Rate of resonant two-photon ionization in the presence of a partially coherent radiation field

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A treatment is given of two-photon ionization in which particular attention is paid to the influence of the bandwidth of the ionizing light on the ionization rate. The model adopted for the atom is a common one, including two bound levels and a continuum. The model of the laser is general enough to allow short-term temporal fluctuations of amplitude or phase, and these fluctuations give rise to the laser bandwidth. It is found that the fluctuations have the most interesting effects on the ionization rate when the laser coherence time is shorter than the atomic memory time, as could be expected. In addition to laser bandwidth, we consider the influence on the ionization rate of the detuning of the laser from the intermediate-state resonance, the laser power, and the finite lifetime of the intermediate state. Comparisons with related earlier calculations are made and similarities and differences are pointed out.

I. INTRODUCTION

The theory of near-resonant two-photon ionization rates[1] and the theory of the near-resonant interaction of few-level atoms with electromagnetic fields[2] have both been treated in the literature in some detail but from quite different points of view. We describe an alternative treatment of resonant two-photon ionization rates that exploits the simplicity of the two-level atom, but also includes the positive-energy continuum states and short-term stochastic fluctuations in the ionizing applied field.

Our approach is formulated in the Heisenberg picture. Besides the usual two-level-atom Heisenberg operators, we introduce explicitly the operators referring to the atomic continuum. By eliminating from the equations of motion the terms related to the electromagnetic self-field as well as the terms referring to the continuum, we obtain transverse and longitudinal damping corresponding to spontaneous emission and photoionization. The use of simple amplitude correlation functions allows us to account for the finite bandwidth of the applied field.[3]

We consider examples where a unique two-photon ionization rate can be defined, and calculate this ionization rate as a function of the detuning from the resonant intermediate state. The effects of power broadening, laser bandwidth, and saturation are included.

II. ATOM-FIELD MODEL

The atom-field model is illustrated in Fig. 1. Ionization is effectuated by a quasimonochromatic radiation field of mean frequency ω_0. The energies of the initial state $|1\rangle$, the resonant intermediate state $|2\rangle$, and the final continuum states $|\omega\rangle$ are, respectively, $\hbar\omega_1$, $\hbar\omega_2$, and $\hbar\omega$. The ionization threshold defines the origin of the energy scale. The dipole transition matrix elements $\vec{d}_{12}$ and $\vec{d}_{2\omega}$ are assumed to be real vectors. The continuum

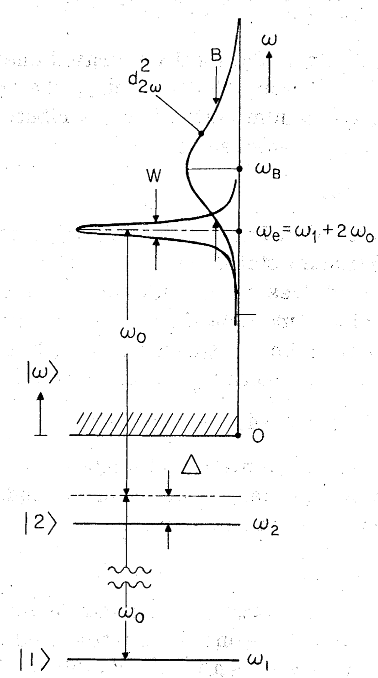

FIG. 1. Atom-field model for resonant two-photon ionization. The mean frequency of the applied field is ω_0; the applied field has a Lorentzian power spectrum of full width at half maximum W. Energy conservation occurs around $\omega_e = \omega_1 + 2\omega_0$. The fictitious ω dependence of $d^2_{2\omega}$, adopted for temporary convenience in the calculations, is shown as a Lorentzian of full width at half maximum $B \gg W$.

states are energy normalized,

$$\langle\omega|\omega'\rangle=\delta(\hbar\omega-\hbar\omega'),\qquad(1)$$

and all have the same angular momentum quantum numbers. So, in this model, free-free dipole transitions are excluded.

The applied field is characterized by a coherent state $|\{\alpha\}\rangle$, such that

$$\langle\{\alpha\}|\hat{\vec{E}}_F(0;t)|\{\alpha\}\rangle=\vec{\epsilon}(\mathcal{E}_t e^{-i\omega_0 t}+\mathcal{E}_t^* e^{i\omega_0 t}),\qquad(2)$$

where $\hat{\vec{E}}_F(0;t)$ is the free electric field operator in dipole approximation, and $\vec{\epsilon}$ is a unit polarization vector. The fluctuating amplitudes $\mathcal{E}_t$ and $\mathcal{E}_t^*$ are particular realizations of a stochastic variate, and we characterize the partial coherence of the field by the following second-order correlation function[3]:

$$\langle\langle\mathcal{E}_t\mathcal{E}_{t-\tau}^*\rangle\rangle=\mathcal{E}^2 e^{-W|\tau|/2}.\qquad(3)$$

The double parentheses denote an ensemble average. The coherence time τ_c is given by W^{-1}, and the spectral density is a Lorentzian of full width at half maximum W:

$$I(\omega)=\frac{c\mathcal{E}^2}{2\pi}\frac{W/2\pi}{(\omega-\omega_0)^2+(\frac{1}{2}W)^2}.\qquad(4)$$

Note that $c\mathcal{E}^2/2\pi$ is the total integrated energy flux (W/cm^2) of the laser. Consequently, the energy-conserving continuum states are distributed over a band of width W centered at

$$\omega_e=\omega_1+2\omega_0,\qquad(5)$$

as is illustrated in Fig. 1. We assume that this band of continuum states does not extend down to threshold, and does not sample any structure in the transition dipole moment $\vec{d}_{2\omega}$. These assumptions can conveniently be formulated explicitly by assigning a fictitious Lorentzian structure to $d_{2\omega}^2$,

$$d_{2\omega}^2=d_{2c}^2B^2/[B^2+4(\omega-\omega_B)^2].\qquad(6)$$

Of course the dipole matrix element $d_{2\omega}$ is usually practically independent of ω, so in the end we will consider only the limiting case[4]

$$W\ll B\ll\omega_B,\qquad(7)$$

and the parameter B will not appear in any of the results. However, if one were interested in a case where the continuum really did exhibit some structure, then (6) could still be used as a model, and one would simply drop the requirement $B\gg W$.

III. EQUATIONS OF MOTION

The usual[2] Heisenberg "transition operators" $\hat{\sigma}_{lm}\equiv|l\rangle\langle m|$ are suitable for discussing the dynamics of the ionization process. In Appendix A we review the use of these operators, and indicate our general approach, by working out briefly the one-photon ionization problem.

In the much more complex two-photon case a convenient set of Heisenberg operators is

$$\hat{r}=\hat{\sigma}_{22}+\hat{\sigma}_{11},\qquad(8)$$

$$\hat{w}=\hat{\sigma}_{22}-\hat{\sigma}_{11},\qquad(9)$$

and the operators $\hat{\sigma}_{ab}$, with $a\neq b$ and $a,b=1,2,\omega$. Note that $\hat{r}$ represents the total population of the discrete states. Given its time evolution, the ionization problem is solved. The operators $\hat{\sigma}_{\omega\omega'}$, $0\leqslant\omega,\omega'<\infty$ are ignored. They are manifestations of the continuum population. Since the ionization process is essentially irreversible, they cannot have any important effect on the dynamics of $\hat{r}$.

Using Heisenberg's equation of motion and eliminating the electromagnetic self-field in favor of the natural damping constants,[5] one readily obtains the equations of motion for the expectation values

$$r=\langle 1|\langle\{\alpha\}|\hat{r}|\{\alpha\}\rangle|1\rangle,\ \text{etc}.\qquad(10)$$

Introducing the slowly varying quantities

$$\rho_{12}=\sigma_{12}e^{i\omega_0 t},\qquad(11)$$

$$\rho_{2\omega}=\sigma_{2\omega}e^{i\omega_0 t},\qquad(12)$$

and

$$\rho_{1\omega}=\sigma_{1\omega}e^{2i\omega_0 t},\qquad(13)$$

and applying the rotating wave approximation,[2] we find

$$\dot{r}=\int_0^\infty d\omega\,(id_{2\omega}\mathcal{E}_t^*\rho_{2\omega}+\text{c.c.}),\qquad(14)$$

$$\dot{w}=-A(w+r)-(\kappa_{12}\mathcal{E}_t^*\rho_{12}+\text{c.c.})+\int_0^\infty d\omega\,(id_{2\omega}\mathcal{E}_t^*\rho_{2\omega}+\text{c.c.}),\qquad(15)$$

$$\dot{\rho}_{12}=-(\tfrac{1}{2}A+i\Delta)\rho_{12}-i\tfrac{1}{2}\kappa_{12}\mathcal{E}_t w+i\mathcal{E}_t^*\int_0^\infty d\omega\,d_{2\omega}\rho_{1\omega},\qquad(16)$$

$$\dot{\rho}_{1\omega}=i(\omega_e-\omega)\rho_{1\omega}+(i/\hbar)d_{2\omega}\mathcal{E}_t\rho_{12},\qquad(17)$$

$$\dot{\rho}_{2\omega}=i(\omega_2+\omega_0-\omega)\rho_{2\omega}+(i/2\hbar)d_{2\omega}\mathcal{E}_t(w+r).\qquad(18)$$

Here $d_{ij}=\vec{d}_{ij}\cdot\vec{\epsilon}$ and

$$\kappa_{12}=2d_{12}/\hbar.\qquad(19)$$

Δ is the detuning (see Fig. 1),

$$\Delta=\omega_2-\omega_1-\omega_0,\qquad(20)$$

and A is the spontaneous decay rate for level $|2\rangle$.

We have ignored the fact that $\rho_{1\omega}$ and $\rho_{2\omega}$ are actually coupled to each other, as well as the fact that $\rho_{2\omega}$ is subjected to natural damping. In Appendix B we show that this is justified, as expected, if the continuum-bound matrix elements $d_{2\omega}$ are sufficiently smooth functions of ω [i.e., if B is large enough in (6)].

