27
3-21-77
75 cy. to NTIS

ORNL/TM-5754

Two-State and Two-State Plus Continuum Problems Associated with the Interaction of Intense Laser Pulses with Atoms

C. W. Choi
M. G. Payne

OAK RIDGE NATIONAL LABORATORY
OPERATED BY UNION CARBIDE CORPORATION FOR THE ENERGY RESEARCH AND DEVELOPMENT ADMINISTRATION

DISCLAIMER

Printed in the United States of America. Available from
National Technical Information Service
U.S. Department of Commerce
5285 Port Royal Road, Springfield, Virginia 22161
Price: Printed Copy $5.50; Microfiche $3.00

ORNL/TM-5754

Contract No. W-7405-eng-26

HEALTH PHYSICS DIVISION

TWO-STATE AND TWO-STATE PLUS CONTINUUM PROBLEMS ASSOCIATED WITH THE INTERACTION OF INTENSE LASER PULSES WITH ATOMS

C. W. Choi and M. G. Payne

Submitted by C. W. Choi as a dissertation to the Graduate School of the University of Kentucky in partial fulfillment of the requirements for the degree of Doctor of Philosophy

Date Published - February 1977

OAK RIDGE NATIONAL LABORATORY
Oak Ridge, Tennessee 37830
operated by
UNION CARBIDE CORPORATION
for the
ENERGY RESEARCH AND DEVELOPMENT ADMINISTRATION

THIS PAGE

WAS INTENTIONALLY

LEFT BLANK

CONTENTS

CONTENTS (continued)

LIST OF FIGURES

THIS PAGE

WAS INTENTIONALLY

LEFT BLANK

LIST OF TABLES

THIS PAGE

WAS INTENTIONALLY

LEFT BLANK

ACKNOWLEDGEMENTS

The authors wish to express gratitude to G. S. Hurst, F. Gabbard, and W. R. Garrett for their support and useful discussions.

During the research work, one of the authors (CWC) received financial support through the Laboratory Graduate Participation Program of the Oak Ridge Associated Universities.

THIS PAGE

WAS INTENTIONALLY

LEFT BLANK

ABSTRACT

Due to the development of high power tunable lasers, a series of interesting new laser interaction problems have emerged. These have to do with phenomena occurring when a high power pulsed laser is tuned near either a one-photon or a multiphoton resonance. In the latter situation, one finds that the time dependence of the a.c. Stark shifts and power broadening leads to several novel effects. For instance, Grischkowsky and Loy have shown that a strong pulsed laser tuned between the ground state and a two-photon resonance can lead to a nearly completely inverted population.

In the present work we have utilized two mathematical methods (one a form of adiabatic approximation, and the other closely related to the Zener method from collision theory) in order to calculate the probability of three-photon ionization when strong counter propagating pulses are tuned very near a two-photon resonant state. In this case the inverted populations predicted by Grischkowsky and Loy for smooth laser pulses lead to larger ionization probabilities than would be obtained for a square pulse of equal peak power and energy per pulse. The line shape of the ionization probability is also quite unusual in the above problem. A sharp onset in the ionization probability occurs

as the lasers are tuned through the exact unperturbed two-photon resonance. Under proper conditions, the change can be from a very small value to one near unity. It occurs in a very small frequency range determined by the larger of the residual Doppler effect and the reciprocal duration of the pulse. Thus, the line shape retains a Doppler-free aspect even at power levels such that power broadening would dwarf even the full Doppler effect in the case of a square pulse of equal energy and peak power. The same mathematical methods have been used to calculate line shapes for the two-photon excitation of fluorescence when the atoms see a pulsed field due to their time of passage across a tightly focused cw laser beam. Thus, the mathematical methods used above permitted accurate analytical calculations under a set of very interesting conditions.

We have also utilized one of the mathematical methods in order to treat two-photon ionization by a pulsed laser tuned near a resonance transition. These works are believed to represent the first nonperturbational treatment of multiphoton ionization due to a pulsed laser tuned near a one-photon or multiphoton resonance.

CHAPTER I

INTRODUCTION

In recent years lasers have been applied in many fields of physics, particularly in atomic physics and precision spectroscopy.[1-9] In addition, there have been many suggestions as to how to use lasers for isotope separation.[10-13] Other applications of lasers have been suggested[14-15] and demonstrated,[16] and numerous theoretical studies[17-20] have been devoted to the use of cw lasers for these applications.

Our present work involves theoretical estimation of photoionization probability and fluorescence due to tunable pulsed lasers, and the development of mathematical methods which enable us to approach the problems with considerable ease. We find that by using proper pulse shapes and high power levels it is possible to enhance photoionization probabilities of atoms or molecules approximately 100% in comparison with the case of using square pulses with identical energy per pulse, resulting in superior efficiency (see Figs. 3 and 4). In addition to higher efficiency, we will show that, in some cases, there is higher selectivity when smooth pulses are used for near resonance multiphoton processes due to a sharp onset of the process at the unperturbed resonance which is totally masked at such power levels with cw lasers because of power broadening. In some cases it is possible to have a photoionization probability of unity with existing high power lasers (see Figs. 3 and 12). Some[21-24] have studied the case with cw lasers, but there has been no thorough understanding of the effect of the pulses with time-dependent amplitude until our present studies.

In addition, we also emphasize the experimental applications in the production of very short but intense pulses of metastable atoms for crossed beam scattering experiments employing abrupt change of pulse amplitude, and in the lifetime measurement of excited states by way of the time-gating technique. Using a similar saturated photoionization concept, Hurst et al.[16] have developed and demonstrated a one-atom detection technique.

Considerable effort has been devoted to verification, through the use of a computer, of the validity of our "Factorization Method," which is simple but powerful in solving a set of coupled differential equations. We have also applied a mathematical technique which we call the "Method of Isolated Curve Crossings" to laser-atom interaction problems. The latter method is mathematically equivalent to Zener's method[25] from collision theory. The Factorization Method and the Method of Isolated Curve Crossings complement each other such that many pulsed laser-atom interactions can be dealt with by applying one of these methods.

CHAPTER II

MATHEMATICAL METHODS FOR LASER-MATTER INTERACTION PROBLEMS

A. The Factorization Method

We begin by describing a non-perturbative method for solving the time dependent Schrödinger equation in cases where the problem reduces to one for a system with a finite basis of state functions. For simplicity, we restrict the discussion to the case where a two-state description is a good approximation. The problems that we will discuss later will show that this simplification is not as restrictive as it first seems.

As we shall see later, many interesting problems involving either collisions or the interaction of a powerful laser pulse with a low density gas involve the solution of a special case of the following set of differential equations:

$$\frac{da_0}{dt} = iu(t)\, e^{iq(t)}\, a_2,$$

$$\frac{da_2}{dt} = -\gamma(t)a_2 + iu^*(t)e^{-iq(t)}\, a_0. \tag{1}$$

In Eqs.(1) we shall assume that $q(t)$ and $\gamma(t)$ are real and that

$$\frac{dq}{dt} = \delta + \alpha_{01}\, g(S_1 t) + \alpha_{02}\, g^2(S,t) + \alpha_{03}\, f^2(S_2 t),$$

$$\gamma(t) = \alpha_{11}\, g(S_1 t) + \alpha_{12}\, g^2(S_1 t) + \alpha_{13}\, f^2(S_2 t) + \gamma_2/2, \qquad (2)$$

$$u(t) = \alpha_{21}\, g(S_1 t) + \alpha_{22}\, g^2(S_1 t) + \alpha_{23}\, g(S_1 t)\, f(S_2 t),$$

where S_1 and S_2 are small positive numbers having units of frequency, the α_{ij}'s are constants having units of frequency, and $g(x)$ and $f(x)$ are smooth functions having a single maximum of unit magnitude at $x = 0$ while dropping off monotonically to zero at $x = \pm\infty$. An example of a suitable $g(x)$ and $f(x)$ is $[1 + x^2]^{-n}$, $n \geq 1$.

Typically, in a laser interaction problem, a_0 and a_2 would be probability amplitudes for being in states $|0\rangle$ and $|2\rangle$ at t, δ would represent an amount of detuning in frequency from a resonance by the laser pulse, $u(t)$ is a laser or collision (or both) induced coupling term and $\gamma(t)$ is partly a rate of collision or laser-induced ionization or dissociation out of state $|2\rangle$ and partly (i.e., $\gamma_2/2$) spontaneous emission. The remainder of dq/dt would usually represent a frequency detuning effect due either to collisional interaction or a.c. Stark shift. We will not be more precise about physical interpretation at this point since examples where Eqs. (1) arise will be discussed in later chapters.

We will now discuss the solution of Eqs. (1) subject to the initial conditions $a_0(-\infty) = 1$, $a_2(-\infty) = 0$. We proceed by eliminating $a_0(t)$ as follows:

$$\frac{d^2 a_2}{dt^2} + F_1(t)\,\frac{da_2}{dt} + F_2(t)a_2 = 0, \tag{3}$$

where

$$\begin{aligned} F_1(t) &= \gamma(t) - \frac{d}{dt}\,\ell n u^*(t) + i\,\frac{dq}{dt}\,, \\ F_2(t) &= \frac{d\gamma}{dt}(t) - \gamma(t)\,\frac{d}{dt}\,\ell n u^*(t) + i\gamma(t)\,\frac{dq}{dt} + |u(t)|^2. \end{aligned} \tag{4}$$

As mentioned earlier, we want to solve in a way that takes full advantage of the fact that S_1 and S_2 are very small. Depending on the problem being investigated, S_1^{-1} and S_2^{-1} are either related to a time of collision or to the length of the laser pulse. Thus, the method would apply to laser interaction problems with long smooth amplitudes for the laser field or to slow collisions.

With very small S_1 and S_2 the functions $F_1(t)$ and $F_2(t)$ are slowly varying functions of t with $F_2(\pm\,\infty) =$

$i\gamma_2\delta/_2$, and $F_1(\pm\infty) = i\delta + \gamma_2/2$. Both are generally complex numbers. We want to take advantage of the slow variation of F_1 and F_2 with t. To do this, we write

$$\left(\frac{d}{dt} + g_1\right)\left(\frac{d}{dt} + g_2\right) a_2 = 0, \qquad (5)$$

where g_1 and g_2 must satisfy

$$g_1 + g_2 = F_1(t),$$
$$\frac{dg_2}{dt} + g_1 g_2 = F_2(t). \qquad (6)$$

One scheme for finding a_2 is immediately obvious. In the limit of exactly constant F_1 and F_2 (or in the limit $S_1 \to 0$ and $S_2 \to 0$), g_1 and g_2 satisfy ($g_{10}(-\infty) = i\delta$, $g_{20}(-\infty) = \frac{\gamma_2}{2}$)

$$g_{10} + g_{20} = F_1(t),$$
$$g_{10}g_{20} = F_2(t). \qquad (7)$$

Thus, when S_1 and S_2 are very small, but not zero, the true g_1 and g_2 are expected to be close to g_{10} and g_{20}. We let

$$g_1 = g_{10} + \varepsilon_1,$$
$$g_2 = g_{20} + \varepsilon_2, \tag{8}$$

and find

$$\varepsilon_1 = -\varepsilon_2,$$

and

$$\frac{d\varepsilon_2}{dt} + (g_{10} - g_{20})\varepsilon_2 - \varepsilon_2^2 = -\frac{dg_{20}}{dt} . \tag{9}$$

Now, g_{20} depends only on S_1t and S_2t and should yield a small dg_{20}/dt; also, $d\varepsilon_2/dt$ and ε_2^2 are expected to be of even higher degree of smallness. The situation is suggestive of a rapidly converging iterative method. Take the zeroth iterate of ε_2 (i.e., $\varepsilon_{2,0}$) to be zero; then the n + 1 iterate, $\varepsilon_{2,n+1}$, satisfies

$$\varepsilon_{2,n+1} = -\frac{dg_{20}/dt}{g_{10}-g_{20}} + \frac{[\varepsilon_{2,n}]^2 - d\varepsilon_{2,n}/dt}{g_{10} - g_{20}} . \tag{10}$$

Typically, one or two iterations are sufficient if

$$S_1/\delta \lesssim 0.1 \text{ and } S_2/\delta \lesssim 0.1, \tag{11}$$

assuming that $|\sum_i \alpha_{0i}|$ is order of $|\sum_i \alpha_{2i}|$. This condition can be relaxed a bit if the α_{ij}'s are very large compared with δ; but, typically, to achieve convergence at all t, one cannot deviate far from Eq. (11). If Eq. (11) is satisfied, the method is valid for large or intermediate α_{ij} with greater accuracy being attained with large α_{ij}. Once g_1 and g_2 are known, we let

$$\frac{da_2}{dt} + g_2 a_2 = Z(t), \tag{12}$$

and find

$$Z(t) = iu^*(t)e^{-iq(-\infty)}\exp\left[\int_{-\infty}^{t}(g_1(t') + \frac{d}{dt'}\ln u^*(t'))dt'\right] . \tag{13}$$

Thus,

$$a_2(t) = ie^{-iq(-\infty)}\exp[-\int_{-\infty}^{t} g_2(t'')dt''] \int_{-\infty}^{t} u^*(t')dt'$$
$$\times \exp\left[\int_{-\infty}^{t'}(g_2(t'')-g_1(t'') - \frac{d}{dt''}\ln u^*(t''))dt''\right]. \tag{14}$$

If t is not extremely large and positive, the same conditions which make valid the method used to find g_1 and g_2

allow the approximate evaluation of $a_2(t)$:

$$|a_2(t)|^2 = |u(t)/D(t)|^2 \exp[2\text{Re}Y_1(t)] \qquad \text{if } \delta \neq 0,$$

(15)

$$|a_2(t)|^2 = |u(t)|^2 \; |\exp[Y_1(t)]/D(t) - \exp[Y_2(t)]/D(-\infty)|^2 \qquad \text{for all } \delta,$$

where

$$D(t) = g_{20}(t) - g_{10}(t),$$

$$Y_1(t) = -\int_{-\infty}^{t} [g_1(t') + \frac{d}{dt} \ln u^*(t')]dt',$$

$$Y_2(t) = -\int_{-\infty}^{t} [g_2(t') + \frac{d}{dt} \ln u^*(t')]dt' .$$

Equations (15) are particularly useful when they are a probability of an ionization or a dissociation that is desired and $\gamma_2 = 0$. In such a case, one desires $1 - |a_0|^2 - |a_2|^2$, and Eqs. (1) imply

$$-\int_{-\infty}^{\infty} (a_0^* \frac{da_0}{dt'} + a \frac{da_0^*}{dt'} + a_2^* \frac{da_2}{dt'} + a_2 \frac{da_2^*}{dt'})dt'$$

$$= 1 - |a_0(\infty)|^2 - |a_2(\infty)|^2 = 2\int_{-\infty}^{\infty} \gamma(t) \; |a_2(t')|^2 \, dt', \qquad (16)$$

$$\equiv R.$$

Thus, one only needs $|a_2(t)|^2$ while $\gamma(t)$ is large; consequently, Eqs. (15) are adequate for calculating R. It should be noted that $|a_2(\infty)|^2$ cannot be calculated from Eqs. (15) since Eqs. (15) predict zero, while the remainder integral that is neglected in integrating by parts is not zero and is consequently no longer negligible. In evaluating Eq. (14) or Eqs. (15), one does not need to integrate from the limiting value $-\infty$, but from the time when very small population changes begin to occur, $a_0 \simeq 1$ and $a_2 \simeq 0$. Of course, for the square pulse cases one needs to integrate only for the time span of the pulse duration. It should be noted that when $u(t) \to 0$, one can assume $\frac{F_2(t)}{F_1(t)} \simeq 0$, and Eq. (14) approaches the first order time-dependent perturbation result.

The method described above will hereafter be designated as the factorization method. It can be used (with more difficulty) in cases where there are more than two simultaneous equations and the equations are of a different form. For instance, it is very useful for obtaining solutions of the equations for the density matrix elements in laser interaction problems.

The above method was first used by Payne[26] in connection with inelastic collisions that are switched by the presence of an intense laser beam. It was subsequently used by Payne and Nayfeh[27] in connection with a scattering problem and the interaction of a laser pulse with a two-level atom.

B. Method of Isolated Curve Crossings

We describe here a method that is closely analogous to the Landau-Zener method[25] in scattering theory. At the time that most of the work described here was done the method had not been used in connection with laser interaction problems. However, in a recent work Lau[28] has used the method to predict the transition probability for two photon excitation of the Na(3s) → Na(5s) transition. In a later chapter we will use the method to solve another laser interaction problem.

In this case we desire a solution to the equations

$$\frac{da_0}{dt} = i\alpha F(t)\, e^{i\phi(t)}\, a_2, \tag{17}$$

$$\frac{da_2}{dt} = -\gamma(t)a_2 + i\alpha^* F(t) e^{-i\phi(t)}\, a_0,$$

where

$$\phi(t) = -\mu \int_{-\infty}^{t} h(t')\, dt' + \delta t, \tag{18}$$

and F(t) as well as h(t) are slowly varying functions which satisfy $F(\pm\infty) = h(\pm\infty) = 0$. Both $F(t)$ and $h(t)$ are real, positive and continuous functions having a single maximum of unit amplitude at $t = 0$. However, $F(t)$ and $h(t)$ need not be symmetric about $t = 0$. We assume $a_0(-\infty) = 1$ and $a_2(-\infty) = 0$.

We deal here with situations where τ_m is a measure of the smaller full width at half maximum of $F(t)$ and $h(t)$ and $|\mu|\tau_m >> 1$ where μ is a real quantity having units of angular frequency. It is typically a measure of the maximum detuning due to the a.c. Stark shifts or, in collision problems, of the detuning due to the atom-atom interaction. When $|\alpha/\mu| << 1$ ($|\alpha|^{-1}$ is a measure of the shortest time over which a_0 or a_2 can change appreciably), we see that, except for points where $\mu h(t) \simeq \delta$, $e^{i\phi(t)}$ will oscillate many times over a time $|\alpha|^{-1}$ so that no change in a_0 or a_2 can occur, even in cases where $|\alpha|\tau_m >> 1$. Consequentially, a_0 and a_2 change only at times very near t_{01} and t_{02}, where these are the two solutions of

$$\mu h(t) - \delta = 0;$$

of course, t_{01} is negative and t_{02} is positive. Thus, when $|\mu|\tau_m >> 1$, $|\alpha/\mu| << 1$, and δ is not too small, changes in a_0 and a_2 come from very short time intervals (compared with τ_m) about crossings which are isolated. The Landau-Zener method is then applicable. The factorization method would require that at points where $\mu h(t)-\delta = 0$, the term $\alpha F(t)$ be large enough so that changes in a_0 and a_2 are saturated. However, in the present method, α can be rather small. Correspondingly, the factorization method does not require $|\alpha/\mu|<<1$ so that the two methods complement each other and enable one to solve a rather wide range of laser interaction problems involving high power pulses which vary smoothly with time.

With the assumptions $|\mu|\tau>>1$ and $|\alpha/\mu|<<1$, we expect that $|a_0|^2$ and $|a_2|^2$ remain 1 and 0, respectively, until just before the negative time at which $\mu h(t) = \delta$. If δ is not of the same sign as μ, such a point will not exist; and $|a_0|^2$ and $|a_2|^2$ will remain near 1 and 0, respectively, throughout the pulse. If μ and δ are of the same sign, we can approximate near the negative time crossing by

$$\frac{da_0}{dV} = i\alpha_1 \, e^{i\beta_1 V^2} \, a_2,$$

$$\frac{da_2}{dV} = i\alpha_1^* \, e^{-i\beta_1 V^2} \, a_0, \qquad (19)$$

where $\alpha_1 = \alpha F(t_{01})e^{i\phi(t_{01})}$, $\beta_1 = -\frac{\mu}{2}\frac{dh(t_{01})}{dt}$, and $V = t-t_{01}$.

Thus, to find a_0 and a_2 just after the first crossing we solve Eqs. (19) at $V \to \infty$ using a_0 $(V = -\infty) = 1$, a_2 $(V = -\infty) = 0$. We have assumed that the time interval over which a_0 and a_2 change is so short that the damping (i.e. $-\gamma(t)a_2$) can be neglected during the crossing. In order to replace F(t) by a constant and h(t) by $h(t_{01}) + \frac{dh(t_{01})}{dt} V$ we must be able to choose a $|V_m| << |t_{01}|$ which satisfies $|2\beta_1 V_m| >> |\alpha_1|$. The latter condition leads to $|\alpha_1/\mu| << |\frac{dh(t_{01})}{dt} t_{01}|$, which will be true for $|\alpha/\mu| << 1$ and $|\mu \tau_m| >> 1$ until either $|\delta|$ becomes very many times smaller than $|\mu|$ or δ approaches μ and t_{01} becomes too small.

We eliminate a_0 from Eqs. (19) and let

$$a_2 = U \exp(-\beta_1 V^2/2) \tag{20}$$

and

$$Z = (2\beta_1)^{1/2} \exp(-i \pi/4) \cdot V ; \tag{21}$$

then

$$\frac{d^2U}{dZ^2} + \left(i \frac{|\alpha_1|^2}{2\beta_1} + \frac{1}{2} - \frac{Z^2}{4}\right)U = 0. \tag{22}$$

Eq. (22) has a general solution in terms of parabolic cylinder functions (Weber functions) $D_n(Z)$, where

$$\frac{d^2D_n(Z)}{dZ^2} + (n + \frac{1}{2} - \frac{Z^2}{4})\ D_n(Z) = 0. \tag{23}$$

Applying the boundary conditions $|a_2(V = -\infty)|^2 = 0$ and $|a_0(V = -\infty)|^2 = 1$, we find the asymptotic solution[25] $|U(V \to \infty)|^2 = 1 - \exp\ [-2\pi|Z_1|^2]$ where $|Z_1|^2 = \frac{|\alpha_1|^2}{2|\beta_1|}$. Thus, just after t_{01} we have

$$|a_2|^2 = 1 - \exp\ [-2\pi|Z_1|^2]. \tag{24}$$

When $t_{01} \le t \le t_{02}$ we can neglect coupling between a_0 and a_2 and

$$\frac{da_2}{dt} \simeq -\ \gamma(t)a_2,$$

$$|a_2|^2 = [1 - \exp\ (-2\pi|Z_1|^2)]\ \exp\ [-2\int_{t_{01}}^{t} \gamma(t')dt']. \tag{25}$$

When t approaches t_{02}, the values of a_0 and a_2 will start to change again and equations analogous to Eqs. (19) will apply.

Then,

$$\frac{da_0}{dV} = i\alpha_2\, e^{i\beta_2 V^2} a_2,$$
$$\frac{da_2}{dV} = i\alpha_2^{*}\, e^{-i\beta_2 V^2} a_0, \tag{26}$$

with $V = t - t_{02}$, $\alpha_2 = \alpha F(t_{02}) \exp\,[i\phi(t_{02})]$ and $\beta_2 = -\frac{\mu}{2}\frac{dh(t_{02})}{dt}$. The relative phase of a_0 and a_2 at the second crossing is very sensitive to the exact details of the first crossing. Different pulses having nearly the same $|Z_1|$ can lead to very different relative phases at the second crossing. Let $|a_2(t_{02}-)|^2$ and $|a_0(t_{02}-)|^2$ be values of $|a_2|^2$ and $|a_0|^2$ just before the second crossing. Averaging over pulses that are repeatable on $|Z_1|$, but a bit different we find for $|a_2|^2$ just after the crossing

$$|a_2(t_{02}+)|^2 = |a_0(t_{02}-)|^2\left(1-e^{-2\pi|Z_2|^2}\right) + |a_2(t_{02}-)|^2\, e^{-2\pi|Z_2|^2}, \tag{27}$$

where $|Z_2|^2 = |\alpha_2|^2/(2|\beta_2|)$ and $|a_2(t_{02}-)|^2$ is determined from Eqs.(25) but $|a_0(t_{02}-)|^2$ depends on the interpretation of $\gamma(t)$ (i.e., whether it repopulates the initial state or represents a loss).

The considerations given above can be generalized considerably, but we have given all that will be needed for our application.

CHAPTER III

DOPPLER-FREE CONTRIBUTION TO THREE-PHOTON IONIZATION BY PULSED LASERS

A. INTRODUCTION

We consider an atom bathed in counter propagating laser beams of frequency w_1' and w_2'. The laser beams are assumed to be pulsed with the counter propagating pulses at frequencies w_1' and w_2' overlapping in both space and time. The laser pulses are very monochromatic with the band width being limited entirely by the pulse length. A fairly tractable mathematical situation is obtained if the pulses are of nearly equal length with the timing being such that one pulse begins to arrive at the atom's location at about the same time as the other and if the peak power density is $\gtrsim 10^5$ w/cm^2 so that the laser field can be treated classically.*

The case where $w_1' = w_2'$ has been studied fairly extensively[17-20] for cw lasers in cases where $2\hbar w_1'$ is nearly equal to $E_2 - E_0 = \hbar(w_2 - w_0)$, with E_2 and E_0 being the energies of two levels of the atom which are connected by

*Semiclassical treatment is valid if $S\lambda^3/C >> 1$, where C is the speed of light and S and λ are the photon flux and the wave length of the laser field. J.J. Sakuri discusses more details in Ch. 2 of his book, Advanced Quantum Mechanics.

an allowed two-photon transition. In such a case, two-photon absorption is dominated by a Doppler-free contribution in the region very near the two-photon resonance, providing (1) the lifetime of the upper state, τ_2, is such that $\tau_2^{-1} << \bar{V}\, w_1'/C$; (2) the peak power is sufficiently low that power broadening does not approach $\bar{V}\, w_1'/C$. Intuitively, the reason for the Doppler-free contribution is obvious. By absorbing one photon going one direction and another going the opposite direction, the total energy intake by an atom of velocity V_z is $\simeq \hbar\, w_1'(1-V_z/C) + \hbar\, w_1'(1+V_z/C) \simeq 2\hbar\, w_1'$. Thus, if $2\hbar\, w_1'$ is very near the excitation energy, this process is resonant for any V_z. On the other hand, the absorption of photons with the same propagation direction leads to an energy mismatch $\simeq (V_z/C)\, 2\hbar\, w_1'$ which, for typical V_z, is sufficient to suppress the contribution unless power broadening or the line width due to the state's lifetime can overcome the energy defect.

