Modulated Exponential Decay in a Two-Level Quantum System*

O. A. Keller and R. T. Robiscoe^{\dagger}

Harrison M. Randall Laboratory, The University of Michigan, Ann Arbor, Michigan 48104

(Received 27 December 1968; revised manuscript received 7 July 1969)

We carry out a Weisskopf-Wigner-type calculation for a system of two quantum levels which are simultaneously coupled by a quantized radiation interaction H and a classical external perturbation V. The results for the level populations properly reduce to the quantum oscillation solution when $H \rightarrow 0$, and to the exponential-decay solution when $V \rightarrow 0$. When neither H nor V is zero, the upper level undergoes a modulated exponential decay. We derive an expression for the perturbed decay rate and discover a new type of modulation factor. The result suggests possibly important modifications to the standard, phenomenological solution of the three-level H + V problem, which is of practical interest.

I. INTE JDUCTION

Experiments¹⁻³ on the system of states 2s, 2p, and 1s in atomic hydrogen reveal many interesting features of, and provide a number of important tests for, the time-dependent perturbation theory of atomic transitions. As indicated in Fig. 1, the transitions occur under the combined effects of an external perturbation V and the radiation interaction H. The metastable 2s state can be coupled to the 2p state via a Stark matrix element |V|, while 2p is coupled to 1s via the radiation coupling |H|, which gives rise to the 2p spontaneous decay rate $\gamma \propto |H|^2$. If the 2p state did not decay (i.e., $\gamma = 0$), then the 2s - 2p coupling by |V| would result in the well-known "quantum oscillation" between these states, at (angular) frequency δ , and with transition amplitude $\propto |V|^2$.⁴ As it is, with $\gamma \neq 0$, one expects in general some sort of quantum oscillation superimposed on an exponential decay.

A phenomenological theory describing transitions within this system due to the coupling (H + V)has been given by Lamb, ⁵ as an extension of earlier work by Bethe. ⁶ Lamb used the theory, under the condition $|V| \ll (\frac{1}{2}\hbar\gamma)$, to derive line-



FIG. 1. Three-level system in hydrogen. The 2s and 2p levels (energy separation $\hbar \delta$) can be coupled by an external (Stark) perturbation V. The 2s level is metastable, but the 2p level decays spontaneously to the 1s ground state, emitting a photon of energy $\hbar \omega_0$. The decay is due to the 2p-1s coupling by the radiation interaction H, which results in the 2p spontaneous decay rate $\gamma \propto H^2$. shapes for the $2s \rightarrow 2p$ quenching resonances⁷ observed to high accuracy during measurements on hydrogen fine structure.¹ The same theory has been used by Wangsness,⁸ under the condition $|V| \gg (\frac{1}{2} \hbar \gamma)$, to account for the temporal intensity variations in various spectral lines of hydrogen observed during beam-foil experiments.^{3,9} Wangsness concludes that for sufficiently large |V|, the $2p \rightarrow 1s$ exponential decay will be modulated by the 2s-2p quantum oscillation. Series has also used the theory to propose a novel method for measuring the Lamb shift.¹⁰

The theory of these transition processes is phenomenological in that the 2p-1s coupling by *H* is accounted for by simply adding a "damping term," proportional to γ , to the ordinary equations of time-dependent perturbation theory which describe the coupling of the (stationary) states by *V*. Thus, for the problem in Fig. 1, the equations for the 2p and 2s state amplitudes α and α are taken to be

$$i\hbar \dot{a} = V_{ps} \otimes \exp(-i\delta t) - \frac{1}{2}i\hbar\gamma \otimes ,$$

$$i\hbar \otimes = V_{ps}^* \otimes \exp(+i\delta t) .$$
(1)

Here, $V_{ps} = \langle 2p | V | 2s \rangle$, and the relatively small 2s-state decay rate is ignored. In neglecting to include the 1s state in this description, it is implicitly assumed that the ground state is sufficiently far away ($\omega_0 \gg \delta$) so as to be only weakly coupled by V. The ground state thus serves only as a sort of reservoir into which the 2p decays are dumped. However, it is generally true that the same V which couples 2p to 2s will also couple 2p to 1s. The effect of this additional coupling can become important for $|V| \sim (\frac{1}{2} \pi \gamma)$, or larger, as in the beam-foil experiments.³

The phenomenological Eqs. (1) can presumably be justified by writing equations of the Weisskopf-Wigner (WW) type.¹¹ However, if we wish to in-

clude the ground state, so as to estimate the effects of the additional 2p-1s coupling by V, as well as to insure that probability is conserved for times such that $\gamma t \gg 1$, it is necessary to carry out a full WW calculation, including a quantized radiation interaction, sums over photons, etc.¹² We expect to find that for sufficiently large |V|, the 2p - 1s exponential decay will be modulated by both the 2p-2s and 2p-1s quantum oscillations. As a first step in understanding this additional modulation, we are led to consider the twolevel (H + V) problem indicated in Fig. 2. The p and s states here resemble the 2p and 1s states of the three-level (H + V) problem in Fig. 1; note, however, that now these states are simultaneously coupled by H and V, a feature which is missing in the problem of Fig. 1.

