Theory of laser-induced transitions between autoionizing states of He

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We present a theory and calculations of transitions between autoionizing states in He. A ^1P resonance is assumed excited by a weak source while a strong laser couples it to other autoionizing states. We calculate the total ionization probability, the line shape, and their dependence on the intensity of the laser. Possible experimental verification is also assessed.

I. INTRODUCTION

Although transitions from bound states to autoionizing states have been the subject of extensive investigations over more than three decades now, there is essentially nothing known about transitions between autoionizing states. Until recently, this question would have been of only academic interest since the excitation of such states by traditional (weak) radiation sources or electron beams does not allow the observation of such transitions. The autoionizing states simply decay (autoionize) too fast for these excitation schemes to be able to cause an additional transition within a time of the order of 10^{-12} sec, or much less in the usual case. The question has, however, now become very pertinent in view of experiments in multiphoton excitation of autoionizing states under strong lasers.[1-4] The available intensity is more than sufficient to cause such transitions. In addition, the tunability available in present day lasers and the multiphoton character of the excitation allows the selective excitation of such transitions. There exists indirect experimental evidence that transitions between autoionizing states may have played a significant role in recent multiphoton experiments on Sr.[1,2]

In this paper we address this question in the simplest possible context which allows the direct evaluation of the relevant probabilities. We have considered transitions from the lowest $^1P_1$ autoionizing state of the He atom to the first higher group of $^1D_2$ states. To keep the context as simple as possible, we assume that the $^1P_1$ state is excited either by a *weak* source (e.g., synchrotron) via a single-photon transition or by a multiphoton transition of the appropriate order (odd). Excitation by electron scattering is also, at least in principle, possible, but an examination of the conditions for the observability of this process shows that it would be highly impractical. The second excitation is assumed to be induced by a laser whose frequency range and necessary intensity are discussed in detail later on. An experiment along similar lines in Ne^- had been reported[5] some years ago, but to the best of our knowledge its results remain inconclusive. In view of the fact that the lower Ne^- resonance in that experiment was excited by electron scattering, the enormity of the difficulty in exciting a higher resonance with a laser is compatible with our findings.

In the following sections we present a brief review of the formalism[6] needed for our calculation, a description of the method[7,8] employed in the construction of the wave functions, and our results, together with an evaluation of their observability and relevance to experiments on other atoms.

II. FORMULATION

The formalism employed in this paper is based on previous work[6] developed for problems of this type. Thus we can be brief in establishing the necessary equations. To keep the formal equations as simple as possible, we assume that the first excitation is caused by single-photon absorption. The modifications that would be necessary for other forms of excitation are straightforward extensions and will be indicated later on.

Let $|g\rangle$ be the ground state of the atom, $|a\rangle$ the first autoionizing state, and $|b\rangle$ the second. An electric dipole transition induced by linearly polarized photons of frequency ω_1 causes the excitation $g \rightarrow a$, while a second photon of frequency ω_2 (also linearly polarized along the same direction) causes the transition $a \rightarrow b$. Each of the autoionizing states a and b autoionizes to a continuum. The presence of the laser photon ω_2, however, can induce additional transitions to these continua as well as to other continua which would not enter the problem under the usual conditions of autoionization. There are four such continua in this problem. Although not all of them are equally important in our calculations, in developing the formalism all four are included; in the specific application we decide which are the dominant ones.

The total Hamiltonian of the problem is written as

$$H = H_0 + V + H_R + D \ , \tag{2.1}$$

where $H_0+V=H_A$ is the atomic part, H_R is the free radiation part, and D is the dipole interaction between the two. In terms of the creation and annihilation operators for a single mode, D is given by $a_k(\epsilon_k\cdot\mathbf{r})-a_k^\dagger(\epsilon_k^*\cdot\mathbf{r})$ where ϵ_k is the polarization vector of the photon mode $\mathbf{k}$. We need not be concerned with multimode and bandwidth aspects here. It is sufficient to assume that two modes of the field, having the same polarization and propagating in the same direction, are occupied with occupation numbers n_1 and n_2 corresponding to the frequencies ω_1 and ω_2. We denote the continua by c_l where l designates the orbital angular momentum (partial wave) of the continuum state. The "atom plus field" states entering the problem are

$$|\bar{g}\rangle=|g\rangle|n_1,n_2\rangle=|g;n_1,n_2\rangle\ ,$$
$$|\bar{a}\rangle=|a;n_1-1,n_2\rangle\ ,$$
$$|\bar{b}\rangle=|b;n_1-1,n_2\pm1\rangle\ ,$$
$$|\bar{c}_0\rangle=|c_0;n_1-1,n_2+1\rangle\ ,$$
$$|\bar{c}'_0\rangle=|c_0,;n_1-1,n_2-1\rangle\ ,$$
$$|\bar{c}_1\rangle=|c_1;n_1-1,n_2\rangle\ ,$$
$$|\bar{c}'_1\rangle=|c_1;n_1-1,n_2-2\rangle\ ,$$
$$|\bar{c}''_1\rangle=|c_1;n_1-1,n_2+2\rangle\ ,$$
$$|\bar{c}_2\rangle=|c_2;n_1-1,n_2+1\rangle\ ,$$
$$|\bar{c}'_2\rangle=|c_2;n_1-1,n_2-1\rangle\ ,$$
$$|\bar{c}_3\rangle=|c_3;n_1-1,n_2\rangle\ ,$$
$$|\bar{c}'_3\rangle=|c_3;n_1-1,n_2-2\rangle\ .$$

The transitions through which the above states enter the problem are shown in Fig. 1. The respective energies are given by the sum of the atomic and corresponding field energies, e.g., $\bar{E}_g=E_g+n_1\hbar\omega_1+n_2\hbar\omega_2$, $\bar{E}_{c_0}=E_{c_0}+(n_1-1)\hbar\omega_1+(n_2+1)\hbar\omega_2$, etc. The continuum states $\bar{c}_l$ and $\bar{c}'_l$ involve the same partial wave but differ in energy because different numbers of photons are absorbed and emitted in reaching these states as illustrated in Fig. 1. For example, the $l=2$ partial wave can be reached from state $|a\rangle$ either by absorption or by stimulated emission of a photon ω_2. These additional paths become significant because the intensity of the laser can make such induced processes as strong as or stronger than autoionization.