If two lasers of frequencies w_1' and w_2' (such that $w_1'+w_2' - (w_2-w_0) \simeq 0$) are used, one achieves far greater versatility.[7-8] Firstly, the coupling for a two-photon transition can be made far greater at a given power level by tuning w_1' to be rather near an intermediate state. Thus, a.c. Stark shifts and two-photon transition rates can be large at moderate power without the necessity of an intermediate state lying almost exactly halfway between the initial and final states. Secondly, if the two laser beams propagate in opposite directions and w_1' and w_2' are different by at least a few percent, then the only resonant situation corresponds to absorbing one photon propagating in one direction and another propagating in the opposite direction. (Generally, absorption of two photons propagating in the same direction is far out of resonance.) In such a situation the only remaining Doppler shift for the two-photon transition is $|w_1'-w_2'|V_z/C$, which is very small if w_1' and w_2' differ by only a few percent. In the present case we write the field in the general form

$$E(z,t) = E_1(t)\cos(w_1't - k_1V_zt) + E_2(t)\cos(w_2't + k_2V_zt + \beta). \quad (28)$$

The Doppler-free aspect of both the one-laser and two-laser situations with counter-propagating beams was first recognized by Vasilenko et al.[5] In the two-laser beam case, Doppler effects can be neglected if (1) $\tau_2^{-1} >> |w_1' - w_2'| \times |\bar{V}_z|/C$; (2) $\tau^{-1} >> |w_1' - w_2'||\bar{V}_z|/C$, where τ is laser pulse length; (3) a.c. Stark shifts or power broadening are large compared with the residual Doppler shift. The Doppler-free two-photon process has been studied experimentally by Levinson and Bloembergen,[3] Biraben et al.,[6] and others.[4-5] In the present work we will assume counter-propagating beams.

However, much of the present work will emphasize high power levels where the line widths for the two-photon process is larger than the full Doppler width. We will show that even in the above case, sharp features in the line shape remain which are characteristic of the near absence of Doppler effect.

The present work differs from other work by simultaneously taking into account the pulsed nature of the laser field (which is necessary if high power levels are to be reached) and the calculation of three-photon ionization in the neighborhood of a two-photon resonance. The ionization provides an extremely sensitive way to monitor the line shape of the two-photon transitions.

B. STATEMENT OF MODEL

The atom interacting with the highly monochromatic counter propagating laser pulses is assumed to be part of a very low density gas or vapor so that it can be considered isolated. We further assume that it has energy levels something like that shown in Fig. 1. In Fig. 1 the states $|0\rangle$ and $|2\rangle$ are assumed, for convenience, to be s states and no p states are closer than a few hundreths of an electron volt to being in resonance for a one-photon transition driven by either pulse. In particular, $\hbar w_1'$ is most resonant for the transition $|0\rangle$ to $|p,j\rangle$, and $\hbar w_2'$ is most resonant for a transition $|p,j\rangle$ to $|2\rangle$. Generally, only a few intermediate states $|p,j\rangle$ will dominate because, by choice, w_1' and w_2' are

ORNL-DWG 76-15990

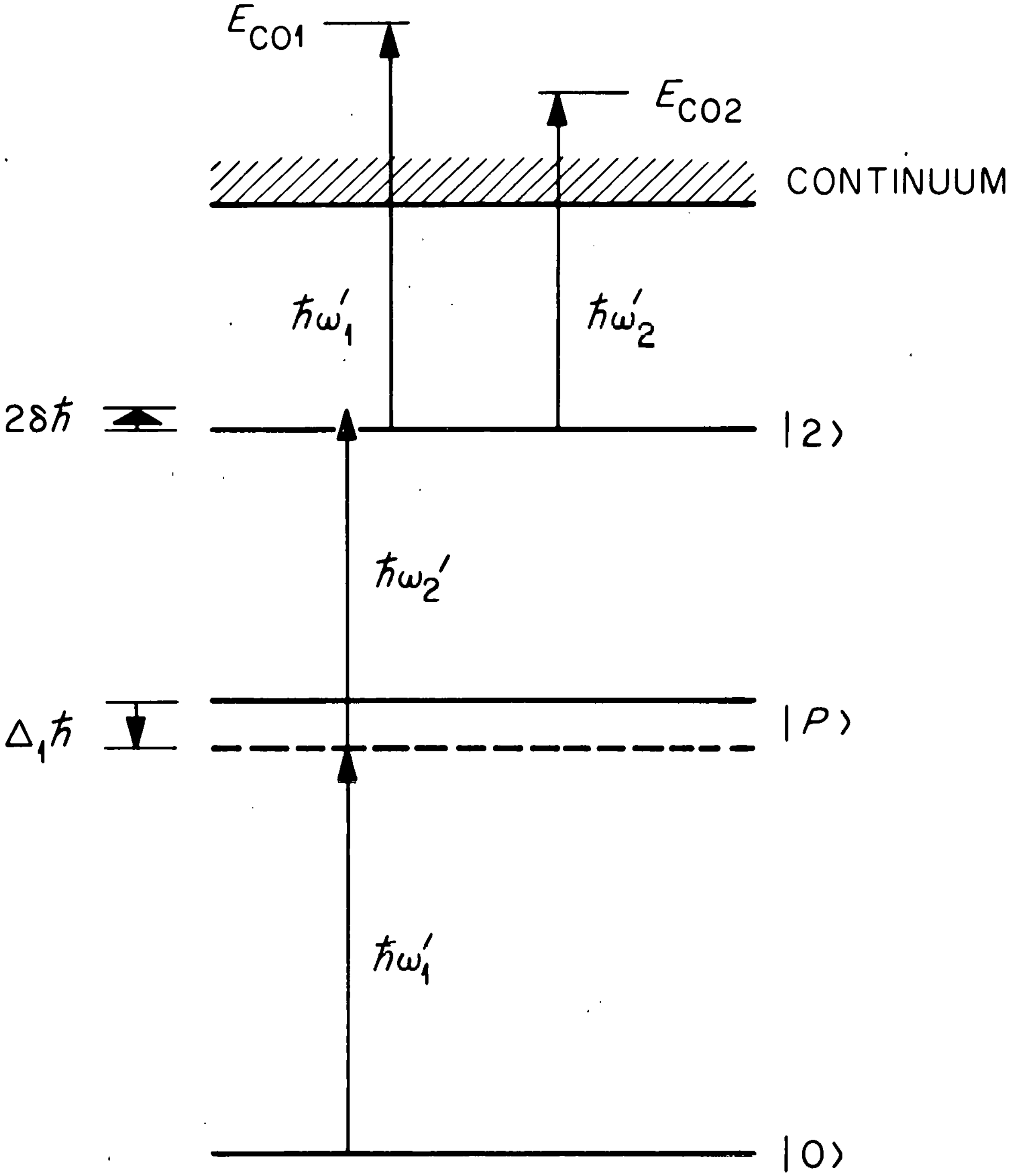

Figure 1. Energy Level Diagram, 3-Photon Transition with Lasers at w_1' and w_2'.

chosen to be rather close to resonance, but far enough away so that line broadening due to the state's lifetime or due to power broadening do not permit real transitions. We write the state vector of the atom as

$$|\psi(t)\rangle = a\ (t)\ e^{-iw_0 t}|0\rangle + a_2(t)\ e^{-iw_2 t}|2\rangle$$

$$+ \sum_j a_{p,j}(t)\ e^{-iw_{pj} t}|p,j\rangle$$

$$+ \sum_k \int dE_c C_{p,k}(E_c,t)\ e^{-iw_c t}|E_c,k\rangle, \qquad (29)$$

where $|0\rangle$, $|2\rangle$, and $|p,j\rangle$ are all orthogonal discrete states and $|E_c,k\rangle$ is a continuum state. The continuum states are normalized so that

$$\langle E_c,k|E_c',k'\rangle = \delta(E_c-E_c')\delta_{k,k'}\ . \qquad (30)$$

Assuming that the wavelength is very large compared with the size of the atom and treating the EM field classically, we have

$$\hat{H} = \hat{H}_0 - \hat{p}_x E(z(t),t) - i\hbar\frac{\gamma_2}{2}|2\rangle\langle 2|. \qquad (31)$$

In Eq. (31), $\hat{H}_0$ is the electronic Hamiltonian of the isolated atom, $\hat{p}_x$ is the x-component of the electric dipole operator for the atom, γ_2 is the spontaneous decay rate of the state $|2\rangle$ (we treat this phenomenologically), $z(t)$ is the z coordinate of the atom evaluated along a straight line classical path, and $|p,j\rangle$ are sufficiently far off resonance so that their spontaneous decay can be neglected (i.e., $|\Delta w_j|$ = amount $|p,j\rangle$ are off resonance satisfies $|\Delta w_j|/\gamma_{pj} >> 1$). The laser fields are assumed to be plane polarized along the x-axis and pulses propagate parallel to the $\pm$ z axis.

Applying the time-dependent Schrödinger equation to Eqs. (29) and (31) and keeping only the least rapidly oscillating coupling terms, we find

$$\frac{da_0}{dt} = \frac{iE_1(t)}{2\hbar} \sum_j \langle 0|p_x|p,j\rangle\, a_{p,j}\, e^{i(w_0-w_{pj}+w_1'-k_1V_z)t}$$

$$\frac{da_{p,j}}{dt} = \frac{iE_1(t)}{2\hbar} \langle p,j|\hat{p}_x|0\rangle\, e^{i(w_{pj}-w_0-w_1'+k_1V_z)t}\, a_0$$

$$+ \frac{iE_2(t)}{2\hbar} \langle p,j|\hat{p}_x|2\rangle\, e^{i\beta}\, e^{i(w_{pj}-w_2+w_2'+k_2V_z)t}\, a_2, \qquad (32)$$

$$\frac{da_2}{dt} = \frac{iE_2(t)}{2\hbar} \sum_j \langle 2|\hat{p}_x|p,j\rangle\, a_{p,j}\, e^{-i\beta}\, e^{-i(w_{pj}-w_2+w_2'+k_2V_z)t} - \frac{\gamma_2}{2} a_2$$

$$+ i \sum_k \int dE_c E_{p,k}(E_c,t) \frac{\langle 2|\hat{p}_x|E_c,k\rangle}{2h} [E_1(t)\, e^{-i(w_c-w_2-w_1'+k_1V_z)t}$$

$$+ E_2(t)\, e^{i\beta}\, e^{-i(w_c-w_2-w_2'-k_2V_z)t}],$$

$$\frac{dC_{p,k}}{dt} = i \frac{\langle E_c,k|\hat{p}_x|2\rangle}{2\hbar} a_2[E_1(t)\, e^{i(w_c-w_2-w_1'+k_1V_z)t}$$

$$+ E_2(t)\, e^{-i\beta}\, e^{i(w_c-w_2-w_2'-k_2V_z)t}].$$

In the above equations it is important to reemphasize that the states $|p,j\rangle$ include only those which are rather close to resonance for allowed dipole transitions driven between $|0\rangle$ and $|p,j\rangle$ by the laser at w_1', and that these states are much farther from resonance for the laser at w_2'. Further, such states must not exist for the transition $|0\rangle$ to $|p,j'\rangle$ for the laser at w_2'. Similar comments apply to the laser at w_2' being much closer to resonance for the transition $|p,j\rangle$ to $|2\rangle$. Without these restrictions, other intermediate states must be included and both $E_1(t)$ and $E_2(t)$ must couple $|0\rangle$ to $|p,j\rangle$ and $|p,j\rangle$ to $|2\rangle$. Further, the rotating wave[29] approximation used above must be refined to generate more accurate a.c. Stark shifts.

We will now show how Eqs. (32) can be simplified. Let $\Delta_{1j} = -w_{pj} + w_0 + w_1' - k_1V_z$, $\Delta_{2j} = -w_{pj} + w_2 - w_2' - k_2V_z$ and integrate the second of these equations

$$a_{p,j} = \frac{i}{2\hbar} \langle p,j|\hat{p}_x|0\rangle \int_{-\infty}^{t} E_1(t')a_0(t')\, e^{-i\Delta_{1j}t'}\, dt'$$

$$+ \frac{i}{2\hbar} \langle p,j|\hat{p}_x|2\rangle\, e^{i\beta} \int_{-\infty}^{t} E_2(t')a_2(t')\, e^{-i\Delta_{2j}t'}\, dt'. \quad (33)$$

If we assume that Δ_m (the smaller of $|\Delta_{1j}|$ and $|\Delta_{2j}|$) is much greater than $E_1(t) \sum_j |\langle 0|\hat{p}_x|p,j\rangle|(2\hbar)^{-1}$, γ_2, τ^{-1}, $E_2(t) \sum_j |\langle 2|\hat{p}_x|p,j\rangle|(2\hbar)^{-1}$, and $\sum_k \int dE_c |\langle 2|\hat{p}_x|E_c,k\rangle|[E_1(t) + E_2(t)](2\hbar)^{-1}$, then we also have $\Delta_m >> |\frac{d}{dt} \ln a_0|$, $|\frac{d}{dt} \ln a_2|$, $|\frac{d}{dt} \ln E_1(t)|$ and $|\frac{d}{dt} \ln E_2(t)|$. Thus, we can do the time integrations by parts and neglect the remaining integrals:

$$a_{p,j}(t) = -\frac{\langle p,j|\hat{p}_x|0\rangle}{2\hbar\Delta_{1j}} E_1(t)\, e^{-i\Delta_{1j}t}\, a_0(t)$$

$$-\frac{\langle p,j|\hat{p}_x|2\rangle}{2\hbar\Delta_{2j}} E_2(t)\, e^{-i(\Delta_{2j}t-\beta)}\, a_2(t). \quad (34)$$

Thus, all of the $a_{pj}(t)$ can be eliminated, leaving a set of equations for a_0, a_2, and C_{pk}. If the continuum matrix elements $\langle E_c,k|\hat{p}_x|2\rangle$ vary so slowly with E_c that they are highly constant over a region of energy $2\Delta E$ such that $\Delta E/\hbar = \Delta_m$, we can eliminate the C_{pk}. We note

$$C_{p,k}(E_c,t) = i\,\frac{\langle E_{c,k}|\hat{p}_x|2\rangle}{2\hbar}\int_{-\infty}^{t} a_2(t')[E_1(t')\, e^{i\Delta_{c1}t'}$$

$$+ E_2(t')\, e^{i(\Delta_{c2}t'-\beta)}]dt', \quad (35)$$

where $\Delta_{c1} = w_c - w_2 - w_1' + k_1V_z$, $\Delta_{c2} = w_c - w_2 - w_2' - k_2V_z$. If $E_c = \hbar w_c$

is such that both $|\Delta_{c1}| > \Delta E/\hbar \equiv \Delta$ and $|\Delta_{c2}| > \Delta$, we can integrate by part and find

$$C_{p,k}(E_c,t) = \frac{\langle E_c,k|\hat{p}_x|2\rangle}{2\hbar} a_2(t) \left[\frac{E_1(t)\, e^{i\Delta_{c1}t}}{\Delta_{c1}} + \frac{E_2(t)\, e^{i(\Delta_{c2}t-\beta)}}{\Delta_{c2}}\right]. \qquad (36)$$

In either the interval $|\Delta_{c1}| \leq \Delta$ or $|\Delta_{c2}| \leq \Delta$, we can replace $\langle E_c,k|\hat{p}_x|2\rangle$ by its value at the center of the interval and otherwise keep the integral form of Eq. (35). If we use the above simplifications in the third of Eqs. (32) and further assume $|w_1'-w_2'| >> \Delta$, we can show that constancy of $\langle E_c,k|\hat{p}_x|2\rangle$ over $|\Delta_{c1}| \leq \Delta$ and $|\Delta_{c2}| \leq \Delta$ leads to a Dirac delta function contribution in the integration over these intervals, while the integration over dE_c outside these intervals leads to a principle value integration. After some algebra, we obtain (A_0 differs only by a phase factor from a_0 and A_2 differs only by a phase factor from a_2)

$$\frac{dA_0}{dt} = i\Omega_4(t)A_2(t) \exp[i \int_{-\infty}^{t} (\Omega_2(t')-\Omega_3(t')+\Delta w(t')+2\delta)dt'],$$

$$\frac{dA_2}{dt} = - (\frac{\gamma_2}{2} + p(t)) A_2 + i\Omega_4^*(t)A_0(t) \times \exp[-i \int_{-\infty}^{t} (\Omega_2(t')-\Omega_3(t') + \Delta w(t') + 2\delta)dt'], \qquad (37)$$

where

$$p(t) = \sum_{\ell=1}^{2} \frac{\pi E_\ell^2(t)}{4\hbar} \sum_k |\langle 2|\hat{p}_x|E_{c0,\ell},k\rangle|^2,$$

$$\Delta w(t) \equiv \sum_{\ell=1}^{2} \frac{E_\ell^2(t)}{4\hbar^2} P \int \frac{\sum_k |\langle E_c,k|\hat{p}_x|2\rangle|^2 dE_c}{\Delta_{c\ell}},$$

$$\Omega_2(t) = -\sum_j \frac{|\langle p,j|\hat{p}_x|2\rangle|^2 E_2^2(t)}{4\hbar^2\Delta_{2j}}, \tag{38}$$

$$\Omega_3(t) = -\sum_j \frac{|\langle 0|\hat{p}_x|p,j\rangle|^2 E_1^2(t)}{4\hbar^2\Delta_{1j}}$$

$$\Omega_4(t) = -\sum_j \frac{\langle 0|\hat{p}_x|p,j\rangle \ \langle p,j|\hat{p}_x|2\rangle}{4\hbar^2\Delta_{2j}} E_1(t)E_2(t)e^{i\beta}$$

and

$$2\delta = w_1'+w_2' - w_2+w_0 - (k_1-k_2)V_g.$$

The quantity $2\hbar\delta$ is a measure of the excess energy available in the two-photon process. Of course, δ must be restricted so that the states $|p,j\rangle$ can be eliminated as described earlier. The quantity $p(t)$ represents a damping of the populations of $|0\rangle$ and $|2\rangle$ due to photoionization, $\Delta w(t)$ is an a.c. Stark shift due to coupling with continuum states, $\Omega_2(t)$ is the a.c. Stark shift of the state $|2\rangle$ due to the states $|p,j\rangle$, $\Omega_3(t)$ is the a.c. Stark shift of state $|0\rangle$ due to states $|p,j\rangle$ and $\Omega_4(t)$ is the two-photon coupling coefficient.

Except for the inclusion of photoionization by a pulsed laser the 3-level atom problem discussed above has been formulated and discussed by several authors.[30-37] In particular if γ_2 and $p(t)$ are neglected even the pulsed nature of the field can be dealt with by using conventional adiabatic perturbation theory of the type used in atom-atom collisions. Much of the above work has been formulated in terms of the optical Bloch equations; which is equivalent to the Schrödinger equation approach except for the possibility of including incoherent sources for the population, as well as collisional relaxation, by phenomenological parameters.

C. APPLICATION OF THE FACTORIZATION METHOD

In this section we will apply the factorization method described in Chapter II to three-photon ionization as described by Eqs. (37).

We take

$$E_1^2(t) = E_{10}^2 g(t),$$

$$E_2^2(t) = E_{20}^2 g(t),$$

where g(t) is a smooth, slow varying function as described in Eq.(2). We then get

$$\frac{dA_0}{dt} = i\alpha g(t) A_2 \exp\left[i\left(2\delta t - \mu\int_{-\infty}^{t} g(t')dt'\right)\right],$$

(39)

$$\frac{dA_2}{dt} = -\left(\frac{\gamma_2}{2} + P_o g(t)\right)A_2 + i\alpha^* g(t) A_0 \exp\left[-i\left(2\delta t - \mu\int_{-\infty}^{t} g(t')dt'\right)\right],$$

where δ and γ_2 are as defined earlier and

$$\mu = -\sum_j \frac{|\langle 0|\hat{p}_x|p,j\rangle|^2 E_{10}^2}{4\hbar^2 \Delta_{ij}} + \sum_j \frac{|\langle p,j|p_x|2\rangle|^2 E_{20}^2}{4\hbar^2 \Delta_{2j}}$$

$$- \sum_{\ell=1}^{2} \frac{E_{\ell 0}^2}{4\hbar^2} P\int \frac{\sum_k |\langle E_c,k|\hat{p}_x|2\rangle|^2 dE_c}{\Delta_{c\ell}}, \qquad (40a)$$

$$\alpha = -\sum_j \frac{\langle 0|\hat{p}_x|p,j\rangle\langle p,j|\hat{p}_x|2\rangle}{4\hbar^2 \Delta_{2j}} E_{10}E_{20}e^{i\beta}, \qquad (40b)$$

$$P_o = \sum_{\ell=1}^{2} \frac{\pi E_{\ell 0}^2}{4\hbar} \sum_k |\langle 2|\hat{p}_x|E_{c0,\ell},k\rangle|^2 {}^{*}. \qquad (40c)$$

---

* P_o is proportional to the photon flux of the laser field times photoionization cross section of the state |2>. The cross section is, in general, order of 10^{-17} cm^2 to 10^{-20} cm^2.

If we let

$$\frac{dq}{dt} = 2\delta - \mu g(t),$$

$$u(t) = \alpha g(t),$$

$$\gamma(t) = \frac{\gamma_2}{2} + p_o g(t),$$

the factorization method is immediately applicable. We get

$$\left(\frac{d}{dt} + g_1(t)\right)\left(\frac{d}{dt} + g_2(t)\right)A_2 = 0, \qquad (41)$$

where

$$g_1+g_2 = i(2\delta-\mu g) - \frac{d}{dt}\,\ell ng + p_o g + \frac{\gamma_2}{2}, \qquad (42)$$

$$g_1 g_2 + \frac{dq_2}{dt} = |\alpha|^2 g^2 + i(2\delta-\mu g)\left(p_o g + \frac{\gamma_2}{2}\right) - \frac{\gamma_2}{2}\frac{d}{dt}\ell ng.$$

We choose: $g_1 = g_{10} + \varepsilon_1$, $g_2 = g_{20} + \varepsilon_2$, where we have defined

$$g_{10} + g_{20} = i(2\delta-\mu g),$$

$$g_{10} g_{20} = |\alpha|^2 g^2.$$

We obtain

$$g_{10} = \frac{i(2\delta-\mu g)+i\varepsilon J}{2} , \tag{43a}$$

$$g_{20} = \frac{i(2\delta-\mu g)-i\varepsilon J}{2} , \tag{43b}$$

where

$$J = \sqrt{(2\delta-\mu g)^2 + 4|\alpha|^2 g^2} , \tag{43c}$$

and

$$\varepsilon = \delta/|\delta| .$$

Intuitively, we expect that if g(t) is a very slowly varying function of time and if $|\alpha|$ is large then ε_1 and ε_2 can be treated as small slowly varying function of time. Thus, neglecting $\varepsilon_1\varepsilon_2$ and $d\varepsilon_2/dt$ compared with $g_{10}\varepsilon_2, g_{20}\varepsilon_1$ and dg_{20}/dt we find

$$\varepsilon_2 = (\frac{\gamma_2}{2} + p_o g)[\frac{1}{2} + \frac{\varepsilon(2\delta-\mu g)}{2J}] + \frac{1}{2}\frac{d}{dt}\ell nJ$$

$$+ [\frac{|\delta|}{J} - \frac{1}{2} + i\frac{\varepsilon\gamma_2}{2J}] \frac{d}{dt}\ell ng, \tag{44a}$$

$$\varepsilon_1 = (\frac{\gamma_2}{2} + p_o g)[\frac{1}{2} - \frac{(2\delta - g)}{2J}] - \frac{1}{2}\frac{d}{dt}\ln J - [\frac{|\delta|}{J} + \frac{1}{2} + \frac{i\varepsilon\gamma_2}{2J}]\frac{d}{dt}\ln g. \quad (44b)$$

The above method is not exactly equivalent to the iterative scheme described in Chapter II, but it is more convenient for analytical work.

The expressions obtained for g_1 and g_2 by using g_{10}, g_{20} and our approximate values of ε_1 and ε_2 are not always accurate. The most severe restrictions are related to obtaining accurate g_1 and g_2 functions when $g(t)$ is small or when t is near a value at which $2\delta=\mu g$. In order to obtain accuracy when $g(t)$ is small, we must restrict δ such that

$$|\delta|\tau > 10, \quad (45)$$

where τ is the full width at half maximum of $g(t)$. The condition $|\delta|\tau > 10$ is not obvious, but its enforcement yields excellent accuracy in a large variety of numerical examples. The approximate g_1 and g_2 functions are accurate near times such that $2\delta=\mu g$, providing that at these times

$$\frac{|\mu|}{2|\alpha|^2} g^{-2}(t)\frac{dg}{dt} << 1$$

$$|P_o/\alpha| << 1, \quad (46)$$

and

$$\frac{\gamma_2}{|\alpha| g(t)} << 1.$$

The above conditions arise because it is obvious that when $2\delta=\mu g$, unless $|\alpha|g$ is large, the values of g_{10} and g_{20} will become small, while ε_1 and ε_2 take on their largest values. Thus, the expressions are obtained by requiring that the neglected terms $\varepsilon_1\varepsilon_2$ and $d\varepsilon_2/dt$ be small compared with $g_{10}\varepsilon_2$, $g_{20}\varepsilon_1$ and dg_{20}/dt which were retained. Thus, if $|\mu|\tau >> 1$, $|\alpha|\tau >> 1$, $|\alpha|/\gamma_2 >> 1$, $|\alpha/P_o| >> 1$ and $|\delta|\tau > 10$, we will obtain accurate solutions as long as $|\delta|$ does not become so small that the validity conditions fail or unless $|\alpha/\mu|$ is very small. Very crudely, we can rewrite conditions (46) as

$$2|\alpha/\mu|^2|\mu_1\tau| = 4|\frac{\alpha}{\mu}|^2|\delta\tau| >> 1,$$

$$|P_o/\alpha| << 1, \qquad (47)$$

and

$$\gamma_2/|\alpha_1| << 1,$$

where t_0 is a time such that $2\delta = \mu g(t_0)$, $|\alpha_1| = |\alpha|g(t_0)$, and $|\mu_1| = |\mu|g(t_0)$. Conditions (45) and (47) are sufficient for all times as long as (47) is satisfied at all "crossings." The validity equations all indicate that the results apply to long pulses and very high power levels. However, we shall see later that the power levels and pulse lengths are easily available with present nitrogen laser pumped dye lasers. Further, the factorization method enables one to calculate accurately almost all of the lineshape for ionization.