Since only the dynamics of the atomic population are of interest, we eliminate any reference to the off-diagonal operators from the equations of motion. Formal integration of (17) and substitution in (16) gives

$$\dot{\rho}_{12} = -(\tfrac{1}{2}A + i\Delta)\rho_{12} - i\tfrac{1}{2}\kappa_{12}\mathcal{E}_t w - \frac{1}{\hbar}\int_0^\infty d\omega\, d_{2\omega}^2 \int_0^t d\tau\, \mathcal{E}_t^* \mathcal{E}_{t-\tau}\rho_{12}(t-\tau) \times \exp[i(\omega_e - \omega)\tau]. \quad (21)$$

The continuum integration is carried out next. In view of (6) and (7) we may extend the lower integration limit to $-\infty$ and write ($\tau \geq 0$):

$$\frac{1}{\hbar}\int_0^\infty d\omega\, d_{2\omega}^2 \exp[i(\omega_e - \omega)\tau] \simeq (\pi d_{2c}^2/\hbar)\tfrac{1}{2}B\exp\{-[\tfrac{1}{2}B + i(\omega_B - \omega_e)]\tau\}. \quad (22)$$

After substituting this in (21), the τ integration is considered. With (4) and (7) one sees that only times

$$\tau \ll B^{-1} \ll (\omega_B - \omega_e)^{-1}, W^{-1}, A^{-1} \quad (23)$$

contribute. Thus, to an excellent approximation that improves as $B \to \infty$, the τ integration in (21) gives

$$\dot{\rho}_{12} = -\{\tfrac{1}{2}(A + (2\pi d_{2c}^2/\hbar)|\mathcal{E}_t|^2) + i\Delta\}\rho_{12} - i\tfrac{1}{2}\kappa_{12}\mathcal{E}_t w, \quad (24)$$

where the exponentially damped term has been dropped. Note that the transverse-damping rate is one-half times the sum of the natural decay rate and the instantaneous one-photon ionization rate out of level $|2\rangle$. Treating the continuum integrations in (14) and (15) as above, one finds, with the formal integrations of (18) and (24):

$$\dot{r} = -\tfrac{1}{2}R_{2ct}(w + r), \quad (25)$$

$$\dot{w} = -[A + \tfrac{1}{2}R_{2ct}](w + r) - \tfrac{1}{2}\kappa_{12}^2\int_0^t dt' \exp\left(-\int_{t'}^t dt''[\tfrac{1}{2}(A + R_{2ct''}) + i\Delta]\right) \times \mathcal{E}_t^*\mathcal{E}_{t'} w(t - t') + \text{c.c.}, \quad (26)$$

where we have introduced the notation

$$R_{2ct} = 2\pi(d_{2c}^2/\hbar)\mathcal{E}_t^*\mathcal{E}_t. \quad (27)$$

R_{2ct} is the quasistatic one-photon ionization rate from level 2 into the continuum. Equations (25) and (26) form the starting point for our subsequent discussion of the two-photon ionization process.

IV. STOCHASTIC TWO-PHOTON IONIZATION RATE

In the case of a monochromatic field, it would be a simple task to solve (25) and (26) by using the Laplace-transform convolution theorem, or by any of several other methods. However, the stochastic character of the time-dependent field amplitude makes it rather difficult to start from these equations and untangle r from w in the general case. Fortunately this is not necessary. Most strong-laser ionization experiments do not have the capacity for time resolution of the ion current. The observed quantity is a smoothed, time-integrated signal, without most of the temporal features that the exact solutions of (25) and (26) contain. In many cases, especially if the ionizing laser is not so strong as to appreciably deplete the supply of ionizable atoms, a theoretical analysis leading to a single rate coefficient for ionization is completely adequate. We therefore look for the rate at which population leaves the bound system.

Equation (25) shows that the rate of loss of population from the bound system is proportional to both R_{2ct} and to $w + r$. The first of these is the one-photon ionization rate from level 2 (see Appendix A, for example); and the second is seen to be exactly twice the population of the second level, by combining Eqs. (8) and (9). If the laser is not to deplete the supply of ionizable atoms, we may begin by assuming that R_{2ct} is small in the sense that r does not change rapidly. Thus, we may use the assumption of approximately constant r and σ_{22} in using Eq. (26) to find σ_{22}.

The first step is to add (25) and (26) to obtain an equation for $\dot{\sigma}_{22}$:

$$\dot{\sigma}_{22} = -(A + R_{2ct})\sigma_{22} - \tfrac{1}{4}\kappa_{12}^2\int_0^t dt'\, \mathcal{E}_t^*\mathcal{E}_{t'}(2\sigma_{22} - r)_{t'} \times \exp\left(-\int_{t'}^t dt''[\tfrac{1}{2}(A + R_{2ct''}) + i\Delta]\right) + \text{c.c.} \quad (28)$$

Then, because of the slow variation postulated for r and σ_{22}, we can remove $w = 2\sigma_{22} - r$ from the integral. In the case of a monochromatic field it is easy to show that this is completely equivalent to the "adiabatic" or "pole" or "Weisskopf-Wigner" approximation.[6]

In carrying out the adiabatic approximation we will encounter two conditions that establish the limits of validity of the resultant rate coefficients. First, we assume that ionization occurs in approximately $[R(\Delta)]^{-1}$ sec, where $R(\Delta)$ will be identified below with the two-photon ionization rate. Temporal features of r and σ_{22} on time scales much smaller than $[R(\Delta)]^{-1}$ are not of interest. That is, we may sensibly adopt the approximation

$$(2\sigma_{22} - r)_{t'} = (2\sigma_{22} - r)_t, \quad t - t' \ll [R(\Delta)]^{-1}. \quad (29)$$

For the purposes of our present argument, in the t' integration in (28) the product $\mathcal{E}_t^* \mathcal{E}_{t'}$ behaves effectively like $\mathcal{E}^2 e^{-W|t-t'|/2}$, and the factor

$$\exp\left(-\tfrac{1}{2}\int_{t'}^{t} dt'' R_{2ct''}\right)$$

behaves like

$$\exp[-\tfrac{1}{2}R_{2c}(t-t')],$$

where R_{2c} is the ensemble average of R_{2ct}:

$$R_{2c} = (2\pi/\hbar) d_{2c}^2 \mathcal{E}^2 . \tag{30}$$

Combining these results, we see that only times

$$t - t' \lesssim |A + W + R_{2c} + 2i\Delta|^{-1} \tag{31}$$

contribute appreciably in the integrand. We may thus pull the factor $2\sigma_{22} - r$ outside the t' integration, provided that

$$R(\Delta) \ll |A + W + R_{2c} + 2i\Delta| . \tag{32}$$

The lower limit of the t' integration leads to a damped oscillatory term which falls rapidly in time intervals of $[R(\Delta)]^{-1}$ sec. This switching-on transient effect can be ignored, which we do by putting the lower limit at $-\infty$.

With these approximations, Eq. (28) reduces to

$$\dot{\sigma}_{22} = -(A + R_{2ct})\sigma_{22} - (2\sigma_{22} - r)R_{12t}(\Delta) , \tag{33}$$

where

$$R_{12t}(\Delta) = \tfrac{1}{4}\kappa_{12}^2 \int_{-\infty}^{t} dt' \, \mathcal{E}_t^* \mathcal{E}_{t'} \times \exp\left(-\int_{t'}^{t} dt''[\tfrac{1}{2}(A + R_{2ct''}) + i\Delta]\right) + \text{c.c.} \tag{34}$$

The notation is intended to suggest that R_{12t} is the generalized stimulated transition rate between levels 1 and 2. This point is clarified in Appendix C.

The second step of the adiabatic approximation is to ignore $\dot{\sigma}_{22}$ in (33). Since the smoothed σ_{22} varies appreciably only over times of the order of $[R(\Delta)]^{-1}$ sec, this is allowed when

$$R(\Delta) \ll R_{12}(\Delta) + A + R_{2c} . \tag{35}$$

This follows by inspection of (33), replacing R_{2ct} and R_{12t} for order-of-magnitude purposes by their ensemble-averaged values R_{2c} and R_{12}. Thus, in the adiabatic approximation, a quasistatic value of σ_{22} follows immediately:

$$\sigma_{22t} = R_{12t}(\Delta)/[2R_{12t}(\Delta) + A + R_{2ct}] . \tag{36}$$

Substitution in (25) immediately leads to the definition of a quasistatic two-photon ionization rate $R_t(\Delta)$:

$$R_t(\Delta) = R_{2ct}R_{12t}(\Delta)/[2R_{12t}(\Delta) + A + R_{2ct}] . \tag{37}$$

This result constitutes the starting point for the subsequent discussion of two-photon ionization. Expression (37) represents a simple yet complete rate characterization of the ionization process. The range of validity of (37) is determined by the two inequalities (32) and (35).

The experimentally relevant quantity is the ensemble average $R(\Delta)$:

$$R(\Delta) = \left(\!\!\left(\frac{R_{2ct}R_{12t}(\Delta)}{2R_{12t}(\Delta) + A + R_{2ct}} \right)\!\!\right) . \tag{38}$$

Only in very specific limiting situations can this ensemble average be calculated easily.

V. SATURATION AND LASER-BANDWIDTH EFFECTS

A global view of saturation and laser-bandwidth effects in two-photon ionization is easily obtained from (38) by retaining only second-order correlation effects. In Appendix D we calculate some of the simpler consequences of retaining higher-order correlations. In the "second-order" approximation we may write (38) as follows:

$$R(\Delta) = \frac{(\!(R_{2ct})\!)(\!(R_{12t}(\Delta))\!)}{(\!(2R_{12t}(\Delta))\!) + A + (\!(R_{2ct})\!)} , \tag{39}$$

where $(\!(R_{12t}(\Delta))\!)$ is given approximately by (C3):

$$R_{12}(\Delta) \simeq \Omega^2(A + W + R_{2c})/[4\Delta^2 + (A + W + R_{2c})^2] . \tag{40}$$

Note that the term "second order" should be understood carefully. We have kept only second-order correlations, but we have treated the numerator and denominator of (38) quite separately in arriving at (39). Thus (39) really represents an "all-orders" ansatz. Its advantages are that it is analytically simple, and it reproduces faithfully the fully saturated strong-field limits of the exact expression.

