Radiation amplification near an autoionizing state: A model in atomic Ca

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We present an internally consistent theory of amplification near the minimum of the absorption profile of autoionizing states, with a quantitative application to calcium. We have included a completely specified three-photon pumping, and have obtained quantitative results for the gain that reveal certain unusual aspects of this system. Our results and their extensions are also discussed in the context of present-day experimental possibilities.

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I. INTRODUCTION

Discrete states embedded in continua represent very interesting quantum mechanical systems whose properties lend themselves to a number of effects exploiting coherence within the atomic continuum. Such coherences may in principle be manipulated by external fields so as to enhance the stability of selected parts of a manifold of decaying (autoionizing) states [1–5]. This manipulation, in addition to its intrinsic fundamental interest, offers the possibility of potentially promising applications in nonlinear optics and the generation of coherent short-wavelength radiation. One class of phenomena related to autoionizing states (AIS's) concerns the possibility of radiation amplification without population inversion (AWoI) [6–10] which has been the subject of much discussion during the last four years.

In pondering the problem of AWoI two aspects, namely an asymmetric absorption profile (or more generally an asymmetry between absorption and emission) and a mechanism for pumping population to the upper state, represent the cornerstones of any scheme. If an autoionizing state is to serve as the upper state, the asymmetric profile is provided by intra-atomic interaction, but the pumping mechanism must be properly chosen. Since most often the autoionizing state of an atom cannot be modeled realistically by one discrete state interacting with a continuum, the question of pumping presents not only practical but also conceptual difficulties. The discrete state entering the formal model studies [7,8] of AWoI through AIS's is not a physical state, even if one discrete state coupled to a single continuum represents the atom adequately. In two recent papers [11,12], we identified some of the difficulties associated with multiphoton pumping of either autoionizing or autoionizing-like states.

It is the purpose of this paper to present a consistent treatment of AWoI through an autoionizing state with a concrete quantitative application to Ca. In addition to including a realistic and quantitative description of the relevant atomic structure, our treatment is self-consistent in that it includes also a complete specification of the pumping.

II. THE MODEL

We consider a sequence of autoionizing doubly excited states of Ca belonging to the $3dnp(^1P_1)$ series which is also coupled to the series $3dnf(^1P_1)$. These series [13,14] decay by autoionization into the continuum $4s\epsilon p$ and cannot be treated as isolated discrete states embedded in a continuum. To obtain the necessary atomic parameters, a separate calculation, which is a project in itself, must be performed. This has been accomplished in a separate work [15], has been compared with available experimental data [16] on photoabsorption, and has provided the parameters employed in this work.

We assume that the atom is pumped from its ground state $4s^2(1S_0)$ to the vicinity of one of the $^1P_1$ autoionizing resonances by a three-photon process. The required wavelength range for such pumping is around 565 nm, which is readily accessible through presently available pulsed lasers. In addition we consider an external probe beam whose amplification or attenuation probes the existence or lack of positive gain under the specified pump conditions.

After having determined the autoionizing spectrum and wave functions over an extended range of energy through techniques described elsewhere [15,17], we consider an appropriately restricted energy range around the desired resonance and by fitting the single- as well as three-photon absorption profiles to the form $(q+\delta)^2/(1+\delta^2)$, where q is the so-called asymmetry parameter and δ the detuning in units of half-width of the autoionizing state (see Ref. [1]), combined with an *ab initio* calculation of the transition to the continuum and the autoionization width, we can determine sets of parameters (Table I) representing an effective discrete state plus continuum models separately for single- and three-photon transitions, which, however, contain all of the necessary complexity of the coupled-channel atomic problem.

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TABLE I. Values of atomic parameters with I_P in units of W/cm^2. The first row of numbers corresponds to state $(3d5p)^1P_1$, and the second to state $(3d6p)^1P_1$.

