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Coherence versus incoherence: Time-independent rates for resonant two-photon ionization

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We study three ways in which two-photon ionization, at high light intensities and near an intermediate-state resonance, may be described. We calculate the instantaneous ionization rate, the total ion count, and a saturable rate constant. We find that the total ion count, which is the experimentally interesting quantity in most cases, is very closely modelled by our saturable rate constant. The instantaneous ionization rate bears no useful relation to the total ion count in many cases. In comparing these three descriptions of ionization we have included the effects of detuning from resonance, finite light bandwidth, finite intermediate-state lifetime, and light intensity; we have varied all of these parameters independently over several orders of magnitude. Our conclusions are in good agreement with, and extend the findings of, Ackerhalt and Shore.

It is well known that the interaction of light with individual atoms and molecules is, in the most general case, poorly described by the Einstein A and B coefficients.

However, in many realistic situations the phase sensitivity of the Schrödinger and Maxwell equations, which in principle govern the light-matter interaction process, can be safely ignored. In these cases the Einstein coefficients for emission and absorption are satisfactory as fundamental parameters of the interaction, and one writes so-called "rate equations" for a radiative transition as follows:

$$\frac{d}{dt} n_2 = -An_2 + R_{12}(n_1 - n_2), \tag{1a}$$

$$\frac{d}{dt} n_1 = An_2 - R_{12}(n_1 - n_2). \tag{1b}$$

Here n_1 and n_2 are the numbers of atoms or molecules in energy levels 1 and 2, A is the decay rate of level 2 due to spontaneous emission, and R_{12} is the stimulated transition rate between the levels (i.e., $R_{12} = \sigma_{12}\Phi$, where σ_{12} is the absorption cross section of the transition and Φ is the photon flux). For simplicity we have taken level 1 to be the ground level, and have assumed that the radiation field is of such wavelength, polarization, ..., etc., that transitions to levels other than 2 may be ignored.

Of course, for the understanding of many ultrashort laser-related optical phenomena[1] it is insufficient to work with rate equations like Eqs. (1), even if allowance is made for the existence of other energy levels, for collisions, for ionization, for ensemble averages due to Doppler velocity distributions, and so on.

Recently, in connection with multiphoton ionization experiments carried out with tunable, narrow-band, high-power lasers,[2] the question has arisen whether rate equations provide a valid theoretical description. Both analytic[3] and numerical[4] studies of ionization have been made in model systems under high-power excitation and a few studies[5] of high-power processes[6] in specific atoms have been completed. In many of these studies[3-5] it is pointed out that the ionization rate (i.e., the negative time derivative of the total number of atoms in bound states) is not a simple numerical constant, but can be strongly time dependent. This is supposed to mean, among other things, that a simple rate-equation treatment of the ionization process is not valid.

In this note we investigate directly the question of the possible validity of a simple rate-equation treatment of high-power ionization.[7] To do this we begin with the fully phase-preserving transition operator or density matrix equations. We then take three approaches to a solution of these equations: (i) "Adiabatic" and approximate elimination of off-diagonal coherence,[8] leading to a single ionization rate constant. This rate is still dependent on resonance detunings, laser power, atomic lifetimes, etc., of course, but is not time-dependent. (ii) Direct numerical solution of the complete set of equations, obtaining the rate of loss of bound state population as a function of time as well as a function of all the parameters mentioned in (i) above. (iii) Numerical integration of the rate found in (ii) to obtain the total number of ions produced between laser turn-on at $t=0$ and any later time t.

As we show below, the first approach leads to the "traditional" (except that it may be saturated) rate constant. The third approach gives, strictly speaking, not a rate but the ion count. It is closest in definition to what is usually measured experimentally. And the second approach obviously gives, at every instant of time, the instantaneous

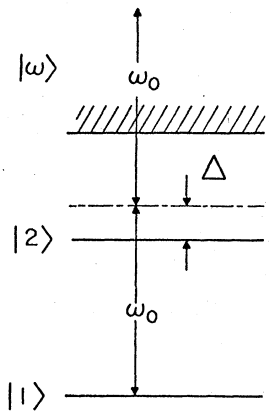

FIG. 1. Two-level atom undergoing two-photon ionization. The two-level model is expected to be accurate if the detuning Δ of the laser from the intermediate state $|2\rangle$ is smaller than the detuning from any of the other states of the real atom.

ionization rate. What is perhaps surprising is that over a broad spread of times and of ionization parameters, the experimental ion count is very closely approached in form, if not always in numerical magnitude, by the traditional rate (multiplied by t). There is, practically speaking, no significant correlation between ion count and instantaneous rate in the high-power regime.