The decorrelation employed to obtain (39) from (38) is also interesting because two entirely different features of the stochastic field $\mathcal{E}_t$ and its interaction with the atom are involved. One of these features is the *memory of the atom* during the course of the interaction, and the other is the presence of *fluctuations in the field* during the time an experimental record of the ionization is being accumulated. These are obviously distinct aspects of the experiment.

One can imagine circumstances in which a sequence of laser shots is required for an experimental record, the laser being very steady during each shot but wandering unpredictably in intensity between shots. In this case, as far as any atom is concerned, the laser is monochromatic. On the other hand, the laser may be operating continuously, with atoms drifting through the laser beam. If the laser fluctuates appreciably while interacting with the atom, on a time scale shorter than the atom's effective lifetime, then the laser cannot be characterized as monochromatic, and the atom's memory of the field's fluctuations will be an important element in the results.

An early investigation[7] of the simultaneous influence of field statistics and saturation on near-resonant multiphoton ionization by Armstrong, Lambropoulos, and Rahman (ALR) considered the first of these two possibilities. In fact, Eq. (2) of ALR can be obtained from (38) in the limit $A \gg W$, R_{2c}. ALR take the view that the memory of the atom can be disregarded, i.e., that while the field may fluctuate, it does not do so rapidly enough for any one atom to notice. This is consistent with the limit $A \gg W$, R_{2c}. Our interest is almost entirely in the opposite limit, where laser-bandwidth effects become important.

Even with the second-order decorrelations of (39) and (40), the two-photon rate formula is relatively complicated. In several important limits $R(\Delta)$ takes simpler forms and a few of these are discussed below. Unless stated otherwise, verification of (32) and (35) in any of the following examples is straightforward and left to the reader.

A. Recovery of perturbation-theory results

For detunings and field strengths such that

$$\Delta \gg A + W + R_{2c} \tag{41}$$

and

$$R_{12}(\Delta) \ll A + R_{2c}, \tag{42}$$

we obtain, with (39) and (40),

$$R_{12}(\Delta) = \Omega^2 (A + W + R_{2c})/4\Delta^2 \tag{43}$$

and

$$R(\Delta) = R_{2c} \frac{1}{A + R_{2c}} R_{12}(\Delta). \tag{44}$$

In Fig. 2(a), the shaded region indicates the range of laser bandwidths for which the perturbation-theory formula (44) is accurate. Obviously, the perturbation-theory region does not contain the highest ionization rates.

If spontaneous emission is negligible, i.e., if

$$A \ll W,\ R_{2c} \tag{45}$$

then these results reduce to

$$R(\Delta) = R_{12}(\Delta), \tag{46}$$

which we write

$$R(\Delta) = \frac{R_{2c}\Omega^2}{4\Delta^2} + R_{2c} \frac{1}{R_{2c}} \frac{W\Omega^2}{4\Delta^2}. \tag{47}$$

The first term is just the second-order perturbation-theory prediction for the "direct" two-photon ionization rate in the presence of a coherent applied field. The second term is of the "two-step" form: (rate) × (lifetime) × (rate), and $W\Omega^2/4\Delta^2$ is the correct first-order perturbation-theory prediction for the transition rate from level 1 to level 2, due to the on-resonance frequency components in the tail of the power spectrum. The lifetime of level 2 is determined here by its one-photon ionization rate.

When the natural decay rate dominates the one-photon ionization rate R_{2c},

$$R_{2c} \ll A, W, \tag{48}$$

we find

$$R(\Delta) = \frac{R_{2c}\Omega^2}{4\Delta^2} + R_{2c} \frac{1}{A} \frac{W\Omega^2}{4\Delta^2}. \tag{49}$$

The two terms are interpreted in a way similar to (47) above. Note that the second term is the origin of the remark that a "two-step" form for the rate is obtained only if spontaneous emission determines the intermediate-state lifetime. However, Eq. (47) above already shows that such a remark can easily be false.

B. Resonant two-photon ionization rates when the applied field is the dominant source of incoherence

The applied field being the dominant source of incoherence, we have

$$W \gg A, R_{2c} \tag{50}$$

and, in view of (40),

$$R_{12}(\Delta) = \Omega^2 W/(4\Delta^2 + W^2). \tag{51}$$

We now distinguish between the following cases.

1. Weak fields

"Weak fields" are defined by the condition that both the bound-bound and bound-free stimulated rates are dominated by the spontaneous emission rate:

$$A \gg R_{12}(0) \tag{52}$$

and

$$A \gg R_{2c}. \tag{53}$$

With (39) and (40) the two-photon near-resonant ionization rate is now

$$R(\Delta) = \frac{\Omega^2 W}{4\Delta^2 + W^2} \frac{1}{A} R_{2c}. \tag{54}$$

The region of validity of this large-bandwith weak-field formula is shown in Fig. 2(b). Note that in the range $\frac{1}{2}W \geq \Delta$ the formula predicts that the ionization rate *increases* with *decreasing* power

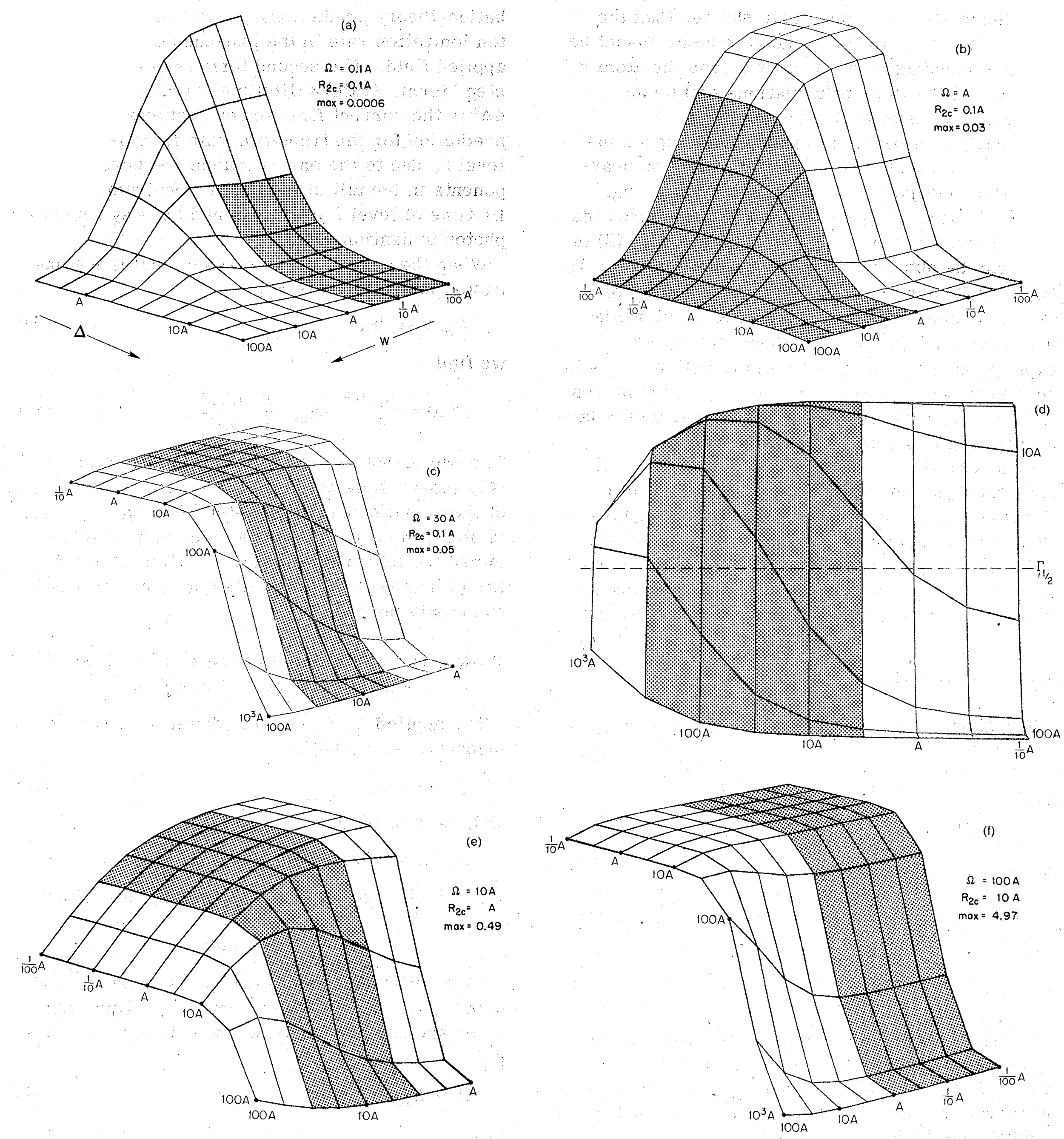

FIG. 2. (a)–(f) Portions of ionization-rate surfaces in perspective view. In every case the axes in the base plane are detuning Δ, and laser bandwidth W, as labeled in (a). The vertical axis is the two-photon ionization rate $R(\Delta)$ of Eq. (39). Each figure is individually normalized to fit into a unit cube, and the viewing point in (a)–(c), (e), and (f) is $X=10$, $Y=10$, $Z=5$. In (d) the viewing point is adjusted to $X=\frac{1}{2}$, $Y=10$, $Z=\frac{1}{2}$ to give a frontal view of the side face of (c). The coordinate grid is logarithmic in both directions, with the spontaneous emission rate A setting the scale. In all cases Δ and W increase toward the front edges of the figures. Each figure contains a legend showing the value of Rabi frequency Ω, and one-photon ionization rate R_{2c}, for which it is drawn, as well as the maximum value of $R(\Delta)$ for those parameters. The shaded portion of each figure illustrates the Δ-W dependence of a particular limit discussed in the text, as follows: (a) Perturbation limit, Eq. (44). (b) Weak field, large bandwidth, Eq. (54). (c) Strong field, bound-bound dominance, Eq. (58). (e) Strong field, power broadened level 2, Eq. (64). (f) Strong field, bound-free dominance, Eq. (68). (d) is a special view of (c) designed to illustrate the bandwidth dependence of the ionization line-width given in Eq. (60). The label of the Δ curve that cuts the $\Gamma_{1/2}$ line gives the ionization half width. In the shaded region these half widths increase with increasing W in rough agreement with Eq. (60).

per unit bandwidth. This is seen clearly in the figure. The interpretation of this result is obvious: even though the spectral density of laser power is made smaller when W increases, the effective power on resonance increases to a maximum at $\frac{1}{2}W = \Delta$. At exact resonance,

$$R(0) = (\Omega^2/W)(1/A)R_{2c}\ , \tag{55}$$

a result first obtained by Lambropoulos.[1] Note that the resonant transition rate $R_{12}(0) = \Omega^2/W$ is proportional to the power per unit bandwidth, as it should be in this case. Note further that (54) and (55) take the form (rate from 1 to 2) $\times$ (lifetime of level 2) $\times$ (rate from 2 to the continuum). In contradistinction to Lambropoulos on this point, we mention that R_{12} in (51) is the correct *incoherent* stimulated rate when $W \gg \Omega$, and so (54) and (55) must be given a "two-step" interpretation.