After applying Eqs. (15) of Chapter II we find

$$|A_2(t)|^2 = [1/2 - \frac{\varepsilon(2\delta-\mu g)}{2J}]$$

$$\times \exp[-2\int_{-\infty}^{t} (\frac{\gamma_2}{2} + p_o g(t'))(\frac{1}{2} - \frac{\varepsilon(2\delta-\mu g(t'))}{2J(t')})\, dt'],$$

$$= |A_{2o}(t)|^2 \exp[-2\int_{-\infty}^{t} (\frac{\gamma_2}{2} + p_o g(t'))|A_{20}(t')|^2 dt'], \quad (48)$$

where

$$|A_{20}(t)|^2 = \frac{1}{2} - \varepsilon(2\delta-\mu g)(2J)^{-1}. \quad (49)$$

$|A_{20}(t)|^2$ is the probability of the upper resonance state being populated at time t in the absence of spontaneous decay or photoionization. $|A_{20}(t)|^2$ has some very interesting properties that are worth pointing out. We write Eq. (49) as

$$|A_{20}(t)|^2 = \frac{1}{2} - \frac{\varepsilon'(\frac{2\delta}{\mu} - g)}{2\sqrt{(\frac{2\delta}{\mu} - g)^2 + 4|\frac{\alpha}{\mu}|^2 g^2}}, \quad (50)$$

where $\varepsilon' = \delta\mu/|\delta\mu|$. From the last equation we see that if $|\frac{\alpha}{\mu}|^2 \leq 0.1$ then $|A_{20}(t)|^2$ remains small at all times if $\delta/\mu < 0$; but that when $\delta/\mu > 0$ it remains zero until the first "crossing" (i.e., $2\delta = \mu g$), then rapidly rises to a value near unity until the second crossing where it falls again to a value near zero. Thus, for reasonably small values of δ a large population inversion is induced and it persists for a large portion of the pulse. When $|\alpha/\mu|^2$ is near unity a population inversion still exists for $\delta/\mu > 0$ but it is smaller.

In order to calculate R, the probability of an atom being ionized, we use

$$R = \int \sum_k | C_{p,k}(E_c,\infty)|^2 dE_c. \tag{51}$$

$C_{p,k}(E_c,t)$ was eliminated, but we can return to the initial coupled equations and with the same requirements for elimination of $C_{p,k}(E_c,t)$ show that

$$R = 2p_o \int_{-\infty}^{\infty} g(t)\,|A_2(t)|^2 dt,$$

$$= 2p_o \int_{-\infty}^{\infty} g(t)\,|A_{20}(t)|^2$$

$$\times \exp[-2\int_{-\infty}^{t} (\gamma_2/2+p_o g(t'))\,|A_{20}(t')|^2\, dt']dt. \tag{52}$$

We choose $g(t) = [1+(t/\tau)^2]^{-3}$ and $V = t/\tau$. Then, noting that $|A_{20}(t)|^2 \equiv F(V)$ we find

$$R = 2p_o\tau \int_{-\infty}^{\infty} dVg(V)F(V)$$

$$\times \exp\left[-2\int_{-\infty}^{V}\left(\frac{\gamma_2\tau}{2} + p_o\tau g(V')\right)F(V')dV'\right],$$

$$= R(p_o\tau, \gamma_2\tau, \frac{\delta}{\mu}, |\frac{\alpha}{\mu}|). \qquad (53)$$

In many cases $\gamma_2\tau \ll 1$ and we find

$$R = 1-\exp[-p_o\tau H(\delta/\mu, |\frac{\alpha}{\mu}|)], \qquad (54)$$

where

$$H(\delta/\mu, |\alpha/\mu|) = 2\int_{-\infty}^{\infty} g(V')F(V')dV', \qquad (55)$$

with

$$F(V) = \frac{1}{2} - \frac{\varepsilon'(\frac{2\delta}{\mu} - g(V))}{2\sqrt{(2\delta/\mu - g(V))^2 + 4|\alpha/\mu|^2 g^2(V)}}. \qquad (56)$$

In the special case $E_{10}=E_{20}$ the ratio $|\alpha/\mu|$ is independent of power and we see that a graph of $-(p_o\tau)^{-1}\ell n(1 - R)$ versus δ/μ should yield the same curve for all peak power and pulse length such that $\gamma_2\tau \ll 1$ and such that the other validity conditions are met. (See Appendix A for tabulation of the H function.)

To illustrate the validity of the above results, we have chosen $\tau = 10^{-8}$ sec, $\mu = 1.38 \times 10^3\ I_o$, and $\alpha = 3.7 \times 10^2\ I_o$. Figure 2 shows a graph of a numerically calculated plot of $-(p_o\tau)^{-1}\ell n(1 - R)$ versus δ/μ for $I_o = 10^7$ w/cm^2 and 10^8 w/cm^2, where I_o is the peak power density of the laser pulse. On the same graph we have shown the result calculated from Eq. (54). Note that the only discrepancy occurs when $|\delta\tau| < 10$. When $\delta = 0$, Eqs. (39) can be solved by letting $u = \int_{-\infty}^{t} g(t')dt'$ (see Appendix B). Using $g(V) = (1 + V^2)^{-3}$, we find

$$R = 1 - \exp\left[-\frac{3\pi}{2} p_o\tau|\alpha/\mu|^2(1 + 4|\alpha/\mu|^2)^{-1}\right] . \tag{57}$$

This simple expression was used to check the numerical calculations at $\delta = 0$, and agreement was excellent.

Eqs. (39) can also be solved exactly if the two lasers give identical square pulses (see Appendix B). Let

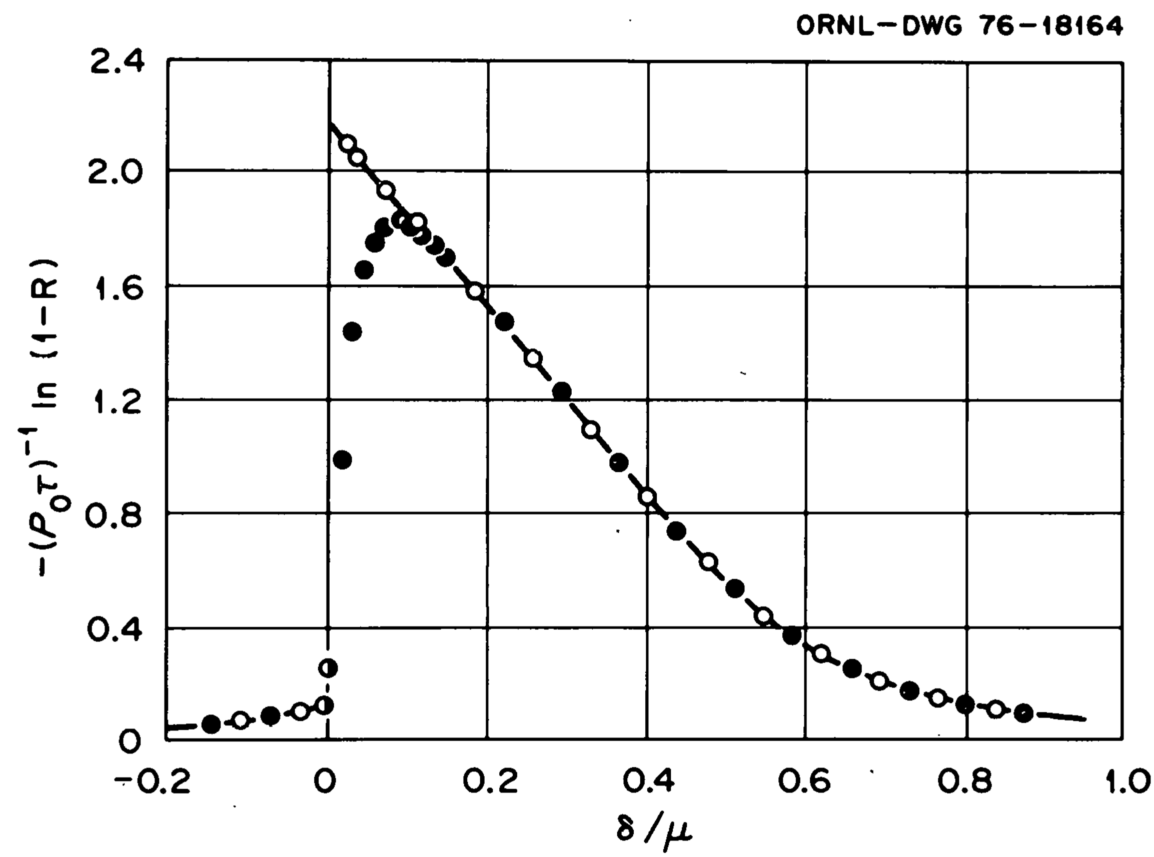

Figure 2. Photoionization Probability Comparison, — for the analytical solution, o for the numerical solution with $I_o = 10^8$ w/cm^2 and ● for the numerical solution with $I_o = 10^7$ w/cm^2.

$$g(t) = 0,\ t < 0;$$

$$= \frac{3\pi}{8},\ 0 \leq t \leq \tau;$$

$$= 0,\ t > \tau.$$

We have solved analytically for R in cases where $\alpha\tau >> 1$. In Figs. 3-8 we have graphed R versus δ for various I_o and $\tau = 10^{-8}$ sec, $\mu = 1.38 \times 10^3\ I_o$, $\alpha = 3.7 \times 10^2\ I_o$ and $p_o = 0.08\ I_o$. Both the square pulse and $g = [1 + V^2]^{-3}$ cases are shown in Figs. 3-5. The reader should note that the square pulse result peaks at $2\delta = \mu g$ and the only width is due to power broadening which is approximately $2|\alpha|$. The line shape for $g(V) = (1+V^2)^{-3}$ is extremely different due to the curve crossings (i.e., $2\delta = \mu g(V)$) which occur for $\delta/\mu > 0$ and for $|\delta| < |\mu/2|$. When $|\mu| >> |\alpha|$ the width is almost entirely determined by $|\mu|$ and is many times wider than the width for the square pulse case. With smooth pulses the values of R are larger due to the fact that inverted populations exist in this case. For square pulses and $|\alpha\tau| >> 1$ Rabi flopping[29] occurs and when $|A_2|^2$ is averaged over several cycles it is always $\leq \frac{1}{2}$; thereby, leading to smaller R.

The analytical expression for $|A_2|^2$ is discontinuous at $\delta = 0$; being small on the $\delta/\mu < 0$ side and larger than 1/2 on the $\delta/\mu > 0$ side. The discontinuity is due to deviations

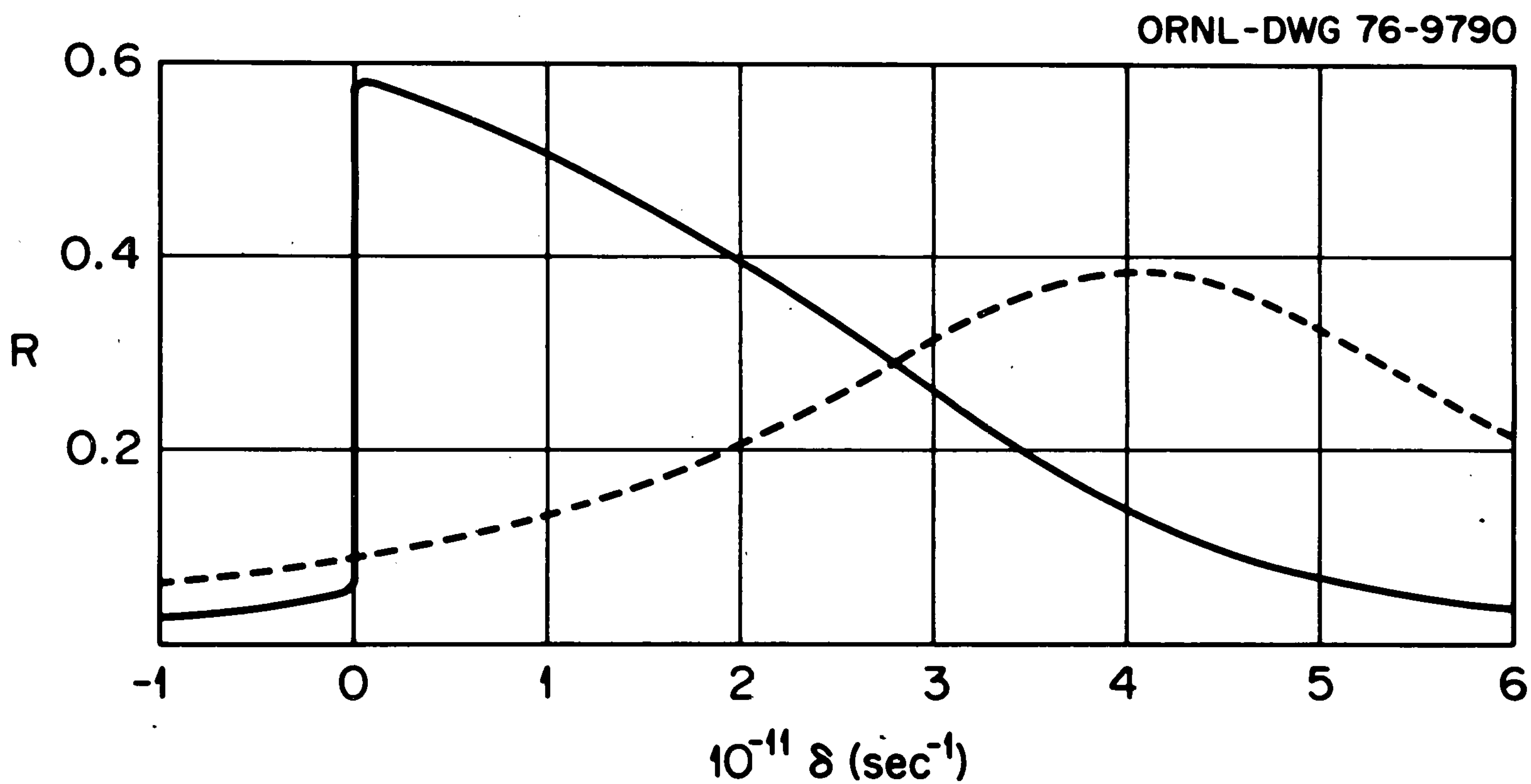

Figure 3. Photoionization Probability ($I_o = 5 \times 10^8$ w/cm^2, $\tau = 10^{-8}$ sec), —— for $g(t) = [1 + (t/\tau)^2]^{-3}$ and --- for $g(t) = 3\pi/8$.

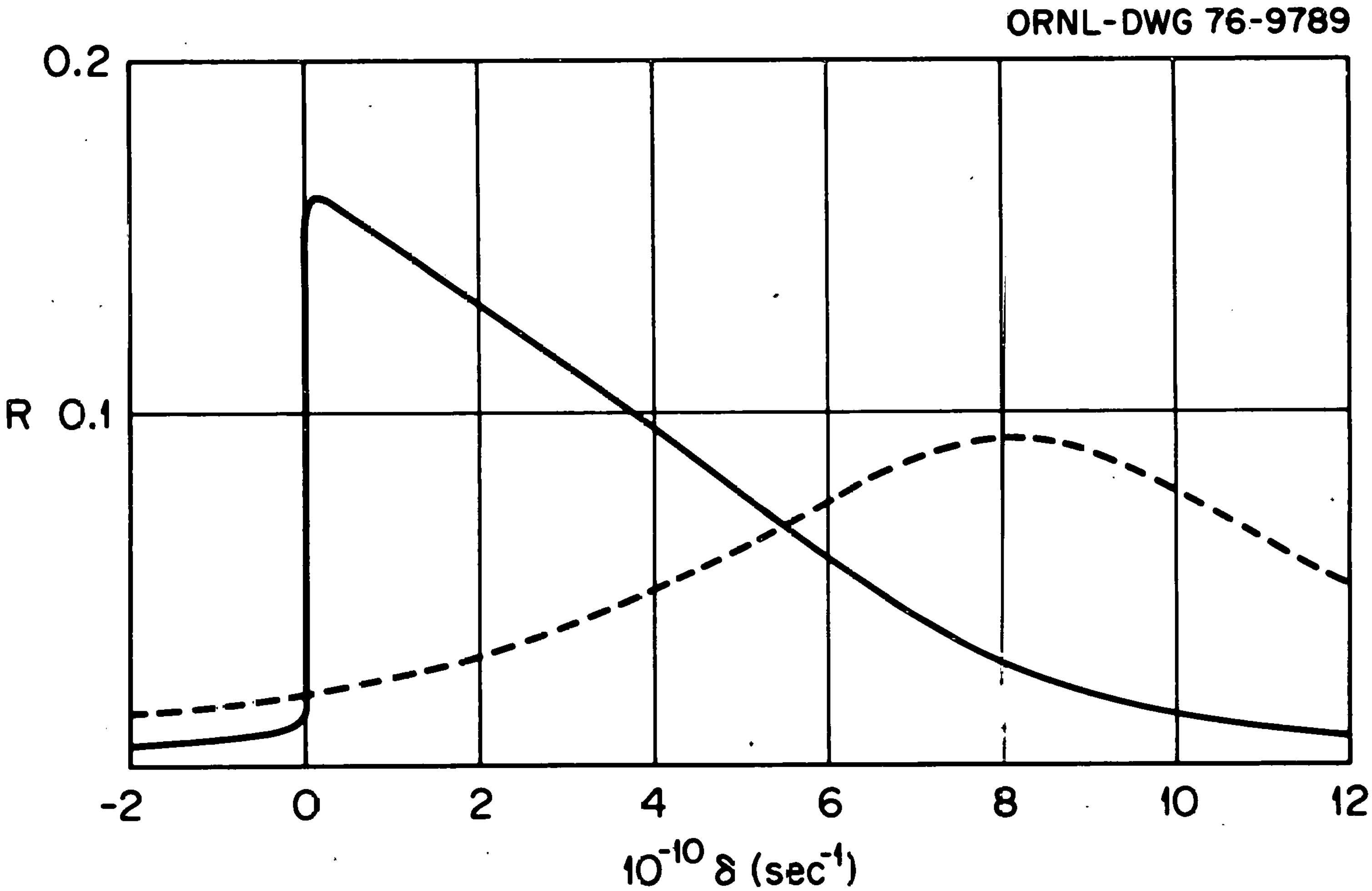

Figure 4. Photoionization Probability ($I_o = 10^8$ w/cm^2, $\tau = 10^{-8}$ sec), —— for $g(t) = [1 + (t/\tau)^2]^{-3}$ and --- for $g(t) = 3\pi/8$.

ORNL-DWG 76-9788

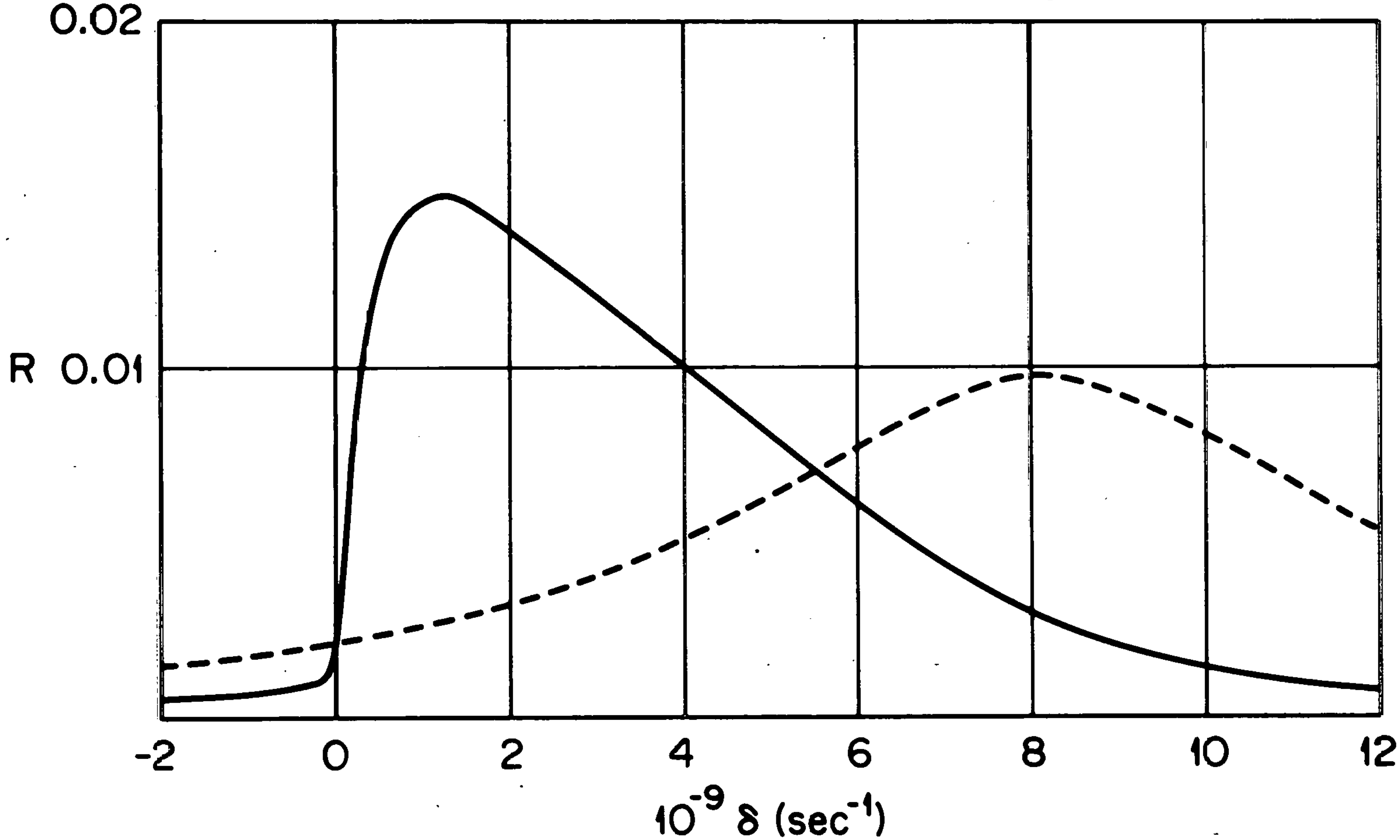

Figure 5. Photoionization Probability ($I_o = 10^7$ w/cm^2, $\tau = 10^{-8}$ sec), —— for $g(t) = [1 + (t/\tau)^2]^{-3}$ and --- for $g(t) = 3\pi/8$.

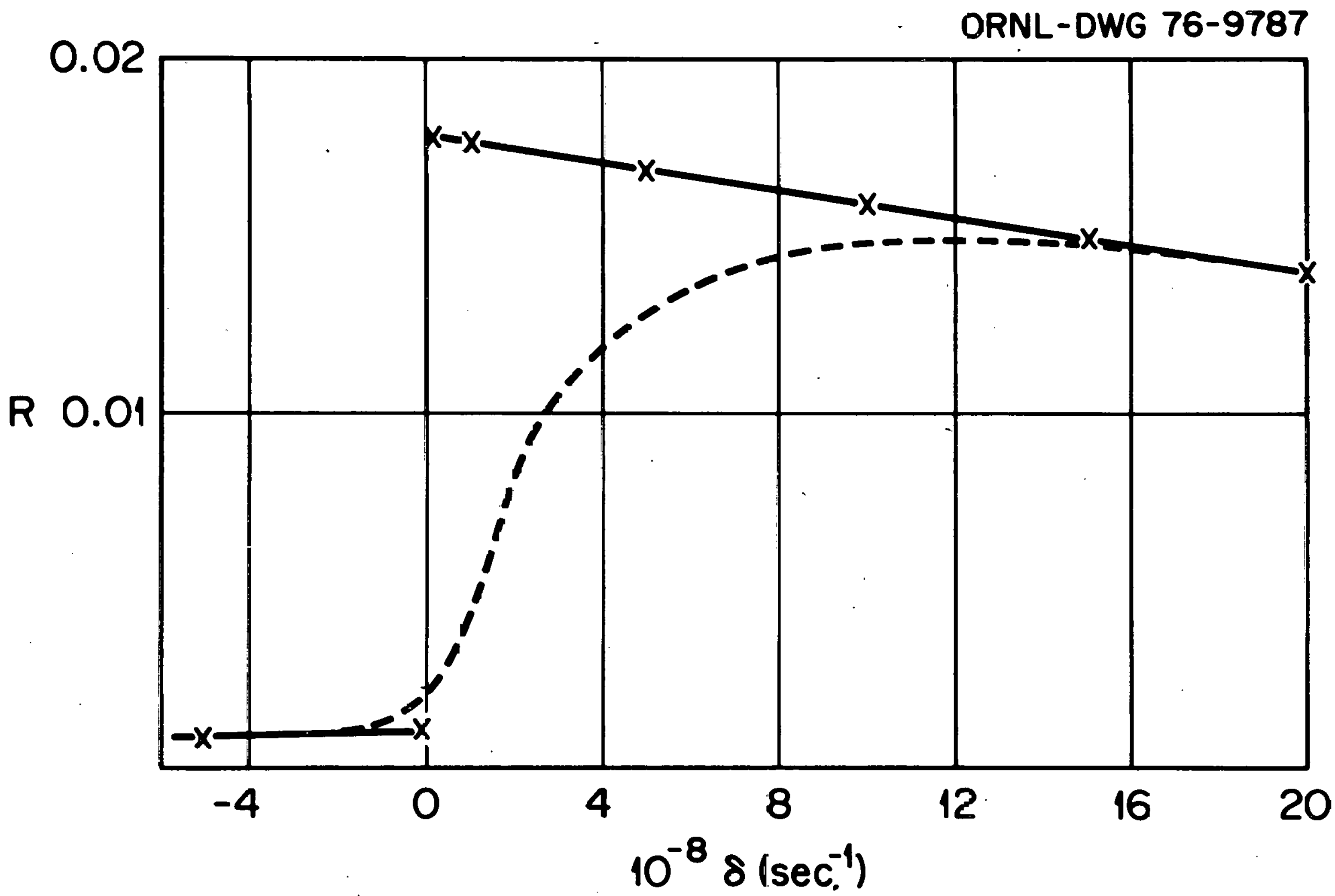

Figure 6. Blow-up Graph Near $\delta = 0$ ($I_o = 10^7$ w/cm^2, $\tau = 10^{-8}$ sec), --- for the numerical solution and x for the analytical solution.