For our purposes the two-level ionization model, studied many times in the past,[3] is adequate. The complete dynamics, after elimination of the degrees of freedom associated with the two broad continua in the problem (the electronic continuum of the atom and the continuum of vacuum radiation modes), is governed by four equations:

$$\dot{\rho}_{12} = -\left[\tfrac{1}{2}(A+W+R_{2c}) + i\Delta\right]\rho_{12} - (\tfrac{1}{2}i)\Omega(2\sigma_{22} - n), \tag{2a}$$

$$\dot{\rho}_{21} = -\left[\tfrac{1}{2}(A+W+R_{2c}) - i\Delta\right]\rho_{21} + (\tfrac{1}{2}i)\Omega(2\sigma_{22} - n), \tag{2b}$$

$$\dot{\sigma}_{22} = -(A+R_{2c})\sigma_{22} - (\tfrac{1}{2}i)\Omega(\rho_{12} - \rho_{21}), \tag{2c}$$

$$\dot{n} = -R_{2c}\sigma_{22}. \tag{2d}$$

For the most part we follow the derivation and notation of de Meijere and Eberly.[3] The model is shown in Fig. 1. The Rabi frequency Ω is linear in the laser field strength and the one-photon ionization rate from level 2 to the continuum, labeled R_{2c}, is linear in laser intensity. The model can be made to apply to more general situations than the one-photon-resonant two-photon ionization shown in Fig. 1 simply by allowing Ω and R_{2c} to be proportional to higher powers of field strength and intensity. The laser bandwidth full width at half-maximum, denoted W, has been inserted without approximation according to the phase fluctuation substitution rule discussed at length by Wódkiewicz.[9] The spontaneous emission rate connecting levels 2 and 1 is denoted A, and the detuning $\Delta = \omega_{21} - \omega$ is the amount by which the laser frequency ω falls short of the effective atomic transition frequency ω_{21}. Also, $n = \sigma_{22} + \sigma_{11}$, so that n represents the fractional number of atoms in either one of the two bound levels. The laser is assumed to have constant amplitude, so both Ω and R_{2c} are constant in time. Finally, for simplicity only two bound levels are retained, so there is no "direct" far off-resonance ionization channel present.

The first approach to a solution involves solving (2a) and (2b) and substituting the results into (2c). The "solution" of (2a) and (2b) is understood in the sense that the (presumably rapid) transients at frequency $\Gamma(\Delta) \equiv |\tfrac{1}{2}(A+W+R_{2c}) \pm i\Delta|$ are allowed to die away and the slowly varying remainders are kept[8]

$$\rho_{12} = -\tfrac{1}{2}\Omega \frac{1}{\Delta - (i/2)(A+W+R_{2c})}(2\sigma_{22} - n), \tag{3a}$$

$$\rho_{21} = -\tfrac{1}{2}\Omega \frac{1}{\Delta + (i/2)(A+W+R_{2c})}(2\sigma_{22} - n). \tag{3b}$$

The same assuption about relatively rapid transients allows (2c) to be "solved":

$$\sigma_{22} = -\frac{i}{2}\Omega \frac{1}{A+R_{2c}}(\rho_{12} - \rho_{21}). \tag{3c}$$

Then the substituion of (3a) and (3b) into (3c) gives

$$\sigma_{22} = -\frac{\Omega^2}{A+R_{2c}} \frac{\tfrac{1}{4}(A+W+R_{2c})}{\Delta^2 + \tfrac{1}{4}(A+W+R_{2c})^2}(2\sigma_{22} - n). \tag{4}$$

The "solution" for the rate entails the solution of (4) for σ_{22} and its insertion into (2c). After dividing both sides by n we find

$$\frac{d}{dt}(\ln n) = -R_{2c}\frac{R_{12}(\Delta)}{2R_{12}(\Delta) + A + R_{2c}} \equiv -R_{\text{ion}}(\Delta), \tag{5}$$

where $R_{12}(\Delta)$ is given by

$$R_{12}(\Delta) = \frac{\Omega^2}{4}\frac{A+W+R_{2c}}{\Delta^2 + \tfrac{1}{4}(A+W+R_{2c})^2}. \tag{6}$$