2. Strong fields, type 1

This domain of field strengths is defined by requiring nominal saturation of the bound-bound transition,

$$R_{12}(0) \gg A\ , \tag{56}$$

with fields weak enough that the intermediate-state level width is still due to the bound-bound decay channel

$$A \gg R_{2c}\,. \tag{57}$$

Together with (50) and (51), these conditions lead to:

$$R(\Delta) = R_{2c}\,\frac{1}{A}\,\frac{\Omega^2 W}{4\Delta^2 + 2\Omega^2 W/A}\,. \tag{58}$$

This behavior is shown in Fig. 2(c). At resonance the two-level system is saturated, $\sigma_{22} = \frac{1}{2}$, and

$$R(0) = \tfrac{1}{2}R_{2c}\,. \tag{59}$$

The two-photon absorption line is power-broadened with width Γ:

$$\Gamma = (2\Omega^2 W/A)^{1/2}\,. \tag{60}$$

So the linewidth depends critically on the ratio W/A, and, except for the factor of 2, is exactly the geometric mean of the laser bandwith W and the nominal bound-bound absorption rate in the absence of laser bandwidth, Ω^2/A. Figure 2(d) shows the bandwidth dependence of Γ given in (60).

3. Strong fields, type 2

In this case the saturating field is strong enough that the one-photon ionization channel determines the level width of the intermediate state:

$$R_{12}(0) \gg R_{2c} \gg A\ . \tag{61}$$

One sees by inspection that $R(\Delta)$, which is approximately $\frac{1}{2}R_{2c}$ at exact resonance, is still of the order of $\frac{1}{3}R_{2c}$ for detunings such that

$$R_{12}(\Delta) \simeq R_{2c}\,. \tag{62}$$

Note that the validity of our rate description is weakest in this region. Criterion (35) is satisfied by less than a factor of 10. For all other values of Δ, we obtain

$$R(\Delta) = R_{2c}R_{12}(\Delta)/[2R_{12}(\Delta) + R_{2c}]\,. \tag{63}$$

This can be written

$$R(\Delta) = \Omega^2 W/[(2\Omega^2 W/R_{2c}) + 4\Delta^2]\,. \tag{64}$$

The width of the ionization-rate line shape is now

$$\Gamma' = (2\Omega^2 W/R_{2c})^{1/2}\,. \tag{65}$$

Note that this width is field independent; i.e., a saturation of the power broadening has occurred. [Formally, the linewidth Γ' cannot be defined exactly, since it is just for detunings $\Delta \simeq \frac{1}{2}\Gamma'$ that criterion (35) is least well satisfied.] The domain of W and Δ relevant to expression (64) is shaded in Fig. 2(e).

C. Superstrong fields; photoionization from the upper discrete level is the dominant source of incoherence

In this example, we consider the resonant two-photon ionization rate when the intensity of the applied field is such that

$$R_{2c} \gg A,\ W. \tag{66}$$

Thus, now, contrary to the situation in the example of Sec. V B, the one-photon ionization from level 2 is the dominant source of incoherence. With (40) and (66) it follows that

$$R_{12}(\Delta) = \Omega^2 R_{2c}/[4\Delta^2 + R_{2c}^2] \tag{67}$$

which leads directly to

$$R(\Delta) = \Omega^2 R_{2c}/(4\Delta^2 + 2\Omega^2 + R_{2c}^2)\,. \tag{68}$$

This formula is illustrated in Fig. 2(f). In the limit $\Omega \gg R_{2c}$, it reduces to case A2 of Beers and Armstrong,[1] if their "direct" ionization channel is ignored.

In view of (68), the ionization-rate line shape is again power broadened, this time with the width

$$\Gamma'' = (2\Omega^2 + R_{2c}^2)^{1/2}\,. \tag{69}$$

Note that Γ'' is qualitatively different from the earlier example of a power-broadened width, Γ in Eq. (60), because at the highest powers[8] Γ'' is proportional to $\mathcal{E}^2$ instead of $\mathcal{E}$. In this regime, the on-resonance two-photon ionization rate is completely independent of the field strength. The explanation for this effect is simple. It signifies not only saturation in the usual sense, but also the development of a "bottleneck" in the atom at level 2. The bottleneck comes about because, as R_{2c} increases, it becomes the linewidth of level 2, and on resonance R_{12} reduces to Ω^2/R_{2c}, which is independent of laser power. This bottleneck has been described recently in several other contexts.[9] Here it leads to a "zero-slope" ionization process, one for which a plot of log(ion rate) vs log(laser power) has zero slope.

Since R_{2c} is not subject to any saturation there exists a domain of "superstrong fields" such that R_{2c} is also the dominant stimulated transition rate (this is the case labeled A1 by Beers and Armstrong[1]):

$$R_{2c} \gg R_{12}(0). \tag{70}$$

It is then clear from (39) and (69) that the resonant two-photon ionization rate is[10]

$$R(\Delta) = R_{12}(\Delta). \tag{71}$$

In all of the cases considered under "strong" or "superstrong" fields we appear to reach different conclusions about saturation from those of Lambropoulos.[1] There are detailed differences that arise from the different assumptions made about the laser line shape. Our interest here is not in these details, however, but in overall qualitative behavior. In the first place we do not find that R_{12} ever plays the role of level *width*, whereas Lambropoulos includes it [under the notation $\gamma_{2(1)}$ in Eq. (5.13c)] among the contributors to the width of level 2. In the second place, we agree that the width of the second level keeps increasing with laser power, but we ascribe this increase to increasingly important one-photon ionization from 2 to the continuum. As a result we do not find that the width of $R(\Delta)$ saturates at a value equal to the laser bandwidth. As expressions (60), (65), and (69) show, we find that the ionization line shape gets broader at the highest powers[8] and is there proportional to R_{2c} itself.

It is difficult to be certain but the differences noted above all appear to arise from insufficient consideration of power broadening in Lambropoulos' formalism. As additional evidence for this conjecture we point out that in both (59) and (68) the peak ionization rate is proportional to intensity and never to intensity squared. In fact the peak rate takes the value $\frac{1}{2}R_{2c}$, exactly what should be expected if the 1-2 transition is fully saturated with one-half of the population in level 2. This is in agreement with Eq. (18) of Beers and Armstrong[1] in the limit $\gamma_g = 0$.

VI. DISCUSSION

We have presented a study of two-photon ionization. Our work has many contacts with earlier work,[1,7,9] but has one main difference. This is our integrated treatment of the effects of laser statistics and of laser bandwidth, allowing both to arise naturally from the assumption that the laser's electric field fluctuates stochastically. We have used a model[3] for the field fluctuations that is simple enough to allow analytic expressions to be found for the ionization rate in a variety of experimentally interesting situations. These include combinations of the following: on-resonance and off-resonance with the intermediate state, power-independent finite lifetime of the intermediate state, weak or strong coupling of the ground state with the intermediate state, dominance of the bound-bound transition or of the bound-free transition, and arbitrarily large or small laser bandwidth.

In order to allow properly for laser fluctuations, one is obliged to take a time-dependent view of the ionization process, even if a cw laser is assumed. Thus, we have first derived the appropriate dynamic transition-operator or density-matrix equations (14)–(18), taking care to treat the final state as an infinitely wide continuum of energy levels. Two approximations were introduced at the outset: all far-off-resonance intermediate states were ignored, thus eliminating the main cause of an ac Stark shift of the intermediate state as well as of a "direct" ground-to-continuum ionization channel; and all free-free transitions in the continuum were ignored. Both of these approximations can be defended easily.

There is no evidence that free-free transitions are very important in multiphoton ionization, and a "direct" far-off-resonance channel can be expected to be significant only at very early times in the ionization process, in the first fraction of the resonant transition's first Rabi period.[11] The absence of ac Stark shifts and widths from the model is also unimportant. They have minimal influence on the saturation properties of the ionization because they are usually of small magnitude; and they have no effect at all on the resonant character of the ionization in the sense that, no matter what the intermediate level's shift is, the level can be located with a tunable laser and the resonance properties of the ionization studied.[12]

We have solved the dynamic equations in the rate regime, first identifying those frequency components in the ionization signal that can be ignored, and then invoking the adiabatic approximation appropriate to the remainder of the signal. No statistical decorrelations are made in these steps, and the ionization-rate constant given in (38) is a general expression for two-photon ionization, valid for a wide variety of laser powers, detunings, and statistical characteristics. Again, we emphasize the need to account not only for the existence of field statistics, something that has been widely done since Mollow's early two-photon-absorption discussion,[13] but also to account for the fact that such statistics imply the existence of an underlying fluctuation time associated with the field.

We have shown in Sec. V that the ionization rate can be quite different if the characteristic coherence time of the field is greater or smaller than the atom's own memory time. This is perhaps most evident in the ionization linewidth formula (60), and in the rate (54) which grows larger as the bandwidth gets larger. These considerations underline the need to treat carefully and completely the relaxation processes that are responsible for atomic memory. Comparison of our results, even in the limit $W=0$, with those of earlier workers, particularly those of Ref. 1, is hampered by their unconventional treatment or neglect of one of these relaxation processes.