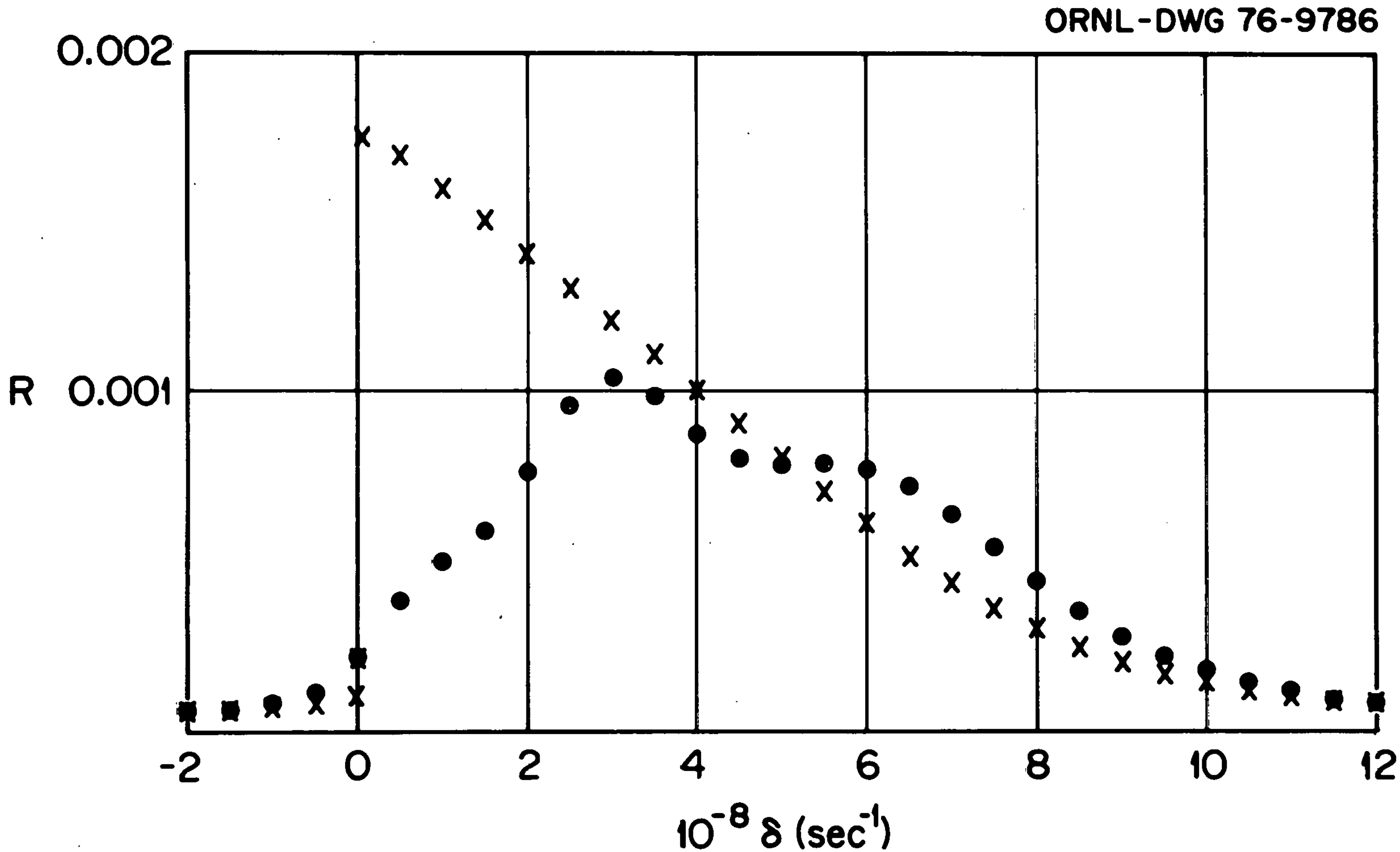

Figure 7. Photoionization Probability ($I_o = 10^6$ w/cm^2, $\tau = 10^{-8}$ sec), x for the analytical solution and ● for the numerical solution with $g(t) = [1 + (t/\tau)^2]^{-3}$.

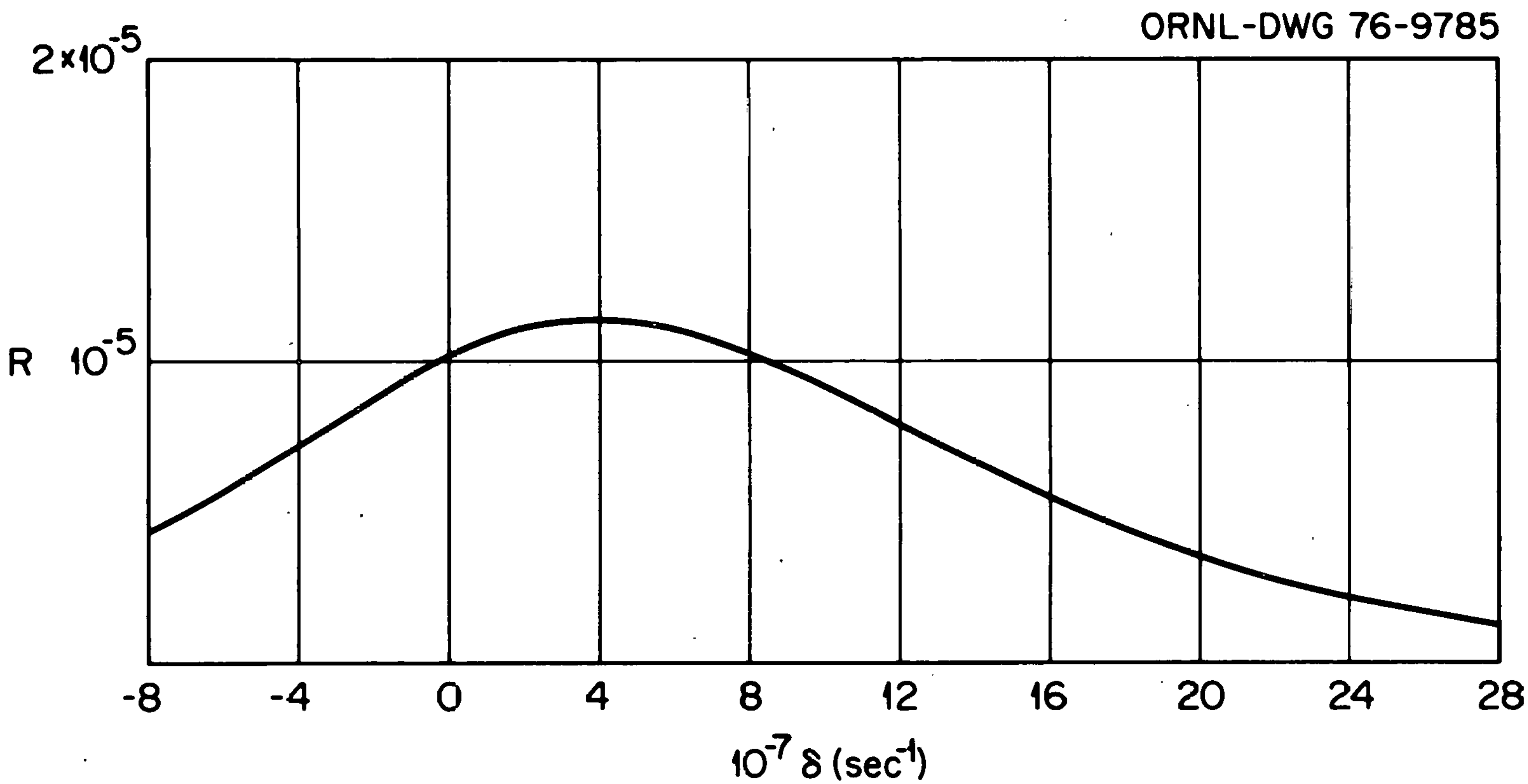

Figure 8. Photoionization Probability ($I_o = 10^5$ w/cm^2, $\tau = 10^{-8}$ sec), numerical solution for $g(t) = [1 + (t/\tau)^2]^{-3}$.

from adiabatic behavior when $|\delta|\tau < 10$. When non-adiabatic effects are taken into account R rises sharply, but continuously in a region $\Delta\delta$ given by $|\Delta\delta| \simeq 10/\tau$. Thus, for long pulses the rise can be very sharp compared with the full Doppler width. In this sense a Doppler-free aspect of the line shape survives even at very large power levels. Making use of this sharp rise, one could selectively ionize one atom in the presence of another with a very nearby two-photon resonance. By most methods high R is impossible without a correspondingly high value for the nearby level due to power broadening effects.

We have noted in condition (47) that the case of small $|\alpha/\mu|$ requires extremely large values of $|\alpha|\tau$ (the present method requires fairly large values of $|\alpha|\tau$ even when $|\alpha/\mu| \simeq 1$). In the next section we require $|\alpha/\mu| << 1$ but we will not require $|\alpha|\tau >> 1$. The latter situation is very interesting if one of the laser pulses is much more powerful than the other.

D. APPLICATION OF THE METHOD OF ISOLATED CURVE CROSSINGS TO THREE-PHOTON IONIZATION

We consider the same situation as in Section C except now $|\alpha/\mu| << 1$ and $|\mu\tau| >> 1$. In comparing with Chapter II, Section B we note that $h(t)=F(t)=[1+(t/\tau)^2]^{-3}=g(t)$. We take $\gamma_2=0$ so that $\gamma(t) = p_o g(t)$. We have for all t

$$|A_2(t)|^2 \simeq 0, \text{ if } \delta/\mu < 0 \text{ or } 2\delta/\mu > 1. \tag{58}$$

When $\delta/\mu > 0$ and $2\delta/\mu < 1$ we obtain

$$|A_2(t)|^2 = 0, \; t < t_{01};$$

$$= T \exp[-2p_o \int_{-t_{02}}^{t} g(t')dt'], \; t_{01} \leq t \leq t_{02}, \tag{59}$$

$$= T(1-T)[1+\exp(-2p_o \int_{-t_{02}}^{t_{02}} g(t')dt')]$$

$$\times \exp[-2p_o \int_{t_{02}}^{t} g(t')dt'], \; t > t_{02};$$

where

$$t_{02} = \tau [(\tfrac{\mu}{2\delta})^{1/3} - 1]^{1/2} = -t_{01},$$

$$|z_1|^2 = \frac{|\mu\tau|}{6} |\tfrac{\alpha}{\mu}|^2 (\tfrac{2\delta}{\mu})^{1/2}[1-(\tfrac{2\delta}{\mu})^{1/3}]^{-1/2}, \tag{60}$$

$$T = 1-\exp(-2\pi |z_1|^2).$$

As explained in the mathematical discussion of Chapter II, the only appreciable changes in $A_2(t)$ occur in a narrow time interval about times where $2\delta = \mu g$. Except in these time intervals the lasers are far out of resonance and only photoionization and spontaneous decay are effective in changing A_2. In our case we assume $\gamma_2\tau << 1$ and the only effect in changing the population of either $|0\rangle$ or $|2\rangle$ is photoionization.

Using Eqs. (58) and (59)-(60) we find

$$R = 0 \text{ if } \delta/\mu < 0 \text{ or } 2\delta/\mu > 1; \tag{61}$$

and for $0 \leq 2\delta/\mu \leq 1$ we have

$$R = T[1-Q^2(V_{02})]+T(1-T)[1+Q^2(V_{02})][1 - \frac{Q(\infty)}{Q(V_{02})}], \tag{62}$$

$$= R(|\frac{\alpha}{\mu}|^2|\mu\tau| \;,\; p_o\tau, \frac{\delta}{\mu})$$

where

$$V = t/\tau$$

$$Q(V) = \exp(-2p_o\tau\, m(V)) \tag{63}$$

and

$$m(V) = \int_0^V \frac{dV'}{(1+V'^2)^3}$$

$$= \frac{1}{8}\left[\frac{3V}{1+V^2} + \frac{2V}{(1+V^2)^2} + 3\tan^{-1} V\right] \qquad (64)$$

Thus, if one fixes $|\alpha|^2\tau/|\mu|$ and $p_o\tau$, plots of R versus $2\delta/\mu$ are the same curve for all μ. (The term $p_o\tau$ is fixed by producing most of the ionization by a third laser pulse $E_3(t)\cos(w_3't-k_3V_zt+\beta_3) = E_{30}g^{1/2}(t)\cos(w_3't-k_3V_zt+\beta_3)$; with w_3' being far from resonance as far as participation in the two-photon resonance is concerned.) This is illustrated in Figs. 9-12.

A second interesting application of the present mathematical problem is to consider a situation identical to that described above except that we now assume $\gamma_2 \neq 0$. Thus for all t:

$$|A_2(t)|^2 = 0, \text{ if } \delta/\mu < 0 \text{ or } 2\delta/\mu > 1 \qquad (65)$$

when $0 \leq 2\delta/\mu \leq 1$ we obtain

$$|A_2(t)|^2 = 0,\ t < t_{01};$$

$$= T\exp[-\gamma_2(t+t_{02}) - 2p_o\int_{-t_{02}}^{t} g(t')dt'],$$

ORNL-DWG 76-18166

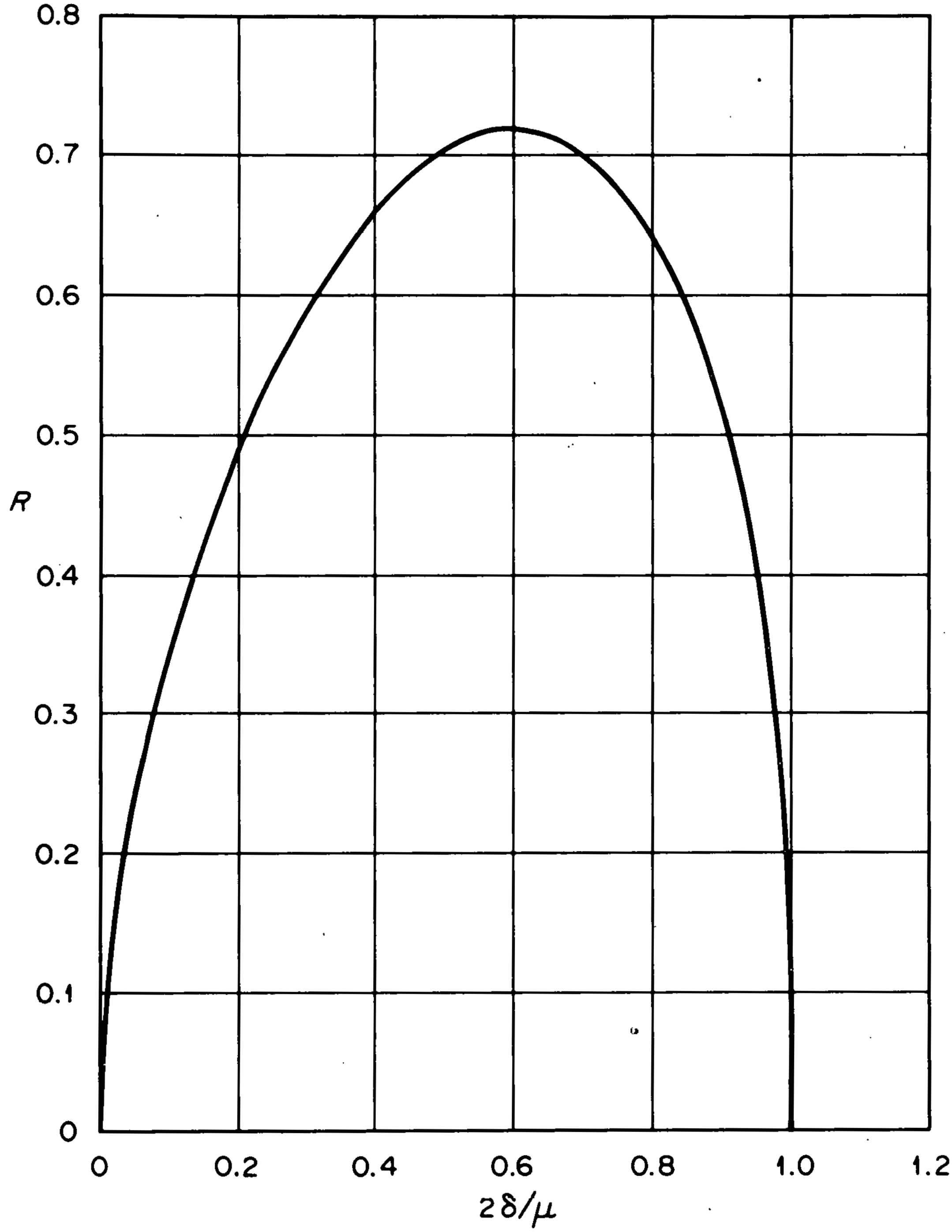

Figure 9. Photoionization Probability (isolated crossings), $|\alpha/\mu|^2|\mu\tau| = 1$, $P_o\tau = 1$ and $g(t)=[1+(t/\tau)^2]^{-3}$.

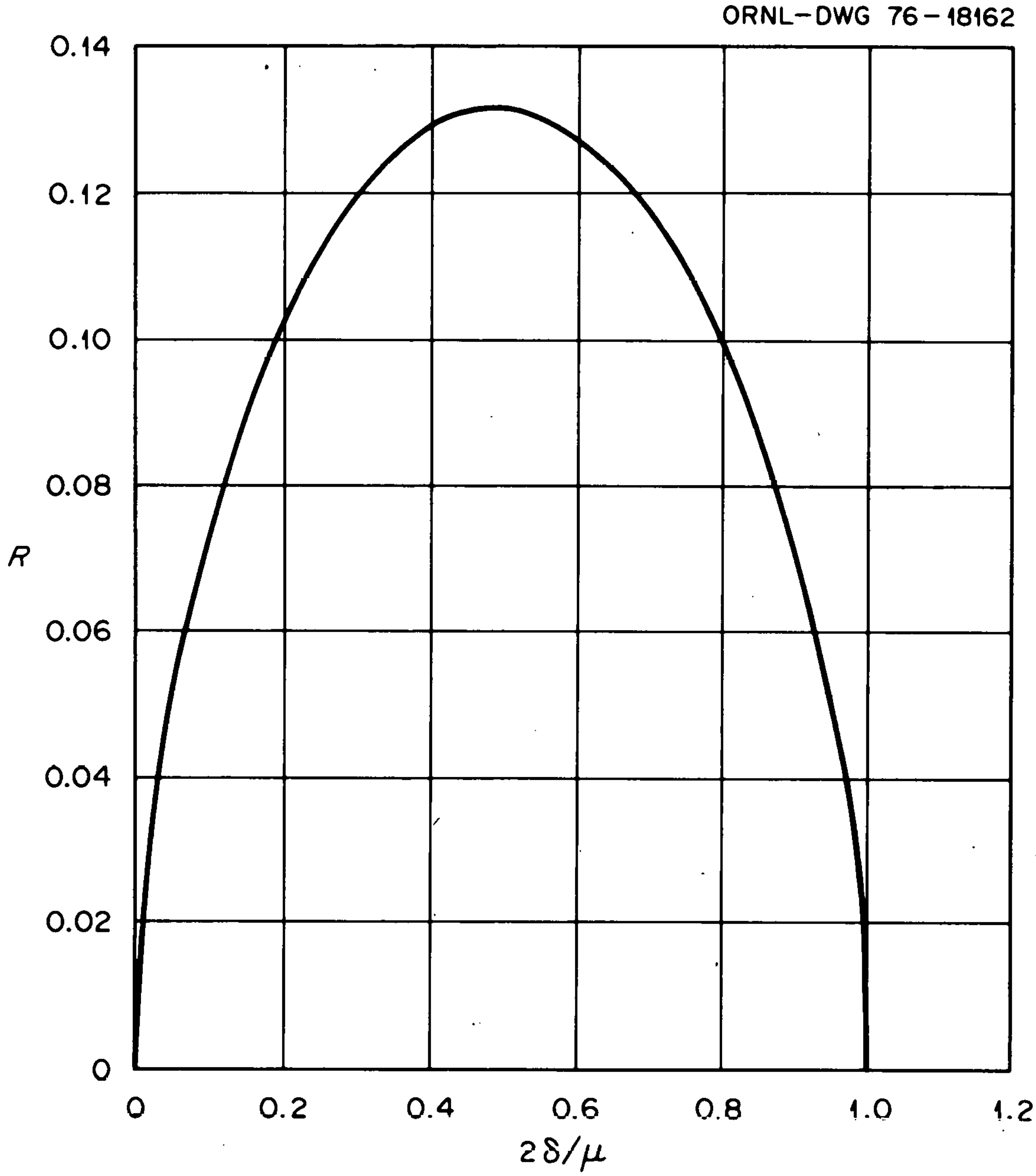

Figure 10. Photoionization Probability (isolated crossings), $|\alpha/\mu|^2|\mu\tau| = 1$, $P_o\tau = 0.1$ and $g(t)=[1+(t/\tau)^2]^{-3}$.

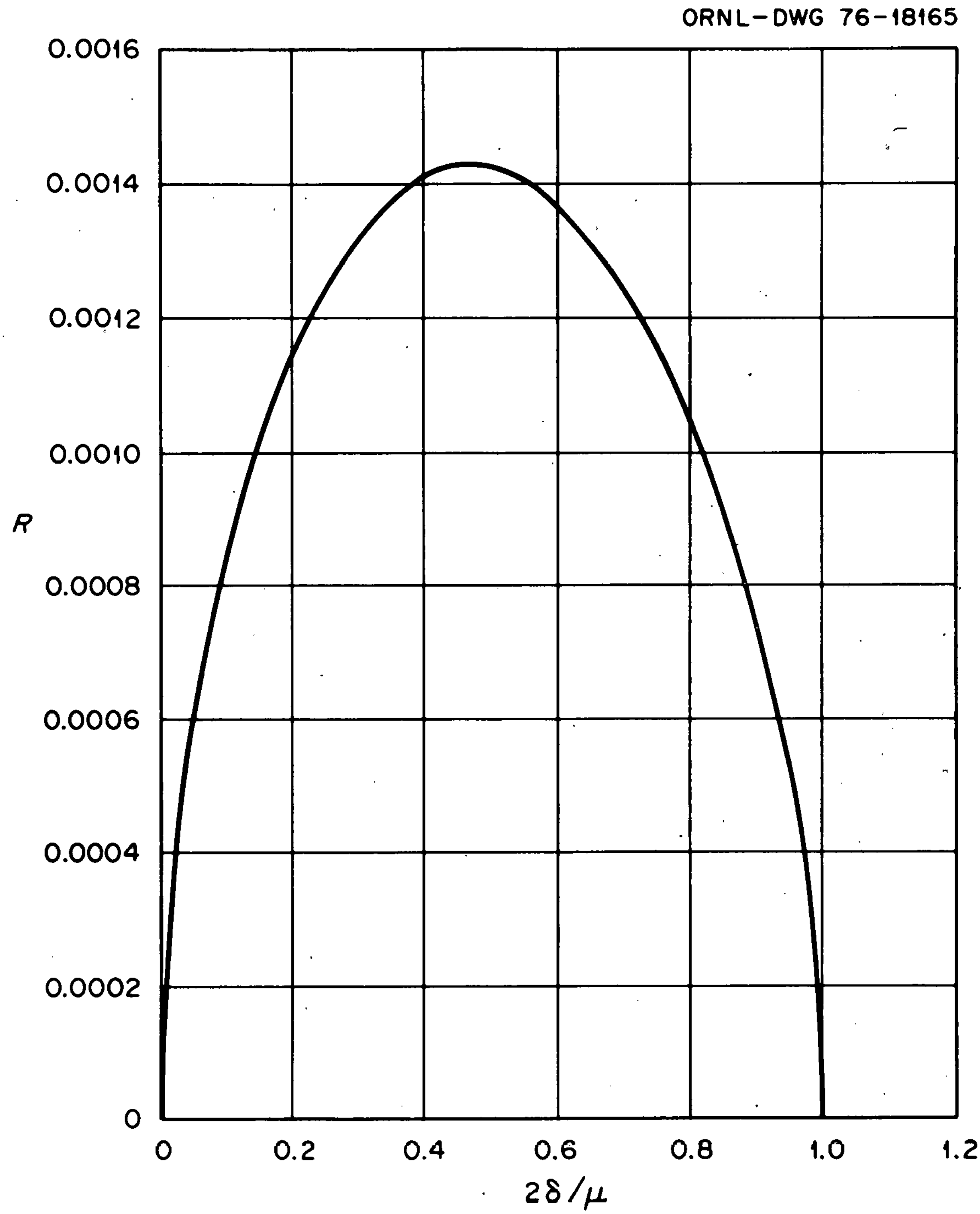

Figure 11. Photoionization Probability (isolated crossings), $|\alpha/\mu|^2|\mu\tau|=1$, $P_o\tau=0.001$ and $g(t)=[1+(t/\tau)^2]^{-3}$.