We introduce the resolvent operator $G(z)=(z-H)^{-1}$ and the projection operators P and $Q=1-P$ with P defined by

$$P=P_1+P_2-P_1P_2\ , \tag{2.2a}$$

where

$$P_i=|\phi_{1s}(i)\rangle\langle\phi_{1s}(i)|\ ,\quad i=1,2 \tag{2.2b}$$

and $\phi_{1s}(i)$ is the hydrogenic wave function (with $Z=2$) for the ith electron. We evaluate the corresponding time evolution operator $U(t)$ whose matrix elements are denoted by $U_{\bar{a}\bar{g}}, U_{\bar{b}\bar{g}}$, etc. In terms of these matrix elements, the ionization probability is given by

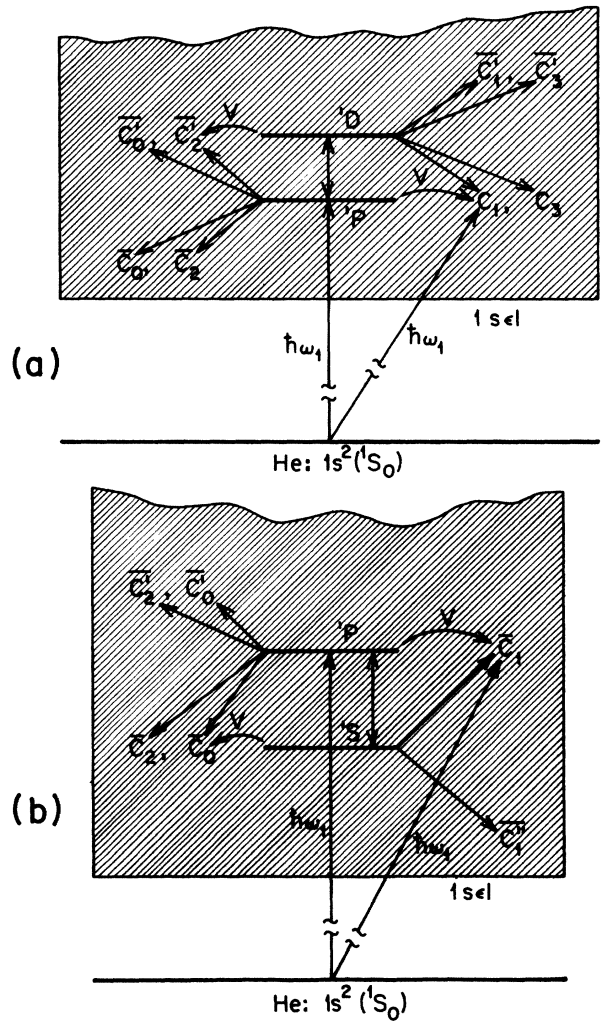

FIG. 1. Schematic representation of the coupling between the ground, the autoionizing states, and the continua entering the problem in each case. The arrow marked $\hbar\omega_1$ represents the weak source photon. All other arrows represent laser photons.

$$W=1-|U_{\bar{g}\bar{g}}(T)|^2\ , \tag{2.3}$$

where T is the time of interaction between atom and field, which in our calculation will be understood to represent the laser pulse duration. The Hamiltonian H_A of the atom is now partitioned as follows:

$$H_A=\tilde{H}_0+\tilde{V}\ , \tag{2.4a}$$
$$\tilde{H}_0=PH_AP+QH_AQ\ , \tag{2.4b}$$
$$\tilde{V}=PH_AQ+QH_AP\ , \tag{2.4c}$$

where we have used the identity

$$H_A=(P+Q)H_A(P+Q)\ .$$

We have

$$H=\tilde{H}_0+H_R+\tilde{V}+D\ . \tag{2.5}$$

With the Hamiltonian partitioned as above we have

$$[z-(\tilde{H}_0+H_R)]G-\tilde{V}G-DG=1 \tag{2.6}$$

and incorporating the initial-state vector $|\bar{g}\rangle$ we obtain the equation

$$(z-\tilde{H}_0-H_R)G|\bar{g}\rangle-\tilde{V}G|\bar{g}\rangle-DG|\bar{g}\rangle=|\bar{g}\rangle \tag{2.7}$$

from which we can obtain matrix elements of the type $G_{\bar{a}\bar{g}}$, $G_{\bar{b}\bar{g}}$, and $G_{\bar{c}_i\bar{g}}$, etc. with all other states of interest in the calculation. After writing these equations, we eliminate the matrix elements involving the continua which give rise to ionization widths and shifts. The formal development of this elimination follows the procedure of Ref. 6 and will not be reproduced here. The reader should note that the definition of P here is somewhat different from the P of Ref. 6. Thus we arrive at the equations

$$\left[z+\frac{i}{2}\gamma_g\right]G_{gg}-\bar{D}_{ga}G_{ag}-\bar{D}^{(2)}_{gb}G_{bg}=1\ , \tag{2.8a}$$

$$-\bar{D}_{ag}G_{gg}+\left[z+\delta_1+\frac{i}{2}(\gamma_a^{(0)}+\gamma_a^{(2)}+\Gamma_a)\right]G_{ag}-\bar{D}_{ab}G_{bg}=0\ , \tag{2.8b}$$

$$-\bar{D}^{(2)}_{bg}G_{gg}-\bar{D}_{ba}G_{ag}+\left[z+\delta_1+\delta_2+\frac{i}{2}(\Gamma_b+\gamma_b^{(3)}+\gamma_b^{(1)})\right]G_{bg}=0\ , \tag{2.8c}$$

where the detunings δ_1 and δ_2 from the two resonance transitions are defined by

$$\delta_1=\hbar\omega_1-[(E_a+S_a^{(2)}+S_a^{(0)}+\Delta_a)-(E_g+S_g)]\ , \tag{2.9a}$$

$$\delta_2=\hbar\omega_2-[(E_b+S_b^{(3)}+S_b^{(1)}+\Delta_b)-(E_a-S_a^{(2)}+S_a^{(0)}+\Delta_a)]\ . \tag{2.9b}$$