Our own treatment of relaxation processes can also be criticized on grounds of neglect because, as mentioned above, we do not include a weak direct-ionization channel from $|1\rangle$ to $|\omega\rangle$, but more seriously because in common with other existing treatments we allow, at most, only radiative decay from $|2\rangle$ to $|1\rangle$. Collisional relaxation of $|2\rangle$ and decay from $|2\rangle$ that is trapped in a metastable level $|1'\rangle$ are both ignored. However, our general result, given in (38), has a structure that suggests how to incorporate these effects *a posteriori*. Let us take the Bloch view that relaxation processes can be separated into two kinds, transverse and longitudinal. Then resonance theory shows[14] that we may include collisional and other forms of relaxation by the following replacement in (37):

$$A+R_{2ct}\rightarrow A+R_{2ct}+1/T_1\,, \tag{72}$$

and the accompanying replacement in (34):

$$\tfrac{1}{2}(A+R_{2ct''})\rightarrow\tfrac{1}{2}(A+R_{2ct''})+1/T_2\,. \tag{73}$$

Here, $1/T_1$ and $1/T_2$ are the longitudinal and transverse rates associated with relaxation processes that may be present in addition to spontaneous decay from $|2\rangle$ to $|1\rangle$ and one-photon ionization from $|2\rangle$ to $|\omega\rangle$. The only constraint between them is $1/T_2\geq\frac{1}{2}1/T_1$. Obviously, the replacements (72) and (73) would not alter the qualitative character of our results, but might change drastically numerical predictions in specific cases.

The absence of Rabi oscillations from our general formula (38) does not mean that Rabi oscillations are absent in the ionization process, but only that under the conditions of validity of (38), given in (32) and (35), any Rabi oscillations are either much too fast or much too slow to have an appreciable effect in the ion current. One would probably want to undertake a nonadiabatic integration of the dynamic equations if the multiphoton-absorption process were being monitored, via fluorescence for example, with detectors having response times in the few nanosecond range, much shorter than ion-counter response times currently available.

Our most general result, Eq. (38), is too general to be really useful. The decorrelation ansatz, expressed in (39), is the key to our main discussion. This decorrelation allows almost any combination of detuning, laser bandwidth, laser power, and atomic-dipole transition strengths to be studied with ease. There is no confusion about the physical meaning of the ionization rate in any regime. The structure of (39) shows that, when expressed properly, the two-photon ionization rate is a natural generalization of very well-known two-level-system expressions.[14,15] That is, if $\langle\langle R_{2ct}\rangle\rangle$ is removed from the numerator, then the remainder is exactly the quasi-steady-state population of level 2 of a two-level system undergoing stimulated emission and absorption $\langle\langle R_{12t}\rangle\rangle$, internal spontaneous relaxation A, and relaxation via population removal $\langle\langle R_{2ct}\rangle\rangle$. Similarly, the expression for $R_{12}(\Delta)$ is intuitively clear, being composed in the traditional way[14] of the Rabi frequency, the overall level width, and the detuning appropriate to the bound-bound transition.

In summary, the new features of our work that appear to be of greatest interest are: the recognition that the two-photon ionization rate $R(\Delta)$ can be cast into an easily interpreted form; the natural appearance of the laser's bandwidth W in the bound-bound transition rate R_{12} on the same footing with the other relaxation rates, A and R_{2c}; and the incorporation of all saturation effects. All of these features follow more or less directly from the ansatz (3) that expresses the laser's bandwidth as a fundamental consequence of the laser field's intrinsic fluctuations. While the Lorentzian shape implied for the laser's spectrum will not be correct in many circumstances, it still seems highly likely that the role of the parameter W is given correctly for a wide variety of cases. Finally, it is obvious in all of our formulas that the smooth laser line shape implies a smooth ionization profile.

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APPENDIX A: ONE-PHOTON IONIZATION RATE VIA HEISENBERG OPERATOR EQUATIONS

The atomic Hamiltonian that accounts for dipole coupling with a classical electromagnetic field is[6]

$$H=\sum_m \hbar\omega_m\hat{\sigma}_{mm}-\sum_{m,n}\vec{d}_{mn}\cdot\vec{E}(t)\hat{\sigma}_{mn}\,, \qquad (A1)$$

where $\hat{\sigma}_{ij}$ is the generalized projector $|i\rangle\langle j|$. We consider a model with only one bound level and a full range of continuum levels. We are interested in the loss of population from the bound level, and so calculate the expectation value $\langle\sigma_{11}(t)\rangle$, the probability of occupation of level 1. From Heisenberg's equation we find

$$\frac{d}{dt}\hat{\sigma}_{11}=-i\sum_m\left(\frac{\vec{d}_{m1}\cdot\vec{E}(t)}{\hbar}\right)\hat{\sigma}_{m1}+\text{H.c.} \qquad (A2)$$

Similarly, we obtain the equation for $\hat{\sigma}_{m1}$:

$$\frac{d}{dt}\hat{\sigma}_{m1}=i\omega_{m1}\hat{\sigma}_{m1}+i\sum_j\left(\frac{\vec{d}_{1j}\cdot\vec{E}(t)}{\hbar}\right)\hat{\sigma}_{mj}-i\sum_j\left(\frac{\vec{d}_{jm}\cdot\vec{E}(t)}{\hbar}\right)\hat{\sigma}_{j1}\,. \qquad (A3)$$

In all of these Heisenberg equations the summations are understood to include the one bound state and all of the continuum states.[16] It is unnecessary to specify the continuum normalization at this point.

The formal solution to (A3) is

$$\hat{\sigma}_{m1}(t)=i\sum_j\int_0^t dt'e^{i\omega_{m1}(t-t')}\left[\left(\frac{\vec{d}_{1j}\cdot\vec{E}(t')}{\hbar}\right)\hat{\sigma}_{mj}(t')-\left(\frac{\vec{d}_{jm}\cdot\vec{E}(t')}{\hbar}\right)\hat{\sigma}_{j1}(t')\right]+\hat{\sigma}_{m1}(0)e^{i\omega_{m1}t}\,. \qquad (A4)$$

The homogeneous term can be ignored. We substitute (A4) into (A2) and use both (2) and (3) to obtain

$$\frac{d}{dt}\hat{\sigma}_{11}=\mathcal{E}^2\sum_{m,j}\left(\frac{\vec{d}_{m1}\cdot\hat{\epsilon}}{\hbar}\right)\int_0^t dt'\exp\{-[\tfrac{1}{2}W-i(\omega_{m1}-\omega_0)](t-t')\}\left[\left(\frac{\vec{d}_{1j}\cdot\hat{\epsilon}}{\hbar}\right)\hat{\sigma}_{mj}(t')-\left(\frac{\vec{d}_{jm}\cdot\hat{\epsilon}}{\hbar}\right)\hat{\sigma}_{j1}(t')\right]+\{\omega_0\to-\omega_0\}+\text{H.c.} \qquad (A5)$$

We note that only the terms on the right involving $\hat{\sigma}_{11}(t')$ make contributions in lowest (second) order and drop all of the other terms.

It is convenient to take the Laplace transform of (A5). We denote the Laplace transform of $\hat{\sigma}_{11}(t)$ by $\tilde{\sigma}_{11}(s)$. The solution for $\tilde{\sigma}_{11}(s)$ is

$$\tilde{\sigma}_{11}(s)=\hat{\sigma}_{11}(0)/[s+R(s)]\,, \qquad (A6)$$

where

$$R(s)=2\sum_m\left|\frac{\vec{d}_{1m}\cdot\hat{\epsilon}}{\hbar}\right|^2\mathcal{E}^2\frac{\tfrac{1}{2}W+s}{(\omega_{m1}-\omega_0)^2+(\tfrac{1}{2}W+s)^2}+\{\omega_0\to-\omega_0\}\,.$$

The summations in $R(s)$ can be evaluated approximately in several ways. The representation given in (6) for the bound-free dipole matrix element allows us to write it as the integral

$$\left(\frac{d_{1c}\mathcal{E}}{\hbar}\right)^2\int_0^\infty d\omega\,\frac{(\tfrac{1}{2}B)^2}{(\omega-\omega_B)^2+(\tfrac{1}{2}B)^2}\times\frac{\tfrac{1}{2}W+s}{(\omega-\omega_1-\omega_0)^2+(\tfrac{1}{2}W+s)^2}\,.$$

The contribution from $\omega_0\to-\omega_0$ is negligible. Then, if $\omega_B\gg B$, $\tfrac{1}{2}W+s$, the lower limit can be put at $-\infty$, in which case we get

$$\pi\left(\frac{d_{1c}\mathcal{E}}{\hbar}\right)^2\frac{\tfrac{1}{2}B(\tfrac{1}{2}B+\tfrac{1}{2}W+s)}{(\omega_0+\omega_1-\omega_B)^2+(\tfrac{1}{2}B+\tfrac{1}{2}W+s)^2}\,. \qquad (A7)$$

In the usual case the matrix element $d_{1\omega}$ is a very smooth function of ω. This implies that our fictitious width $\tfrac{1}{2}B$ must be very large. If we ignore $\tfrac{1}{2}W+s$ in comparison, then we find

$$\tilde{\sigma}_{11}(s)=\hat{\sigma}_{11}(0)/(s+R)\,, \qquad (A8)$$

where the constant R is given by

$$R=2\pi\left(\frac{\vec{d}_{1\omega}\cdot\hat{\epsilon}}{\hbar}\right)\mathcal{E}^2\Big|_{\omega=\omega_1+\omega_0}\,. \qquad (A9)$$

A more conventional approach that gives the same result, but does not rely on the specific shape function given in (6), proceeds as follows. First one recognizes that d^2_{1m} is practically independent of m, especially by comparison with the Lorentzian function multiplying it. This Lorentzian can simply be regarded as a representation of $\pi\delta(\omega_{m1}-\omega_0)$. Thus, one finds

$$R=2\pi\sum_m\left|\frac{\vec{d}_{1m}\cdot\hat{\epsilon}}{\hbar}\right|^2\mathcal{E}^2\delta(\omega_m-\omega_1-\omega_0)\,. \qquad (A10)$$

In this form R is immediately recognized as the golden rule one-photon ionization rate.[17]

Finally, it is easy to see that R does play the role of ionization rate in the Heisenberg picture. This follows from the Laplace inverse of (A6):

$$\hat{\sigma}_{11}(t)=\hat{\sigma}_{11}(0)e^{-Rt} \ . \tag{A11}$$

That is, R is the rate at which bound-state population disappears.