Γ_2 (sec^{-1})	$\gamma_1(P)$ (sec^{-1})	$\gamma_1(F)$ (sec^{-1})	$M_{12}^{(3)}$ (a.u.)	$M_{12}^{(1)}$ (a.u.)
1.19×10^{14}	$4.37\times10^{-21}I_P^3$	$2.35\times10^{-25}I_P^3$	$5.51\times10^4(1-i/0.12)$	$0.402(1+i/4.38)$
8.33×10^{12}	$6.43\times10^{-23}I_P^3$	$5.73\times10^{-22}I_P^3$	$1.41\times10^5(1-i/9.58)$	$0.144(1-i/19.8)$

III. TIME-DEPENDENT DYNAMICS

The atomic system can now be described in terms of the time-dependent density matrix from which we can calculate all quantities needed in our problem as a function of pump frequency ω_P and field strength ε_P and of probe frequency. Extending the formalism that we have employed in previous work [8,18], we derive a set of equations in the absence of the probe for the reduced density matrix, which can eventually be reduced to

$$\left[\frac{\partial}{\partial t}+\gamma_1\right]\sigma_{11}=-2\,\mathrm{Im}[M_{21}^{(3)}(\varepsilon_P)^3\sigma_{12}]\ , \tag{1}$$

$$\left[\frac{\partial}{\partial t}+\Gamma_2\right]\sigma_{22}=2\,\mathrm{Im}[M_{21}^{*(3)}(\varepsilon_P)^3\sigma_{12}]\ , \tag{2}$$

$$\left[\frac{\partial}{\partial t}+i\Delta_{12}+\frac{\gamma_1+\Gamma_2}{2}\right]\sigma_{12} =-i(M_{12}^{*(3)}\sigma_{22}-M_{12}^{(3)}\sigma_{11})(\varepsilon_P^*)^3\ , \tag{3}$$

where

$$M_{21}^{(3)}=M_{12}^{(3)}\equiv\sum_{l,m}\frac{\mu_{1l}\mu_{lm}\left[\mu_{m2}+\int d\omega_c\frac{\mu_{mc}V_{c2}}{E_{mc}-\omega_P}\right]}{(E_{1l}-\omega_P)(E_{1m}-2\omega_P)} \tag{4}$$

is the effective three-photon dipole transition moment between $|1\rangle$ and $|2\rangle$, with μ_{ij} denoting the usual electric dipole matrix element, and V_{c2} the configuration interaction matrix element (coupling $|2\rangle$ to the continuum $|c\rangle$) which is responsible for autoionization. The summation $\sum_{l,m}$ extends over a complete set of states, and the integral contains a principal value part and an imaginary part at the pole. This matrix element is a very essential part of the pumping process, containing the interference between three-photon transitions and autoionization. $\Delta_{12}=3\omega_P-E_{21}$ is the detuning of $3\omega_P$ from the three-photon resonance between $|2\rangle$ and $|1\rangle$, with E_{21} being the energy difference between $|2\rangle$ and $|1\rangle$, including the ac Stark shift of each state.

$$\gamma_1=2\pi\left[|\varepsilon_P|^6\left|\sum_{l,m}\frac{\mu_{1l}\mu_{lm}\mu_{mc}}{(E_{1l}-\omega_P)(E_{1m}-2\omega_P)}\right|^2_{E_c=E_1+3\omega_P}\right]$$

is the three-photon ionization width of $|1\rangle$ by the absorption of $3\omega_P$ photons, leading directly into the continuum, where E_c denotes the energy of the continuum state $|c\rangle$. $\Gamma_2=\pi|V_{c2}|^2$ is the autoionization width of $|2\rangle$.

The solution of Eqs. (1)–(3) under prescribed laser pulses provides the state of the atomic system as a function of time. We solve these equations numerically with a pump pulse whose time dependence is given by

$$I(t)=I_0\mathrm{sech}^2(t/\tau)\ , \tag{5}$$

with I being the intensity and τ what will be referred to as the pulse duration. It is important to stress here that the equations do not imply pumping to a discrete part, but a transition (pumping) involving the correct wave function at the energy $3\omega_P$.