Note that from the definition of Rabi frequency: $\Omega = 2d_{12}\mathcal{E}/\hbar$, and intensity: $I = (c/4\pi)[E^2(t)]_{\text{av}} = (c/2\pi)\mathcal{E}^2$, we can rewrite $R_{12}(\Delta)$ in more conventional form

$$R_{12}(\Delta) = \sigma_{12}(\Delta)\Phi. \tag{7}$$

Here $I = \hbar\omega\Phi$ and $\sigma_{12}(\Delta)$ is the dipole absorption cross section:

$$\sigma_{12}(\Delta) = \frac{4\pi d_{12}^2 \omega_{21}}{\hbar c}\frac{\tfrac{1}{2}\Gamma_{12}}{\Delta^2 + (\tfrac{1}{2}\Gamma_{12})^2}, \tag{8}$$

where $\Gamma_{12} = A + W + R_{2c}$ is the total off-diagonal linewidth of the 1-2 transition. The expression for R_{12} in (7) is a generalization of the R_{12} in (1) because it is not linear in the intensity of the light: the width Γ_{12} is intensity dependent.

The completely saturated limit of (5) is clear. When $R_{12} \gg A + R_{2c}$ we find

$$R_{\text{ion}} = \tfrac{1}{2} R_{2c} . \tag{9}$$

In other words, the stimulating field is so strong that the fractional population in level 2 is $\frac{1}{2}$, and the net ionization rate is determined only by the one-photon rate from 2 to be continuum. Incomplete saturation presents a much more complicated picture, especially in the neighborhood of resonance. This case may be analyzed by rationalizing the denominator of R_{12} in (5). We find

$$R_{\text{ion}}(\Delta) = \frac{\frac{\Omega^2}{4}\,\frac{A+W+R_{2c}}{A+R_{2c}}}{\Delta^2 + \left(\frac{A+W+R_{2c}}{2}\right)^2 + \frac{\Omega^2}{2}\,\frac{A+W+R_{2c}}{A+R_{2c}}} . \tag{10}$$

Various limiting expressions that follow from (10) when the laser power or bandwidth are large or small or when the laser is on, or far off, resonance have been explored by de Meijere and Eberly.[3] They have exhibited R_{ion} as a simultaneous function of Δ and W for several values of Ω and R_{2c}, and we reproduce three of their rate surfaces in Fig. 2.

The second approach to an ionization rate solution of Eqs. (2) is simply to integrate them numerically, and display the results for $\dot{n}(t)$ as a function of the various parameters in the equations. Such an integration is trivial in principle, and complicated in practice only by the need to find the complex roots of a quartic polynomial repeatedly for many values of the ionization parameters. If we let λ_j, $j = 1, \ldots, 4$, be the roots of the determinant of coefficients of Eqs. (2), then the solution for $n(t)$ is

$$n(t) = \sum_{j=1}^{4} a_j e^{-\lambda_j t} , \tag{11}$$

and the instantaneous rate at which ions appear (bound state atoms disappear), given by $-\dot{n}(t)$:

$$-\dot{n}(t) = \sum_{j=1}^{4} \lambda_j a_j e^{-\lambda_j t} . \tag{12}$$

The initial conditions, $n(0) = 1$, $\sigma_{22}(0) = \rho_{12}(0) = \rho_{21}(0) = 0$, determine the coefficients a_j as functions of A, W, Ω, R_{2c}, and Δ.

We have found the λ's and a's numerically for a wide range of values of W, Ω, R_{2c}, and Δ relative to A. In Fig. 3 we show a typical series of results for $-\dot{n}(t)$ vs t. One sees very clearly the pronounced Rabi oscillations that are characteristic of coherent high-power laser-atom interactions.