APPENDIX B: BOUND-FREE RELAXATION EFFECTS

In the text below Eq. (20), we claim that the equations of motion (17) and (18) for $\rho_{1\omega}$ and $\rho_{2\omega}$ can be used, in spite of the fact that these equations are incomplete. Expressions for $\rho_{1\omega}$ and $\rho_{2\omega}$ were obtained by formal integration of (17) and (18). Substitution of these expressions into the terms

$$i\mathcal{E}_t\int_0^\infty d\omega\, d_{2\omega}\rho_{j\omega}(t), \quad j=1,2 \tag{B1}$$

which occur in (14), (15), and (16), resulted in transverse- and longitudinal-damping terms describing the effect of ionization. To justify this procedure we establish the conditions under which the solutions of the complete equations of motion for $\rho_{1\omega}$ and $\rho_{2\omega}$ leads, upon substitution in (B1), to the same damping terms.

The complete equations of motion for $\rho_{1\omega}$ and $\rho_{2\omega}$ are

$$\dot{\rho}_{1\omega}=i(\omega_e-\omega)\rho_{1\omega}+(i/\hbar)d_{2\omega}\mathcal{E}_t\rho_{12}-\tfrac{1}{2}i\kappa\mathcal{E}_t\rho_{2\omega} \tag{B2}$$

and

$$\dot{\rho}_{2\omega}=\{i(\omega_e+\Delta-\omega)-\tfrac{1}{2}A\}\rho_{2\omega} + (i/2\hbar)d_{2\omega}\mathcal{E}_t(w+r)-\tfrac{1}{2}i\kappa\mathcal{E}_t^*\rho_{1\omega}\ , \tag{B3}$$

where we used (5) and (20) to write

$$\omega_2+\omega_0-\omega=\omega_e+\Delta-\omega\ . \tag{B4}$$

Now we evaluate the third term of (16), which is given by (B1) with $j=1$, and compare the result with the corresponding damping term

$$-(\pi d^2/\hbar)|\mathcal{E}_t|^2\rho_{12}(t) \tag{B5}$$

in (24). Upon formal integration of (B2) and (B3), and elimination of $\rho_{2\omega}$, one obtains an integral equation for $\rho_{1\omega}$. By iteration, the formal solution for $\rho_{1\omega}$ follows. We find

$$\begin{aligned}\rho_{1\omega}(t)=\frac{i}{\hbar}d_{2\omega}\sum_{n=0}^{\infty}\Bigg\{\left(\frac{i\kappa}{2}\right)^{2n}\int_0^t dt_1\int_0^{t_1}dt_2\cdots\int_0^{t_{2n}}dt_{2n+1}\Bigg[\left(\prod_{j=1}^{2n+1}\mathcal{E}_{t_j}\right)\rho_{12}(t_{2n+1})\exp[i(\omega_e-\omega)(t-t_{2n+1})]\\ \times\left(\prod_{j=0}^{n-1}\exp[(i\Delta-\tfrac{1}{2}A)(t_{2j+1}-t_{2j+2})]\right)\Bigg]\Bigg\}\\ +\frac{\kappa}{4\hbar}d_{2\omega}\sum_{n=0}^{\infty}\Bigg\{\left(\frac{i\kappa}{2}\right)^{2n}\int_0^t dt_1\int_0^{t_1}dt_2\cdots\int_0^{t_{2n+1}}dt_{2n+2}\Bigg[\left(\prod_{j=1}^{2n+2}\mathcal{E}_{t_j}\right)(w+r)_{t_{2n+2}}\exp[i(\omega_e-\omega)(t-t_{2n+2})]\\ \times\left(\prod_{j=0}^{n}\exp[(i\Delta-\tfrac{1}{2}A)(t_{2j+1}-t_{2j+2})]\right)\Bigg]\Bigg\}.\end{aligned} \tag{B6}$$

Here, and in the remainder of Appendix B, for the sake of notational simplicity, we ignore the distinction between $\mathcal{E}_t$ and $\mathcal{E}_t^*$.

Here, the notation

$$(w+r)_t=w(t)+r(t) \tag{B7}$$

has been used; and the product

$$\prod_{j=0}^{n-1}\exp[(i\Delta-\tfrac{1}{2}A)(t_{2j+1}-t_{2j+2})] \tag{B8}$$

in the first series is to be set equal to 1 for $n=0$. With $\rho_{1\omega}$ given as a series expansion, the continuum integration in (B1) can be carried out term by term using (22). For example, considering the first term of (B6), we have

$$\begin{aligned}\frac{1}{\hbar}\int_0^\infty d\omega\, d_{2\omega}^2\exp[i(\omega_e-\omega)(t-t_{2n+1})] &\simeq(\pi d_{2c}^2/\hbar)\tfrac{1}{2}B\exp\{-[\tfrac{1}{2}B+i(\omega_B-\omega_e)](t-t_{2n+1})\}\\ &=(\pi d_{2c}^2/\hbar)\tfrac{1}{2}B\prod_{j=0}^{2n}\exp\{-[\tfrac{1}{2}B+i(\omega_B-\omega_e)](t_j-t_{j+1})\}\ .\end{aligned} \tag{B9}$$

As in Eq. (23), we assume that the continuum width is much larger than the other rates that influence the time integrations in (B6):

$$B\gg(\omega_B-\omega_e),\Delta,W,A\ . \tag{B10}$$

In view of (B10) we can then ignore the product (B8) and the factor $\exp[i(\omega_B-\omega_e)(t_j-t_{j+1})]$ wherever they arise in (B6). For the continuum integration involving the second term of (B6) we find

$$i\mathcal{E}_t\int_0^\infty d\omega\, d_{2\omega}\rho_{1\omega}(t)=S_1(t)+S_2(t)\,, \tag{B11}$$

with

$$S_j(t)=\sum_{n=0}^{\infty} S_{j,n}(t)\,,\quad j=1,2 \tag{B12}$$

and

$$S_{1,n}(t)\simeq-\frac{\pi d^2}{\hbar}\mathcal{E}_t\frac{B}{2}\left(\frac{i\kappa}{2}\right)^{2n}\int_0^t dt_1\int_0^{t_1}dt_2\cdots\int_0^{t_{2n}}dt_{2n+1}\left[\left(\prod_{j=1}^{2n+1}\mathcal{E}_{t_j}\right)\rho_{12}(t_{2n+1})\left(\prod_{j=0}^{2n}\exp[-\tfrac{1}{2}B(t_j-t_{j+1})]\right)\right], \tag{B13}$$

$$S_{2,n}(t)\simeq i\,\frac{\pi d^2}{\hbar}\mathcal{E}_t\frac{B\kappa}{4}\left(\frac{i\kappa}{2}\right)^{2n}\int_0^t dt_1\int_0^{t_1}dt_2\cdots\int_0^{t_{2n+1}}dt_{2n+2}\left[\left(\prod_{j=1}^{2n+2}\mathcal{E}_{t_j}\right)\tfrac{1}{2}(w+r)_{t_{2n+2}}\left(\prod_{j=0}^{2n+1}\exp[-\tfrac{1}{2}B(t_j-t_{j+1})]\right)\right]. \tag{B14}$$

The t_{2n+1} integration in (B13) can be evaluated as follows:

$$\int_0^{t_{2n}}dt_{2n+1}\mathcal{E}_{t_{2n+1}}\rho_{12}(t_{2n+1})\exp[-\tfrac{1}{2}B(t_{2n}-t_{2n+1})]\simeq\mathcal{E}_{t_{2n}}\rho_{12}(t_{2n})\int_0^{t_{2n}}dt_{2n+1}\exp[-\tfrac{1}{2}B(t_{2n}-t_{2n+1})]$$
$$=\mathcal{E}_{t_{2n}}\rho_{12}(t_{2n})(\tfrac{1}{2}B)^{-1}(1-e^{-Bt_{2n}})\,. \tag{B15}$$

This approximation, which will be referred to as the "adiabatic approximation",[6] was discussed in the text above Eq. (24). Substitution of (B15) in (B13) gives

$$S_{1,n}(t)=-\frac{\pi d^2}{\hbar}\mathcal{E}_t\frac{B}{2}\left(\frac{i\kappa}{2}\right)^{2n}\int_0^t dt_1\int_0^{t_1}dt_2\cdots\int_0^{t_{2n-1}}dt_{2n}\left[\left(\prod_{j=1}^{2n}\mathcal{E}_{t_j}\right)\mathcal{E}_{t_{2n}}(\tfrac{1}{2}B)^{-1}\rho_{12}(t_{2n})\right.$$
$$\left.\times\left(\prod_{j=0}^{2n-1}\exp[-\tfrac{1}{2}B(t_j-t_{j+1})]\right)\right]$$
$$+e^{-Bt/2}\left\{\frac{\pi d^2}{\hbar}\mathcal{E}_t\frac{B}{2}\left(\frac{i\kappa}{2}\right)^{2n}\int_0^t dt_1\int_0^{t_1}dt_2\cdots\int_0^{t_{2n-1}}dt_{2n}\left[\left(\prod_{j=1}^{2n}\mathcal{E}_{t_j}\right)\mathcal{E}_{t_{2n}}\left(\frac{B}{2}\right)^{-1}\rho_{12}(t_{2n})\right]\right\}. \tag{B16}$$

The second term of (B16) is strongly damped and can therefore be neglected. In the first term the t_{2n} integration as well as any of the consecutive time integrations can be done in the same way as the t_{2n+1} integration. Thus, by applying the adiabatic approximation each time and dropping the exponentially damped term, we can reduce $S_{1,n}(t)$ to

$$S_{1,n}(t)\simeq-(\pi d^2/\hbar)\mathcal{E}_t^2\rho_{12}(t)(i\kappa\mathcal{E}_t/B)^{2n}\,. \tag{B17}$$

With (B12) and (B17) it follows that

$$S_1(t)\simeq-\frac{\pi d^2}{\hbar}\mathcal{E}_t^2\rho_{12}(t)\,\frac{1}{1+\lambda^2}\,, \tag{B18}$$

where

$$\lambda=\kappa\mathcal{E}_t/B\,. \tag{B19}$$

In the same way, we find for $S_2(t)$

$$S_2(t)\simeq i\,\frac{\pi d^2}{\hbar}\mathcal{E}_t^2\tfrac{1}{2}(w+v)_t\,\frac{\lambda}{1+\lambda^2}\,. \tag{B20}$$

Hence, for field strengths such that

$$\lambda\ll 1\,, \tag{B21}$$

or, in other words, as long as no appreciable continuum structure is sampled by means of power-broadening, we have

$$|S_2(t)|\ll|S_1(t)|\,, \tag{B22}$$

and with (B11) and (B18):

$$i\mathcal{E}_t\int_0^\infty d\omega\, d_{2\omega}\rho_{1\omega}(t)\simeq-\frac{\pi d^2}{\hbar}\mathcal{E}_t^2\rho_{12}(t)\,. \tag{B23}$$

This is just the factor (B5), occurring in (24).