Here Γ and Δ denote the nonradiative autoionization widths and shifts. The radiative widths γ and the shifts S are defined by equations of the form

$$\tfrac{1}{2}\gamma_\alpha^{(i)}=\pi(|D_{c_i\alpha}|^2_{\bar{E}_{c_i}=\bar{E}_g}+|D_{c_i'\alpha}|^2_{\bar{E}_{c_i'}=\bar{E}_g})\ , \tag{2.10a}$$

$$S_a^{(i)}=\mathrm{P}\int d\bar{E}_{c_i}\frac{|D_{c_i\alpha}|^2}{\bar{E}_g-\bar{E}_{c_i}}+\mathrm{P}\int d\bar{E}_{c_i'}\frac{|D_{c_i'\alpha}|^2}{\bar{E}_g-\bar{E}_{c_i'}}\ , \tag{2.10b}$$

where P here means "principal value" and where $j=a,b$ and the superscript (i) (which assumes the values 0,1,2,3) indicates the continuum into which ionization occurs. It should be noted that ionization from either a or b can here occur by either the emission or the absorption of a laser photon as indicated in Fig. 1. Thus the term ionization width in this context has a meaning that is somewhat more general than usual.

The quantities $\bar{D}_{ga}$, $\bar{D}_{ab}$, and $\bar{D}^{(2)}_{gb}$ are not simple dipole matrix elements between the respective discrete states since they also involve indirect couplings of these states through the continua, as discussed in Ref. 6. Finally, direct dipole, continuum-continuum couplings have been neglected in deriving Eqs. (2.8) as they are not expected to be significant in this problem (see also related comments in Ref. 6). All shifts S are proportional to the laser intensity and as a result the detuning will in general vary with laser intensity; they are dynamical detunings. All widths are also proportional to the laser intensity. The couplings $\bar{D}_{ga}$ and $\bar{D}_{ab}$ are proportional to the square root of the intensity while $\bar{D}^{(2)}_{gb}$ representing a two-photon coupling is proportional to the intensity.

With a given choice of wave functions, all of the above parameters can be calculated and from the solution of the algebraic equations (2.8) the matrix elements of G are expressed as functions of z. From those expressions through the inversion integral of the Laplace transform, we obtain the matrix elements of $U(T)$ needed for the calculation of ionization. It will be noticed that we have dropped the bars over the subscripts in Eqs. (2.8). This should not cause any confusion because after having eliminated the continua, the photon numbers appear only as multiplicative constants of the coupling parameters. Thus the matrix elements of G can now be identified unambiguously with the three discrete states of the problem.

III. WAVE FUNCTIONS

The continuum wave function is approximated by $\psi_c^+(E)$ defined by

$$\psi_c^+(E)=\frac{1}{\sqrt{2}}A[\phi_{n_1l_1m_1}(\mathbf{r}_1)F_{l_2}(\mathbf{r}_2)]\ , \tag{3.1}$$

where $\phi_{n_1l_1m_1}(\mathbf{r}_1)$ is a bound hydrogenic wave function for electron 1 corresponding to the ion He^+ and A denotes the antisymmetrization operator. The continuum wave function $F_{l_2}(\mathbf{r}_2)$ has the usual general form

$$F_{l_2}(\mathbf{r}_2)=\frac{1}{4\pi}i^{l_2}(2l_2+1)e^{i(\delta_{l_2}+\eta_{l_2})}P_{l_2}(\cos\alpha)\frac{u_{l_2}(r_2)}{r_2} \tag{3.2}$$

with $u_{l_2}(r_2)$ being the solution of the equation (in atomic units)

$$\left[\frac{d^2}{dr_2^2}-\frac{l_2(l_2+1)}{r_2^2}+k^2+\frac{2Z}{r_2}\right]u_{l_2}(r_2)=0\ , \tag{3.3}$$

where in Eq. (3.3) we set $Z=1$. The energy of the ejected electron is $\frac{1}{2}k^2$ and the asymptotic form of $u_{l_2}(r_2)$ is

$$u_{l_2}(r_2)\underset{r_2\to\infty}{\to}\left[\frac{2}{\pi k}\right]^{1/2}\sin\theta(r_2)\ ,$$

where $\theta(r)$ is the phase of the pure Coulomb wave function. This condition assumes the normalization

$$\langle\psi_c^+(E)|\psi_c^+(E')\rangle=\delta(E-E')\delta(\Omega_k-\Omega_{k'})\ ,$$

where Ω_k and $\Omega_{k'}$ are the directions of propagation of the electron and E,E', the respective energies. The angle α between the electron wave vector $\mathbf{k}$ and $\mathbf{r}_2$ is given by $(\mathbf{r}_2\cdot\mathbf{k})/kr_2$.

The method of calculation of the autoionizing states has been given in Ref. 8. Here we simply summarize some of the general features of the calculation.