In this paper, we shall solve the two-level problem of Fig. 2, when neither H nor V is zero. The solution of this problem must reduce to the quantum oscillation (QO) solution⁴ when $H \rightarrow 0$, and to the WW solution¹¹ when $V \rightarrow 0$; these separate solutions are well-known exercises in the elementary quantum mechanics of a two-level system. We shall solve the two-level (H + V) problem by carrying out a WW-type calculation in which the external perturbation V is treated classically, and the radiation interaction H is quantized (as in the or-



FIG. 2. Hypothetical two-level system. The p and s levels here (energy separation $\hbar \omega_0$) are analogous to the 2p and 1s levels in Fig. 1. However, now they are simultaneously coupled by the external perturbation V and radiation interaction H. Separately, V gives a quantum oscillation between p and s, while H gives an exponential decay of the upper state, at decay rate γ .

dinary WW theory). We do this not only as a pedagogical extension of the elementary quantummechanical solutions, but also as a first step in understanding the modulated exponential decay of the three-level problem, which is of practical interest. The calculation is straightforward, but rather involved. The results show a new type of modulation factor, which suggests that important modifications to the standard, phenomenological solutions of the three-level problem may result when it is treated by a complete WW calculation.

II. ELEMENTARY SOLUTIONS

In this section, we review the standard solutions to the elementary two-level problems, namely, (A) H=0 but $V \neq 0$, which gives the (QO) solution, and (B) $H \neq 0$, but V=0, which - by means of the WW calculation - gives the WW solution, i.e., an exponential decay of the upper state. In both cases, we assume that an interaction U, either V or H, effects a superposition of states:

$$\Psi(x,t) = A(t)\psi_p(x)\exp(-iE_pt/\hbar) + B(t)\psi_s(x)\exp(-iE_st/\hbar) , \qquad (2)$$

where the $\psi_n(x)$ are orthonormal eigenfunctions of the Hamiltonian \mathcal{K} which binds the two-level system, $\mathcal{K}\psi_n = E_n\psi_n$, and the time-dependent amplitudes A and B are to be found from the Schrödinger equation, $i\hbar(\partial\Psi/\partial t) = (\mathcal{K}+U)\Psi$. In both cases, we shall choose the initial conditions: A = 1, B = 0, at t = 0 (i.e., the system is initially in the upper state). The notation defined here will be used later in solving the combined (H + V) problem.

A. QO Solution

For the case where the radiation interaction H=0, but the unquantized, external perturbation $V \neq 0$, the amplitude equations are

$$i\hbar\dot{A} = V_{ps}B\exp(+i\omega_0 t), \qquad i\hbar\dot{B} = V_{ps}^*A\exp(-i\omega_0 t), \qquad (3)$$

where $\omega_0 \equiv (E_p - E_s)/\hbar$ and $V_{pp} \equiv V_{ss} \equiv 0$. We write $V_{ps} \equiv \langle \psi_p | V | \psi_s \rangle \equiv V \exp(-i\varphi)$,

where V is real and time-independent (by assumption), and φ is an arbitrary phase. It is convenient to define the dimensionless quantities

188

$$Q = \left[1 + (2V/\hbar\omega_0)^2\right]^{1/2}, \qquad N = \left[2Q/(Q+1)\right]^{1/2} = (1+\mu^2)^{1/2}, \qquad (4)$$

where $\mu = [(Q-1)/(Q+1)]^{1/2} \simeq V/\hbar \omega_0$, as $V \to 0$.

The parameter μ measures the strength of V relative to the binding interaction.

1. Exact Solution in Terms of the Eigenfunctions of *H*

The solutions to Eq. (3), subject to the initial conditions A = 1, B = 0, at t = 0 are exactly

$$A(t)\exp(-iE_{p}t/\hbar) = (1/N^{2})[e^{-iPt} + \mu^{2}e^{-iSt}], \qquad B(t)\exp(-iE_{s}t/\hbar) = (\mu e^{i\varphi}/N^{2})[e^{-iPt} - e^{-iSt}], \tag{5}$$

where P and S are the perturbed eigenfrequencies,

$$P = (E_{p} + \Delta)/\hbar , \quad S = (E_{s} - \Delta)/\hbar , \quad \text{and} \quad \Delta = \frac{1}{2}(Q - 1)\hbar \omega_{0} \simeq V^{2}/(E_{p} - E_{s}), \quad \text{as } V \to 0.$$
(6)

The state populations, as functions of time, are therefore exactly

$$\mathcal{O}(t) = |A(t)|^2 = 1 - (4\mu^2/N^2)\sin^2(\frac{1}{2}\omega't) ,$$

$$s(t) = |B(t)|^2 = (4\mu^2/N^2)\sin^2(\frac{1}{2}\omega't) ,$$
(7)

where $\omega' = P - S = Q\omega_0$ is the perturbed frequency difference. Probability is conserved, since $\mathscr{O} + \$ = 1$, for all time.

2. Exact Solution in Terms of the Eigenfunctions of $(\mathcal{K} + V)$

In addition to the above solution, it will be useful later to have the solution expressed in terms of the eigenfunctions of $(\mathcal{K}+V)$. We diagonalize the problem $(\mathcal{K}+V)\phi_n = E'_n \phi_n$, where $E'_n \equiv E_n \pm \Delta$ are the perturbed energies. The complete eigenfunctions are

$$\Phi_{p}(x,t) = \phi_{p}(x)e^{-iPt}, \quad \Phi_{s}(x,t) = \phi_{s}(x)e^{-iSt},$$
where $\phi_{p}(x) = \frac{1}{N}[\psi_{p}(x) + \mu e^{i\varphi}\psi_{s}(x)], \quad \phi_{s}(x) = \frac{1}{N}[e^{i\varphi}\psi_{s}(x) - \mu\psi_{p}(x)].$
(8)

