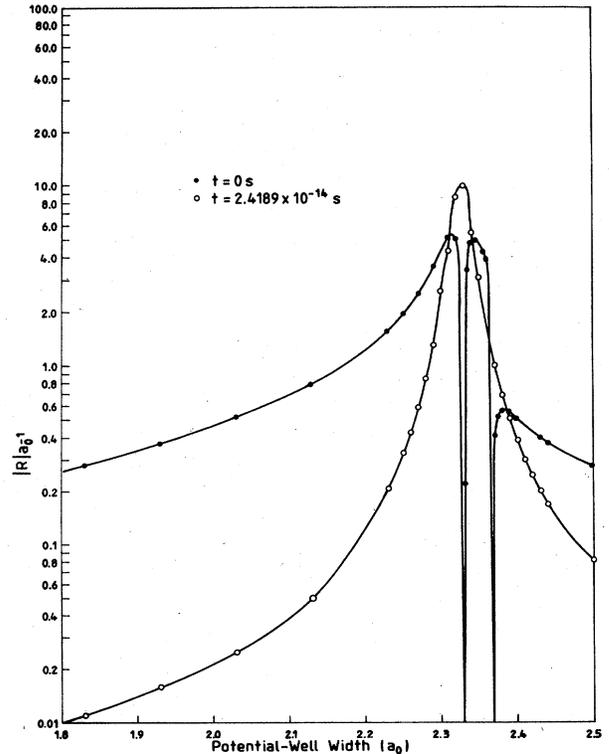


FIG. 2. $|R(t)|$ vs potential-well width for $t = 0$ and 1 a.u.

known¹⁵ to produce a p -wave shape resonance near $ka=0.7$ (or $a=2.33a_0$). The inequality is seen to hold fairly well for small or large a , i.e., as the p wave approaches nonresonant behavior. The accuracy of the numerical integration used to evaluate Eq. (9) is checked at $t=0$ and $V_0=0$, in which case

$$R(0)=k_0 j_1(k_0 r) n_1(k_0 r') . \quad (10)$$

This result is proved analytically by Lambropoulos and

$$|a_{2s}|^2 = e^{-t(\gamma_{2s} + \gamma_{3s})/2} \left[P_1(t) + \frac{(\Delta - \Delta S)^2 + (\Delta \gamma)^2}{\Delta_1^2} P_2(t) - \Delta_1^{-2} P_3(t) \right], \quad (11a)$$

$$|a_{3s}|^2 = e^{-t(\gamma_{2s} + \gamma_{3s})/2} \frac{|\Omega|^2}{\Delta_1^2} P_2(t), \quad (11b)$$

$$P_1(t) = \cos^2(\frac{1}{2}\Omega_+ t) \cosh^2(\frac{1}{2}\Omega_- t) + \sin^2(\frac{1}{2}\Omega_+ t) \sinh^2(\frac{1}{2}\Omega_- t), \quad (11c)$$

$$P_2(t) = \sin^2(\frac{1}{2}\Omega_+ t) \cosh^2(\frac{1}{2}\Omega_- t) + \cos^2(\frac{1}{2}\Omega_+ t) \sinh^2(\frac{1}{2}\Omega_- t), \quad (11d)$$

$$P_3(t) = \{ [\Theta(x) - \Theta(-x)] (\Delta - \Delta S) \Omega_+ + \frac{1}{2} \Delta \gamma \Omega_- \} \sinh \Omega_- t \\ - \{ [\Theta(x) - \Theta(-x)] (\Delta - \Delta S) \Omega_- - \frac{1}{2} \Delta \gamma \Omega_+ \} \sin \Omega_+ t, \quad (11e)$$

$$\Delta S = S_{2s} - S_{3s}, \quad (11f)$$

$$\Delta \gamma = \gamma_{2s} - \gamma_{3s}, \quad (11g)$$

$$\Delta_1^2 = \left[\left[(\Delta - \Delta S)^2 + \frac{(\Delta \gamma)^2}{4} \right]^2 + |\Omega|^4 + 2 \left[(\Delta - \Delta S)^2 - \frac{(\Delta \gamma)^2}{4} \right] [(4S_0^2 - \gamma_0^2)] + 8(\Delta - \Delta S) \Delta \gamma S_0 \gamma_0 \right]^{1/2}, \quad (11h)$$