Employing the operators P and Q defined in the previous section, we define the resonant wave function $\chi_s^{L,S}$ as the solution of the equation[7]

$$(QH_AQ-\xi_s)\chi_s^{L,S}=0\ , \tag{3.4}$$

where H_A is the total nonrelativistic Hamiltonian of the atomic system with L,S being the total orbital angular momentum and total spin, respectively. The energy of the state is $E_s=\xi_s+\Delta_s$ where Δ_s is the shift introduced by the configuration interaction coupling the resonant state $\chi_s^{L,S}$ with the continuum and is expressed as

$$\Delta_s=\langle\chi_s^{L,S}|(QH_AP)\mathrm{P}(\xi_s-PH_AP)^{-1}(PH_AQ)|\chi_s^{L,S}\rangle\ , \tag{3.5}$$

where P indicates the principal value. Since Δ_s turns out to be of the order of magnitude of the width, we shall neglect it in this work. The overall scope of this calculation, moreover, is not the prediction of the exact resonances but rather their behavior under laser excitation.

We expand $\chi_s^{L,S}$ on a basis of hydrogenic functions as

$$\chi_s^{L,S}=\sum_j a_j u_j^{L,S}\ , \tag{3.6a}$$

where (with δ_{ij} the Kronecker delta)

$$u_j^{L,S}=\frac{1}{[2(1+\delta_{n_1n_2}\delta_{l_1l_2})]^{1/2}} \times A[R_{n_1l_1}(r_1)R_{n_2l_2}(r_2)y^{L,M}(1,2)]\ , \tag{3.6b}$$

$R_{nl}(r)$ represents the radial part of the hydrogenic function $\phi_{nlm}(\mathbf{r})$, and

$$y^{L,M}(1,2)=\sum_m (l_1l_2L\mid m,M-m)Y_{l_1}^m(1)Y_{l_2}^{M-m}(2) \tag{3.7}$$

with Y_l^m being the usual spherical harmonic and $(l_1l_2L\mid m,M-m)$ a Clebsch-Gordan coefficient. Given the normalization condition adopted above, the width of the autoionizing state can be expressed as

$$\Gamma_s=2\pi\int|\langle\chi_s^{L,S}|QH_AP|\psi_s^+(E_s)\rangle|^2d\Omega_k\ , \tag{3.8}$$

where we have integrated over the solid angle of the outgoing electron and where

$$(H'-E_s)\psi_s^+(E_s)=0 \tag{3.9}$$

and

$$H'=PH_AP+\sum_{n(\neq s)}\frac{PH_AQ|\chi_n^{L,S}\rangle\langle\chi_n^{L,S}|QH_AP}{E_s-\xi_n}\ . \tag{3.10}$$

(ξ_n is the energy of the generic resonant state in Q space, the subscript s indicates the specific resonance of interest.) We approximate the function $\psi_s^+(E)$ by $\psi_c^+(E)$ in Eq. (3.8), which means that the bound electron is assumed to fully screen the nuclear charge. Noting that $P|\psi_c^+(E_s)\rangle=|\psi_c^+(E_s)\rangle$ and $Q|\chi_s^{L,S}(E_s)\rangle=|\chi_s^{L,S}(E_s)\rangle$, the operator QHP in the matrix element of Eq. (3.8) practically reduces to $1/r_{12}$. Finally, the matrix elements coupling the resonant states $\chi_s^{L,S}$ with the continuum can be shown to involve integrals of the form

$$J_s^l(\beta)=\int_0^\infty dr\,r^{s+l+1}e^{-r(\beta+ik)} \times{}_1F_1(iZ/k+l+1,2l+2,2ikr)\ , \tag{3.11}$$

where ${}_1F_1(a,b,x)$ is the confluent hypergeometric function and the integrals J_s^l are easily expressed through recursion relations.[9]

IV. CALCULATIONS

We consider first the excitation of a $^1P^o$ autoionizing resonance. There are several such resonances for which we adopt the general notation $^{2s+1}L^{o,e}(n)$ where s is the total spin, L is the total orbital angular momentum $(S,P,D,\ldots)$, (o,e) indicate odd or even parity, and n is the numbering of the resonances within the L series under consideration. The energy of the resonance increases with increasing n. The atom, initially in the ground $^1S^e$ state, is excited to the $^1P^o(1)$ resonance by the absorption of a photon from a weak field whose frequency is near-resonant with the transition. A second photon from a strong source (laser) couples $^1P^o(1)$ with either $^1D^e(3)$ or $^1S^e(1)$ and these two cases will be discussed separately. In the notation of Sec. II, $^1P^o(1)$ corresponds to state $|a\rangle$ while $^1D^e(3)$ or $^1S^e(1)$ correspond to $|b\rangle$. In Table I we list some of the atomic parameters of these states as obtained in this calculation together with values obtained by Bhatia and Temkin[10] as well as an experimental value for Fano's shape parameter q obtained by Madden and Codling.[11] Our values for the autoionization widths Γ and the energy positions E of the resonances are in reasonably good agreement with the results of Bhatia and Temkin (see Table I). Our value for q, on the other hand, differs significantly from the experimental result. The cause of this discrepancy can be attributed mainly to our overestimation of the dipole coupling between the ground state and the resonance $^1P^o(1)$. This overestimation is most probably due to the inaccuracy of our ground-state wave function which is constructed in terms of a hydrogenic basis set. We have used a set of between 20 and 30 elements $u^{L,S}$ for the calculations of the ground and resonant autoionizing states. Since the main purpose of this work is to study the coupling between autoionizing states under a strong laser, the above discrepancy does not introduce any major difficulties. The transition from the ground state to $^1P^o(1)$ is merely a weak probe for our purposes. We note that values of q to the other resonances are neither known nor meaningful in our context, because

TABLE I. Autoionization widths Γ, energy positions E, and shape parameter q of the resonances entering the problem. D_{ab} is the dipole coupling between ^1P and the resonances ^1S and ^1D. All the quantities are in atomic units (a.u.).