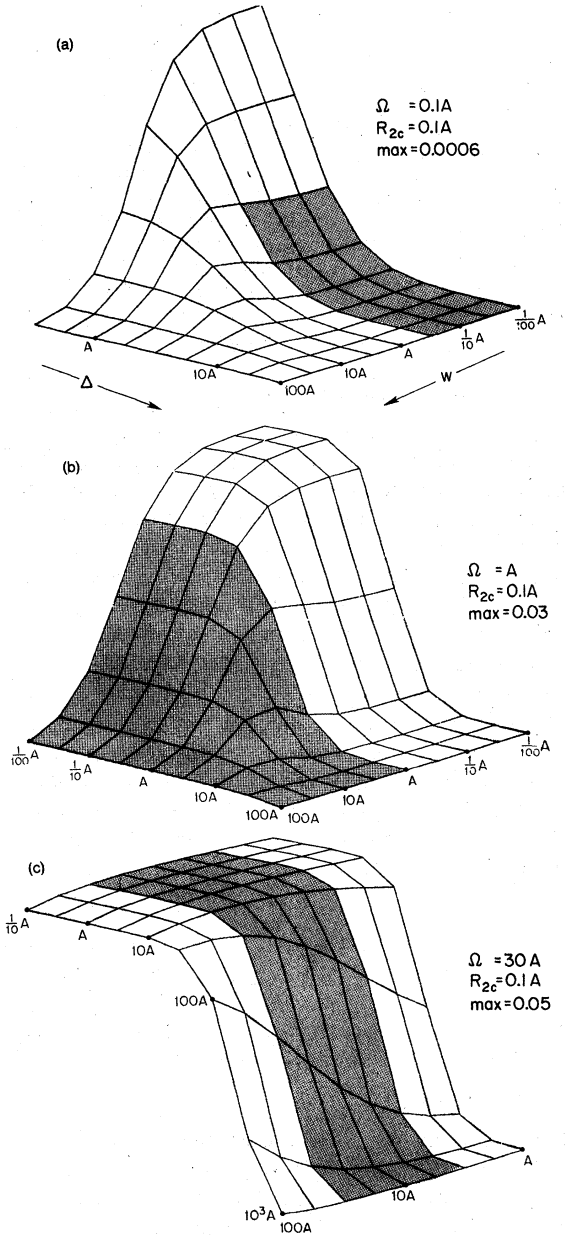

FIG. 2. Ionization rate surfaces from de Meijere and Eberly (Ref. 3). In each case the vertical height of the surfaces gives point by point values of the ionization rate R_{ion} [Eq. (10)] as a function of both detuning Δ and laser bandwidth W. Both detuning and bandwidth are scaled in terms of the Einstein spontaneous emission rate A appropriate to the intermediate state $|2\rangle$, and vary over several orders of magnitude. The Rabi frequencies increase by a factor of 300 from (a) and (c). The vertical scale of the three graphs is not the same. The changes in scale are indicated by the maxima indicated on the graphs. These maxima are the ionization rates, in units of A, associated with the highest points on the graphs. The half-tone regions are where a single time-independent rate might naively be expected to describe the ionization process (see de Meijere and Eberly, Ref. 3).

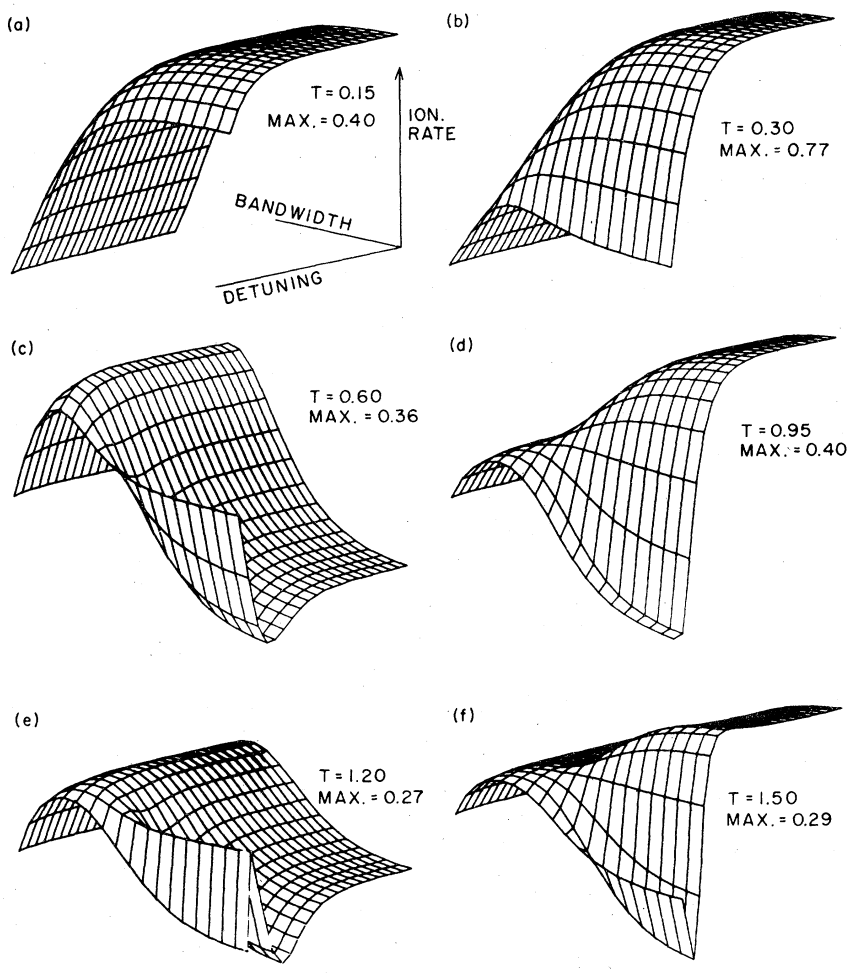

FIG. 3. Ionization rate surfaces similar to those shown in Fig. 2. In this case the instantaneous ionization rate is shown as a function of time for the fixed values $\Omega = 10A$ and $R_{2c} = A$. The pronounced Rabi oscillations are confined to the region of the surface where the bandwidth is not much larger than the Rabi frequency.

