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Induced transition probabilities and energies for the strongly coupled two-level system

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An exact method of solution to the time-dependent wave equation for a system interacting with a sinusoidal field, which formally treats time and the phase of the field on an equal footing, is used to discuss (1) the importance of averaging the properties of a system over the phase of the applied field and (2) the nature and behavior of the characteristic exponents derived from the Floquet solution (with particular emphasis on the frequency-sweep experiment). The results for the Zeeman tuning experiment are used to resolve recent discrepancies in the literature which relate to the validity of Shirley's important result for the two-level average induced transition probability involving the derivative of the characteristic exponents with respect to the Zeeman splitting parameter ω_0. The explicit calculations included in this work are for the single-photon two-level problem. Important implications can be inferred from them with respect to both the importance of phase averaging and the usefulness of the characteristic exponents as a quantitative means of obtaining resonance frequency shifts and half-widths for multiphoton and multilevel problems.

I. INTRODUCTION

The probability of a transition occurring between two states of an atom or molecule is, in general, a function of time and the phase δ of the sinusoidal field inducing the transition. In most experiments (see for example papers by Shirley[1] and Gush and Gush[2]) the initial phase of the field seen by the atom or molecule is not well defined and then the phase-averaged transition probability is of interest. Finally, the long-time (steady-state) average of this latter result corresponds to the physically observed spectrum of the system of interest if the effects of the perturbing field occur over time intervals short relative to the important relaxation times involved in the system.[1,3] For weak fields the phase dependence of the transition probability is negligible. However, for strong applied fields this is not the case and the failure to recognize this point can and has led to difficulties associated with evaluating the relevant transition probabilities for intense fields (see Sec. II below). It is well known[1,4-6] that the characteristic exponents or dressed-atom energies associated with the solution of the time-dependent Schrödinger equation yield the physically important resonance shifts and half-widths for the two-level Zeeman tuning experiment without the necessity of computing the phase- and time-averaged transition probability. Apparently, little attention has been given to the analogous use of the characteristic exponents for the corresponding frequency-sweep experiment.

Recently,[7] an exact solution has been obtained for the time-dependent wave equation of an N-level system in a sinusoidal field of arbitrary strength, frequency, and phase. This approach exploits the time periodicity of the wave equation, and the solution over the initial period of the Hamiltonian, obtained by a matching power series technique,[8] is used in an iterative way to obtain the complete solution over arbitrary times and phases. The solution can also be recast[7] in Floquet form to permit the evaluation of the important time- and/or phase-averaged properties of the system on an equally efficient basis. Hence the approach is ideally suited for discussing the importance of phase averaging in the evaluation of transition probabilities and is used in this paper.

The main purpose of this work is to stress the importance of averaging the properties of a system over the phase of the applied sinusoidal field and to discuss the nature and behavior of the characteristic exponents derived from the Floquet solution with particular emphasis on the frequency sweep experiment. In addition, discrepancies recently raised by Ahmad[9] between his results for the average induced transition probability and characteristic exponents for the two-level Zeeman-tuning experiment and those obtained earlier by Shirley[1] are resolved. Difficulties in evaluating average transition probabilities, analogous to those encountered in Ref. 9, will also arise if some[10,11] of the other available methods are used for intense applied fields. While the calculations discussed in this paper are for the single-photon two-level Zeeman-tuning and frequency-sweep experiments, analogous problems arise in the evaluation of multi-photon and multilevel transition probabilities, as discussed briefly in Sec. III.