ORNL-DWG 76-18167

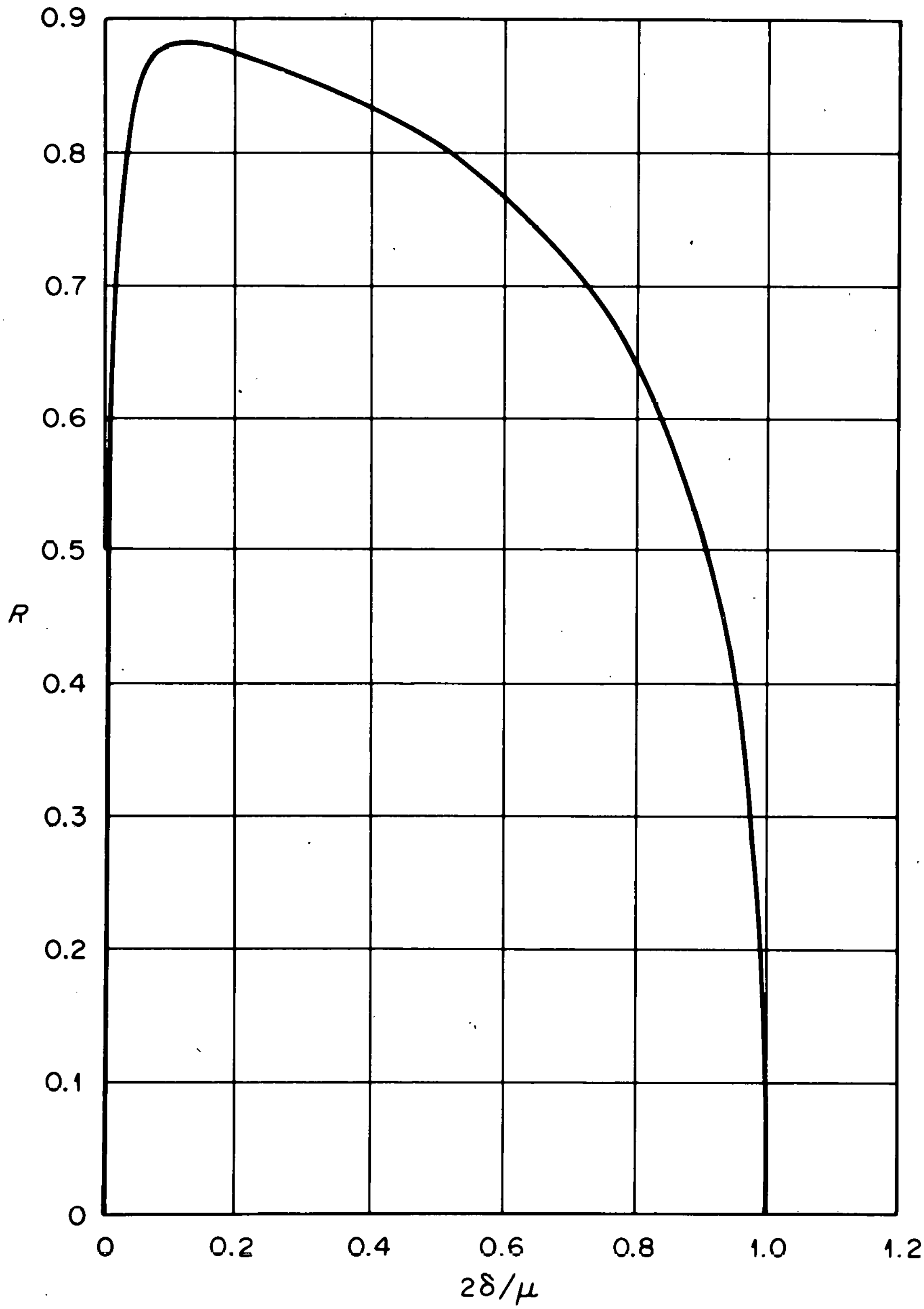

Figure 12. Photoionization Probability (isolated crossings), $|\alpha/\mu|^2|\mu\tau| = 10$, $P_o\tau = 1$ and $g(t)=[1+(t/\tau)^2]^{-3}$.

$$-t_{02} \leq t \leq t_{02};$$

$$= T(1-T)[1+\exp(-2\gamma_2 t_{02}-2p_o \int_{-t_{02}}^{t_{02}} g(t')dt')]$$

$$\times \exp[-\gamma_2(t-t_{02})+2p_o \int_{t_{02}}^{t} g(t')dt'], \qquad (66)$$

$t > t_{02}$. The probability of emission of a fluorescence photon per unit time at t is then

$$S(t) = \gamma_2 |A_2(t)|^2. \qquad (67)$$

If $|\alpha/\mu|^2 \frac{|\mu\tau|}{6} >> 1$, we see that $T \simeq 1$ for nearly all δ satisfying $0 \leq 2\delta/\mu \leq 1$. In such a circumstance nearly all fluorescence occurs in the time interval $-t_{02} \leq t \leq t_{02}$. By adjusting δ/μ the time interval during which the fluorescence occurs can be changed as indicated by Eqs. (60). If $\gamma_2\tau << 1$ and $p_o\tau << 1$ we have a situation where the entire population jumps from the state $|0\rangle$ to the state $|2\rangle$ at time $-t_{02}$, fluoresces until time t_{02}, and then almost all atoms return to the ground state. All fluorescence occurs in a time short compared with the excited states lifetime; but during this time all atoms in the state $|2\rangle$ are fluorescing. Such an effect should be relatively easy to demonstrate with commercially available electronic time gating equipment.

In the situation $|\frac{\alpha}{\mu}|^2 \frac{|\mu\tau|}{6} >> 1$ the population is completely inverted in passing through the first crossing. By using slowly growing pulses which are terminated abruptly it may be possible to have situations where T is nearly unity at a first crossing and small compared with unity at a second. In such a circumstance one should be able to produce very large populations of metastable atoms in elements such as those in the second column of the periodic table. Such a possibility might be of interest as a way to produce very short but intense pulses of metastable atoms for crossed beam scattering experiments.

E. CROSSING EFFECTS DUE TO TIME OF PASSAGE ACROSS TIGHTLY FOCUSED LASER BEAMS

In this section we consider two counter propagating cw laser beams at frequencies w_1' and w_2'. The beams are brought to a common focus in a small cell occupied by a low concentration of atoms having energy levels similar to those described in Section A of the present chapter. If the laser at w_1' is much more powerful as well as more tightly focused, we have a situation in which two-photon excitation is only possible in the region of space where both laser fields are strong. This region can be as small in radius as $\sim 10^{-4}$ cm; so that at thermal equilibrium the time of passage of atom across the high field region is $\sim 10^{-8}$ second. Thus, in such a situation every atom moving through the beam sees a pulsed field of very high intensity. With the most powerful tunable cw dye lasers, the peak power density may reach a level $\gtrsim 10^6$ w/cm^2 at the beam center.

We begin by assuming that the laser at w_2' is much less tightly focused so that in the region of overlap its amplitude can be taken as a constant given by E_{20}. The laser beams propagate parallel to the Z axis and the radial dependance of the amplitude of the laser at w_1' is

$$E_1(t) = E_{10} \exp [- r^2/2\sigma_1{}^2].$$

Thus, an atom approaching the beam sees $E_1(t)$ given by

$$E_1(t) = E_{10} \exp [- \frac{b^2}{2\sigma_1{}^2} - \frac{V_x{}^2 t^2}{2\sigma_1{}^2}],$$

where b is the distance of closest approach to the beam axis and V_x is the component of velocity perpendicular to the Z axis. Obviously the probability of being in $|2\rangle$ at time t, if initially in $|0\rangle$, can be obtained by solving Eqs. (37) with the present interpretation of $E_1(t)$ and $E_2(t)$. Further, here we can have $|\mu\tau| >> 1$ but $|\alpha/\mu|^2 << 1$ so that the method of isolated curve crossing can be applied. The same restrictions (for permitting the derivation of Eqs. (37)) apply here as in the previous sections of the present chapter.

The number of photons emitted per second per unit length of the laser beam is given by (using collisionless kinetic theory and assuming thermal equilibrium)

$$R = \frac{2mN_o}{kT} \int_0^\infty dV_x \int_0^{b_m} dbV_x^2 \exp(-mV_x^2/2kT)$$

$$\times \; P(V_x, b, E_{20}, E_{10}, \sigma_1, \gamma_2, 2\delta) \qquad (68)$$

where N_o is the atom concentration, m is the mass of an atom, T is absolute temperature, k is the Boltzmann's constant and b_m is the maximum value of b, $p(V_x, b, E_{20}, E_{10}, \sigma_1, \gamma_2, 2\delta)$ is the probability that a passage at V_x and b will lead to the emission of a photon with the laser parameters E_{20}, E_{10}, σ_1 and 2δ and with the spontaneous decay rate. Here we have assumed that photoionization does not occur. It remains for us to find p and to use it in Eq. (68). Using the same considerations as in Section D of this chapter we find P and obtain

$$R = 4N_o\sqrt{\frac{2kT}{m}}\, \sigma_1 \left(\ln\frac{K_1}{\Delta}\right)^{1/2} H(|z_o|^2, \Gamma), \qquad (69)$$

where

$$|z_o|^2 = \frac{K_2\sigma_1}{2\left(\ln\frac{K_1}{\Delta}\right)^{1/2}\sqrt{\frac{2kT}{m}}},$$

$$\Gamma = \frac{\gamma_2 \sigma_1}{\sqrt{\frac{2kT}{m}}} \left(\ell n \frac{K_1}{\Delta}\right)^{1/2} , \quad \text{note } \tau = \frac{\sigma_1}{\sqrt{\frac{2kT}{m}}} ;$$

$$K_1 = - \frac{|\langle 0|\hat{P}_x|P\rangle|^2 E_{10}^{\ 2}}{4\hbar^2 \Delta_1} ,$$

$$K_2 = - \frac{|\langle P|\hat{P}_x|2\rangle|^2 E_{20}^{\ 2}}{4\hbar^2 \Delta_2} .$$

Note that we have assumed that there is a single dominant virtual state and $k_1 V_s$ and $k_2 V_s$ in Δ_1 and Δ_2 are ignored; and also note that $\mu = K_1 \exp(-b^2/\sigma_1^{\ 2})$ and $|\alpha| = \sqrt{K_1 K_2} \exp(-b^2/2\sigma_1^{\ 2})$,

$$\Delta = K_2 + 2\delta,$$

$$K_2/K_1 << 1,$$

$$0 < \Delta/K_1 < 1,$$

$$H(|Z_o|^2, P) = 2 \int_0^\infty y^2 e^{-y^2} \int^1 \frac{udu}{\sqrt{1 - u^2}} \left(1 - e^{-\frac{2\pi |Z_o|^2}{yu}}\right)$$

$$\times \left(1 - e^{-\frac{2Pu}{y}} + e^{-\frac{2Pu}{y} - \frac{2\pi |Z_o|^2}{yu}}\right) dy$$

if the spontaneous decay feeds the ground state immediately (case 1) and

$$H(|Z_o|^2,\Gamma) = \int_0^\infty y^2 e^{-y^2} \int_0^1 \frac{u\,du}{\sqrt{1-u^2}}\left(1-e^{\frac{-2\pi|Z_o|^2}{yu}}\right)$$

$$\times \left(1-e^{\frac{-2\Gamma u}{y}} + e^{\frac{-2\pi|Z_o|^2}{yu}} + e^{\frac{-2\Gamma u}{y} - \frac{2\pi|Z_o|^2}{yu}}\right) dy$$

if the spontaneous decay does not appreciably repopulate |0> state during the duration of the laser pulse (case 2).

Fig. 13 and Fig. 18 show $H(|Z_o|^2,\Gamma)$ for the two cases. Figs. 14-17 and Figs. 19-22 show some of the examples of $R/(4N_o\sigma_1\sqrt{2kT/m})$ versus Δ/K_1 for various values of $\Gamma_{oo} = \gamma_2\sigma_1/\sqrt{2kT/m}$ and $Z_{oo} \doteq \frac{1}{2}K_2\sigma_1/\sqrt{2kT/m}$. The general line shapes are similar to the one which was observed by Liao and Bjorkholm.[7] Fig. 14 and Fig. 19 show an interesting aspect which was discussed earlier in Section D of this chapter. With large Z_{oo} and small Γ_{oo} a large population inversion occurs at the first crossing, a small decay occurs between crossings and |2> population becomes nearly null at the second crossing, thereby leaving almost nothing to decay thereafter. With smaller Z_{oo} (but not too small), there remains more upper state population just after the second crossing. Thus, the latter case produces more fluorecence than the former case.

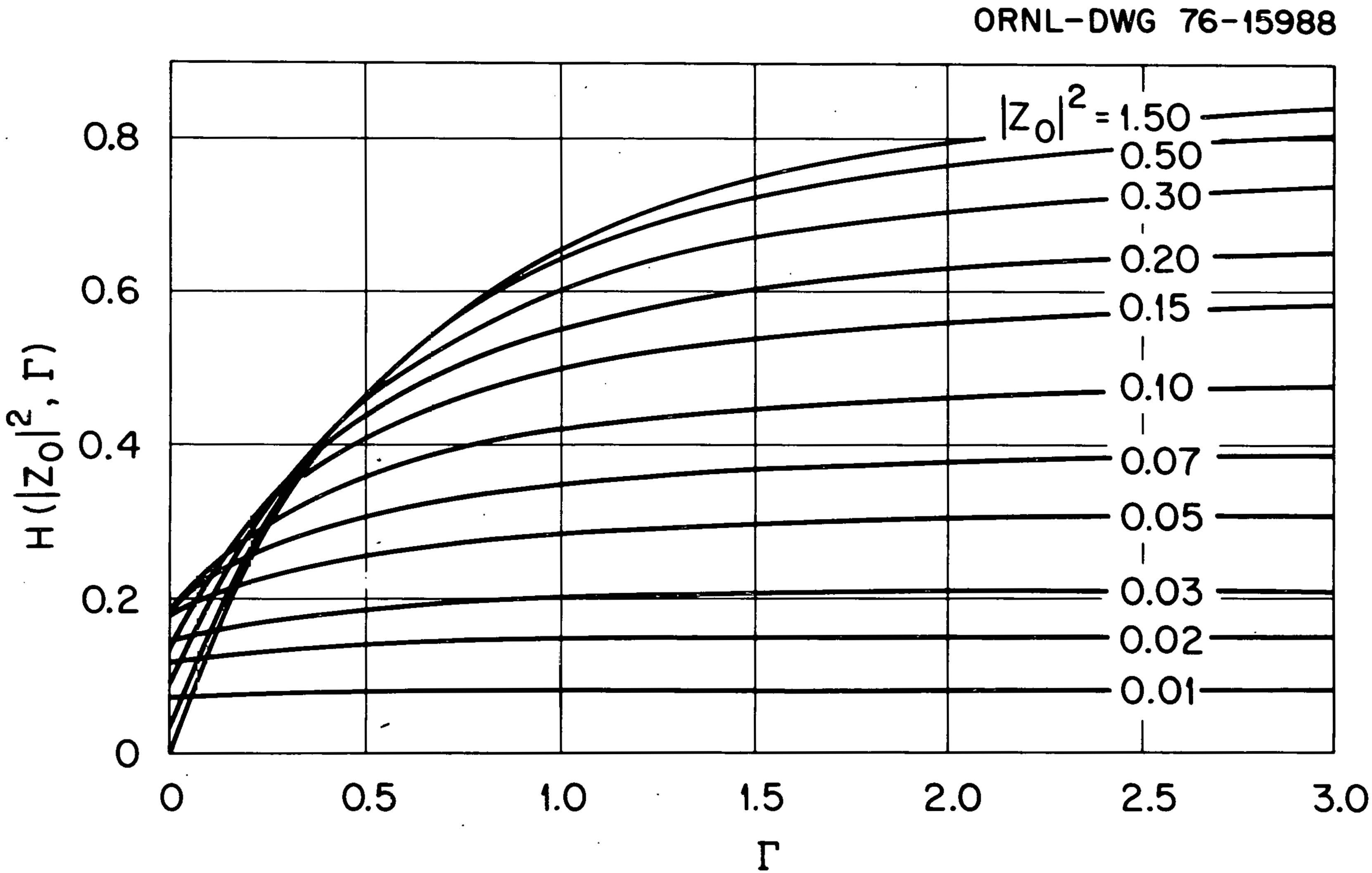

Figure 13. $H(|Z_0|^2, \Gamma)$. Spontaneous decay feeds |0> state without time delay (case 1).

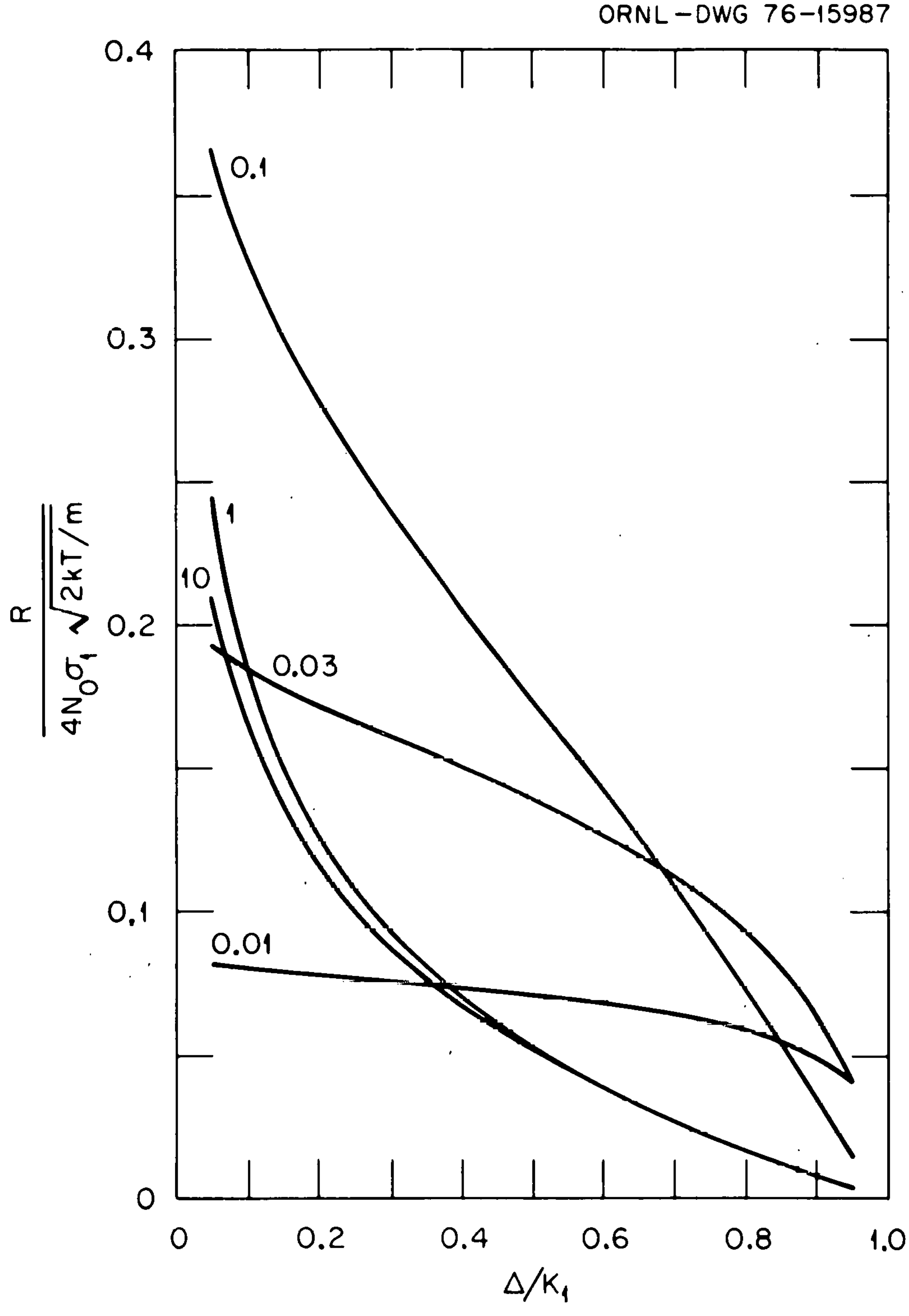

Figure 14. Fluorescence (case 1 with Γ_{oo} = 0.05 and Z_{oo} = 0.01 - 10).

ORNL-DWG 76-15992

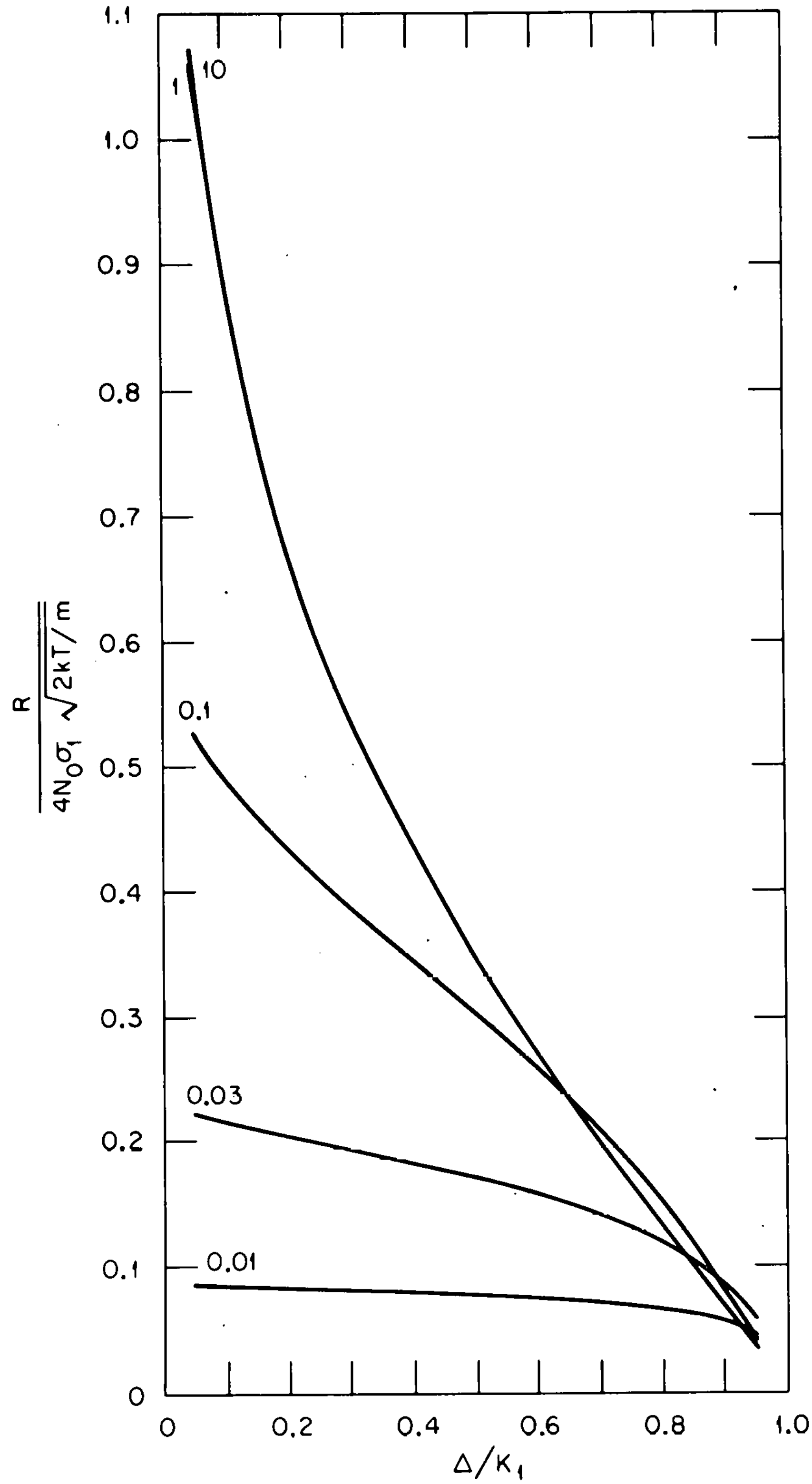

Figure 15. Fluorescence (case 1 with Γ_{oo} = 0.5 and Z_{oo} = 0.01 - 10).

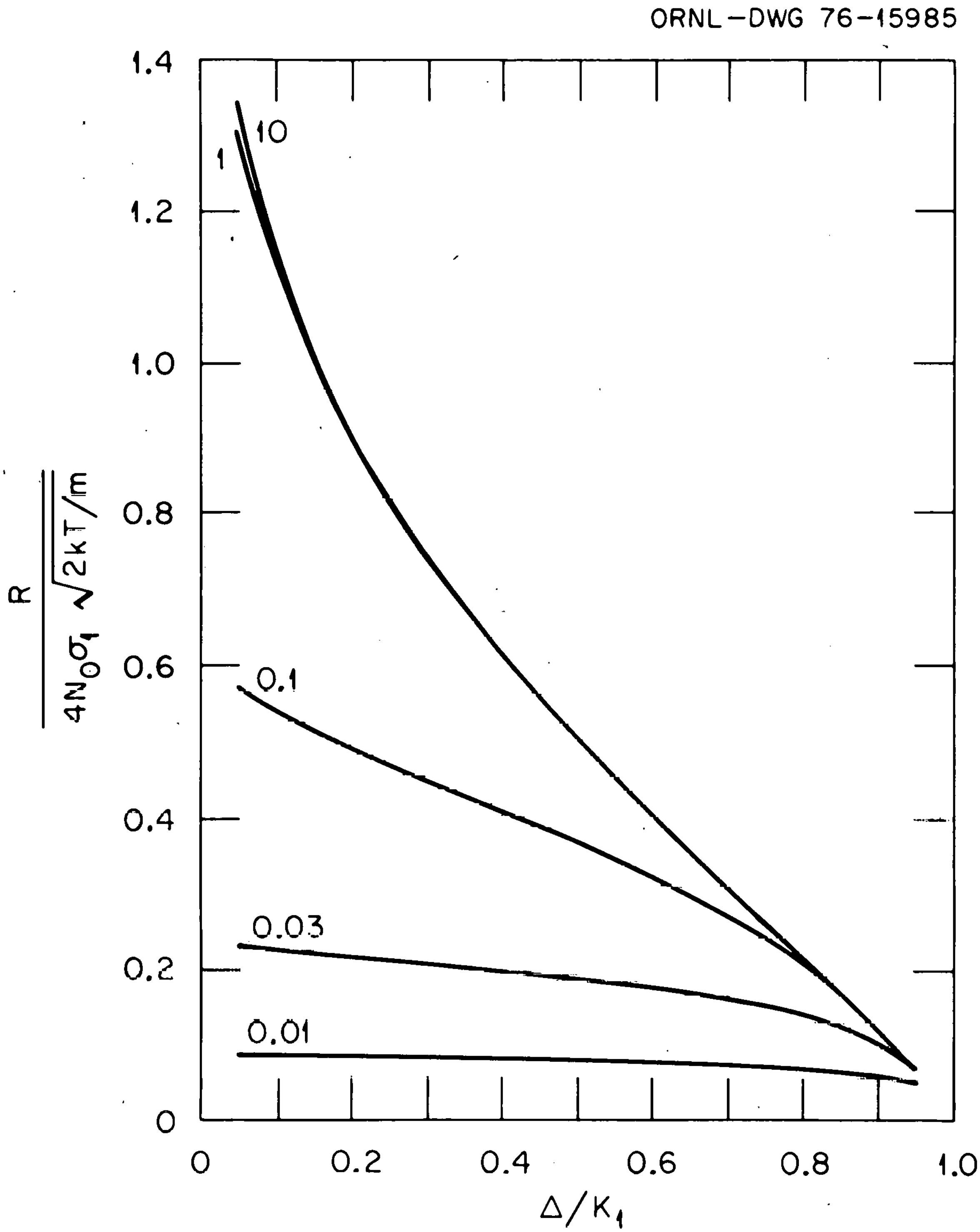

Figure 16. Fluorescence (case 1 with $\Gamma_{oo} = 1.0$ and $Z_{oo} =$ 0.01 - 10).