It should be noted that in this diagonalization process initial conditions have not been specified, as they previously were. However, linear combinations of Φ_p and Φ_s can be formed which do correspond to the system being entirely in the p state or the s state at t=0:

$$\Phi_{1} = \frac{1}{N} (\Phi_{p} - \mu \Phi_{s}), \quad \text{for pure } \psi_{p} \text{ at } t = 0,$$

$$\Phi_{2} = \frac{1}{N} (\mu \Phi_{p} + \Phi_{s}), \quad \text{for pure } \psi_{s} \text{ at } t = 0.$$
(9)

The Φ_n , and the ϕ_n , are orthonormal if the ψ_n are normalized. Later, in solving the (H+V) problem, we shall use

$$\Phi(x,t) = a(t) \Phi_1(x,t) + b(t) \Phi_2(x,t) , \qquad (10)$$

as the general superposition of eigenstates of the V problem, which is then perturbed by $H \neq 0$.

3. Comparison of the Two Solutions

By substituting Eqs. (9) and (8) into Eq. (10), we can show that the superposition of Eq. (10) is identical to that of Eq. (2) if the ψ_n and Φ_n amplitudes are related by

$$A(t)\exp(-iE_{p}t/\hbar) = (1/N^{2})[(a+\mu b)e^{-iPt} + \mu(\mu a - b)e^{-iSt}], \qquad (11)$$

$$B(t) \exp(-iE_{s}t/\hbar) = (e^{i\varphi}/N^{2}) \left[\mu(a+\mu b)e^{-iPt} - (\mu a - b)e^{-iSt} \right].$$

Comparing this with Eq. (5) shows that the QO solution, with the system initially in the upper state, corresponds to the choice a = 1, b = 0 for all time.

B. WW Method

For the case where the unquantized, external perturbation V=0, but the quantized radiation interaction $H \neq 0$, the amplitude equations are¹³

$$i\hbar\dot{A} = \sum_{\vec{k},\lambda} H_{sp}^{*}(\vec{k},\lambda) B e^{+i(k_{0}-k)ct}, \quad i\hbar\dot{B} = H_{sp}(\vec{k},\lambda) A e^{-i(k_{0}-k)ct}, \quad (12)$$

where $k_0 = \omega_0/c$, $k = \omega/c$ = photon wave number, and

$$\sum_{\mathbf{k},\lambda} \simeq \sum_{\lambda} \int_{4\pi} d\Omega_{\mathbf{k}} \int_0^\infty dk \, w(k) \; .$$

The sum over photons (wave vector \vec{k} and polarization λ) involves an integration over solid angles as well as integration over the photon density of states, w(k), which is $vk^2/(2\pi)^3$ for photons quantized in a box of volume v. The sum occurs in the first amplitude equation, but not in the second, as the absorption $s \rightarrow p$ can result from a variety of photons in the radiation field, while the emission $p \rightarrow s$ gives rise to only a single photon.

The WW calculation proceeds by assuming that the upper state decays exponentially, so that the amplitude A may be given by $A(t) = \exp(-\frac{1}{2}\gamma_0 t)$, where γ_0 is a time-independent decay constant to be found. We write $\langle \psi_S | H | \psi_p \rangle \equiv H_{Sp} \equiv H \exp(+i\theta)$, where H is real and time-independent, and θ is an arbitrary phase. Putting this Ansatz into Eqs. (12) gives

$$B(t) = (He^{i\theta}/\hbar)z_0^{-1} [1 - e^{+iz_0t}], \qquad \gamma_0 = (2i/\hbar^2) \sum_{\vec{k}, \lambda} H^2 z_0^{-1} [e^{-iz_0t} - 1], \qquad (13)$$

where $z_0 \equiv (k - k_0) c + \frac{1}{2} i \gamma_0$.

The integral equation for γ_0 can be solved approximately by assuming: (i) γ_0 is small with respect to k_0c , i.e., the radiation interaction is weak compared to the binding energy; then γ_0 may be neglected in the integrand. (ii) H and w(k) vary slowly with k compared to the sharp resonance at k_0 exhibited by the exponential terms. Then H and w(k) may be evaluated at $k = k_0$ and taken outside the k integral. The result for the decay rate, for time such that $k_0ct \gg 1$, is

$$\gamma_0 \simeq (2\pi/\hbar^2 c) \sum_{\lambda} \int_{4\pi} d\Omega_{\vec{k}} H^2(\vec{k},\lambda) w(k) \Big|_{k=k_0}$$
(14)

The population of the lower state can be found by summing over all photons which can cause this state to be populated. Using the approximation (ii), but *not* neglecting γ_0 in the integrand, gives

$$\delta(t) = \sum_{\vec{k},\lambda} |B(t)|^2 \simeq \sum_{\lambda} \int_{4\pi} d\Omega_{\vec{k}} \left[H(\vec{k},\lambda)/\hbar \right]^2 w(k) \Big|_{k=k_0} \times \int_0^\infty dk |z_0^{-1}(1-e^{iz_0t})|^2 .$$

$$(15)$$

Carrying out the integration, again for $k_0 ct \gg 1$, gives the state populations

$$\mathbf{s}(t) = 1 - \exp(-\gamma_0 t) , \quad \mathcal{O}(t) = |A(t)|^2 = \exp(-\gamma_0 t) . \tag{16}$$

As in the QO case, probability is conserved, since $\mathcal{O} + \mathcal{S} = 1$, for all time.