We can show that the introduction of a weak probe beam leads to the replacement of $M_{21}^{(3)}(\varepsilon_P)^3$ by $M_{21}^{(3)}(\varepsilon_P)^3+M_{21}^{(1)}\varepsilon_T e^{i(\omega_T-3\omega_P)t-i(k_T-3k_P)z}$ and σ_{12} by $\sigma_{12}+\sigma'_{12}e^{-i(\omega_T-3\omega_P)t+i(k_T-3k_P)z}$, which in combination with Eqs. (1)–(3) leads to

$$\sigma'_{21}=\frac{i(M_{12}^{(1)}\sigma_{22}-M_{12}^{(1)*}\sigma_{11})\varepsilon_T}{-i\Delta_{12}^{(1)}+\Gamma_{12}} \tag{6}$$

where ω_T, k_T, and ε_T are the frequency, wave vector, and amplitude of the probe field.

Then we derive the following expression for the linear gain/loss coefficient for the probe:

$$\kappa\equiv-\frac{N\omega_T}{2\epsilon_0 c n_T}[\alpha''_{1T}\sigma_{11}-\mathrm{Im}(M_{12}^{(1)*}\sigma'_{21})/\varepsilon_T]\ , \tag{7}$$

where $\alpha''_{1T}\equiv\pi|\mu_{1c}|^2_{E_c=E_1+\omega_T}$. Substituting σ'_{21} from Eq. (6) and setting $M_{21}=\mu_{21}(1+i/q)$ (which defines q), we obtain

$$\kappa=-\frac{N\omega_T}{2\epsilon_0 c n_T}\frac{2(\mu_{12})^2}{\Gamma_2}\frac{[(\delta+q)^2+(\eta-1)(q^2+\eta)]\sigma_{11}-\eta(1+q^2)\sigma_{22}}{q^2(\delta^2+\eta^2)}\ . \tag{8}$$

Equation (8) above, which is one of our main formal results, shows that κ depends explicitly on two important parameters: q (the asymmetry parameter of the resonance) and $\eta=1+\gamma_1/\Gamma_2$, where γ_1 is the total ionization width of $|1\rangle$ (due to both ε_P and ε_T) directly into the continuum. μ_{12} is the electric dipole matrix element between $|1\rangle$ and $|2\rangle$, N the atomic density, and n_T the index of refraction at ω_T. The above expression is valid for both coherent and incoherent pumping, which would only affect the values of σ_{11} and σ_{22}. No steady state is implied in Eq. (8). The gain κ can vary in time following the evolution of σ_{11} and σ_{22} under a pulse. The quantity δ expresses the detuning of ω_T from the transition frequency $|1\rangle \rightarrow |2\rangle$ in units of $\frac{1}{2}\Gamma_2$. It must be stressed here that the κ in Eq. (7) is not Raman gain but the gain of an amplifier due to the populations of the active medium, as clearly seen in Eq. (8). For $\eta=1$ and $q\rightarrow\infty$, Eq. (8) reduces to the usual gain, which is proportional to the inversion $\sigma_{22}-\sigma_{11}$. For $\eta=1$, $\sigma_{11}=1$, and $\sigma_{22}=0$, Eq. (8) reduces to the well-known absorption line shape of an autoionizing resonance. For $\eta>1$ and $q<\infty$, we have a situation created by the coherent interaction of a discrete state with a continuum.

IV. RESULTS

Using the above theory, we have explored the existence of positive gain as a function of pump pulse duration and intensity, with the pump tuned on three-photon resonance with $|2\rangle$, i.e., $\Delta_{12}=0$. Positive gain results only when the probe is tuned around the vicinity of the interference minimum of the autoionizing resonance [6–8]. Surprisingly, we have discovered that a broad resonance may produce much more gain than a narrower one. We have traced this to the fact that the three-photon ionization directly into the F continuum (see Table I), which does not enter in the interference channels but represents an independent incoherent loss (of atomic population) mechanism, happens to be smaller for the broad resonance. This is documented below with a specific example of two resonances. In general, we have found that both the maximum gain and the frequency range of positive gain increase with increasing pump intensity, up to a point, beyond which the medium is depleted because of ionization.