$$\Omega_{\pm} = \sqrt{\frac{1}{2}} \left[\Delta_1^2 \pm \left[(\Delta - \Delta S)^2 - \frac{(\Delta \gamma)^2}{4} + 4S_0^2 - \gamma_0^2 \right] \right]^{1/2}, \quad (11i)$$

$$x = \frac{(\Delta - \Delta S) \Delta \gamma + 4S_0 \gamma_0}{|(\Delta - \Delta S)^2 - (\Delta \gamma)^2/4 + 4S_0^2 - \gamma_0^2|}, \quad (11j)$$

where $\Theta(x)$ is the usual unit-step function.

III. RESULTS AND DISCUSSION

Table I shows the values of the matrix elements of Eq. (5) as coefficients (in units of cm^2) of F_q (diagonally) or of $(F_1 F_2)^{1/2}$ nondiagonally. The various contributions have the same order as given in Eqs. (6)–(8). Note the opposite signs in the absorption and emission contributions, tending to give a significantly smaller result for the shifts and the coupling term than obtained when the emission contributions (which are nonresonant) are omitted. The nonresonant contributions to the shifts, occurring for virtual absorption and emission below the ionization threshold, are as important as the contributions which are in resonance with the continuum. For example, the coefficient of F_2 in S_{2s} is wholly nonresonant. This result was surprising at first since it contradicted our first inclination to consider only the resonant continuum contributions to the process. This result should be understood by considering that the resonant contributions for transitions to the continuum are obtained as residues at the poles which occur in the k integrations. Thus they are not resonant in the usual sense of bound-bound transitions

Zoller.² See their Eqs. (3.22), (3.25a), and (3.26). When the detuning δ from the autoionizing resonance is large, their s and γ , in Eq. (3.22) reduce to the constant shift and width for the coupling of a bound level to a nonresonant continuum [such that Eq. (3.22) then gives a result identical to that obtained for a_{2s} or a_{3s} from our Eq. (5) when we set $\frac{1}{2}\Omega_{2s3s}=0$].

Equation (5) is exactly soluble and the bound-state probabilities are given by

due to the severe cancellation which results through the poles in the k integrations. This result was checked numerically by independently obtaining the continuum contributions from Eq. (4) for positive $E_{ns} + E_q$ (the continuum energy) and evaluation of an integral of the type $\int_0^\infty dr u_{ns}(r) r u_{ns+q}$ [as used in the nonresonant contributions in Eqs. (6) and (7)]. Although the Eq. (4) solution has exponentially decaying boundary conditions and does not give an imaginary contribution, this procedure

TABLE I. Matrix elements of Eq. (5) [see Eqs. (6)–(8)]. The coefficients of laser flux are in units of cm^2 .

$$S_{2s} = (1.000 \times 10^{-17} - 0.581 \times 10^{-17}) F_1 \\ + (1.663 \times 10^{-17} - 0.471 \times 10^{-17}) F_2 \\ \gamma_{2s} = 0.187 \times 10^{-17} F_1 \\ S_{3s} = (5.019 \times 10^{-17} - 4.553 \times 10^{-17}) F_1 \\ + (5.456 \times 10^{-17} - 4.518 \times 10^{-17}) F_2 \\ \gamma_{3s} = 0.019 \times 10^{-17} F_1 + 0.474 \times 10^{-17} F_2 \\ \frac{1}{2}\Omega = (-0.646 \times 10^{-17} + 0.5413 \times 10^{-17} \\ - i0.149 \times 10^{-17}) (F_1 F_2)^{1/2}$$