For future reference, we note that the quantization of the radiation field is accounted for, without using direct product states, by associating¹³

 $\sum_{\mathbf{k},\lambda} e^{-ikct}$ with the absorption (s - p) matrix element, $H_{sp}^* = \langle \psi_p | H | \psi_s \rangle$,

86 and

$$e^{+ikct}$$
 with the emission $(p-s)$ matrix element, $H_{sp} = \langle \psi_s | H | \psi_p \rangle$.

These rules, the exponential Ansatz, the approximations (i) and (ii) given above, and the imposition of a time scale by $k_0 ct \gg 1$ comprise the WW method which we apply to the combined (H + V) problem in Sec. III.

III. TWO-LEVEL (H + V) PROBLEM

In this section, we solve the two-level (H + V)problem shown in Fig. 2, using the methods and notation outlined in Sec. II. We carry out a WWtype calculation, using the state superposition of Eq. (10). This superposition and the WW ansatz $a(t) = e^{-\Lambda t/2}$ stipulate that the system is in an eigenstate of $(\mathcal{K} + V)$ when H is turned on. As in the previous section, \mathcal{K} is the binding interaction, V is the external perturbation, and H is the radiation interaction. We later remove this arbitrary time-ordering by averaging over an assumed random phase between H and V. As it must, the solution for H = 0, and to the WW solution for V = 0. When neither H nor V vanishes, the solution shows a new type of modulation factor, in addition to the expected QO modulation of the exponential decay.^{3,8}

We make several approximations during the calculation. First, we consider single-photon processes only. Next, assuming both V and Hare weak compared to *K*, we carry terms of order $V^{2}H$, and no higher, in the amplitude equations. We also assume that the perturbed decay rate Γ is not much different than γ_0 , except for having a slight time-dependence. Then, just as in the H problem, the upper level decays approximately exponentially. We solve the problem for a time scale $\Gamma t \gg 1$ and "near resonance" for the photons involved in the $p \leftrightarrow s$ transitions. Finally, we pass to the "isolated atom," by letting the photon quantization volume $v \rightarrow \infty$. All of these approximations are reasonable in typical physical situations.

A. Differential Equations

Using the state superposition of Eq. (10) in the Schrödinger equation

$$i\hbar \frac{\partial \Phi}{\partial t} = \left[(\mathcal{K} + V) + \hat{H} \right] \Phi$$
(18)

gives the amplitude equations to be solved for the time-dependent coefficients a and b,

$$i\hbar \dot{a} = \langle \Phi_2 | \hat{H} | \Phi_1 \rangle^* b + \langle \Phi_1 | \hat{H} | \Phi_1 \rangle a ,$$

$$i\hbar \dot{b} = \langle \Phi_2 | \hat{H} | \Phi_1 \rangle a + \langle \Phi_2 | \hat{H} | \Phi_2 \rangle b .$$
(19)

Here, the caret over H indicates that the photon sums are to be included in accordance with Eq. (17). The matrix elements can be calculated in a straightforward manner, using Eqs. (8)

and (9), the notation of Sec. II, and the rules of Eq. (17). If we define $\delta \equiv \theta - \varphi$ as the phase difference between *H* and *V*, and keep terms of order V^2H and no higher, we obtain, for example,

$$N^{4} \langle \Phi_{2} | \hat{H} | \Phi_{1} \rangle \simeq e^{i\delta} H(e^{-i\omega't} + 2\mu^{2}) e^{+ikct} + 2\mu^{2} e^{-i\delta} \sum_{\vec{k}, \lambda} H(1 - \cos\omega't) e^{-ikct},$$

where $\mu \simeq V/\hbar\omega_0$, $N^4 \simeq 1 + 2\mu^2$, (20)

and $\omega' \equiv Q\omega_0 \simeq (1+2\mu^2)k_0c$.

We make the WW Ansatz by assuming that the amplitude of the "upper state" Φ_1 , can be represented by $a(t) = \exp(-\frac{1}{2}\Lambda t)$. The real part of Λ will be the perturbed decay rate and will approach γ_0 as $V \rightarrow 0$. Putting this Ansatz and the expressions for the matrix elements into the second of Eqs. (19) gives the equation to be solved for b(t). As a convenience in solving for b(t), we write

$$b(t) \equiv (1/N^4) b_0(t) f(t) ,$$

where $b_0(t) \equiv (He^{i\delta}/\hbar) z^{-1} (1 - e^{izt}) ,$ (21)
and $z \equiv (k - Qk_0) c + \frac{1}{2} i\Lambda .$

The function $b_0(t)$ is analogous to the unperturbed WW amplitude B(t) of Eq. (13); it is obtained by letting $\mu \rightarrow 0$ in the equation for b(t) and solving it with the assumption that z is independent of time. With this assumption, the equation for b(t) can be converted into an equation for f(t). After rewriting one term, we find

$$\dot{f} - [i\nu e^{i\delta}(1 - e^{-i\omega' t})e^{ikct}]f$$

$$\simeq [\exp(-\frac{1}{2}\Lambda t)/b_0(t)]\{\}, \qquad (22)$$

where $\nu \equiv \mu H/N^4 \hbar$.

The braces on the right-hand side contain complicated terms of order V, H, and V^2H . Fortunately, we can ignore the entire right-hand side for times such that $\gamma_0 t \gg 1$; thus we can solve Eq. (22)" exactly." Using f(t=0)=1, evaluating the resulting expression at resonance, $k = Qk_0$, and ignoring a term of order $\nu/\omega' \ll 1$, we get

$$f(t) \simeq \exp(-i\nu t e^{i0}) . \tag{23}$$

This, with Eq. (21), is the solution for b(t).