Concluding, we see that only the zeroth-order term $S_{1,0}(t)$ of $S_1(t)$ in (B11) contributes, provided that (B21) is satisfied. This fact justifies the use of (17) instead of (B2). In the same way, the use of (18) can be justified.

APPENDIX C: GENERALIZED STIMULATED ABSORPTION RATE

It may be helpful to clarify both the notation R_{12t} adopted in Eq. (34), and the relation between our transition rates, written in terms of Rabi frequencies, and the usual rates written in terms of photon flux. For this purpose we can ignore laser correlations beyond second order, and easily obtain an approximate expression for

$$R_{12}(\Delta) \equiv ((R_{12t}(\Delta))) : \tag{C1}$$

$$R_{12}(\Delta) \simeq \tfrac{1}{2}\kappa_{12}^2 \int_{-\infty}^{t} dt'\, \mathcal{E}^2 \exp[-\tfrac{1}{2}(A+W+R_{2c})(t-t')] \times \cos\Delta(t-t') . \tag{C2}$$

This gives

$$R_{12}(\Delta) = \frac{\Omega^2(A+W+R_{2c})}{4\Delta^2+(A+W+R_{2c})^2} , \tag{C3}$$

where Ω is the Rabi frequency:

$$\Omega = \kappa_{12}\mathcal{E} = (2d_{12}/\hbar)\mathcal{E} . \tag{C4}$$

We easily verify that $(c/2\pi)\mathcal{E}^2$ is the field intensity $\hbar\omega_0\Phi$, where Φ is the photon flux. If we put these factors together then (C3) can be rewritten in a familiar way:

$$R_{12}(\Delta) = \sigma(\Delta)\Phi . \tag{C5}$$

Here $\sigma(\Delta)$ is the usual off-resonance absorption cross section,

$$\sigma(\Delta) = \frac{4\pi|d_{12}|^2\omega_0}{\hbar c}\, \frac{\tfrac{1}{2}\Gamma_{12}}{\Delta^2+(\tfrac{1}{2}\Gamma_{12})^2} , \tag{C6}$$

and Γ_{12} is the sum of all the incoherent rates affecting the 1-2 transition,

$$\Gamma_{12} = A + W + R_{2c} . \tag{C7}$$

We may emphasize here a point made before,[9] that an on-resonance transition rate—$R_{12}(0)$ for example—is precisely the ratio of the square of the Rabi frequency to the total transition linewidth.

APPENDIX D: HIGHER-ORDER CORRELATION EFFECTS

In Sec. V we adopted a special kind of "second-order" decorrelation in order to study saturation and laser-bandwidth effects in general. Although this decorrelation is extremely useful in studying the strong-field limits of (39), it is not numerically precise in its treatment of weaker fields. In this appendix we point out some of the errors of formulas (39) and (40), and give some of the lowest-order corrections for $R(\Delta)$ in the cases of thermal and phase-fluctuating fields.

Phase fluctuations

Let us begin with a phase-fluctuating field. This is a good model for the field of a stabilized laser operating well above threshold. In this case we denote the field amplitude by $\mathcal{E}_t^{(\varphi)}$:

$$\mathcal{E}_t^{(\varphi)} = \mathcal{E} e^{i\varphi_t} , \tag{D1}$$

where $\mathcal{E}$ is constant and

$$((e^{i\varphi_t} e^{-i\varphi_{t'}})) = e^{-W|t-t'|/2} . \tag{D2}$$

Obviously in this case the intensity $\mathcal{E}_t^{(\varphi)*}\mathcal{E}_t^{(\varphi)}$ is a constant nonfluctuating quantity. As a consequence, the rate of one-photon ionization from level 2 is also constant:

$$R_{2ct}^{(\varphi)} = 2\pi(d_{2c}^2/\hbar)\mathcal{E}^2 \equiv R_{2c} . \tag{D3}$$

This fact simplifies the calculation of some averages. For example, the lack of fluctuation of $R_{2ct''}$ in the exponent of (34) allows $R_{12}^{(\varphi)}(\Delta) \equiv ((R_{12t}^{(\varphi)}(\Delta)))$ to be evaluated trivially, and the result given in (C3) is reproduced:

$$R_{12}^{(\varphi)}(\Delta) = \frac{\Omega^2(A+W+R_{2c})}{4\Delta^2+(A+W+R_{2c})^2} . \tag{D4}$$

Similarly, one has $((R_{12t}^{(\varphi)}R_{2ct}^{(\varphi)})) = ((R_{12t}^{(\varphi)}))((R_{2ct}^{(\varphi)}))$.

In the weak-field or perturbation-theoretic limit, studied in Sec. V A under the restriction $A+R_{2c} \gg R_{12t}(\Delta)$, the full two-photon rate was shown to reduce to

$$R(\Delta) = R_{2c}\frac{1}{A+R_{2c}}((R_{12t}(\Delta))) . \tag{D5}$$

Now we compute the first correction to this result by expanding the denominator of (38):

$$R^{(\varphi)}(\Delta) = \left(\left(\frac{R_{2c}R_{12}^{(\varphi)}t}{A+R_{2c}}\left\{1-\frac{2R_{12t}^{(\varphi)}}{A+R_{2c}}+\cdots\right\}\right)\right) . \tag{D6}$$

The new correlation function $((R_{12t}^{(\varphi)}R_{12t}^{(\varphi)}))$ is required for the evaluation of this correction term. For simplicity, we adopt the Brownian motion phase-diffusion model[18] for φ_t, taking the "irreversible" limit in which $\tfrac{1}{2}Wt \gg \exp(-\tfrac{1}{2}Wt)$. Then we have

$$((\dot\varphi_t\dot\varphi_{t'})) = W\delta(t-t') , \tag{D7}$$

which leads, as postulated in (3), to

$$((e^{i\varphi_t} e^{-i\varphi_{t'}})) = e^{-W|t-t'|/2} , \tag{D8}$$

as well as to the fourth-order result

$$((e^{2i\varphi_t}e^{-i\varphi_{t'}}e^{-i\varphi_{t''}})) = \exp[-W(|t-t'|+|t-t''|-\tfrac{1}{2}|t'-t''|)] . \tag{D9}$$

One consequence of (D7)–(D9) is a complicated expression for $((R_{12t}^{(\varphi)}R_{12t}^{(\varphi)}))$ that becomes simple if $W \gg A+R_{2c}$. In this case one finds

$$((R_{12t}^{(\varphi)}R_{12t}^{(\varphi)})) \to \frac{\Omega^2}{A+R_{2c}}((R_{12t}^{(\varphi)})) . \tag{D10}$$

In passing we may note, from an inspection of (50), (51), and (D10) that the effect of correlations, calculated consistently to second order, is somewhat greater than allowed in the "second-order" ansatz (39). That is, under the conditions assumed, it follows that

$$((R_{12t}^{(\varphi)}R_{12t}^{(\varphi)})) \gg ((R_{12t}^{(\varphi)}))^2 . \quad (D11)$$

Finally, to lowest order, the corrected version of (D5) and (44) may be written:

$$R^{(\varphi)}(\Delta) = R_{2c}\frac{1}{A + R_{2c} + \Omega^2/(A + R_{2c})} R_{12}^{(\varphi)}(\Delta) . \quad (D12)$$

We have assumed $W \gg A + R_{2c}$, so this is also the lowest-order corrected version of the weak-field result (54) in the case of a phase-diffusing laser field.

Amplitude fluctuations

The case of a light field with amplitude fluctuations, denoted $\mathcal{E}_t^{(a)}$, perhaps appropriate to a noisy multimode laser, presents for study a different kind of correction to the ansatz in (39). Whenever Gaussian amplitude fluctuations occur one expects to encounter the "factorial effect" discussed by Agarwal and others.[19] From the known property of a complex Gaussian variate V_i,

$$((V_1^* V_2^* \cdots V_n^* V_1 V_2 \cdots V_n)) = ((V_a^* V_b))((V_c^* V_d)) \cdots ((V_p^* V_q)) + \cdots , \quad (D13)$$

where each of the $n!$ combinations of pairs of the indices $1, \ldots, n$ contribute a term to the sum, we have $((V^{*n}V^n)) = n!((V^*V))$. We are considering only two-photon ionization, but, because of the occurrence of saturation, factorials considerably higher than 2! may be encountered. We see this possibility as soon as we attempt to evaluate $((R_{12t}^{(a)}))$.