	Γ (a.u.)	E (a.u.)	q	D_{ab} (a.u.)
$^1P^o(1)$	0.127×10^{-2}	-0.6878	-4.7	
	$(0.133\times10^{-2})^a$	$(-0.6929)^a$	$(-2.8)^b$	
			$(-2.3)^a$	
$^1S^e(1)$	0.508×10^{-2}	-0.7750		
	$(0.459\times10^{-2})^a$	$(-0.7788)^a$		1.56
$^1D^e(3)$	0.262×10^{-4}	-0.5560		
	$(0.213\times10^{-4})^a$	$(-0.5564)^a$		1.29

[a]Our results are compared to those of Bhatia and Temkin (Ref. 10).

[b]Experimental result of Madden and Codling (Ref. 11).

the resonances cannot be reached by photoabsorption directly from the ground states. As we show subsequently, the line shape and the probability of autoionization through the even parity resonances depends on the sequence of transitions, the strength of the coupling D_{ab} (also listed in Table I) between $|a\rangle$ and $|b\rangle$ and the laser intensity to which $|D_{ab}|^2$ is proportional.

Having taken $^1P^o(1)$ as state $|a\rangle$, we consider now the coupling of $|a\rangle$ with one of the even parity D resonances, choosing specifically the state $^1D^e(3)$. This choice is in part dictated by practical considerations. For example, $^1D^e(1)$ is energetically so close to $^1P^o(1)$ that it would require a laser source of wavelength $\lambda=27000$ Å to couple the two. On the other hand, although $^1D^e(2)$ is energetically at a more convenient energy, it turns out that its dipole coupling to $^1P^o(1)$ is exceedingly small owing to a cancellation of the contributions of the main a_j components [see Eq. (3.6a)]. This not only makes difficult the observability of this process, it also prevents the precise theoretical determination of the coupling matrix element D_{ab}. It is perhaps worth noting that this problem of cancellation arises also in other autoionizing resonances of He as, for example, when the autoionization width of $^1P^o(4)$ is calculated. The smallness of the coupling D_{ab} would also require rather large intensities (much larger than 10^{12} W/cm^2) for the excitation of $^1D^e(2)$. At such intensities, the picture would be complicated by additional effects requiring a much more elaborate treatment which would obscure the main objective of this work as well as any experimental attempt.

A. Strong coupling of $^1P^o(1)$ with $^1D^e(3)$

With the resonance $^1D^e(3)$ taken as state $|b\rangle$, we note that in addition to its coupling to the continuum $1s\epsilon l$, this resonance can be also coupled to the continua $2s\epsilon l$ and $2p\epsilon l$ by absorbing one more photon. This is possible because the energy of the photon ($h\nu=3.574$ eV) necessary to couple $^1P^o(1)$ with $^1D^e(3)$ is also sufficient to cause a transition of $^1D^e(3)$ into the continuum above the $n=2$ ionization threshold. Note that this would not be the case for $^1D^e(1)$ or $^1D^e(2)$. It is also important for our calculations to note that the dipole coupling between the resonant states and the continua of the form $1s\epsilon l$ is rather small (of the order of $E_0 10^{-2}$ a.u. with E_0 being the strength of the electric field). A consequence of this feature is the absence of significant broadening of the resonant states originating from laser-induced transitions into the continua $1s\epsilon l$. The situation is, however, quite different when transitions into the continua $2s\epsilon l$ and $2p\epsilon l$ are energetically possible. In that case, the resonant states can be ionized via these additional channels as well, thus acquiring additional laser-induced widths which can be quite significant above certain laser intensities (approximately 10^{12} W/cm^2). Ionization through these channels, moreover, leaves the ion in an excited state. In addition to its importance in this paper, the distinction between transitions into continua with different core states is always important when one considers the broadening of autoionizing states due to laser-induced transitions. As the above discussion shows such transitions may or may not be significant depending on the continua that are energetically accessible. This point seems to have escaped the attention of Andryushin *et al.*[12] who have written several papers based on the premise that this broadening is always present.

Let us assume now that a laser is at first tuned exactly on resonance with the transition $|a\rangle \rightarrow |b\rangle$ which implies $\delta_2=0$. Since δ_2 is a dynamic detuning affected by the Stark shifts of resonances, it depends on the laser intensity. For simplicity of presentation, we assume that at each intensity, the frequency is adjusted so as to satisfy the condition $\delta_2=0$, which is also the easiest arrangement for the experiment. The results can always be easily related to the zero-field detunings because the magnitude of the shift is explicitly included in our calculation. With $\delta_2=0$, we assume that a weak source[13] with frequency ω_1 tunable around the transition $|g\rangle \rightarrow |a\rangle$ excites the resonance $|a\rangle = {}^1P^o(1)$ which is coupled to $^1D^e(3) = |b\rangle$ by the laser. We calculate the total ionization as a function of ω_1 for different values of the laser intensity. We present the results in terms of δ_1 [which is uniquely related to ω_1 through Eq. (2.9a)] measured in units of Γ_a the autoionization width of $^1P^o(1)$. Total ionization comes from autoionization of either of the two resonances and possibly from laser-induced transitions to the various continua, if the intensity is sufficiently large. In any case, all of these channels are included in the seemingly simple expression of Eq. (2.3) and have been taken into account in the reported results. The complexity of the problem is of course hidden in the expressions for the matrix elements of $U(T)$.