(17)

The next step is to substitute b(t) and the WW Ansatz into the first of Eqs. (19) to get an equation for Λ . Using the convenient notation

$$\tau \equiv \omega' t, \quad G \equiv \Lambda \tau / 2 \omega',$$

$$r \equiv \epsilon e^{i\delta}, \quad \text{where} \quad \epsilon \equiv \nu / \omega' = \mu H / N^4 \hbar \omega',$$

$$\gamma \equiv (Q^3 / N^8) \gamma_0 \simeq (1 + 2\mu^2) \gamma_0, \quad (24)$$

allowing Λ to be time-dependent, and evaluating the first two terms at resonance $(k = Qk_0)$, gives

$$\frac{dG}{d\tau} \simeq \frac{\gamma}{2\omega'} (1 + 2\mu^2 e^{-i\tau}) e^{-ir\tau} + 4\epsilon^2 \sin^2\left(\frac{\tau}{2}\right) \\ \times \left[\frac{\tau}{G} \left(e^G - 1\right)\right] e^{i(1-r)\tau} + ir(1 - e^{-i\omega't}) e^{ikct} \\ + i \sum_{\vec{k},\lambda} r^*(1 - e^{i\tau}) e^{-ikct} .$$
(25)

The solution for G will give the desired Λ , which is analogous to the WW decay rate in the unperturbed ($\mu = 0$) case. The approximations made in obtaining Eq. (25) are mainly that $\gamma_0 t \gg 1$, and that the various terms are evaluated near resonance, $k = Qk_0$.

B. Perturbed Decay Rate

The first two terms on the right-hand side of Eq. (25) are the dominant ones, as both exhibit factors which grow exponentially with time. We shall ignore the last two terms on the right-hand side which are small by comparison.¹⁴ Of the remaining two terms, the second is small by comparison with the first in the limit of an isolated atom, that is, when we let the photon quantization volume v become infinite. This can be seen with the help of Eq. (14), by which we estimate the value of the radiation interaction matrix element H to be $H^2 \sim \hbar^2 c \gamma_0 / v k_0^2$. Then the parameter ϵ defined in Eq. (24) vanishes when $v \to \infty$, as

$$\epsilon = |r| = \mu H / N^4 \hbar \omega' \sim (\mu / \omega_0^2) (c^3 \gamma_0 / v)^{1/2} \to 0.$$
 (26)

Therefore, we can consider the term in ϵ^2 in Eq. (25) to be a perturbation on the solution for G. We set G = F + g, where F and g satisfy

$$\frac{dF}{d\tau} = \frac{\gamma}{2\omega'} (1 + 2\mu^2 e^{-i\tau}) e^{-i\tau\tau} ,$$
$$\frac{dg}{d\tau} \simeq 4\epsilon^2 \sin^2\left(\frac{\tau}{2}\right) \left(\frac{\tau}{G} (e^G - 1)\right) e^{i(1 - \tau)\tau} . (27)$$

We can solve for F exactly. Imposing the initial condition, F = 0 at $\tau = 0$, gives

$$F(\tau) = \frac{\gamma}{2\omega'} \left[\frac{1 - e^{-ir\tau}}{ir} + 2\mu^2 \left(\frac{1 - e^{-i}(1 + r)\tau}{i(1 + r)} \right) \right]$$
$$r \xrightarrow{\sim}{\rightarrow} 0 \frac{\gamma\tau}{2\omega'} \left(1 + \frac{2\mu^2}{\tau} \left[\sin\tau - i(1 - \cos\tau) \right] \right)$$
$$\equiv F_a(\tau) \mu \xrightarrow{\sim}{\rightarrow} 0 \frac{\gamma\tau}{2\omega'} , \qquad (28)$$

where the useful approximate forms are for $r \rightarrow 0$ and $\mu \rightarrow 0$. To solve for the relatively small term g, we approximate $G \simeq F$ on the right-hand side of the g equation; in fact, we use the final approximate form of F. Imposing g=0 at $\tau=0$ gives

$$g(\tau) = -\epsilon^2 \alpha^2 \int_0^{\gamma \tau/2\omega'} (e^x - 1) e^{-\beta x} (1 - e^{i\alpha x})^2 dx ,$$
(29)

where $\alpha \equiv 2\omega'/\gamma > 1$, $\beta \equiv 2ie^{-i\delta}(\nu/\gamma)$.

After performing the indicated integrations, we use the facts that $\beta \rightarrow 0$ as $v \rightarrow \infty$ (isolated atom limit), and $\alpha > 1$, to obtain the approximate result

$$g(\tau) \simeq \epsilon^2 \alpha^2 (\beta^{-1} - e^{\tau/\alpha}) . \tag{30}$$

Combining the solutions for F and g give the approximate solution to Eq. (25)

$$G(\tau) \simeq F_{a}(\tau) - \frac{1}{2} (\epsilon \gamma / 2\omega') \tau^{2} i e^{i\delta} - \epsilon^{2} \alpha^{2} e^{\tau / \alpha}.$$
(31)

The second term on the right-hand side comes from expanding the exact solution for F; it vanishes in the limit $\epsilon \rightarrow 0$. The last term is the dominant exponential term in Eq. (30) for g, which also vanishes as $\epsilon \rightarrow 0$.