It is also clear that there is no consistent form for the instantaneous ionization rate, considered as a function of Δ, W, and time.

Finally, we show in Figs. 4–6 the time development of the total ion count, i.e., $1 - n(t)$, again considered as a simultaneous function of Δ and W. In Figs. 4–6 we have chosen the same values of Ω and R_{2c} as Fig. 2, values that correspond, respectively, to unsaturated, intermediate, and highly saturated excitation of the bound-bound transition on resonance. The last graph in each of Figs. 4–6 reproduces the saturable rate of Fig. 2. That is, it shows R_{ion} [given by (10)], for comparison with the total ion count.

The conclusion that is suggested by Figs. 4–6, in comparison with Fig. 2, is that the "traditional" (but saturable) rate constant $R_{\text{ion}}(\Delta)$ gives a surprisingly good picture of the ionization process, measured by total ion count, after a suitable time has elapsed. In strong contrast, the instantaneous rate is practically useless for estimating $1 - n(t)$. Note that $R_{\text{ion}}t$ is not actually equal to $1 - n(t)$, but is very nearly proportional to it. In Fig. 5, for example, the ratio is about 1.24, and the ratio is remarkably independent of Δ and W over several orders of magnitude of variation. To emphasize the closeness of functional agreement we show $1 - n(t)$ and R_{ion} superposed in Fig. 7.

We do not have an explanation for the apparent wide validity of R_{ion} as a quantitative measure of ion count for low and high laser intensity, and near to and far from resonance. Our finding does agree qualitatively with an observation (Ackerhalt and Shore, Ref. 4) that the instantaneous ionization rate tends to oscillate with an average value very close to that predicted by rate equations similar to (1a) and (1b). However, our result is stronger because we have reduced our rate analysis to a *single* constant R_{ion}, whereas the rate equations of Ackerhalt and Shore [their Eqs. (8) and (9)] are characterized by two decay constants.