If the laser is sufficiently weak, resonance $|a\rangle$ autoionizes before any excitation to $|b\rangle$ can occur. Then, scanning of δ_1 will simply produce the autoionization line shape of $|a\rangle$ since the laser has no effect whatsoever. This is almost the case in Fig. 2(a) which represents the results of a calculation for laser-field intensity $E_2=10^{-4}$ a.u. (corresponding roughly to 10^9 W/cm^2). At this intensity, the effect of the laser is barely beginning to appear, causing some excitation from $|a\rangle$ to $|b\rangle$, but it still is weak and the overall line shape is that of the $^1P^o(1)$ resonance. The significance of the very narrow dip in the middle (too narrow to be resolved on the scale of the graph) becomes evident as we follow the development of the line-shape (with increasing intensity) from Fig. 2(a) to 2(d) which corresponds to $E_2=3\times10^{-3}$ a.u. The profile has split into two lines as a result of the oscillation between $|a\rangle$ and $|b\rangle$. The separation between the two peaks is equal to about $6\Gamma_a$ and in the parlance of double optical resonance (DOR) it is also equal to the Rabi frequency $\Omega=2D_{ab}$. Physically, it represents the frequency with which the transition $|a\rangle \leftrightarrow |b\rangle$ takes place at this intensity. Thus we can infer that an atom excited to $|a\rangle$ in the presence of this laser intensity will be excited to $|b\rangle$, and deexcited back to $|a\rangle$, by stimulated emission more than once before it ionizes. Still, ionization is dominated by autoionization from $|a\rangle$ because its width is about 50 times larger than that of $|b\rangle$ (see Table I). For the same reason, in Fig. 2(d), it is the width Γ_a that dominates the breadth of the two components.

The reader familiar with double resonance phenomena

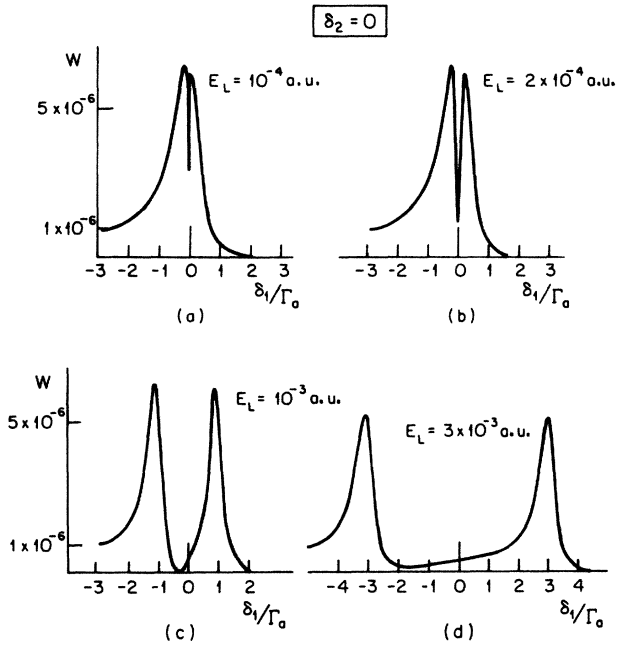

FIG. 2. The line shape of total ionization as a function of $\delta_1=\hbar\omega_1-(E_D-E_g)$ in units of Γ_a the autoionization width of the state ^1P. The strong laser is tuned exactly on resonance with the transition $^1P\rightarrow{}^1D$ which implies $\delta_2=0$. W is the total ionization probability which, owing to the weakness of the first source (synchrotron), is proportional to its intensity I_s and the time T, i.e., $W\sim I_sT$. For the calculations, we have taken $T=10^6$ a.u. and $I_s=1.4\times10^5$ W/cm^2. E_L is the field strength of the laser in atomic units. Note that 1 a.u. of E_L corresponds to 1.4×10^{17} W/cm^2.

in spectroscopy may wonder why the doublet of peaks in Fig. 2 have not equal heights even though the laser is tuned exactly on resonance with the transition $|a\rangle\leftrightarrow|b\rangle$. The reason is that the original (unperturbed by the field) line shape is slightly asymmetric. It is this asymmetry that is manifested in the unequal peak heights. As usual in this type of interactions, the original line-shape asymmetry is due to the coupling of the resonant state with the continuum. In any case, the separation between the two peaks is given by $2\Omega_0=2|D_{ba}|=2E_2|\langle b|z|a\rangle|$ which is a direct measure of the coupling between the two autoionizing states. The presence of the splitting is therefore direct evidence of the excitation of $|b\rangle$ and its magnitude provides the transition probability per unit time.

Excitation of $|b\rangle$ from $|a\rangle$ will occur even when the laser is detuned from exact resonance with that transition. An example of that case is shown in Fig. 3 calculated for detuning $\delta_2=\Gamma_a$. We have again the development of a splitting which in this case is given by 2Ω where now Ω is the generalized Rabi frequency $\Omega=(\delta_2^2+\Omega_0^2)^{1/2}$ with Ω_0 being the Rabi frequency for $\delta_2=0$ as defined above. The asymmetry of the peak heights is more pronounced now because, even if the initial line shape is symmetric (as in traditional DOR), excitation off resonance will result to asymmetric splitting. In terms of dressed states, one can say that one of the two dressed states is excited more. There is, however, one new aspect here which is not found in the typical DOR case, namely the widths of the two peaks are not equal in Fig. 3. This has again to do with the initial asymmetric shape. The width of each peak is determined by autoionization as well as the laser-induced ionization of the upper state to the higher continuum $2p\epsilon l$ and $2s\epsilon l$ by the absorption of an additional photon thus leaving the ion in an excited state. The ionization widths for these channels are $\gamma_{2s\epsilon l}\cong2\pi E_2^2 0.63$ a.u. and $\gamma_{2p\epsilon l}\cong2\pi E_2^2 4.4$ a.u. Most of the ionization does again come from autoionization of $^1P^o(1)$ because of its dominant width. As the intensity rises, however, the upward ionization of $^1D^e(3)$ begins to play an increasing role. That is why the peak heights in Fig. 2(d) are reduced while the widths are increased. Thus we have a rather novel and unconventional DOR in that at intensities below 10^{12} W/cm^2, the laser couples strongly a broad $^1P^o(1)$ with a narrow $^1D^e(3)$ autoionizing state. With increasing intensity, the narrow state begins to broaden because of the increasing contribution of other channels leading to continua that do not interfere with the autoionization ones. To the extent that these continua contribute, the atom is prevented from autoionizing driven instead to higher continua resulting in excited state ions.