We have now obtained approximate solutions of the amplitude Eqs. (19). The solution for b(t) is, from Eqs. (21) and (23),

$$b(t) = (He^{i\delta}/N^{4}\hbar)z^{-1}(1-e^{izt})\exp(-i\nu t e^{i\delta}).$$
 (32)

The solution for a(t) is, from Eqs. (24) and (31),

$$\begin{aligned} a(t) &= \exp[-G(t)] \\ &= e^{(\Lambda_a/2)t} \exp(\frac{1}{4}\epsilon_\gamma \omega' t^2 i e^{i\delta} + \epsilon^2 \alpha^2 e^{(\gamma/2)t}) , \end{aligned}$$

here
$$\Lambda_a \equiv \gamma \left\{ 1 + 2\mu^2 \left[\left(\frac{\sin \omega' t}{\omega' t} \right) - i \left(\frac{1 - \cos \omega' t}{\omega' t} \right) \right] \right\},$$
 (33)

w

with
$$\mu \simeq V/\hbar\omega_0$$
, $\omega' \simeq (1+2\mu^2)\omega_0$

and $\gamma \simeq (1+2\mu^2)\gamma_0$.

 Λ_a is the desired expression for the (complex) perturbed WW decay rate of the "upper state" Φ_1 , in the superposition of Eq. (10). We can see this by noting that when $\epsilon \rightarrow 0$ (isolated atom limit), the Φ_1 amplitude becomes $a(t) \simeq \exp(-\frac{1}{2}\Lambda_a t)$. The approximations involved in these solutions are: (i) matrix elements are evaluated to order V^2H ; (ii) the time scale is fixed by $\gamma_0 t \gg 1$; (iii) only resonance photons occur, i.e., $\omega \simeq \omega' = Qk_0c$; and (iv) the parameter $\epsilon \ll 1$. We have yet to pass to the limit $\epsilon \propto (\sqrt{v})^{-1} \rightarrow 0$ and to average over the relative phase $\boldsymbol{\delta}$.

C. Average State Populations

We now return to Eqs. (11) which give the (ψ_D, ψ_S) state amplitudes A and B in terms of the (Φ_1, Φ_2) amplitudes a and b. By substituting the solutions for a and b into Eqs. (11), we shall derive expressions for the p and s state populations $\mathcal{O} = |A|^2$ and $\mathfrak{S} = |B|^2$. We shall average these expressions over the phase δ , pass to the isolated atom limit $\epsilon \rightarrow 0$, and sum over the photons which can cause the lower state to be populated. In this way, we shall derive expressions for the time dependence of the transitions $p \leftrightarrow s$ when neither the

$$\left|\tilde{b}\right|^{2} = \sum_{\vec{k}, \lambda} (H/N^{4}\hbar)^{2} \left| \frac{1-e^{izt}}{z} \right|^{2} \exp(2\nu t \sin\delta),$$

where $z \simeq (k - Qk_0)c + \frac{1}{2}i\Gamma$.

...

 $\epsilon \rightarrow 0$. The phase-dependent factors in the We approximate $\Lambda \simeq \Gamma$ in Eq. (21) cross term in Eqs. (34) are

$$\chi \propto e^{i\theta} \exp\left\{-\epsilon \omega' t \left[\left(\frac{1}{4} \gamma t - 1\right) \sin \delta + i \left(\frac{1}{4} \gamma t + 1\right) \cos \delta \right] \right\}.$$
(38)

Now we average each of the expressions in Eqs. (36)-(38) over the relative phase δ . We do this by operating with $(1/2\pi)\int d\delta \times$, where the integration extends from 0 to 2π . The phase averages in both Eqs. (36) and (37) involve integrals of the form

$$\frac{1}{2\pi} \int_0^{2\pi} \exp(C\sin\delta) d\delta = I_0(C), \tag{39}$$

where C is a constant $\propto \epsilon$, and I_0 is the modified Bessel function of order zero.¹⁵ Since $I_0(0) = 1$, we readily obtain, in the limit $\epsilon \rightarrow 0$,

$$|a|^{2}_{\text{av}} \equiv \lim_{\epsilon \to 0} \frac{1}{2\pi} \int_{0}^{2\pi} |a|^{2} d\delta \simeq \exp(-\Gamma t) \quad .$$

$$\tag{40}$$

Carrying out the phase average, and a photon sum in the same fashion as in Eqs. (15) and (16), we also get

$$|\tilde{b}|_{av}^{2} \simeq 1 - \exp(-\Gamma t) \simeq 1 - |a|_{av}^{2}$$
 (41)

radiation interaction H nor external perturbation V is zero.

Taking the absolute value of both sides in Eqs. (11) and summing over photons, we find (to order μ²)

$$N^{4}|A|^{2} \simeq (1 + 2\mu^{2}\cos\omega't)|a|^{2} + 2\mu^{2}(1 - \cos\omega't)|\tilde{b}|^{2} + 2\mu \operatorname{Re}\chi ,$$

$$N^{4}|B|^{2} \simeq (1 + 2\mu^{2}\cos\omega't)|\tilde{b}|^{2} + 2\mu^{2}(1 - \cos\omega't)|a|^{2} - 2\mu \operatorname{Re}\chi ,$$
where $x(t) = (1 - e^{i\omega't})a^{*}(t)\tilde{b}(t)$. (34)

The tilde over b indicates that the photon sum is to be done. The various parameters (i.e., μ , N, ω') are to be approximated to order V^2 , as in Eq. (20). We note that if probability is conserved in one representation, then it is conserved in the other, as, to order μ^2 .

$$|A|^{2} + |B|^{2} = |a|^{2} + |b|^{2} .$$
(35)