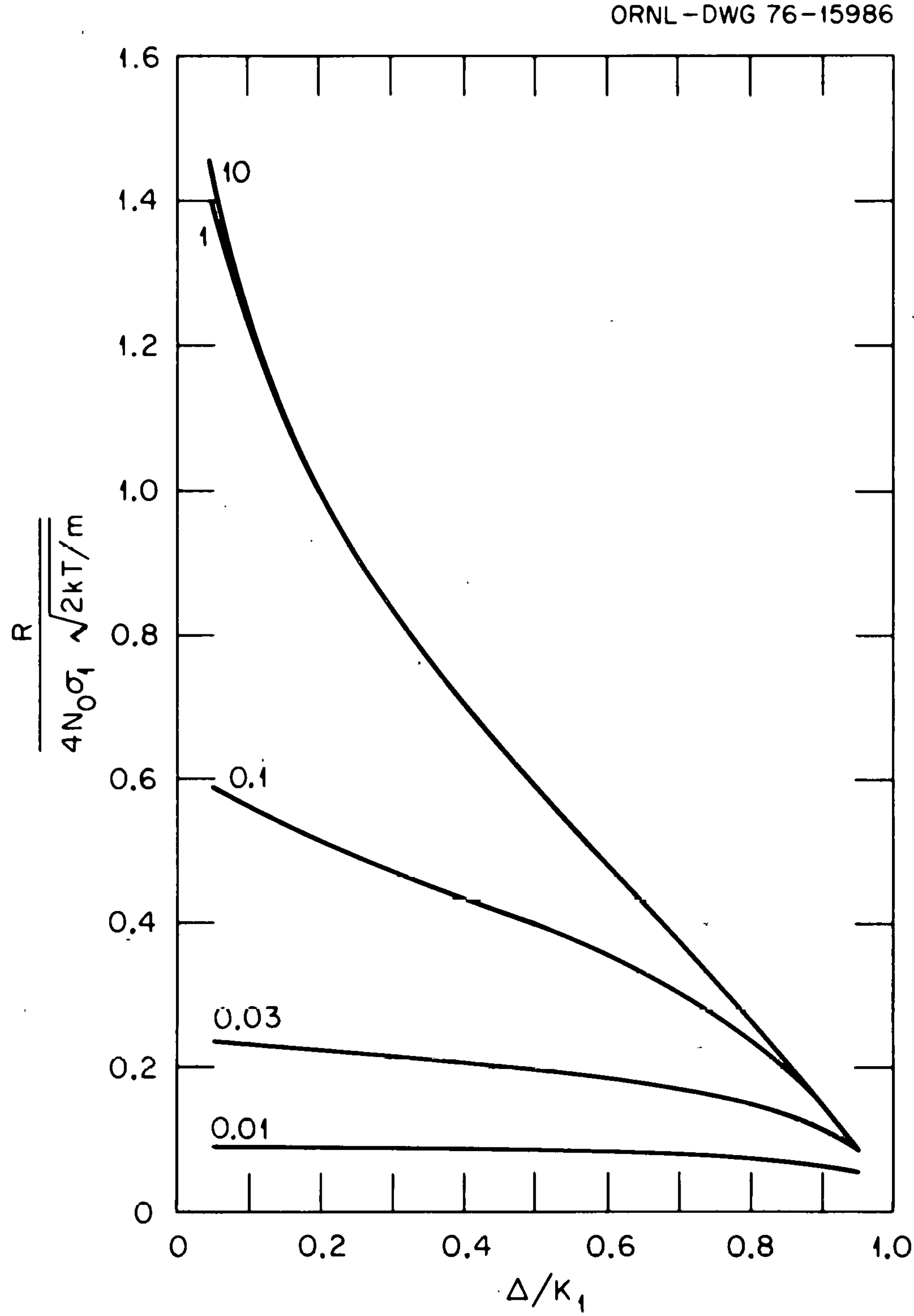

Figure 17. Fluorescence (case 1 with Γ_{oo} = 1.5 and Z_{oo} = 0.01 - 10).

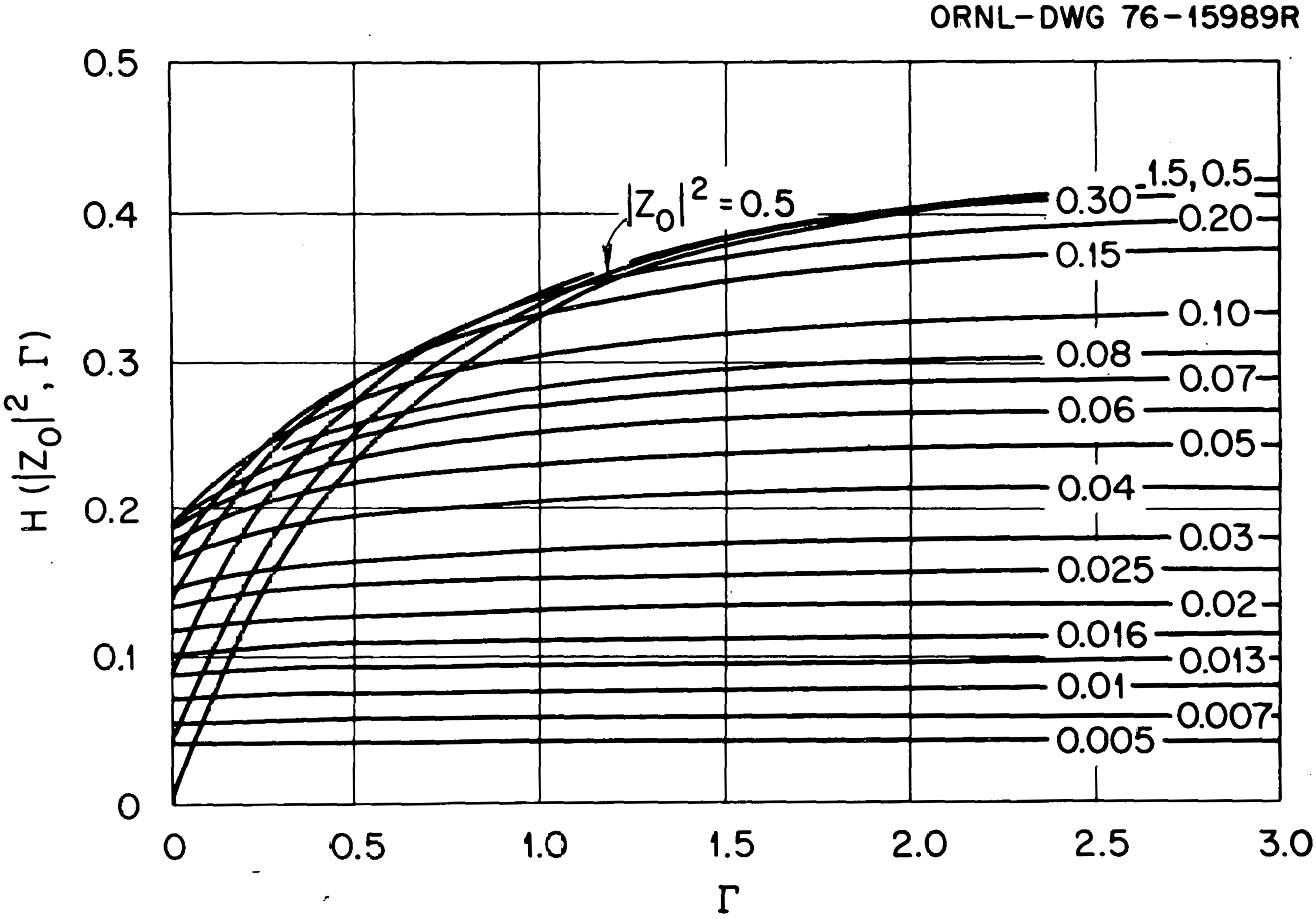

Figure 18. $H(|Z_0|^2, \Gamma)$. Spontaneous decay does not repopulate $|0\rangle$ state (case 2).

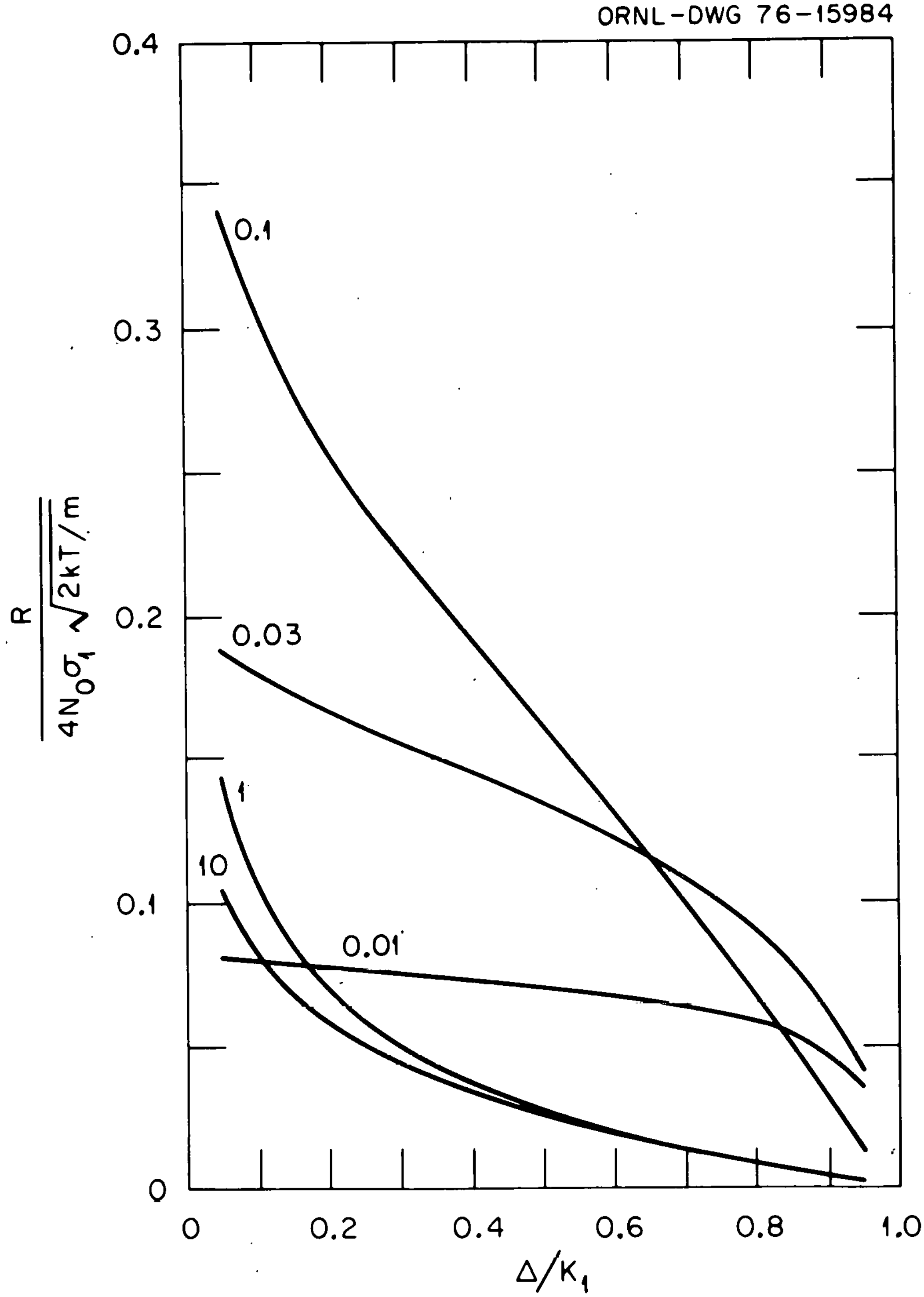

Figure 19. Fluorescence (case 2 with Γ_{oo} = 0.05 and Z_{oo} = 0.01 - 10).

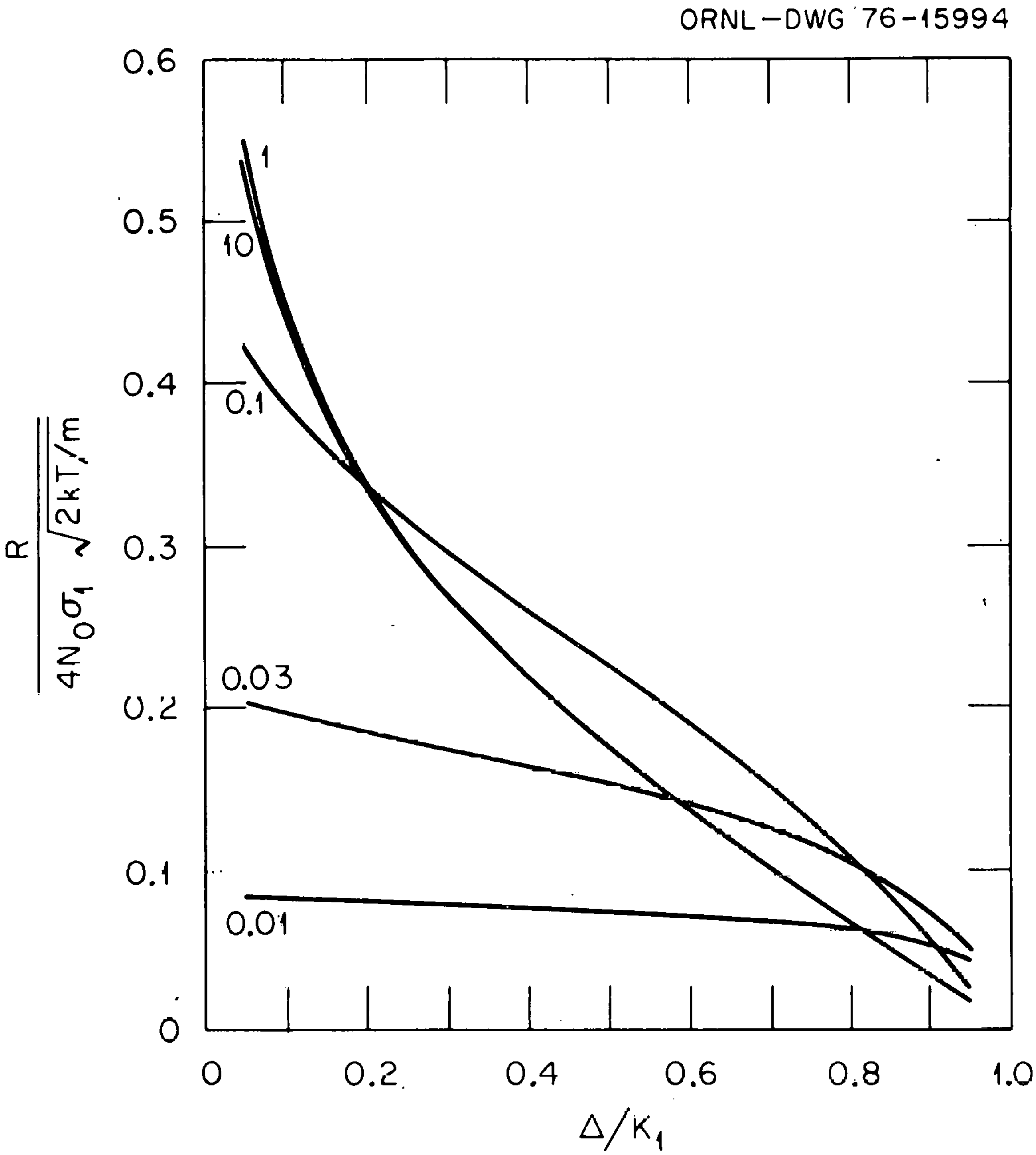

Figure 20. Fluorescence (case 2 with Γ_{oo} = 0.5 and Z_{oo} = 0.01 - 10).

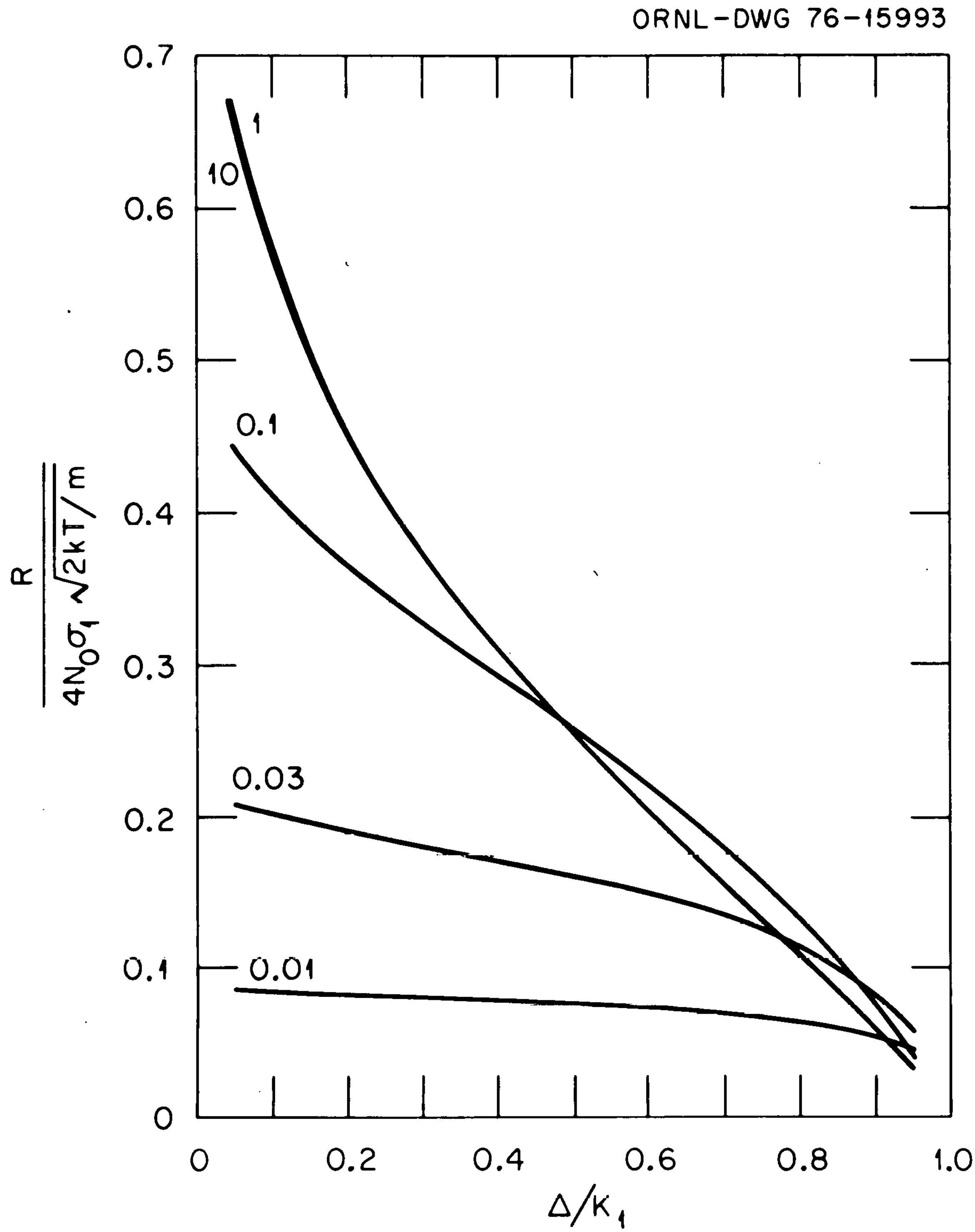

Figure 21. Fluorescence (case 2 with Γ_{oo} = 1.0 and Z_{oo} = 0.01 - 10).

ORNL-DWG 76-15983

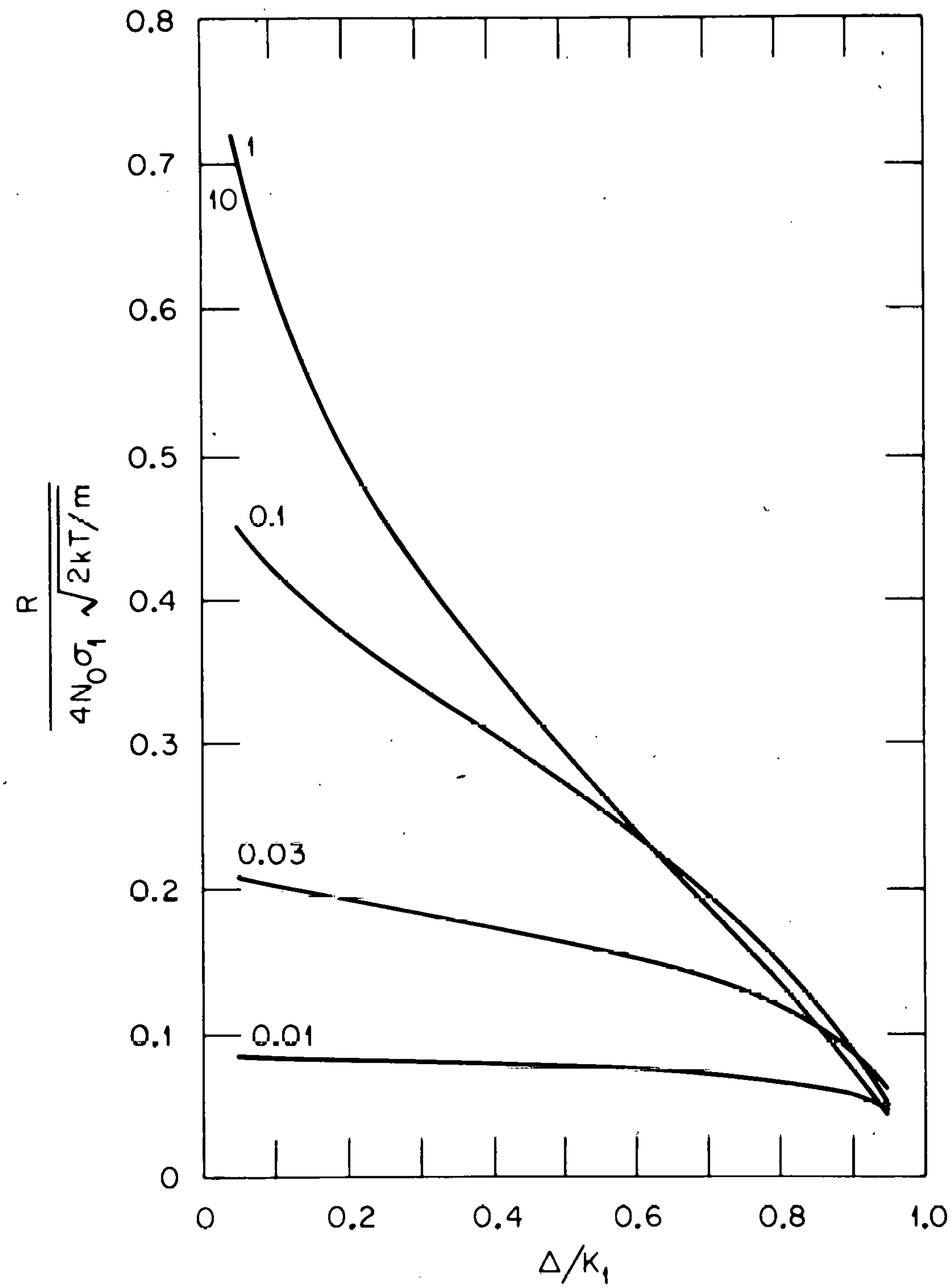

Figure 22. Fluorescence (case 2 with Γ_{oo} = 1.5 and Z_{oo} = 0.01 - 10).

CHAPTER IV

TWO-PHOTON RESONANCE IONIZATION WITH PULSED LASERS

We consider a problem solved earlier by Beers and Armstrong[24] for a fully quantitized single mode electromagnetic field of the square pulse type. In the present work the atom is treated quantum mechanically but the EM field is treated classically. This approximation should be valid for the high power levels that will be emphasized here and which were emphasized in Ref. 24 . The present work allows for the more general pulses of the form

$$\vec{E} = \text{field intensity at } (x_0, y_0, z_0) \text{ at time } t$$

$$= E_0 \vec{i}\ g(S_1 t) \cos(wt - kz_0 + \beta).$$

where $g(S_1 t)$ is a function of the type described (and named the same way) in Section A of Chapter II. The atoms that interact with the laser beam are assumed to be very dilute in density (so that propagation effects and attenuation can be neglected) and to have no component of velocity parallel to the direction of propagation of the laser beam. The atoms are initially in the ground state (i.e. $|0\rangle$) and $\hbar\omega$ is nearly resonant for excitation of a state $|1\rangle$, and a second photon can photoionize state $|1\rangle$ as in Fig. 23. In the latter situation, $|\psi(t)\rangle$ is approximately of the form

$$|\psi(t)\rangle = a_0(t)e^{-iw_0t}|0\rangle + a_1(t)e^{iw_1t}|1\rangle + \sum_\mu \int dE_c C_\mu(E_c,t)e^{-iw_ct}|E_c,\mu\rangle \tag{70}$$

where

$$\langle 0|1\rangle = 0, \langle E_c,\mu|E_c',\mu'\rangle = \delta(E_c-E_c')\delta_{\mu,\mu'}, \langle i|E_c,\mu\rangle = 0.$$

Now,

$$i\hbar \frac{\partial}{\partial t} |\psi(t)\rangle = \hat{H}|\psi(t)\rangle, \tag{71}$$

where

$$\hat{H} = \hat{H}_0 - \hat{P}_x E_0 g(S_1t) \cos(wt-kz_0+\beta) - i\hbar \frac{\gamma_1}{2} |1\rangle\langle 1|, \tag{72}$$

and $\hat{H}_0$ is the electronic Hamiltonian of the isolated atom, $\hat{P}_x$ is the x component of the electric dipole operator and $\vec{E}$ is plane-polarized parallel to the x-axis. We do not follow Beers and Armstrong in including other states which are far off resonance by an effective Hamiltonian method. This will, of course, lead to errors on the far wings of the line shape for photoionization.