In closing this case, we present in Fig. 4 the calculation for $\delta_2=-\Gamma_a$. Everything said above applies here as well, except that the details of the line shapes are now different because of the original asymmetry. It should be evident by now that by detuning to the other side of the asymmetric resonance, a significant difference from Fig. 2 is to be expected, as is indeed the case.

B. Strong coupling of $^1P^o(1)$ with $^1S^e(1)$

We now take as state $|b\rangle$ the autoionizing resonance $^1S^e(1)$ whose energy[14] lies below that of $|a\rangle=^1P^o(1)$ by

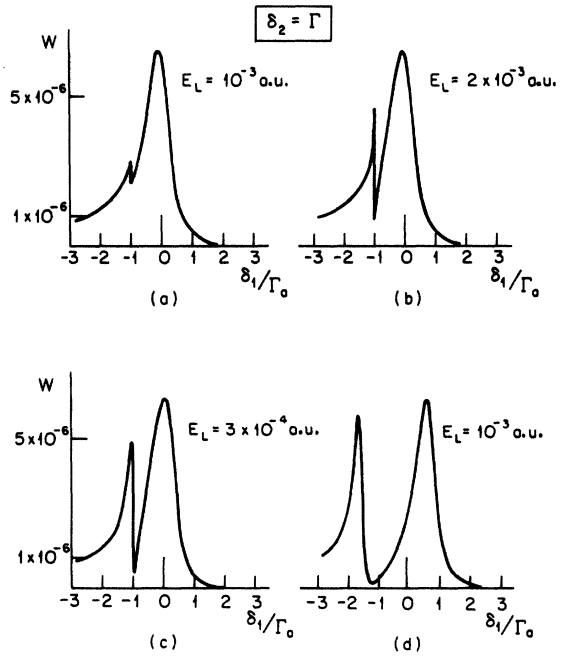

FIG. 3. Same as in Fig. 2 except that $\delta_2=\Gamma_a$.

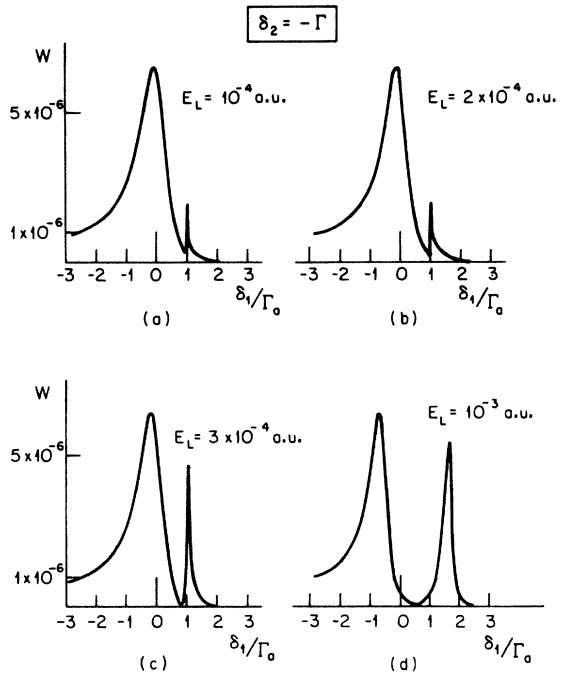

FIG. 4. Same as in Fig. 2 except that $\delta_2 = -\Gamma_a$.

about 2.365 eV or 19028 cm^{-1}. Thus a laser-induced transition from $|b\rangle$ to $|a\rangle$ involves the stimulated emission and not the absorption of a photon. There are in addition two other important differences from the previous case: (a) The continua $2s\epsilon l$ and $2p\epsilon l$ are not accessible through the absorption of one photon. (b) The autoionization widths of both resonances are comparable in magnitude which means that photoelectrons will be produced through the autoionization of both, even when they are strongly coupled by the laser.

The results of the calculation for the line shape are shown in Fig. 5. For low intensity [Fig. 5(a)], we again see the line shape of the $^1P^o(1)$ resonance because the laser is too weak to induce transition to $^1S^e(1)$. With increasing intensity, we have at first some broadening of the field-free line shape [Fig. 5(b)] and eventually the development of the two peaks [Figs. 5(c) and 5(d)]. Again the fact that we obtain a splitting larger than the autoionization width, demonstrates that at that laser intensity ($\sim 5\times 10^{11}$ W/cm^2) we have significant transitions between $|b\rangle$ and $|a\rangle$. The atom, once excited to $^1P^o(1)$ by the weak source (chosen exactly on resonance), undergoes a few transitions to $^1S^e(1)$ and back before autoionizing. The amount of ionization through $^1S^e(1)$ is larger than ionization through $^1P^o(1)$, roughly by a factor of 3.5 which represents the ratio of the respective autoionization widths. The slight asymmetry persisting through all four of the figures has again its origin in the slight asymmetry of the field-free line shape.

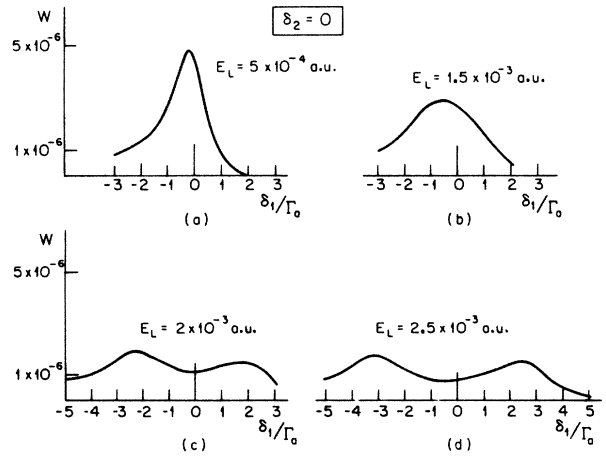

FIG. 5. The laser is now tuned exactly on resonance ($\delta_2 = 0$) with the transition $^1P \rightarrow {}^1S$ corresponding to Fig. 1(b).