The probabilities are, from Eqs. (32) and (33),

~

$$|a|^{2} = e^{-\Gamma t} \exp(2\epsilon^{2} \alpha^{2} e^{(\gamma/2)t} - \frac{1}{2}\epsilon \gamma \omega' t^{2} \sin \delta),$$

where
$$\Gamma \equiv \operatorname{ReA}_{a} = \gamma \left(1 + 2\mu^{2} \frac{\sin \omega' t}{\omega' t} \right) ;$$
 (36)

for
$$z$$
; this is correct as

(37)

At this point, we have shown that probability is conserved in the context of this calculation. Finally, by integrating the right-hand side of Eq. (38), we find

$$\chi_{av} \propto -\frac{1}{2} i (\gamma t)^{1/2} \lim_{\epsilon \to 0} J_1[\epsilon \omega' t (\gamma t)^{1/2}] = 0 \quad , \tag{42}$$

where J_1 is the ordinary Bessel function of order one.

By putting the results given by Eqs. (40)-(42) into Eq. (34), using the definition (36) of Γ , and retaining terms of order μ^2 , we obtain the populations of the *p* and *s* states

$$\mathcal{O}(t) \equiv |A(t)|^2$$
, $S(t) \equiv |B(t)|^2 = 1 - \mathcal{O}(t)$

where
$$\varphi(t) \simeq [1-\mathfrak{Q}(t)] e^{-\gamma t} e^{-\rho \mu^2 \sin \omega' t} + \mathfrak{Q}(t) [1-e^{-\gamma t} e^{-\rho \mu^2 \sin \omega' t}],$$

and $\mathfrak{Q}(t) \equiv 4\mu^2 \sin^2 \frac{\omega' t}{2}, \quad \rho \equiv 4 \frac{\hbar \gamma_0/2}{\hbar \omega_0},$ (43)

with $\mu \simeq V/\hbar \omega_0$, $\omega' \simeq (1+2\mu^2)\omega_0$, and $\gamma \simeq (1+2\mu^2)\gamma_0$, as before. We call $\mathfrak{L}(t)$ a "QO function", as it appears in the QO solution of Eq. (7). The parameters μ and ρ measure, respectively, the strength of the external perturbation V and radiation interaction H with respect to the unperturbed binding energy $\hbar \omega_0$. Finally, ω' and γ are the perturbed counterparts of the *p*-*s* state separation ω_0 and the *p*-state decay rate γ_0 . We can easily see that when $H \rightarrow 0$, we get the QO solution of Eq. (7), namely, $\mathfrak{P} \simeq 1-\mathfrak{L}$, while, when $V \rightarrow 0$, we get the WW solution of Eq. (16), namely, $\mathfrak{P} \simeq \exp(-\gamma_0 t)$.

IV. CONCLUSIONS

The expressions we have derived for the state populations in the two-level (H+V) problem show the combined effects of a quantum oscillation superimposed on an exponential decay. The over-all dependence on time is indicated in Fig. 3. These solutions are valid so long as the effects of the coupling due to both the external perturbation V and the radiation interaction H are small compared to the level separation $\hbar\omega_0$, i.e., the parameters μ and ρ in Eq. (43) are small. Thus, our solution is *not* valid near a level crossing point. On the other hand, the solutions are valid for V either



FIG. 3. Average (fractional) level populations for the problem shown in Fig. 2. The functions \mathcal{O} and \mathcal{S} are derived in the text. At early times, the exponential decay of the upper level is modulated by two quantum oscillation factors. Later, the levels exhibit a characteristic quantum oscillation of amplitude $4\mu^2$, at (angular) frequency $\omega' \simeq (1+2\mu^2)\omega_0$.

large or small compared to $\frac{1}{2}(\hbar \gamma_0)$.

Our results for the two-level p-s system (Fig. 2) cannot be compared directly with previous results^{8, 10} for the three-level 2s-2p-1s system (Fig. 1). There are two reasons for this. First, the three-level calculations are done in the context of the phenomenological theory discussed in Sec. I, rather than the full WW formalism we have used. Second, in the three-level calculations, no allowance is made for any *pair* of levels to be simultaneously coupled by H and V. We can speculate that the second difference here might lead to different factors of order V^2 appearing in the results. For example, if the three-level problem were done by a WW-type calculation, including the 2*p*-1*s* quantum oscillation (at frequency ω_0) as well as the 2s-2p quantum oscillation (at frequency δ), one might expect to see QO functions $\mathfrak{Q}(t, \omega_0)$ as well as $\mathfrak{Q}(t, \delta)$ occurring as factors. On the other hand, the first difference here -adifference in method of calculation - might well lead to the appearance of entirely different terms of order $V^2\gamma$. This is indicated by the observation that in a phenomenological description such as Eqs. (1), an implicit choice has been made with regard to the relative phase of V and H (the latter being represented by its associated decay rate γ_0 only); consequently, the cross terms are likely to be different.

Series¹⁰ has solved Eqs. (1) for the three-level problem in Fig. 1. If we let the unperturbed 2p-2s separation be ω_0 rather than $-\delta$, so as to draw an analogy with the *p*-*s* problem in Fig. 2, Series's result for the population of the upper level may be quoted as¹⁶

$$+ 2\mu^{2}(1 - 2\rho \sin \omega' t) \exp(-\frac{1}{2}\gamma_{0}t), \qquad (44)$$

in the same notation, and to the same degree of approximation, as in Eq. (43). We observe that this expression properly reduces to the QO solution for $\gamma_0 \rightarrow 0$, and to the WW solution for $V \rightarrow 0$, just as in Eq. (43). In the intermediate case, however, it is clear that the terms in Eq. (43) are qualitatively different from those of Eq. (44), particularly in the appearance of the term in $\exp(-\rho\mu^2 \sin\omega' t)$. While this comparison cannot be valid for times such that $\gamma_0 t \gg 1$, ¹⁷ it does suggest a new type of modulation of the exponential decay of the upper level. This new modulation factor is of order $V^2\gamma$; presumably, it will persist when the three-level problem is done by a complete WW calculation. We note also that there is no "threshold condition"

*Work supported in part by the U.S. Atomic Energy Commission.