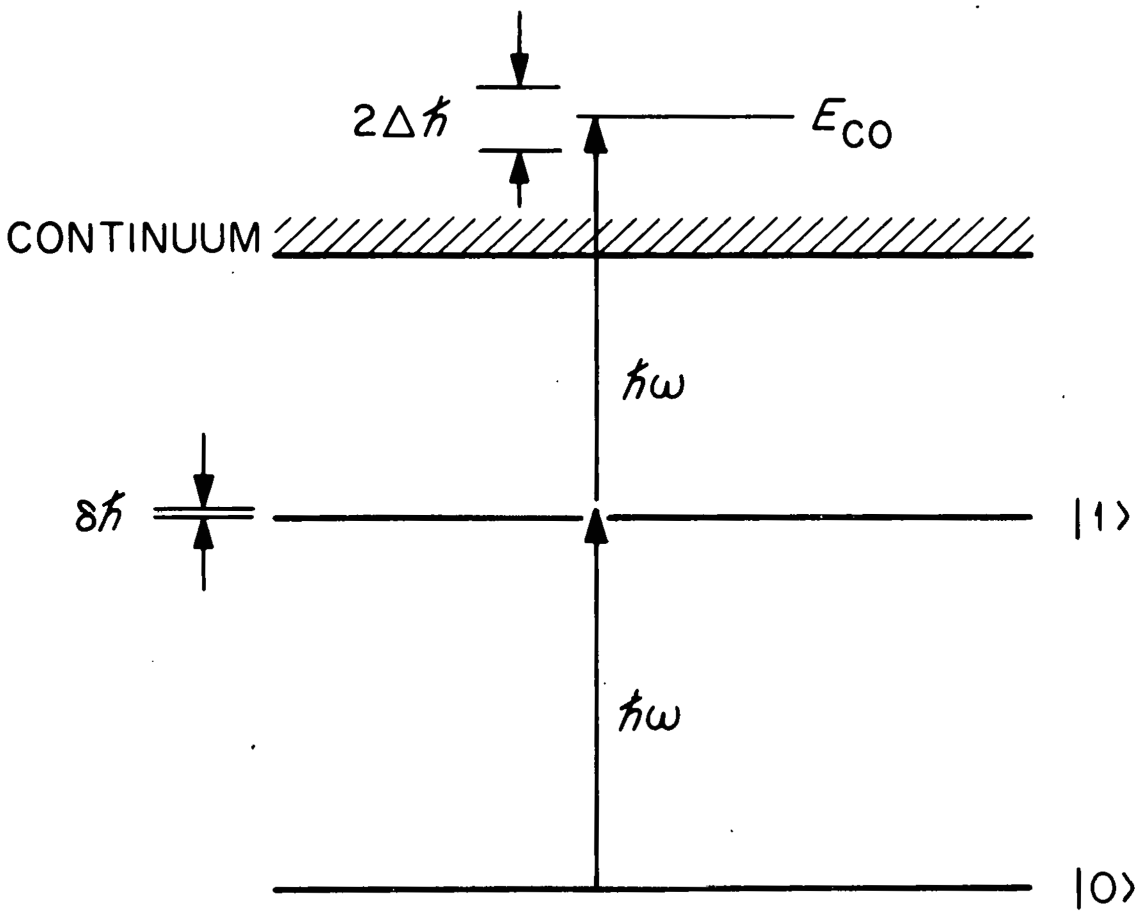

Figure 23. Energy Level Diagram of 2-Photon Transition with the Laser at w.

Using the rotating wave approximation [i.e., keeping only the low frequency oscillating part when $\cos x = (e^{ix} + e^{-ix})/2$ combines with the complex exponentials], we find

$$\frac{da_0}{dt} = iW_p g(S_1 t) e^{i(w-w_1+w_0)t} a_1,$$

$$\frac{da_1}{dt} = -\frac{\gamma_1}{2} a_1 + W_p^* g(S_1 t) e^{-i(w-w_1+w_0)t} a_0$$
$$+ i \sum_\mu \int dE_c C_\mu(E_c,t) e^{i(w_1-w_c+w)t} \alpha_\mu^*(E_c) g(S_1 t), \tag{73}$$

$$\frac{dC_\mu}{dt}(E_c,t) = i\alpha_\mu(E_c) g(S_1 t) e^{-i(w_1-w_c+w)t} a_1$$

where $W_p = \langle 0|\hat{P}_x|1\rangle E_0/2\hbar$, $\alpha_\mu(E_c) = \langle E_c,\mu|\hat{P}_x|1\rangle E_0/2\hbar$. Choosing Δ such that $\alpha_\mu(E_{c0} \pm \hbar\Delta) \simeq \alpha_\mu(E_{c0})$ where $E_{c0} = \hbar w_0 + 2\hbar w$ and $|\Delta| >> |da_1/dt|$, we show that $C_\mu(E_c,t)$ can be eliminated.

To eliminate $C_\mu(E_c,t)$, we integrate the last of Eq. (73)

$$C_\mu(E_c,t) = i\alpha_\mu(E_c) \int_{-\infty}^{t} g(S_1 t') e^{-i(w_1-w_c+w)t'} a_1(t')dt'. \tag{74}$$

We now consider $|w_c - w_1 - w| \geq \Delta$. With the restrictions $|da_1(t)/dt| << \Delta$ and $S_1/\Delta << 1$ we can integrate by parts and neglect the remaing integral.

$$C_\mu(E_c,t) \simeq \frac{\alpha_\mu(E_c)g(S_1t)a_1(t)}{w_c-w_1-w}\, e^{i(w_c-w_1-w)t} \quad , \qquad (75)$$

if $|w_c-w_1-w| \geq \Delta$. When $|w_c-w_1-w| < \Delta$ we can treat $\alpha_\mu(E_c)$ as a constant given by $\alpha_\mu(E_{c0})$. We have

$$C_\mu(E_c,t) = i\alpha_\mu(E_{c0}) \int_{-\infty}^{t} g(S_1t')e^{-i(w_1-w_c+w)t}\, a_1(t')dt', \qquad (76)$$

$$\text{if } |w_c-w_1-w| < \Delta.$$

On substituting Eqs. (75) and (76) into second Eq. (73) the term involving $C_\mu(E_c,t)$ becomes

$$I = \sum_\mu \int dE_c\, C_\mu(E_c,t)e^{i(w_1-w_c+w)t}\, \alpha^*_\mu(E_c)g(S_1t)$$

$$= \sum_\mu \int \frac{|\alpha_\mu(E_c)|^2 dE_c}{w_c-w_1-w}\, g^2(S_1t)a_1(t) \qquad (77)$$

$$+\, i\sum_\mu |\alpha_\mu(E_{c0})|^2 g(S_1t)\int_{-\infty}^{t} g(S_1t')a_1(t')dt' \int_{E_{c0}-\hbar\Delta}^{E_{c0}+\hbar\Delta} dE_c e^{i(w_1-w_c+w)(t-t')} ,$$

where $⨍$ means to delete the region of E_c implied by $|w_c - w_1 - w| < \Delta$. Since $\alpha_\mu(E_c)$ is extremely constant over the latter interval, $⨍$ can be replaced by a principal value integration. Thus,

$$
\begin{aligned}
I &= \overline{W} g^2(S_1 t) a_1(t) \\
&\quad + 2i\hbar \sum_\mu |\alpha_\mu(E_{c0})|^2 g(S_1 t) \int_{-\infty}^{t} g(S_1 t') a_1(t') e^{-i(w_{c0} - w_1 - w)(t-t')} \\
&\quad \times \frac{\sin \Delta(t-t')}{t-t'} \, dt'
\end{aligned}
\tag{78}
$$

with

$$\overline{W} = P \int \frac{dE_c \sum_\mu |\alpha_\mu(E_c)|^2}{w_c - w_1 - w} . \tag{79}$$

With the assumptions that $|\delta| \ll \Delta$ ($\delta = w_{c0} - w_1 - w$) and that $(t-t')^{-1} \sin \Delta(t-t')$ behaves like a Dirac delta function with its peak at $t' = t$, we find

$$\int_{-\infty}^{t} g(S_1 t') a_1(t') e^{-i\delta(t-t')} \frac{\sin \Delta(t-t')}{t-t'} \, dt' \approx \frac{\pi}{2} g(S_1 t) a_1(t) .$$

Thus,

$$I = \overline{W} g^2(S_1 t) a_1(t) + i P_0 g^2(S_1 t) a_1(t) , \tag{80}$$

where

$$P_o = \hbar\ \pi\sum_{\mu}|\alpha_\mu(E_{c0})|^2\ . \qquad (81)$$

In several other problems in the previous chapter we were faced with the inclusion of continuum states. A procedure which is completely analogous to the one described above was used to eliminate the continuum amplitude in each case.

With Eqs. (73) and (80)

$$\frac{da_0}{dt} = iW_p g(S_1t)e^{i\delta t}\ a_1,$$

$$\frac{da_1}{dt} = iW_p^* g(S_1t)e^{-i\delta t}a_0 + (i\overline{W}-p_o)g^2(S_1t)a_1 - \frac{\gamma_1}{2}\ a_1\ ,$$

with $\delta = w - (w_1 - w_0)$. Thus, we now have an effective two-state problem. Let $a_1(t) = b_1(t)\exp[i\overline{W}\int_{-\infty}^{t} g^2(S_1t')dt']$, and

$$\frac{da_0}{dt} = iW_p g(S_1t)e^{i\delta t}\ \exp[i\overline{W}\int_{-\infty}^{t} g^2(S_1t')dt']b_1,$$

$$\frac{db_1}{dt} = -(\frac{\gamma_1}{2} + P_o g^2(S_1t))b_1 + iW_p^*\ g(S_1t)e^{-i\delta t} \qquad (82)$$

$$\times\ \exp[-i\overline{W}\int_{-\infty}^{t} g^2(S_1t')dt']a_0.$$

The coupling terms $W_p g \exp[i\delta t + i\overline{W}\int_{-\infty}^{t} g^2(S_1 t')dt']$ have the property that the oscillating exponential is ineffective in decreasing the effectiveness of the coupling if $|W_p g| >> |\delta + \overline{W}g^2|$. Essentially, this means that $|W_p g|$ is so big that a_o and b_1 can be changed by large amounts in a time which is small compared with the time for $\exp[i\delta t + i\overline{W}\int_{-\infty}^{t} g^2(S_1 t')dt']$ to oscillate once. Thus, if $|W_p| >> |\overline{W}|$ the a.c. Stark shift due to coupling with the continuum states can be neglected for all δ. We note further that unles $\langle 0|\hat{P}_x|1\rangle$ is extremely small we will have both $|W_p| >> |\overline{W}|$ and $|W_p| >> P_o$ in any problems of this type. We also take the peak power to be very large so that $|W_p| >> \gamma_1$ and $|W_p|/S_1 >> 1$. In this situation the factorization method of Chapter II can be used as follows:

$$\left(\frac{d}{dt} + g_1\right)\left(\frac{d}{dt} + g_2\right) b_1 = 0,$$

with

$$g_1 + g_2 = i\delta - \frac{d}{dt}\ell n g + \left(\frac{\gamma_1}{2} + P_o g^2\right), \tag{83}$$

$$g_1 g_2 + \frac{dg_2}{dt} = |W_p|^2 g^2 + i\delta\left(\frac{\gamma_1}{2} + P_o g^2\right) - \left(\frac{\gamma_1}{2} + P_o g^2\right)\frac{d}{dt}\ell n g + P_o\frac{d}{dt}g^2.$$

If $\gamma_1/|W_p| << 1$ and $P_o/|W_p| << 1$ we can neglect some of the terms and use

$$g_1 + g_2 = i\delta - \frac{d}{dt}\ell n g + (\frac{\gamma_1}{2} + P_o g^2),$$

$$g_1 g_2 + \frac{dg_2}{dt} \simeq |W_p|^2 g^2 + i\delta(\frac{\gamma_1}{2} + P_o g^2). \tag{84}$$

With our assumptions the following g_1 and g_2 are adequate for good accuracy (we take $\gamma_1/S_1 << 1$ for simplicity)

$$g_1 = \frac{i\delta+i\epsilon K}{2} - (\frac{1}{2} + \frac{|\delta|}{2K}) \frac{d}{dt}\ell n g + (\frac{1}{2} - \frac{|\delta|}{2K})P_o g^2 - \frac{1}{2}\frac{d}{dt}\ell n K,$$

$$g_2 = \frac{i\delta-i\epsilon K}{2} - (\frac{1}{2} - \frac{|\delta|}{2K}) \frac{d}{dt}\ell n g + (\frac{1}{2} + \frac{|\delta|}{2K})P_o g^2 + \frac{1}{2}\frac{d}{dt}\ell n K, \tag{85}$$

where

$$\epsilon = \delta/|\delta|$$

$$K - \sqrt{\delta^2 + 4|W_p|^2 y^2}\ .$$

Eq. (15) of Chapter II yields

$$|a_1(t)|^2 = (\frac{1}{2} - \frac{|\delta|}{2K})\exp[-P_o\int_{-\infty}^{t} (1 - \frac{|\delta|}{K})g^2 dt'].$$

The ionization probability R is given by

$$R = 1 - \exp\ [-P_o\tau\int_{-\infty}^{\infty} (1 - \frac{|\delta|}{K})g^2 dV], \tag{86}$$

where $\tau = 1/S_1$, $V = tS_1$. R can be put in the form

$$R = 1 - \exp\,[-P_o \tau L_g(|\delta/(2W_p)|)], \qquad (87)$$

where; assuming $g(-V) = g(V)$,

$$L_g(x) = 2\int_0^\infty (1 - x/\sqrt{x^2 + g^2}\,)\, g^2 dV. \qquad (88)$$

Any smoothly varying function having a single maximum of unit amplitude at t = 0 and approaching zero sufficiently rapidly at $t \to \pm\infty$ can be used. We take $g(v) = [1+V^2]^{-3/2}$ and tabulate $L_g(x)$. A graph of $(-P_o\tau)^{-1}\ell n(1-R)$ versus $|\delta/(2W_p)|$ is a graph of the function L_g. For our choice of g a graph of L_g is given in Fig. 24. We note that $L_g(0) = 3\pi/8$ and for $X \gtrsim 6$ we have $L_g(x) \approx 0.3866/x^2$. Thus, at high power levels the entire line shape has been calculated accurately by the factorization method.

With the same approximations and a square pulse

$$\begin{aligned} g^2(S_1 t) &= g^2(V), \\ &= 0, \quad V < 0; \\ &= \frac{3\pi}{8}, \quad 0 \le V \le 1; \\ &= 0\,, \quad V > 1; \end{aligned}$$

we find

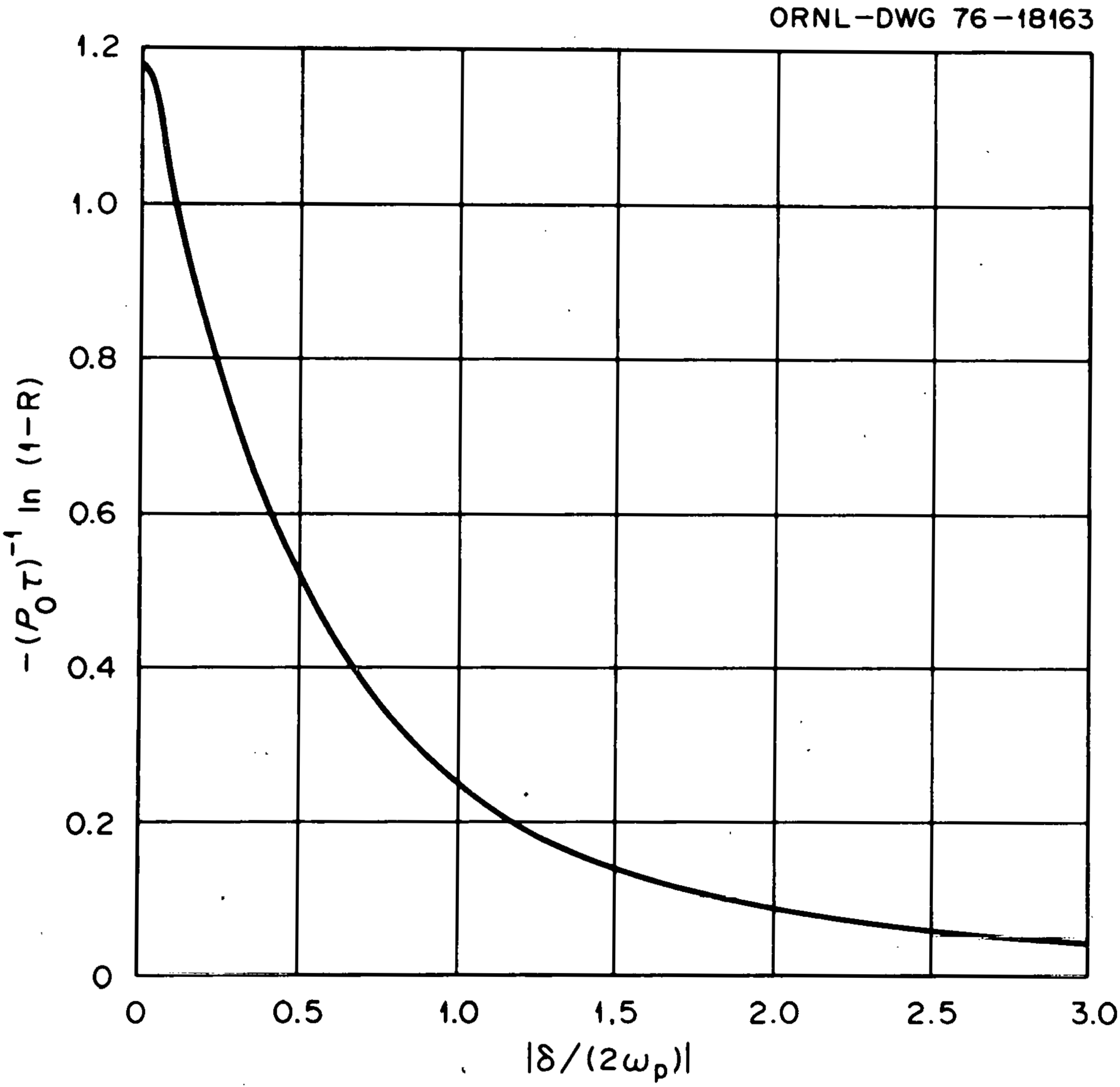

Figure 24. Photoionization Probability, $g(t) = [1 + (t/\tau)^2]^{-3/2}$, 2-Photon Transition.

$$R = \frac{1}{2}\,\frac{P_o g^2 \tau}{1 + x^2}\,\left[\frac{1 - e^{-A}}{A} + \frac{1 - e^{-B}}{B}\right] \qquad (89)$$

where

$$x = |\delta/(2W_p)|,$$

$$A = P_o g^2 \tau \;[1 - x/\sqrt{1 + x^2}],$$

$$B = P_o g^2 \tau \;[1 + x/\sqrt{1 + x^2}].$$

If $x >> 1$, we obtain an asymptotic solution

$$R = 1 - \exp[-P_o g^2 \tau/(2x^2)].$$

In the square pulse case, γ_1 and P_o can be included without the approximation $\gamma_1/|W_p|$ and $P_o/|W_p| << 1$ [see Eqs. (91), Appendix B].

It is interesting to note that if $P_o \tau >> 1$, then both the smooth pulse and the square pulse yield similar line shapes— R being $\simeq 1$ for both cases if $x << \sqrt{P_o \tau}$, with the smooth pulse R dropping a bit faster on the far wing. Also, we note that when $P_o \tau >> 1$ we can use the asymptotic forms of Eqs. (87) and (89) for all δ.

APPENDIX A

TABULATION OF THE H FUNCTION

In Section C of Chapter III we have derived analytical expression for photoionization probability

$$R = 1 - \exp\,[-P_o \tau H(\tfrac{\delta}{\mu},\ |\tfrac{\alpha}{\mu}|)],$$

where

$$H(\tfrac{\alpha}{\mu},\ |\tfrac{\alpha}{\mu}|) = \int_{-\infty}^{\infty} [1 - \frac{\varepsilon'(2\delta/\mu - g(v))}{\sqrt{(2\delta/\mu - g(v))^2 + 4|\alpha/\mu|^2\ g^2(v)}}]\,g(v)\,dV,$$

$$\text{if } \delta \neq 0;$$

$$= \frac{3\pi}{2}\ |\alpha/\mu|^2 (1+4|\alpha/\mu|^2)^{-1},$$

$$\text{if } \delta = 0;$$

and $\varepsilon' = \delta\mu/|\delta\mu|$.

We have tabulated $H(\frac{\delta}{\mu},\ |\frac{\alpha}{\mu}|)$ for $g(v) = (1+V^2)^{-3}$ and the results are given in the following table in the order $\frac{\delta}{\mu}$, $|\frac{\alpha}{\mu}|$ and $H(\frac{\delta}{\mu},\ |\frac{\alpha}{\mu}|)$.

TABLE 1

$H(\frac{\delta}{\mu}, |\frac{\alpha}{\mu}|)$

$\frac{\delta}{\mu}$	$\lvert\frac{\alpha}{\mu}\rvert$	$H(\frac{\delta}{\mu}\ \lvert\frac{\alpha}{\mu}\rvert)$
0.1000000D 01	0.1000000D 00	0.9712551D-02
0.1000000D 01	0.2000000D 00	0.3658152D-01
0.1000000D 01	0.3000000D 00	0.7524913D-01
0.1000000D 01	0.4000000D 00	0.1200089D 00
0.1000000D 01	0.5000000D 00	0.1665224D 00
0.1000000D 01	0.6000000D 00	0.2121147D 00
0.1000000D 01	0.7000000D 00	0.2553891D 00
0.1000000D 01	0.8000000D 00	0.2957536D 00
0.9600000D 00	0.1000000D 00	0.1126695D-01
0.9600000D 00	0.2000000D 00	0.4202336D-01
0.9600000D 00	0.3000000D 00	0.8534175D-01
0.9600000D 00	0.4000000D 00	0.1343135D 00
0.9600000D 00	0.5000000D 00	0.1841085D 00
0.9600000D 00	0.6000000D 00	0.2320302D 00
0.9600000D 00	0.7000000D 00	0.2768452D 00
0.9600000D 00	0.8000000D 00	0.3181550D 00
0.9200000D 00	0.1000000D 00	0.1322724D-01
0.9200000D 00	0.2000000D 00	0.4873299D-01
0.9200000D 00	0.3000000D 00	0.9744327D-01
0.9200000D 00	0.4000000D 00	0.1510110D 00
0.9200000D 00	0.5000000D 00	0.2041698D 00
0.9200000D 00	0.6000000D 00	0.2543275D 00

0.9200000D 00	0.7000000D 00	0.3005113D 00
0.9200000D 00	0.8000000D 00	0.3425718D 00
0.8800000D 00	0.1000000D 00	0.1574763D-01
0.8800000D 00	0.2000000D 00	0.5711692D-01
0.8800000D 00	0.3000000D 00	0.1120644D 00
0.8800000D 00	0.4000000D 00	0.1705727D 00
0.8800000D 00	0.5000000D 00	0.2270846D 00
0.8800000D 00	0.6000000D 00	0.2792937D 00
0.8800000D 00	0.7000000D 00	0.3266019D 00
0.8800000D 00	0.8000000D 00	0.3691653D 00
0.8400000D 00	0.1000000D 00	0.1906250D-01
0.8400000D 00	0.2000000D 00	0.6774637D-01
0.8400000D 00	0.3000000D 00	0.1298612D 00
0.8400000D 00	0.4000000D 00	0.1935585D 00
0.8400000D 00	0.5000000D 00	0.2532756D 00
0.8400000D 00	0.6000000D 00	0.3072351D 00
0.8400000D 00	0.7000000D 00	0.3553390D 00
0.8400000D 00	0.8000000D 00	0.3980990D 00
0.8000000D 00	0.1000000D 00	0.2354120D-01
0.8000000D 00	0.2000000D 00	0.8143481D-01
0.8000000D 00	0.3000000D 00	0.1516726D 00
0.8000000D 00	0.4000000D 00	0.2206224D 00
0.8000000D 00	0.5000000D 00	0.2832051D 00
0.8000000D 00	0.6000000D 00	0.3384720D 00
0.8000000D 00	0.7000000D 00	0.3869473D 00
0.8000000D 00	0.8000000D 00	0.4295357D 00

0.7600000D 00	0.1000000D 00	0.2979077D-01
0.7600000D 00	0.2000000D 00	0.9935251D-01
0.7600000D 00	0.3000000D 00	0.1785566D 00
0.7600000D 00	0.4000000D 00	0.2525107D 00
0.7600000D 00	0.5000000D 00	0.3173647D 00
0.7600000D 00	0.6000000D 00	0.3733295D 00
0.7600000D 00	0.7000000D 00	0.4216491D 00
0.7600000D 00	0.8000000D 00	0.4636337D 00
0.7200000D 00	0.1000000D 00	0.3885827D-01
0.7200000D 00	0.2000000D 00	0.1231885D 00
0.7200000D 00	0.3000000D 00	0.2118172D 00
0.7200000D 00	0.4000000D 00	0.2900458D 00
0.7200000D 00	0.5000000D 00	0.3562585D 00
0.7200000D 00	0.6000000D 00	0.4121267D 00
0.7200000D 00	0.7000000D 00	0.4596575D 00
0.7200000D 00	0.8000000D 00	0.5005438D 00
0.6800000D 00	0.1000000D 00	0.5265032D-01
0.6800000D 00	0.2000000D 00	0.1553558D 00
0.6800000D 00	0.3000000D 00	0.2529955D 00
0.6800000D 00	0.4000000D 00	0.3340881D 00
0.6800000D 00	0.5000000D 00	0.4003766D 00
0.6800000D 00	0.6000000D 00	0.4551620D 00
0.6800000D 00	0.7000000D 00	0.5011686D 00
0.6800000D 00	0.8000000D 00	0.5404048D 00