A quantitative assessment of the above findings is provided in Figs. 1 and 2. Figure 1 demonstrates the dependence of the averaged gain per pulse (for a 1-ps pulse) on the pump laser intensity for the states $3d5p(^1P_1)$ and

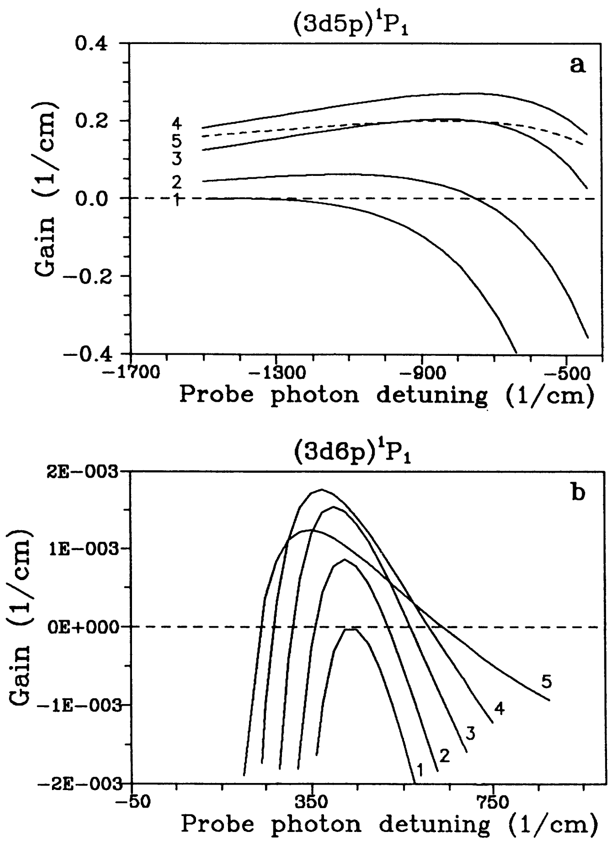

FIG. 1. Averaged gain over a 1-ps pulse as a function of the detuning for (a) $(3d5p)^1P_1$ with different pump laser intensities: curves (1) zero intensity, (2) 3×10^{11} W/cm^2, (3) 4×10^{11} W/cm^2, (4) 5×10^{11} W/cm^2, and (5) 7×10^{11} W/cm^2; and (b) $(3d6p)^1P_1$ with different pump laser intensities: curves (1) zero intensity, (2) 5×10^{10} W/cm^2, (3) 7×10^{10} W/cm^2, (4) 1×10^{11} w/cm^2, and (5) 2×10^{11} W/cm^2. Pump laser detuning Δ_{12} is zero.

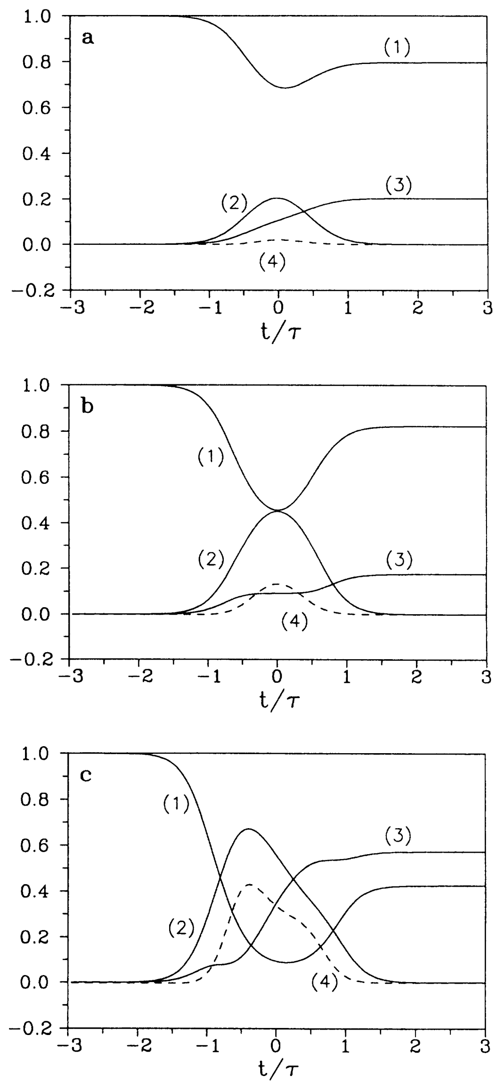

FIG. 2. Time-dependent behavior of (1) ground-state population, (2) upper-state population, (3) ionization, and (4) $\kappa(\mathrm{cm}^{-1})$ in the case of the $(3d5p)^1P_1$ state as the upper state for a 1-ps pulse at three different pump laser intensities: (a) 2.0×10^{11} W/cm^2, (b) 3.0×10^{11} W/cm^2, and (c) 5.5×10^{11} W/cm^2. Pump laser detuning Δ_{12} is zero, and the probe is tuned to the zero-field absorption minimum $\delta=-q$.