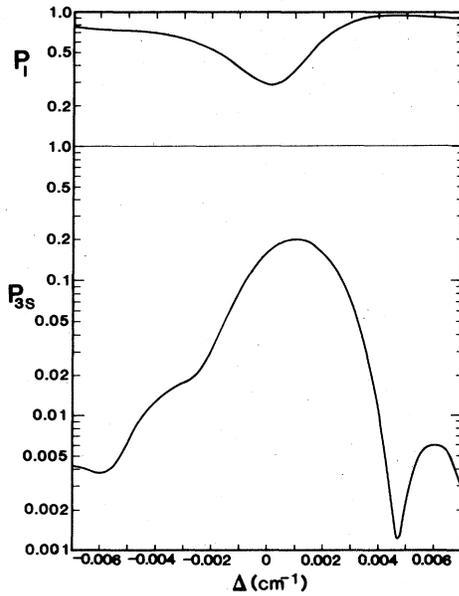


FIG. 3. Ionization probability vs Δ . $F_1=10^{26} \text{ cm}^{-2} \text{ s}^{-1}$ ($I_1=105 \text{ MW cm}^{-2}$); $F_2=10^{26} \text{ cm}^{-2} \text{ s}^{-1}$ ($I_2=51.6 \text{ MW cm}^{-2}$). The photon energies are $\epsilon_1=6.566 \text{ eV}$ and $\epsilon_2=3.222 \text{ eV}$. The photoelectron energy is $\epsilon_c=1.224 \text{ eV}$.

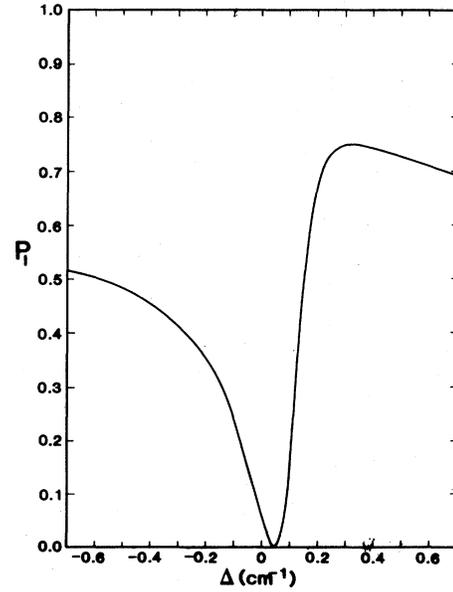


FIG. 4. Ionization probability vs Δ . $F_1=10^{26} \text{ cm}^{-2} \text{ s}^{-1}$; $F_2=10^{28} \text{ cm}^{-2} \text{ s}^{-1}$. The other quantities are the same as in Fig. 3.

nevertheless gives contributions to the shifts which agree to within a few percent with those obtained from the use of continuum orbitals and the evaluation of the k integrals by the principal-value theorem.

Equations (11) have been evaluated for $k=0.3a_0^{-1}$ (corresponding to 1.224 eV continuum energy) and $F_1=10^{26} \text{ cm}^{-2} \text{ s}^{-1}$ ($I_1=105 \text{ MW cm}^{-2}$), $F_2=10^{26} \text{ cm}^{-2} \text{ s}^{-1}$ ($I_2=51.6 \text{ MW cm}^{-2}$) in Fig. 3; and $F_1=10^{26} \text{ cm}^{-2} \text{ s}^{-1}$, $F_2=10^{28} \text{ cm}^{-2} \text{ s}^{-1}$ in Fig. 4, for an assumed square pulse of width 10 and 5 ns, respectively. For the last set of values, analysis shows that Eq. 11(a) is given approximately by

$$|a_{2s}|^2 \simeq e^{-t(\gamma_{2s} + \gamma_{3s})/2} e^{\pm \Omega_- t}, \quad (12)$$

where $+$ is taken if $\Delta\gamma < 0$ (as it is here) and $-$ is taken if $\Delta\gamma > 0$ [occurring for large and small F_2 relative to F_1 , respectively; see Eq. 11(g)]. Essentially, this result is the product of a very small and very large factor to give a probability fairly close to the exact result given by Eq. 11(a). For reasonably large $|\Delta - \Delta S|$ (magnitude of the dynamic detuning), Ω_- is given approximately by [see Eqs. 11(h) and 11(i)]

$$\Omega_- \simeq \frac{1}{\sqrt{2}} \left[\frac{(\Delta\gamma)^2}{2} + \frac{4\Delta\gamma S_0 \gamma_0}{\Delta - \Delta S} \right]^{1/2}. \quad (13)$$