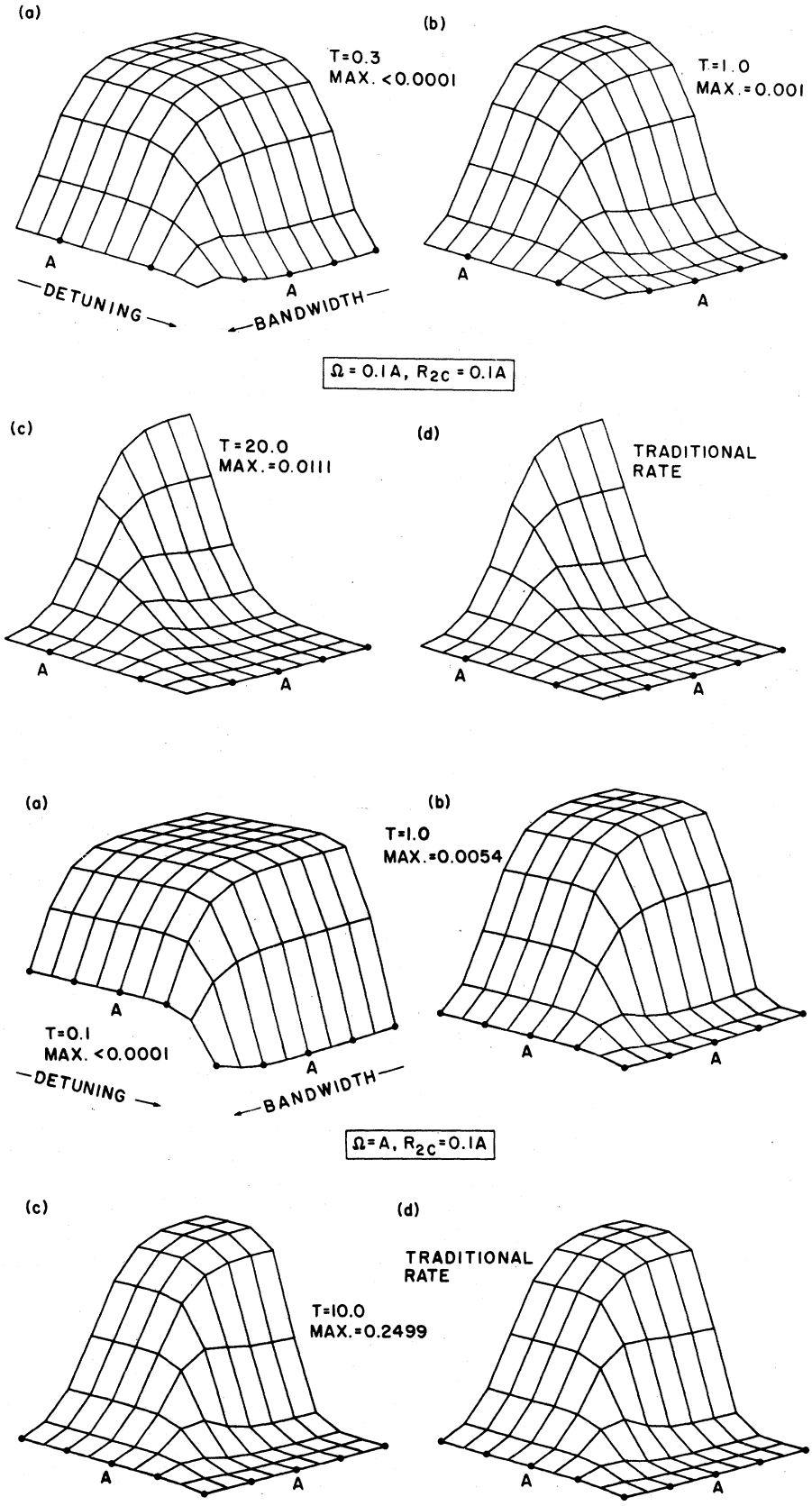

FIG. 4. Ionization rate surfaces similar to those shown in Fig. 2. In this case the total ion count $1-n(t)$ is shown as a function of time in graphs (a)–(c); and in graph (d) the corresponding "traditional" rate shown in Fig. 2 is reproduced for comparison. The points $\Delta=A$ and $W=A$ are labeled on the detuning and bandwidth axes. The black dots on the A and W axes label values of Δ and W that are one or more orders of magnitude larger or smaller than A. The range of variation of both Δ and W is over several orders of magnitude in each graph. The constant values of Rabi frequency and one-photon ionization rate are shown in the figure.

FIG. 5. Ionization rate surfaces showing the total ion count $1-n(t)$ as a function of time in (a)–(c); and the "traditional" rate of Fig. 2 in (d) for comparison. See Fig. 4 for further details.

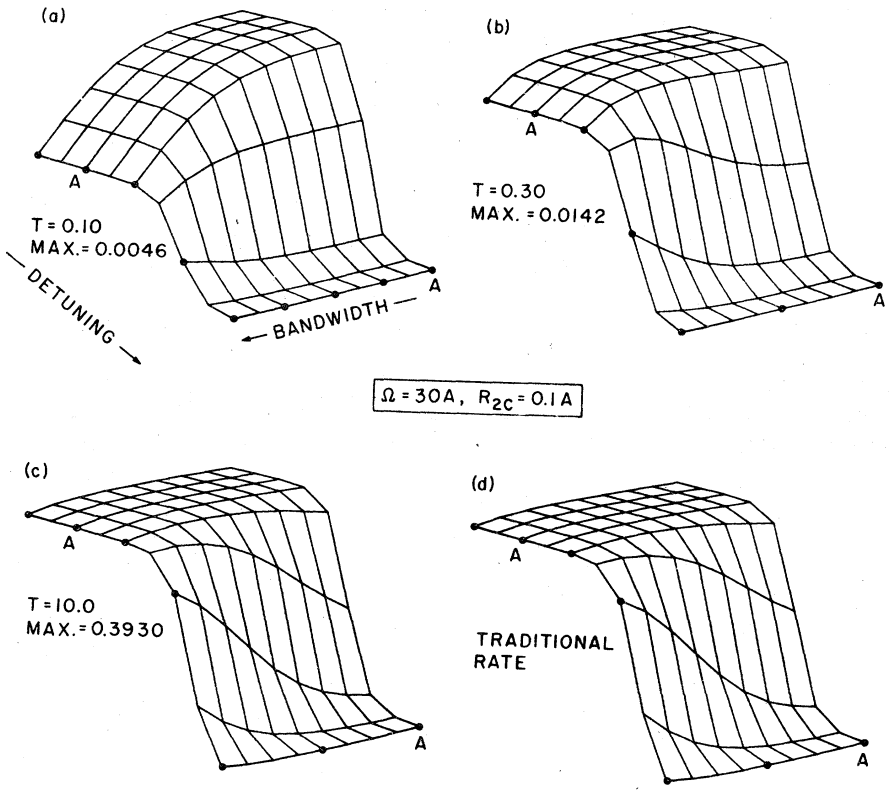

FIG. 6. Ionization rate surfaces showing the total ion count $1-n(t)$ as a function of time in (a)–(c); and the "traditional" rate of Fig. 2 in (d) for comparison. See Fig. 4 for further details.