V. CONCLUDING REMARKS

With our calculations in this work, we have explored the strength with which autoionizing states can be coupled by a strong radiation source (laser). Choosing states that can be coupled by lasers of readily available photon energies (from about 2 to 4 eV), we have shown that these autoionizing states, once excited by whatever means, can be induced to make transitions to other autoionizing states to which dipole transitions are allowed. At laser intensities around 10^{12} W/cm^2, such induced transitions compete with autoionization and eventually dominate. Eventual ionization of the atom is of course the inevitable outcome, but in the process the atom can be excited to higher autoionizing states if the frequency and intensity of the laser are chosen appropriately. As we have shown, one of the signatures of such excitations is the change of the autoionization line shape. Thus if an experiment were to study the excitation of a $^1P^o$ resonance by scanning the frequency of the exciting source, the broadening and splitting of the line shape can be related directly to the strength of the coupling between the autoionizing states. Of course another signature of this effect is the photoelectron energy spectrum because the strong field redistributes the probability of excitation among the coupled resonances and the participating continua. The involvement of the continua under strong coupling is, as we have seen, far from straightforward, especially when excitations above a higher threshold are energetically possible.

The weakest point of this calculation is the dipole transition from the ground state to the $^1P^o(1)$ resonance. As pointed out earlier, this is mainly due to our representing the ground state in terms of hydrogenic wave functions. This dipole transition, however, is rather peripheral to our objective here since it simply serves as the mechanism of excitation of the initial resonance. We could have simply assumed, in fact, that the initial resonance is somehow excited and take it from there. One of the reasons we did not do this is that we want to present a self-contained theory of the process. Our continuum wave functions can also be criticized since they are expressed in terms of Coulomb wave functions. Although they are not expected to be satisfactory in general, they are sufficiently appropriate for our purposes in the energy range of interest. This is borne out by a calculation of the photoionization

cross section of He which agrees reasonably well with data and other calculations in the energy range 50—100 eV. It does not give a good result at very low and high energies, as expected. This will affect somewhat the accuracy of our calculation of photoionization of the autoionizing state $D^e(3)$ to the continuum $2s\epsilon l$ because of the low energy of the ejected electron. Our calculation of the shifts and widths of the resonances (due to the coupling with the continuum) is in good agreement with the results of Bhatia and Temkin.[10]

Experiments in which the effects of this paper will be of relevance must involve obviously a strong laser and a way of exciting the $^1P^o(1)$ or an equivalent resonance. The same laser can in principle serve that purpose through a multiphoton process of relatively high order which has to be odd for the transition to be allowed. For the frequencies to match both the excitation and the coupling of $^1P^o(1)$ to the $^1D^e$ resonance, the selection of a resonance higher than the $^1D^e(3)$ would be required. This arrangement is quite pertinent to presently ongoing studies of multiphoton ionization under strong lasers.[1-4] We cannot, however, discuss the details of such an experiment here since we have not dealt with the multiphoton excitation part which we will leave for a follow-up paper.

We can discuss the possibility of single-photon excitation by synchrotron radiation in the presence of a laser. The details of the synchrotron beam are here important. Thus we can only give a rough general estimate assuming average conditions. Taking a photon flux of $F_s = 10^{14}$ photons/cm^2 sec (which is rather optimistic) and the excitation cross section which is about $\sigma = 10^{-17}$ cm^2, we have an excitation transition probability of $\sigma F_s = 10^{-3}$ sec^{-1}. A strong laser is pulsed. If we take pulse duration of about 10^{-9} sec, we find that 10^{-12} of the atoms in the interaction volume will be excited to the higher state, since as we showed this coupling can be made so strong as to have probability unity. The laser must also be focused which makes the interaction volume about 10^{-3} cm^3 or less. We are assuming here that the synchrotron beam is broader, as is certain to be the case. It all depends now on the density of atoms per cm^3 in the interaction volume. Taking 10^{15}/cm^3 (simply as a point of reference) we obtain one event per laser pulse ($10^{12} \times 10^{-3} \times 10^{15}$). We are not in the position to assess how unrealistic this density may be. The repetition rate of both laser and synchrotron pulses is of critical importance here as is of course the detection requirements about which we do not know much. According to our understanding, it is the repetition rate of the laser that is the limiting one since that of the synchrotron can be quite high (30 MHz). From general considerations and information we have pieced together by talking to experimentalists, we would consider such an experiment as marginal at best. The final word of course belongs to the experimentalists.

Excitation by electron scattering can be easily ruled out because of the short duration of the laser pulse. Note that the electron does not interact with the field here. It simply excites the target. Any electron beam is too weak to excite enough atoms within the laser pulse to make the effect observable. It is orders of magnitude beyond observability.

Although we have dealt specifically with He here, a comparison of our results with related work[15] in other two-electron atoms such as C and Sr shows that general features such as the range of laser intensities and the strength of the coupling between autoionizing states are more or less common. The range of frequencies and other details do of course depend on the structure of the particular atom.

ACKNOWLEDGMENTS

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[13] Weak must be understood in context. If the source is the synchrotron, it is indeed weak. If on the other hand, the resonance $|a\rangle$ were to be excited by a multiphoton process, the source must be strong by conventional standards so as to be capable of creating a sufficient population in $|a\rangle$. It must, however, be weak in the sense of not saturating the transition $|g\rangle \rightarrow |a\rangle$; i.e., the multiphoton Rabi frequency between $|g\rangle$ and $|a\rangle$ must be much smaller then Γ_a which can be easily achieved if the order of the process is higher than 2.
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