0.6400000D 00	0.1000000D 00	0.7483823D-01
0.6400000D 00	0.2000000D 00	0.1991815D 00
0.6400000D 00	0.3000000D 00	0.3037919D 00
0.6400000D 00	0.4000000D 00	0.3854716D 00
0.6400000D 00	0.5000000D 00	0.4501622D 00
0.6400000D 00	0.6000000D 00	0.5026971D 00
0.6400000D 00	0.7000000D 00	0.5463540D 00
0.6400000D 00	0.8000000D 00	0.5833401D 00
0.6000000D 00	0.1000000D 00	0.1127930D 00
0.6000000D 00	0.2000000D 00	0.2588850D 00
0.6000000D 00	0.3000000D 00	0.3658756D 00
0.6000000D 00	0.4000000D 00	0.4449117D 00
0.6000000D 00	0.5000000D 00	0.5059723D 00
0.6000000D 00	0.6000000D 00	0.5549406D 00
0.6000000D 00	0.7000000D 00	0.5953537D 00
0.6000000D 00	0.8000000D 00	0.6294547D 00
0.5600000D 00	0.1000000D 00	0.1809611D 00
0.5600000D 00	0.2000000D 00	0.3389329D 00
0.5600000D 00	0.3000000D 00	0.4405632D 00
0.5600000D 00	0.4000000D 00	0.5128952D 00
0.5600000D 00	0.5000000D 00	0.5680391D 00
0.5600000D 00	0.6000000D 00	0.6120330D 00
0.5600000D 00	0.7000000D 00	0.6482703D 00
0.5600000D 00	0.8000000D 00	0.6788332D 00

0.5200000D 00	0.1000000D 00	0.3014181D 00
0.5200000D 00	0.2000000D 00	0.4423623D 00
0.5200000D 00	0.3000000D 00	0.5284195D 00
0.5200000D 00	0.4000000D 00	0.5895770D 00
0.5200000D 00	0.5000000D 00	0.6364382D 00
0.5200000D 00	0.6000000D 00	0.6740363D 00
0.5200000D 00	0.7000000D 00	0.7051656D 00
0.5200000D 00	0.8000000D 00	0.7315391D 00
0.4800000D 00	0.1000000D 00	0.4842734D 00
0.4800000D 00	0.2000000D 00	0.5685455D 00
0.4800000D 00	0.3000000D 00	0.6289377D 00
0.4800000D 00	0.4000000D 00	0.6747142D 00
0.4800000D 00	0.5000000D 00	0.7110726D 00
0.4800000D 00	0.6000000D 00	0.7409302D 00
0.4800000D 00	0.7000000D 00	0.7660609D 00
0.4800000D 00	0.8000000D 00	0.7876164D 00
0.4400000D 00	0.1000000D 00	0.7009434D 00
0.4400000D 00	0.2000000D 00	0.7123428D 00
0.4400000D 00	0.3000000D 00	0.7404925D 00
0.4400000D 00	0.4000000D 00	0.7676687D 00
0.4400000D 00	0.5000000D 00	0.7916772D 00
0.4400000D 00	0.6000000D 00	0.8126167D 00
0.4400000D 00	0.7000000D 00	0.8309406D 00
0.4400000D 00	0.8000000D 00	0.8470935D 00

0.4000000D 00	0.1000000D 00	0.9137497D 00
0.4000000D 00	0.2000000D 00	0.8660309D 00
0.4000000D 00	0.3000000D 00	0.8606446D 00
0.4000000D 00	0.4000000D 00	0.8674847D 00
0.4000000D 00	0.5000000D 00	0.8778473D 00
0.4000000D 00	0.6000000D 00	0.8889336D 00
0.4000000D 00	0.7000000D 00	0.8997617D 00
0.4000000D 00	0.8000000D 00	0.9099893D 00
0.3600000D 00	0.1000000D 00	0.1107712D 01
0.3600000D 00	0.2000000D 00	0.1022407D 01
0.3600000D 00	0.3000000D 00	0.9866663D 00
0.3600000D 00	0.4000000D 00	0.9730233D 00
0.3600000D 00	0.5000000D 00	0.9690859D 00
0.3600000D 00	0.6000000D 00	0.9696770D 00
0.3600000D 00	0.7000000D 00	0.9724665D 00
0.3600000D 00	0.8000000D 00	0.9763241D 00
0.3200000D 00	0.1000000D 00	0.1282320D 01
0.3200000D 00	0.2000000D 00	0.1176572D 01
0.3200000D 00	0.3000000D 00	0.1116046D 01
0.3200000D 00	0.4000000D 00	0.1083118D 01
0.3200000D 00	0.5000000D 00	0.1064864D 01
0.3200000D 00	0.6000000D 00	0.1054632D 01
0.3200000D 00	0.7000000D 00	0.1049002D 01
0.3200000D 00	0.8000000D 00	0.1046133D 01

0.2800000D 00	0.1000000D 00	0.1440778D 01
0.2800000D 00	0.2000000D 00	0.1325978D 01
0.2800000D 00	0.3000000D 00	0.1246806D 01
0.2800000D 00	0.4000000D 00	0.1196715D 01
0.2800000D 00	0.5000000D 00	0.1164687D 01
0.2800000D 00	0.6000000D 00	0.1143608D 01
0.2800000D 00	0.7000000D 00	0.1129348D 01
0.2800000D 00	0.8000000D 00	0.1119488D 01
0.2400000D 00	0.1000000D 00	0.1586389D 01
0.2400000D 00	0.2000000D 00	0.1469730D 01
0.2400000D 00	0.3000000D 00	0.1377603D 01
0.2400000D 00	0.4000000D 00	0.1312980D 01
0.2400000D 00	0.5000000D 00	0.1268159D 01
0.2400000D 00	0.6000000D 00	0.1236488D 01
0.2400000D 00	0.7000000D 00	0.1213547D 01
0.2400000D 00	0.8000000D 00	0.1196527D 01
0.2000000D 00	0.1000000D 00	0.1721868D 01
0.2000000D 00	0.2000000D 00	0.1607914D 01
0.2000000D 00	0.3000000D 00	0.1507717D 01
0.2000000D 00	0.4000000D 00	0.1431382D 01
0.2000000D 00	0.5000000D 00	0.1375064D 01
0.2000000D 00	0.6000000D 00	0.1333289D 01
0.2000000D 00	0.7000000D 00	0.1301765D 01
0.2000000D 00	0.8000000D 00	0.1277503D 01

0.1600000D 00	0.1000000D 00	0.1849404D 01
0.1600000D 00	0.2000000D 00	0.1741172D 01
0.1600000D 00	0.3000000D 00	0.1636952D 01
0.1600000D 00	0.4000000D 00	0.1551762D 01
0.1600000D 00	0.5000000D 00	0.1485454D 01
0.1600000D 00	0.6000000D 00	0.1434256D 01
0.1600000D 00	0.7000000D 00	0.1394378D 01
0.1600000D 00	0.8000000D 00	0.1362872D 01
0.1200000D 00	0.1000000D 00	0.1970886D 01
0.1200000D 00	0.2000000D 00	0.1870550D 01
0.1200000D 00	0.3000000D 00	0.1765856D 01
0.1200000D 00	0.4000000D 00	0.1674480D 01
0.1200000D 00	0.5000000D 00	0.1599834D 01
0.1200000D 00	0.6000000D 00	0.1540048D 01
0.1200000D 00	0.7000000D 00	0.1492156D 01
0.1200000D 00	0.8000000D 00	0.1453465D 01
0.8000000D-01	0.1000000D 00	0.2088237D 01
0.8000000D-01	0.2000000D 00	0.1997643D 01
0.8000000D-01	0.3000000D 00	0.1895569D 01
0.8000000D-01	0.4000000D 00	0.1800782D 01
0.8000000D-01	0.5000000D 00	0.1719585D 01
0.8000000D-01	0.6000000D 00	0.1652185D 01
0.8000000D-01	0.7000000D 00	0.1596705D 01
0.8000000D-01	0.8000000D 00	0.1550928D 01

0.4000000D-01	0.1000000D 00	0.2204289D 01
0.4000000D-01	0.2000000D 00	0.2125541D 01
0.4000000D-01	0.3000000D 00	0.2029397D 01
0.4000000D-01	0.4000000D 00	0.1934192D 01
0.4000000D-01	0.5000000D 00	0.1848482D 01
0.4000000D-01	0.6000000D 00	0.1774603D 01
0.4000000D-01	0.7000000D 00	0.1712022D 01
0.4000000D-01	0.8000000D 00	0.1659231D 01
0.0	0.1000000D 00	0.4531139D-01
0.0	0.2000000D 00	0.1624960D 00
0.0	0.3000000D 00	0.3118490D 00
0.0	0.4000000D 00	0.4597449D 00
0.0	0.5000000D 00	0.5890481D 00
0.0	0.6000000D 00	0.6952699D 00
0.0	0.7000000D 00	0.7800908D 00
0.0	0.8000000D 00	0.8471703D 00
-0.4000000D-01	0.1000000D 00	0.1680226D-01
-0.4000000D-01	0.2000000D 00	0.6295332D-01
-0.4000000D-01	0.3000000D 00	0.1283782D 00
-0.4000000D-01	0.4000000D 00	0.2023793D 00
-0.4000000D-01	0.5000000D 00	0.2770011D 00
-0.4000000D-01	0.6000000D 00	0.3476276D 00
-0.4000000D-01	0.7000000D 00	0.4121711D 00
-0.4000000D-01	0.8000000D 00	0.4700692D 00

-0.8000000D-01	0.1000000D 00	0.1359383D-01
-0.8000000D-01	0.2000000D 00	0.5144832D-01
-0.8000000D-01	0.3000000D 00	0.1063996D 00
-0.8000000D-01	0.4000000D 00	0.1703532D 00
-0.8000000D-01	0.5000000D 00	0.2367458D 00
-0.8000000D-01	0.6000000D 00	0.3013095D 00
-0.8000000D-01	0.7000000D 00	0.3617491D 00
-0.8000000D-01	0.8000000D 00	0.4171083D 00
-0.1200000D 00	0.1000000D 00	0.1135018D-01
-0.1200000D 00	0.2000000D 00	0.4328351D-01
-0.1200000D 00	0.3000000D 00	0.9048288D-01
-0.1200000D 00	0.4000000D 00	0.1466447D 00
-0.1200000D 00	0.5000000D 00	0.2063089D 00
-0.1200000D 00	0.6000000D 00	0.2656157D 00
-0.1200000D 00	0.7000000D 00	0.3222447D 00
-0.1200000D 00	0.8000000D 00	0.3750238D 00
-0.1600000D 00	0.1000000D 00	0.9668576D-02
-0.1600000D 00	0.2000000D 00	0.3709245D-01
-0.1600000D 00	0.3000000D 00	0.7821542D-01
-0.1600000D 00	0.4000000D 00	0.1280378D 00
-0.1600000D 00	0.5000000D 00	0.1819917D 00
-0.1600000D 00	0.6000000D 00	0.2366283D 00
-0.1600000D 00	0.7000000D 00	0.2896957D 00
-0.1600000D 00	0.8000000D 00	0.3399124D 00

-0.2000000D 00	0.1000000D 00	0.8358339D-02
-0.2000000D 00	0.2000000D 00	0.3222259D-01
-0.2000000D 00	0.3000000D 00	0.6843412D-01
-0.2000000D 00	0.4000000D 00	0.1129718D 00
-0.2000000D 00	0.5000000D 00	0.1619953D 00
-0.2000000D 00	0.6000000D 00	0.2124440D 00
-0.2000000D 00	0.7000000D 00	0.2621847D 00
-0.2000000D 00	0.8000000D 00	0.3098955D 00
-0.2400000D 00	0.1000000D 00	0.7310148D-02
-0.2400000D 00	0.2000000D 00	0.2829575D-01
-0.2400000D 00	0.3000000D 00	0.6045593D-01
-0.2400000D 00	0.4000000D 00	0.1005190D 00
-0.2400000D 00	0.5000000D 00	0.1452413D 00
-0.2400000D 00	0.6000000D 00	0.1919171D 00
-0.2400000D 00	0.7000000D 00	0.2385568D 00
-0.2400000D 00	0.8000000D 00	0.2838431D 00

APPENDIX B

EXACT SOLUTIONS FOR SQUARE PULSES, AND FOR THE GENERAL PULSES WITH $\delta=0$ AND $\gamma_2=0$

Eqs.(39) for square pulses can be written as

$$\frac{dA_0}{dt} = i\alpha A_2 \exp\,[i(2\delta-\mu)t],$$

$$\frac{dA_2}{dt} = -\gamma A_2 + i\alpha^* A_0 \exp\,[-i(2\delta-\mu)t]. \tag{90}$$

where $\gamma = \frac{\gamma_2}{2} + P_o$.

With the boundary conditions $A_0(0) = 1$ and $A_2(0) = 0$ we solve the second order differential equation and find the exact solutions

$$A_2 = \frac{i\alpha^*}{m_1-m_2}\,(e^{m_1 t} - e^{m_2 t}),$$

$$A_0 = \frac{e^{i(2\delta-\mu)t}}{m_1-m_2}\,[(m_1+\gamma)e^{m_1 t} - (m_2+\gamma)e^{m_2 t}], \tag{91}$$

where

$$m_{\substack{1\\2}} = \frac{-[\gamma+i(2\delta-\mu)] \pm i\sqrt{4|\alpha|^2 - [\gamma-i(2\delta-\mu)]^2}}{2},$$

and we choose $\sqrt{}$ such that its real value is positive.

If $\gamma << |\alpha|$, then we can make an approximation and find

$$|A_2(t)|^2 = \frac{2|\alpha|^2 e^{-\gamma t}}{(2\delta-\mu)^2+4|\alpha|^2} \left[\cosh \frac{\gamma(2\delta-\mu)t}{\sqrt{4|\alpha|^2+(2\delta-\mu)^2}} \right. \tag{92}$$

$$\left. - \cos \sqrt{4|\alpha|^2+(2\delta-\mu)^2}\, t\right] .$$

If $\frac{\gamma(2\delta-\mu)\tau}{\sqrt{4|\alpha|^2+(2\delta-\mu)^2}} << 1$, $(0 \le t \le \tau)$, then $\cosh \frac{\gamma(2\delta-\mu)t}{\sqrt{4|\alpha|^2+(2\delta-\mu)^2}}$ $\simeq 1$ dropping 2nd order smallness.

In the case of the pulse shape $g(t)$ with $\gamma_2 = 0$ and $\delta = 0$ and with the substitution $U = \int_{-\infty}^{t} g(t')dt'$ Eq. (39) becomes

$$\frac{dA_0}{dU} = i\alpha\, A_2 \exp(-i\mu U),$$

$$\frac{dA_2}{dU} = - P_0 A_2 + i\alpha^* A_0 \exp(i\mu U), \tag{93}$$

which is the same type as Eqs. (90).

REFERENCES

1. Vasilenko, L.S., V.P. Chebotaev and A.V. Shishaev, JETP Letters 12, 113 (1970).

2. Cagnac, B., G. Grynberg and F. Biraben, Phys. Rev. Lett. 32, 643 (1974).

3. Levenson, M.D. and N. Bloembergen, Phys. Rev. Lett. 32, 645 (1974).

4. Hänsch, T.W., S.A. Lee, R. Wallenstein and C. Wieman, Phys. Rev. Lett. 34, 307 (1975).

5. Hänsch, T.W., K. Harvey, G. Meisel and A.L. Schawlow, Opt. Comm. 11, 50 (1974).

6. Biraben, F., B. Cagnac and G. Grynberg, C. R. Acad. Sc. Paris 279, B51 (1974).

7. Liao, P.F. and J.E. Bjorkholm, Phys. Rev. Lett. 34, 1 (1975).

8. Bjorkholm, J.E. and P.F. Liao, Phys. Rev. Lett. 33 128 (1974).

9. Patel, C.K.N., Laser Spectroscopy, ed. R.G. Brewer and A. Mooradian (Plenum Press, N.Y., 1974).

10. Ambartzumian, R.V., V.S. Letokhov, N.V. Chekalin, E.A. Ryabov Lett to JET Ph. 20, 597 (1974).

11. Ambartzumian, R.V., Yu. A. Gorokhov, V.S. Letokhov, and G.N. Makarov Lett to JET Ph. 21, 375 (1975).

12. Lyman, J.L., R.J. Jensen, J. Rink, C.P. Robinson, and S.D. Rockwood, Preprint LA-UR 75707.

13. Ambartzumian, R.V., V.S. Doljikov, N.V. Chekalin, V.S. Letokhov, E.A. Ryabov, Chem. Phys. Lett. 25, 515 (1974), JET Ph. 68, N7 (1975).

14. Steinfeld, J. I., to appear in CRC Critical Reviews in Analytical Chemistry.

15. Fairbank, W.M., T.W. Hänsch, and A.L. Schawlow, J.O.S.A. 65, 199 (1975).

16. Hurst, G.S., M.G. Payne, M.H. Nayfeh, J.P. Judish, and E.B. Wagner, Phys. Rev. Lett. 35, 82 (1975), Payne, M.G., G.S. Hurst, M.H. Nayfeh, J.P. Judish, C.H. Chen, E.B. Wagner, and J.P. Young, Phys. Rev. Lett. 35, 1154 (1975); Hurst, G.S., M.H. Nayfeh, and J.P. Young, "A Demonstration of One-Atom Detection," Appl. Phys. Lett. (in press); Hurst, G.S., M.H. Nayfeh, and J.P. Young, "One-Atom Detection Using Resonance Ionization Spectroscopy," (to be published).

17. Hänsch, T. and P. Toschek, IEEE J. Quant. Electron QE-4, 467 (1968) and Z. Physik 236, 213 (1970).

18. Holt, H.K., Phys. Rev. Lett. 20, 410 (1968) and Phys. Lett. 19, 1275 (1967); Notkin, G.E., S.G. Rautian and A.A. Feoktistov, Zh. Eksp. Teor. Fiz. 52, 1673 (1967) [Sov. Phys. JETP 25, 1112 (1967)].

19. Brewer, R.G., Science 178, 247 (1972); see also Baklanov, E.V. and V.P. Chebotayev, Zh. Eksp. Teor. Fiz. 61, 922 (1972) [JETP 34, 490 (1972)].

20. Hall, J.L., Lectures in Theoretical Physics (Gordon and Breach, N.Y., 1973), p. 161.

21. Bebb, H.B. and A. Gold, Phys. Rev. 143, 1 (1966); Gontier, Y. and M. Trahin, Phys. Rev. A 7, 1599 (1972).

22. Lambropoulos, P., Phys. Rev. A 9, 1992 (1974).

23. Armstrong, L. and B.L. Beers, Phys. Rev. A 12, 1903 (1975).

24. Beers, B.L. and L. Armstrong, Phys. Rev. A 12, 2447 (1975); see also the references therein.

25. Zener, C., Proc. Roy. Soc. (London) A137, 696 (1932).

26. Payne, M.G., unpublished work.

27. Payne, M.G. and M.H. Nayfeh, Phys. Rev. A 13, 595 (1976).

28. Lau, A.M.F., Phys. Rev. A 14, 279 (1976).

29. Rabi, I.I., Phys. Rev. 49, 324 (1936); Rabi, I.I., Phys. Rev. 51, 652 (1937).

30. Tan-no, N., Y. Kenichi and H. Inaba, J. Phys. B: Atom. Molec. Phys. 8, 339 (1975); Tan-no, N., Y. Kenichi and H. Inaba, J. Phys. B: Atom. Molec. Phys. 8, 349 (1975).

31. Matsuoka, M., Optics Communications 15, 84 (1975).

32. Salomaa, R. and S. Stenholm, J. Phys. B: Atom. Molec. Phys. 8, 1795 (1975).

33. Yatisv, S., W.G. Wagner, G.S. Picus and F.J. McClung, Phys. Rev. Lett. 15, 614 (1965).

34. Grischkowsky, D. and M.M.T. Loy, Phys. Rev. A 12, 1117 (1975).

35. Ward, J.F. and A.V. Smith, Phys. Rev. Lett. 35, 653 (1975).

36. Takatsuji, M., Phys. Rev. A 4, 808 (1971).

37. Semibalomut, V.M. and R.I. Sokolovskii, Opt. Spectrosc.. 38, 618 (1975).

THIS PAGE

WAS INTENTIONALLY

LEFT BLANK

ORNL/TM-5754

INTERNAL DISTRIBUTION

1. Biology Library
2-3. Central Research Library
4. Reactor Division Library
5-6. Laboratory Records Dept. (LRD)
7. Laboratory Records (ORNL-RC)
8. ORNL Patent Office
9. ORNL Y-12 Technical Library, Document Reference Section
10. B. K. Annis
11. J. A. Auxier
12. C. F. Barnett
13. C. E. Bemis
14. R. D. Birkhoff
15. T. A. Carlson
16. C. H. Chen
17-21. C. W. Choi
22. L. G. Christophorou
23. R. N. Compton
24. S. Datz
25. J. S. Faulkner
26. W. R. Garrett
27. L. W. Grossman
28-32. G. S. Hurst
33. J. P. Judish
34. C. E. Klots
35. S. D. Kramer
36. C. H. Krause
37. M. H. Nayfeh
38-42. M. G. Payne
43. H. Postma
44. C. R. Richmond
45. R. H. Ritchie
46. H. C. Schweinler
47. J. E. Turner
48. T. A. Welton
49. J. P. Young

EXTERNAL DISTRIBUTION

50. Lloyd Armstrong, Jr., Department of Physics, Johns Hopkins University, Baltimore, Maryland 21218
51. B. L. Beers, Department of Physics, Johns Hopkins University, Baltimore, Maryland 21218
52. J. E. Bjorkholm, Bell Telephone Laboratories, Holmdel, N.J. 07733
53. N. Bloembergen, Department of Engineering and Applied Physics, Harvard University, Cambridge, Massachusetts 02138
54. J. E. Boring, Research Laboratory of Engineering Science, University of Virginia, Charlottesville, Virginia 22901
55. Keith Boyer, Laser Research and Technology Division, LASL, Los Alamos, N.M. 87545
56. W. Brandt, Physics Department, New York University, New York, N.Y. 10003
57. R. S. Caswell, Radiation Physics Laboratory, NBS, Washington, D.C. 20234
58. L. W. Cochran, Vice President, University of Kentucky, Lexington, Kentucky 40506
59. G. Cowper, Radiation Dosimetry Branch, Atomic Energy of Canada Limited, Chalk River, Ontario, Canada
60. W. C. DeMarcus, Department of Physics and Astronomy, University of Kentucky, Lexington, Kentucky 40506

61. James Dubrin, Lawrence Livermore Laboratory, P. O. Box 808, Livermore, California 94550
62-63. F. Gabbard, Department of Physics and Astronomy, University of Kentucky, Lexington, Kentucky 40506
64. S. Geltman, Joint Institute for Laboratory Astrophysics, University of Colorado, Boulder, Colorado 80309
65. T. F. George, Department of Chemistry, University of Rochester, Rochester, N.Y. 14627
66. B. R. Gossick, Department of Physics and Astronomy, University of Kentucky, Lexington, Kentucky 40506
67. D. Grischkowsky, IBM Thomas J. Watson Research Center, Yorktown Heights, N.Y. 10598
68. J. K. Hancock, Advanced Isotopes Separations Program, ERDA, Washington, D.C. 20545
69. T. W. Hansch, Department of Physics, Stanford University, Stanford, California 94305
70. S. E. Harris, Edward L. Ginzton Laboratory, W. W. Hansen Laboratories of Physics, Stanford University, Stanford, California 94305
71. M. Inokuti, Radiation Physics Division, Argonne National Laboratory, 9700 South Cass Avenue, Argonne, Illinois 60439
72. O'Dean Judd, Laser Research and Technology Division, Los Alamos Scientific Laboratory, P. O. Box 1663, Los Alamos, N.M. 87545
73. R. E. Knight, Department of Physics and Astronomy, University of Kentucky, Lexington, Kentucky 40506
74. P. P. Lambropoulos, Department of Physics, University of Southern California, University Park, Los Angeles, California 90007
75. A. M. F. Lau, Molecular Physics Center, Stanford Research Institute, Menlo Park, California 94025
76. G. W. Lehman, Department of Physics and Astronomy, University of Kentucky, Lexington, Kentucky 40506
77. P. K. Leichner, Department of Physics and Astronomy, University of Kentucky, Lexington, Kentucky 40506
78. P. F. Liao, Bell Telephone Laboratories, Holmdel, N.J. 07733
79. W. Lichten, Department of Physics and Engineering and Applied Science, Yale University, 15 Prospect Street, New Haven, Connecticut 06520
80. J. L. Liverman, Assistant Administrator for Environment and Safety, ERDA, Washington, D.C. 20545
81. D. C. Lorents, Stanford Research Institute, Menlo Park, California 94025
82. M. M. T. Loy, IBM Thomas J. Watson Research Center, Yorktown Heights, N.Y. 10598
83. M. T. McEllistrem, Department of Physics and Astronomy, University of Kentucky, Lexington, Kentucky 40506
84. J. E. Parks, Department of Physics and Astronomy, Western Kentucky University, Bowling Green, Kentucky 42101
85. A. V. Phelps, Joint Institute for Laboratory Astrophysics, University of Colorado, Boulder, Colorado 80302

86. C. P. Robinson, Applied Photochemistry Division, LASL, P. O. Box 1663, Los Alamos, N.M. 87545
87. W. C. Royster, Dean of the Graduate School, University of Kentucky, Lexington, Kentucky 40506
88. R. L. Schriever, Division of Laser Fusion, ERDA, Washington, D.C. 20545
89. R. Schrils, Department of Physics and Astronomy, University of Kentucky, Lexington, Kentucky 40506
90. Maxine Sheets, Division of Biomedical and Environmental Research, ERDA, Washington, D.C. 20545
91. R. F. Stebbings, Department of Space Physics, Box 1892, Rice University, Houston, Texas 77001
92. T. D. Strickler, Department of Physics, Berea College, Berea Kentucky 40403
93. J. E. Talmage, 114 Nasson Lane, Oak Ridge, Tennessee 37830
94. G. K. Walters, Department of Physics, Rice University, Houston, Texas 77001
95. W. H. Wing, Department of Physics, University of Arizona, Tucson, Arizona 85721
96. R. W. Wood, Division of Biomedical and Environmental Research, ERDA, Washington, D.C. 20545
97. ORAU Library

98-99. ORAU Fellowship Office

100. Research and Technical Support Division, ORO

101-127. Technical Information Center (TIC)