The introduction of the concept of rate linearization in the Laplace transform domain suggests a rate-constant derivation procedure that is different from the traditional "adiabatic" elimination of off-diagonal coherence,[8] the procedure that was employed above to derive R_{ion}. In the appendix we explain rate linearization in the Laplace domain and use it to justify the significance of R_{ion}, but only in a special case. Nevertheless, the use of R_{ion} in analyzing multiphoton absorption and ionization data appears to be strongly indicated. This is particularly true in those cases where functional behavior rather than an absolute rate is being measured. Two applications, to the near-resonant power law of three-photon-resonant ionization in cesium, and the laser-bandwidth-dependence of two-photon absorption in sodium,[10] will be published elsewhere.

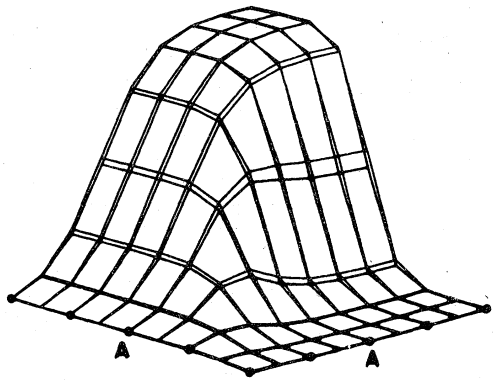

FIG. 7. Superposition of the rate surfaces showing the total ion count $1-n(t)$ and the traditional rate for the case already shown in Figs. 5(c) and 5(d).

ACKNOWLEDGMENTS

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APPENDIX

One usually says that a rate constant provides an accurate picture of a complicated process such as is modeled by Eqs. (2) if one of the eigenvalues λ_j has a substantially smaller real part than any of the others. Let λ' be the one with smallest real

part. Then, after a time, the solution for $n(t)$ will be well represented by

$$n(t) \to a' e^{-\lambda' t} , \tag{A1}$$

where a' is that coefficient a_j corresponding to $\lambda_j = \lambda'$. The negative logarithmic derivative of $n(t)$ gives

$$-\frac{d}{dt} \ln n(t) \to \lambda' , \tag{A2}$$

which is the analog of R_{ion}.

The question is: can one determine the properties of the smallest eigenvalue in any simple way? To answer this question in a limited sense we compute the Laplace transforms of Eqs. (2). Let the Laplace parameter be s. Then the transform of σ_{22} may easily be shown to be

$$\tilde{\sigma}_{22}(s) = \tfrac{1}{4}\Omega^2 [(2s+\Gamma)/D(s)] , \tag{A3}$$

where $D(s)$ is the determinant of the coefficient matrix of Eqs. (2):

$$D(s) = \begin{vmatrix} s+\frac{1}{2}\Gamma + i\Delta & 0 & i\Omega & -\frac{1}{2}i\Omega \\ 0 & s+\frac{1}{2}\Gamma - i\Delta & -i\Omega & \frac{1}{2}i\Omega \\ \frac{1}{2}i\Omega & -\frac{1}{2}i\Omega & s+\Gamma - W & 0 \\ 0 & 0 & 0 & s \end{vmatrix} . \tag{A4}$$

Of course, in terms of the eigenvalues λ_j we also have $D(s) = (s+\lambda_1)(s+\lambda_2)(s+\lambda_3)(s+\lambda_4)$; and we can obviously write

$$D(s) = A + Bs + Cs^2 + Ds^3 + s^4 . \tag{A5}$$

Now, if there is one very small root of $D(s)$, it should be sufficient, as far as that root goes, to approximate $D(s)$ by its linearized counterpart

$$[D(s)]_{\text{lin}} = A + Bs . \tag{A6}$$

In other words, the linearized smallest root is

$$\lambda'_{\text{lin}} = -A/B . \tag{A7}$$

This allows $\tilde{\sigma}_{22}(s)$ to be inverted trivially. From the resulting $\sigma_{22}(t)$ and Eq. (2d) it follows that the "linear" approximation to $n(t)$ is

$$[n(t)]_{\text{lin}} = e^{-(A/B)t} . \tag{A8}$$

That is, the coefficient $a_j = a'$ corresponding to $\lambda_j = \lambda'$ is exactly one in this linearized limit. Thus it is clear what the connection between $1 - n(t)$ and λ' is in this case. Since λ' is very small, by hypothesis, we therefore have simply:

$$1 - n(t) = 1 - [1 - (A/B)t + \cdots] \approx (A/B)t , \tag{A9}$$

so A/B is to be identified with R_{ion}.