$3d6p(^1P_1)$ whose autoionization widths are 634 and 35 cm^{-1}, respectively. As mentioned above, we have here a clear case where the narrow state produces gain three orders of magnitude smaller than the much broader one. In Fig. 2, we present the temporal behavior of four key quantities: gain, populations of $|2\rangle$ and $|1\rangle$, and ionization, for different pump intensities. First, we note an increase of the gain with the pump intensity. Second, we note that in Fig. 2(c) during part of the pulse (from about -0.9τ to $+0.9\tau$) we have inverted populations ($\sigma_{22}>\sigma_{11}$), while otherwise we have ($\sigma_{22}<\sigma_{11}$). Therefore we have amplification without inversion, but surprisingly under certain conditions we also have inversion; this is rather unexpected since our pumping occurs through an electromagnetic transition.

Here we should note a fundamental and rather counterintuitive feature of an autoionizing state excited by a single strong field (in the sense that the field-induced transition is comparable to Γ_2). It can reach a steady state, although the system is connected to the continuum. This is most easily illustrated analytically by taking an intensity such that $\gamma_1/\Gamma_2=1$, with the pump laser tuned on $\Delta_{12}=0$. Then, for a square pulse we obtain $\sigma_{11}+\sigma_{22}=\frac{1}{2}(1+\epsilon^{-2\Gamma_2 t})$ and $\sigma_{11}-\sigma_{22}=e^{-\Gamma_2 t}\cos(\Gamma_2 qt)$, which demonstrate that, for $t\rightarrow\infty$, half of the population is lost to ionization and the other half is shared equally between the two states. This phenomenon (whose origins are implicit in Refs. [1] and [2]) plays some role in the behavior seen in Fig. 2(b), where ionization and populations stabilize at the end of the pulse. A more general implication that we noted in our studies within a certain range of intensities is that, up to a point, ionization (loss) decreases with increasing intensity.

V. CONCLUDING DISCUSSION

Within the framework of an internally consistent formalism, we have shown that multiphoton (coherent or incoherent) pumping can provide a realistic scheme of amplification with or without inversion near the minimum of an AIS. On the basis of realistic atomic parameters and a time-dependent analysis, we have obtained the gain for a probe pulse of an appropriate duration. For the calculation of the gain, we have assumed an atomic density of 10^{17} cm^{-3}, which can be obtained in a heatpipe. Our intention here is not to provide a working setup for a practical amplifier, but a realistic situation for testing the validity of the fundamentals of the model with presently accessible experimental means. The overall accuracy of our atomic model (as tested by comparison with whatever experimental or theoretical data are available) provides, we believe, sufficient confidence not to deter us from proposing this (or a closely related scheme) for immediate experimental study.

The usual question of whether a basis exists in which the populations of $|2\rangle$ and $|1\rangle$ are inverted is irrelevant here because there is no dressing by external fields. The asymmetry is provided by intraatomic interactions which determine the only physically meaningful basis. In any case, we prefer to place the emphasis on the novelty of these amplification (and eventually lasing) schemes, with reduced requirements on inversion, rather than on the complete absence of inversion.

Experimental realization of our scheme in a medium of density 10^{17} cm^{-3} may also lead to the generation of the third harmonic of the pump photon. Depending on phase-matching conditions (which are externally controllable) this may be substantial; and if tuned to the minimum of the AIS can, at least in principle, serve as the radiation to be amplified through the mechanism of this paper. Here we have chosen the probe as a separate external source in order to concentrate on the basic mechanism of pumping and amplification. In a separate publication we will discuss the problem with the third harmonic included. Experimentally one can always decouple it from the amplification through phase matching and/or detuning the pump away from the minimum, as has been the case here.

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