The two-level bound-bound transition rate, in the presence of ionization from the upper level, is interesting in its own right. From (34) we see that it requires the evaluation of the infinite-order correlation $C_{12}^{(a)}(t,t')$, where

$$C_{12}^{(a)}(t,t') = \left(\left(\mathcal{E}_t^{(a)*}\mathcal{E}_{t'}^{(a)}\left[\exp -\tfrac{1}{2}\int_{t'}^{t} dt'' R_{2ct''}^{(a)}\right]\right)\right) \quad . \quad (D14)$$

Again, we restrict ourselves to the first correction to the partially decorrelated result (C3), and expand the exponential, keeping only two terms. One finds

$$C_{12}^{(a)}(t,t') = ((\mathcal{E}_t^{(a)*}\mathcal{E}_{t'}^{(a)})) - \frac{\pi d_{2c}^2}{\hbar} \times \int_{t'}^{t} dt'' ((\mathcal{E}_t^{(a)*}\mathcal{E}_{t'}^{(a)}\mathcal{E}_{t''}^{(a)*}\mathcal{E}_{t''}^{(a)})) + \cdots . \quad (D15)$$

The result is

$$C_{12}^{(a)}(t,t') = \mathcal{E}^2 e^{-W|t-t'|/2}\left(1 - (2!)\pi\frac{d_{2c}^2}{\hbar}\mathcal{E}^{2(t-t')} + \cdots\right). \quad (D16)$$

Under the assumption already introduced, that ionization is not occurring rapidly, the second-order term can be replaced into the exponent, and the remaining integral in (34) can be evaluated, giving the following bound-bound transition rate:

$$R_{12}^{(a)}(\Delta) = \frac{\Omega^2(A + 2R_{2c}^{(a)} + W)}{4\Delta^2 + (A + 2R_{2c}^{(a)} + W)^2} . \quad (D17)$$

We note that, due to the laser's amplitude fluctuations, the influence of ionization (the $R_{2c}^{(a)}$ term) on bound-bound transitions is twice as great as given in (C3). The factor of 2 is the same as the 2! in (D16).

Now let us consider the two-photon ionization rate itself. There are a number of interesting limits, the simplest being the weak-field limit of Sec. V A. In that case, we take $A \gg R_{2ct} + R_{12t}$ and find

$$R^{(a)}(\Delta) = ((R_{2ct}^{(a)}(1/A)R_{12t}^{(a)}(\Delta))) . \quad (D18)$$

In this case the $R_{2ct''}^{(a)}$ term in (34) can be neglected altogether compared with A, leaving a trivial fourth-order correlation. The result is found to be

$$R^{(a)}(\Delta) = (2!)R_{2c}^{(a)}(1/A)R_{12}^{(a)}(\Delta) , \quad (D19)$$

where $R_{12}^{(a)}(\Delta)$ is now given by (D17) in the limit $R_{2c} \to 0$. This is the completely standard result for the two-photon rate given a thermal light field, except that the laser's bandwidth is now included in $R_{12}^{(a)}$ via (D17).

A simple example of a correction due to a correlation of higher than fourth order occurs in the limit $1/T_1 \gg R_{2c} \gg R_{12}$. This particular limit was not discussed in the text, but it might occur in several ways. For example, a bound-bound quadrupole transition could lead to a very small R_{12} and collisions could provide the dominant longitudinal relaxation mechanism [recall Eqs. (72) and (73)]. In this case we would have

$$R^{(a)}(\Delta) = \left(\left(\frac{R_{2ct}^{(a)}R_{12t}^{(a)}(\Delta)}{1/T_1 + R_{2ct}^{(a)}}\right)\right) \simeq \left(\left(\frac{R_{2ct}^{(a)}R_{12t}^{(a)}}{1/T_1}\left[1 - \frac{R_{2ct}^{(a)}}{1/T_1} + \cdots\right]\right)\right) . \quad (D20)$$

The result is

$$R^{(a)}(\Delta) \simeq \frac{(2!)R_{2c}^{(a)}R_{12}^{(a)}}{1/T_1} - \frac{(3!)R_{2c}^{(a)}R_{12}^{(a)}R_{2c}^{(a)}}{(1/T_1)^2} + \cdots \simeq \frac{2R_{2c}^{(a)}R_{12}^{(a)}(\Delta)}{1/T_1 + \frac{3}{2}R_{2c}^{(a)}} . \quad (D21)$$

As a final example, we choose to look again at super-strong fields, discussed in the text in Sec. V C. In this case, we have

$$R^{(a)}(\Delta) \simeq ((R_{12t}^{(a)})) . \quad (D22)$$

From (D17) we obtain

$$R^{(a)}(\Delta) \simeq \tfrac{1}{2}\Omega^2 R_{2c}^{(a)}/[\Delta^2 + (R_{2c}^{(a)})^2] . \quad (D23)$$

The effects of the laser's amplitude fluctuations are to be contrasted with (67), which is the expression one finds for a phase-diffusing field. The maximum rate on resonance is half as great for an amplitude-fluctuating field. What is also interesting is that the ionization linewidth is twice as great, making the total number of ions in the ionization profile for an amplitude-fluctuating field equal to that for a phase-diffusing laser field in this superstrong-field case.

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[2]The one-photon Bloch equations for a two-level atom are well known. See, for example, L. Allen and J. H. Eberly, *Optical Resonance and Two-Level Atoms* (Wiley, New York, 1975). The two-laser analogs, suitable for a three-level atom near or far from resonance and incorporating arbitrary longitudinal and transverse relaxation, are given by J. R. Ackerhalt and J. H. Eberly [Phys. Rev. A 14, 1705 (1976), Appendix]. See also D. M. Larsen and N. Bloembergen, Opt. Commun. 17, 254 (1976); B. W. Shore and J. R. Ackerhalt, Phys. Rev. A 15, 1640 (1977); M. D. Fedorov and A. E. Kazakov, Opt. Commun. 22, 42 (1977); Z. Białynicka-Birula *et al.*, Phys. Rev. A 16, 2048 (1977); and V. S. Letokhov and A. A. Makarov (unpublished).

[3]J. H. Eberly, Phys. Rev. Lett. 37, 1387 (1976). For references to related work, and a comment on differences due to different statistical models, see P. Zoller and F. Ehlotzky, J. Phys. B 10, 3023 (1977). See also the work on ionization reported at the International Conference on Multiphoton Processes, June 1977, by P. Agostini *et al.*, p. 95; K. Wódkiewicz, p. 203; and J. L. F. de Meijere and J. H. Eberly, p. 208, in ICOMP Abstracts (available from Department of Physics and Astronomy, University of Rochester).

[4]It is not necessary to choose $B \gg \omega_e$, but it renders continuum integrations trivial. See, for example, Eq. (22).

[5]J. R. Ackerhalt and J. H. Eberly, Phys. Rev. D 10, 3350 (1974).

[6]See, for example, P. W. Milonni, Phys. Rep. 25C, 1 (1976); G. S. Agarwal, *Quantum Statistical Theories of Spontaneous Emission and Their Relation to Other Approaches* (Springer, Heidelberg, 1974); or P. W. Milonni and J. H. Eberly, J. Chem. Phys. (to be published).

[7]L. Armstrong, P. Lambropoulos and N. K. Rahman, Phys. Rev. Lett. 36, 952 (1976).

[8]The term "highest powers" does not refer to an unreachable asymptotic regime, but simply implies that the one-photon ionization rate R_{2c} is somewhat larger than the bound-bound Rabi frequency Ω. This might occur if the bound-bound matrix elements were "anomalously" small, as they could easily be if the bound-bound transition were quadrupole or two-photon in character. A discussion of the latter case has been given recently, including laser-bandwidth effects, by P. Agostini *et al.* [J. Phys. B (to be published)].

[9]N. C. Wong and J. H. Eberly, Opt. Lett. 1, 211 (1977); and J. R. Ackerhalt and J. H. Eberly, Phys. Rev. A 14, 1705 (1976).

[10]The nonfluctuating monochromatic field limit of this resonant two-photon ionization rate has been obtained and discussed by B. L. Beers and L. Armstrong, Jr., and by A. E. Kazakov *et al.*, Ref. 1. Note that a weak "direct channel" for ionization from level 1 to the continuum, such as was considered by Beers and Armstrong, could easily be incorporated into (14) and (15). In the near-resonance and long-time limits discussed here we expect its effects would be inconsequential.

[11]It is pointed out by Kazakov *et al.*, Ref. 1, and by D. L. Andrews [J. Phys. B 10, L659 (1977)] that the resonant ionization channel is populated with a probability proportional to t^3 for very short times. This is a consequence of the well-known short-time t^2 dependence for the Rabi oscillation in the 1-2 transition (Ref. 2) and the linear t dependence of the transition from 2 to the continuum. Obviously, at short enough times this t^3 behavior will be dominated by the linear t behavior of a direct transition from 1 to the continuum.

[12]If the poorly controlled turn-on and turn-off for the laser are comparable in duration to the effective atomic relaxation time, then the ac Stark shifts will be time dependent. Under the same conditions, however, the entire problem becomes time dependent in an uninteresting way, and we do not consider it.

[13]B. R. Mollow, Phys. Rev. 175, 1555 (1968).

[14]See, for example, L. Allen and J. H. Eberly, Ref. 2, Secs. 6.2 and 6.3.

[15]See, for example, C. P. Slichter, *Principles of Magnetic Resonance* (Harper and Row, New York, 1963), Sec. 1.3, Eq. (21), where $\frac{1}{2}(1-n/n_0)$ is the occupation probability for the upper level, W is the stimulated absorption rate (our R_{12}), and $1/T_1$ is the population relaxation rate (our $A+R_{2c}$). After these notation changes our Eq. (36) is reproduced exactly.

[16]A similar use of the Heisenberg equations to discuss one-photon ionization has been made by K. Wódkiewicz, ICOMP Abstracts, Ref. 3, and by J. L. F. de Meijere, Ph.D. thesis (University of Rochester, 1977).

[17]See, for example, L. I. Schiff, *Quantum Mechanics*, 3rd ed. (McGraw-Hill, New York, 1968), pp. 285 and 420.

[18]See, for example, M. C. Wang and G. E. Uhlenbeck, in *Selected Papers on Noise and Stochastic Processes*, edited by N. Wax (Dover, New York, 1954). This same model has recently been exploited by K. Wódkiewicz in a study of statistical incoherence in the Bloch equations (private communication).

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