The precise connection with our numerical observation that $1 - n(t) \approx R_{\text{ion}} t$ is made by working out the value of A/B and comparing it with R_{ion}. One finds

$$\frac{A}{B} = \frac{\Omega^2}{4} \frac{\dfrac{A + W + R_{2c}}{A + R_{2c}}}{\Delta^2 + \left(\dfrac{\Gamma}{2}\right)^2 + \dfrac{\Omega^2}{2} \dfrac{A + W + 2R_{2c}}{A + R_{2c}}} R_{2c} . \tag{A10}$$

This is insignificantly different from R_{ion} [Eq. (10)] under the present assumption that A/B is small.

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[1]For example, equations like (1) completely fail to describe photon echoes, self-induced transparency, superradiance, and the operation of the laser itself. See H. M. Nussenzveig, *Introduction to Quantum Optics* (Gordon and Breach, New York, 1973); M. Sargent III, M. O. Scully, and W. E. Lamb, Jr., *Laser Physics* (Addison-Wesley, Reading, Mass., 1974); and L. Allen and J. H. Eberly, *Optical Resonance and Two-Level Atoms* (Wiley, New York, 1975).

[2]See reviews and references in *Multiphoton Processes*, edited by J. H. Eberly and P. Lambropoulos (Wiley, New York, 1978).

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[6]Throughout this paper the terms high power and high intensity have a specific, but not absolute, meaning, namely that the laser field is strong enough to saturate one of the atomic transitions. This means that at least one level in addition to the ground level will be significantly populated.

[7]In a previous study of this specific question (J. R. Ackerhalt and J. H. Eberly, Ref. 3) a set of conditions on laser power were imposed. These are removed in the present work.

[8]See, for example, P. W. Milonni and J. H. Eberly, J. Chem. Phys. 68, 1602 (1978).

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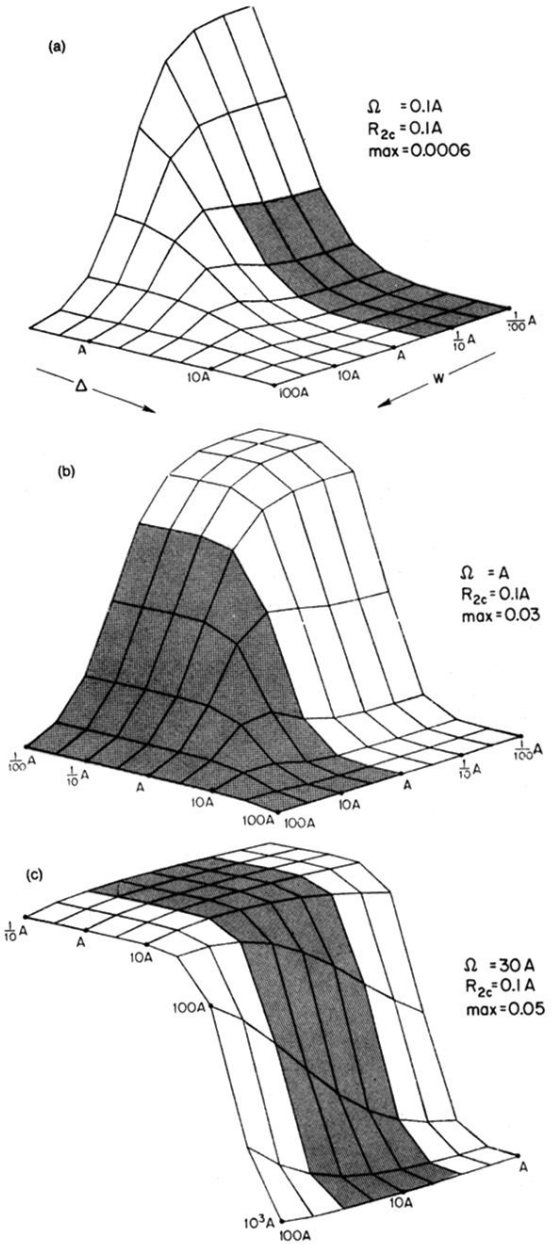

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