[†]Alfred P. Sloan Foundation Fellow. Present address: Department of Physics, Montana State University, Bozeman, Mont. 59715.

¹W. E. Lamb, Jr., and R. C. Retherford, Phys. Rev. <u>79</u>, 549 (1950); R. C. Retherford and W. E. Lamb, Jr., *ibid.* 81, 222 1951.

²R. T. Robiscoe, Phys. Rev. <u>138</u>, A22 (1965); <u>168</u>, 4 (1968); R. T. Robiscoe and B. L. Cosens, Phys. Rev. Letters 17, 69 (1966).

³S. Bashkin, W. S. Bickel, D. Fink, and R. K. Wangsness, Phys. Rev. Letters 15, 284 (1965).

⁴See, for example, L. I. Schiff, <u>Quantum Mechanics</u> (McGraw-Hill Book Co., New York, 1955), 2nd ed., Sec. 29.

⁵See Appendix II of the first paper in Ref. 1.

⁶H. A. Bethe, <u>Handbuch der Physik</u> (Julius Springer-Verlag, Berlin, 1933), 2nd ed., Vol. 24/1, pp. 452-460. An updated version of this work is available in H. A. Bethe and E. E. Salpeter, <u>Quantum Mechanics of</u> <u>One and Two Electrons Atoms</u> (Academic Press Inc., New York, 1957), 1st ed., Sec. 67.

⁷In addition to the work in Ref. 1, the theory of the so-called "sharp transitions" among magnetic substates of the 2s state is developed in W. E. Lamb, Jr., Phys. Rev. <u>85</u>, 259 (1952), Secs. 68-72.

⁸R. K. Wangsness, Phys. Rev. 149, 60 (1966).

⁹It should be remarked that the beam-foil experiments in Ref. 3 did not measure the effects of (H+V) on the $2s \rightarrow 2p \rightarrow 1s$ system, but rather on the (n'=3, 4, 5, ...) $\rightarrow (n=2)$ transitions in hydrogen. However, the results of Ref. 8 apply as well to $(n'=2) \rightarrow (n=1)$ transitions.

¹⁰G. W. Series, Phys. Rev. <u>136</u>, A684 (1964).

for this modulation to occur, as in the calculation by Wangsness.¹⁸

These remarks are speculative in lieu of a detailed treatment of the three-level (H + V) by the WW method. It is clear, however, that this problem should be investigated, since experiments on this system can be done to high accuracy – in measurements of WW (Lorentzian) line shapes for the transitions involved^{1,2} and of details of the modulated exponential decay.³ If the modulation characteristics of the two-level problem persist, they might well be detected in such experiments.

ACKNOWLEDGMENTS

The authors wish to thank Professor H. S. Mani and Professor G. L. Kane for several illuminating discussions. Professor W. L. Williams also provided some useful insights.

¹¹This is pointed out in Appendix II of the first paper in Ref. 1. For references to the original WW calculation, carried out with V=0, see E. Wigner and V. Weisskopf, Z. Physik <u>63</u>, 54 (1930); and G. Wentzel, <u>Handbuch der Physik</u> (Julius Springer-Verlag, Berlin, 1922). Set ed. Vel. 24 (1990).

1933), 2nd ed., Vol. 24/1, pp. 752-755.

¹²This problem is discussed in the work by G. Wentzel, cited in Ref. 11; see pp. 764-768.

¹³The original WW work is cited in Ref. 11. The calculation is discussed in detail in Sec. 18 by W. Heitler, in <u>Quantum Theory of Radiation</u> (Oxford University Press, Cambridge, 1954), 3rd ed., pp. 181-189; and by J. J. Sakurai, in <u>Advanced Quantum Mechanics</u> (Addison-Wesley Publishing Co., Inc., Reading, Mass., 1967), pp. 37-39 and p. 65.

¹⁴Contributions from these terms would also vanish in the phase averaging carried out in Sec. III C.

¹⁵See I. S. Gradshteyn and I. M. Ryzhik, <u>Table of Integrals, Series and Products</u> (Academic Press Inc., New York, 1966), p. 488; also <u>Handbook of Mathematical</u> <u>Functions</u>, edited by M. Abramovitz and I. A. Stegun (U.S. Department of Commerce, National Bureau of Standards, Government Printing Office, Washington, D.C., 1965) p. 375.

¹⁶See Eq. (8) of Ref. 10, which is an exact result. The approximation in Eq. (44) above is carried out for the *H* and *V* relative strength parameters ρ and $\mu \ll 1$.

¹⁷For the problem of Eq. (1), probability is not conserved for the amplitudes α and β , as $|\alpha|^2 + |\beta|^2 \sim \exp(-\gamma_0 t)$.

¹⁸See Ref. 8. Wangsness concludes that in the threelevel problem of Fig. 1, there will be no modulation of the 2p exponential decay unless $V > \frac{1}{2} (\hbar \gamma_0/2)$. No such condition is imposed in Eq. (43).