On the far wings of the resonance, therefore, $\Omega_- \rightarrow \frac{1}{2}(\gamma_+ - \gamma_-)$ [where γ_+ (γ_-) is the greater (lesser) of γ_{2s} or γ_{3s}], such that $|a_{2s}|^2 \rightarrow e^{-\gamma_{2s} t}$, since $\gamma_+ = \gamma_{3s}$ for $\Delta\gamma < 0$ [$+$ sign in Eq. (12)] or $\gamma_+ = \gamma_{2s}$ for $\Delta\gamma > 0$ [$-$ sign in Eq. (12)]. Near resonance (i.e., when $\Delta - \Delta S = 0$), Ω_- goes through a minimum for the $F_2 \gg F_1$ case. For this minimum in Ω_- , $|a_{2s}|^2$ goes

through a minimum, and the ionization probability $P_I = 1 - |a_{2s}|^2 - |a_{3s}|^2$ goes through a maximum. $|a_{3s}|^2$ remains very small for all cases studied, except that for $F_2 \simeq F_1$, where its maximum is about 0.20 at $\Delta - \Delta S = 0$ (Fig. 3). This is the only example of "trapping" the electron in the 3s state. For all other cases F_2 is sufficiently large so that, upon recombination into 3s, the atom is immediately reionized by absorption by the 3s electrons. Thus in Fig. 4, probability is exchanged primarily between 2s and the $\epsilon_c P$ continuum. P_I could reach 1 for sufficiently small Ω_- near $\Delta - \Delta S = 0$ because

$$|a_{2s}|^2 \simeq e^{-t(\gamma_{2s} + \gamma_{3s})/2} \simeq e^{-t\gamma_{3s}/2} \simeq 0,$$

since $\gamma_{3s} \gg \gamma_{2s}$. Essentially, the 2s ionization probability is enhanced upon recombination into 3s because the 3s ionization-rate constant is much larger than that for 2s.

The occurrence of the minimum in Fig. 4 can also be understood from the behavior of Ω_- . Ω_- passes through a maximum to the left of the P_I maximum (Ω_- minimum), causing $|a_{2s}|^2$ to pass through a maximum and P_I through a minimum.

Thus for $F_2 \gg F_1$, the ionization probability is given approximately by $P_I \simeq 1 - |a_{2s}|^2$, and the 2s probability is given approximately by Eq. (12). Differentiating P_I with respect to time gives an ionization rate which depends on a single rate constant,

$$k_I = \frac{1}{2}(\gamma_{2s} + \gamma_{3s}) \mp \Omega_-. \quad (14)$$

In general, the ionization rate is more complicated.

IV. CONCLUSIONS

Our conclusions can be summarized as follows. First, the polelike approximation¹¹ which contributes to an exactly soluble set of equations for two bound levels radiatively coupled through the ionization continuum [Eqs. (11)] appears to be reasonably accurate (Fig. 2) when the continuum wave function is nonresonant. Otherwise the theory is analogous to the Autler-Townes theory¹²⁻¹⁴ for double resonance involving three bound levels. Second, the details of the "induced resonance" (Figs. 3 and 4) depend critically on the relative power levels of the two lasers. For example, in Fig. 3 (for $F_2 \simeq F_1$), significant population (up to 0.20) is temporarily "trapped" in the excited state; however, in Fig. 4 (for $F_2 \gg F_1$) the excited-

state population remains very small for all times due to its very rapid ionization by the second laser. Third, as a consequence of rapid excited-state ionization for $F_2 \gg F_1$, the theory reduces to a simple exponential decay law characterized by a single rate constant [Eq. (14)]. When $F_2 \simeq F_1$, the population behavior with time is more complicated, and, in fact, damped Rabi oscillations (not shown here) are observed.

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