The Hamiltonian H describing the interaction of a spin-$\frac{1}{2}$ system with a static Zeeman field B_0 and a transverse sinusoidal magnetic field $2B\cos(\omega t+\delta)$ is given by[12,13]

$$H = H_0 - \gamma B_0 I_z - 2\gamma B \cos(\omega t+\delta) I_x , \tag{1}$$

where I_z and I_x are the z and x components of the

spin angular momentum I, γ is the gyromagnetic ratio, H_0 is the Hamiltonian describing the system in the absence of external fields, and ω and δ are the frequency and phase of the applied sinusoidal field. In the Schrödinger representation, the coupled differential equations which govern the time evolution of the spin-$\frac{1}{2}$ state amplitudes $a_j(t)$, $j=1,2$, are given in matrix form by

$$\left[i\frac{\delta}{\delta t} - \begin{pmatrix} E_1 & 0 \\ 0 & E_2 \end{pmatrix} + 2B\cos(\omega t+\delta)\begin{pmatrix} 0 & \mu_{12} \\ \mu_{12} & 0 \end{pmatrix}\right]\begin{pmatrix} a_1(t) \\ a_2(t) \end{pmatrix} = 0, \quad (2)$$

where $\mu_{12} = -\gamma\langle -\frac{1}{2}|I_x|\frac{1}{2}\rangle = -\gamma/2 = \mu_{21}$ and E_i, $i=1,2$, are the Zeeman split levels of the system, $E_1 = E_0 + \frac{1}{2}\gamma B_0$ and $E_2 = E_0 - \frac{1}{2}\gamma B_0$, where E_0 is the energy of the initially degenerate spin-$\frac{1}{2}$ states: $H_0|\pm\frac{1}{2}\rangle = E_0|\pm\frac{1}{2}\rangle$. It is relevant to note the analogy [7,10(b)] between this spin-$\frac{1}{2}$ problem and the corresponding problem concerning the interaction of two electric-dipole-connected levels with an applied electric sinusoidal field.

II. INDUCED TRANSITION PROBABILITIES

The exact state amplitudes for the spin-$\frac{1}{2}$ system can be written in Floquet form[7,14]

$$\underline{a}(t) = \underline{Z}(\omega t+\delta)\, e^{\underline{\Delta}(\omega t+\delta)}\, \underline{b}_0(\delta) \quad (3)$$

where the periodic matrix $\underline{Z}$, the real diagonal characteristic exponent matrix $\underline{\Delta}$ and the column vector $\underline{b}_0$, which contains the initial condition information, are defined and discussed in detail in Refs. 7 and 15. In order to discuss the problems associated with phase averaging and to relate the work of Ahmad[9] and others[10,11] to that of Shirley[1] we define the phase-dependent and the phase-averaged[3,7] steady-state induced transition probabilities $\bar{P}_{jj}(\delta)$ and $\bar{P}_{jj}$ respectively:

$$\bar{P}_{jj}(\delta) = \frac{\omega}{2\pi}\int_0^{2\pi/\omega}\sum_{k=1}^{2}|Z_{jk}(\omega t+\delta)|^2\,dt\,|b_k(\delta)|^2, \quad (4)$$

$$\bar{P}_{jj} = \frac{1}{\pi}\int_0^{\pi}\bar{P}_{jj}(\delta)\,d\delta. \quad (5)$$

To compare directly with the results in Ref. 9 the coupling parameter $s = -\gamma B/(2\omega_0)$ is introduced where $\omega_0 = E_2 - E_1 = -\gamma B_0$ represents the splitting of the energy levels caused by the applied static magnetic field. While it is not difficult, using the approach of Refs. 7 and 15, to evaluate $\underline{a}(t)$, $\bar{P}_{jj}(\delta)$, and $\bar{P}_{jj}$ as a function of ω_0 and ω, we will limit ourselves in this section to the case studied explicitly by Ahmad,[9] namely $\bar{P}_{jj}(\delta)$ (and $\bar{P}_{jj}$) for $\omega = \omega_0$. Figure 1 shows $\bar{P}_{22}(\delta)$, for $\delta = 0$, $\pi/4$, and $\pi/2$, together with $\bar{P}_{22}$ as a function of s; a brief discussion of the calculational methods can be found in Ref. 7, details are in Ref. 15. The oscillatory behavior of $\bar{P}_{22}(0)$, including the values of $s = 0$, 1.23–1.24 and 2.07 where $\bar{P}_{22}(0) = 0.5$, coincides precisely with Ahmad's[9] description of his computed average transition probability. Even though both Shirley[1] and Ahmad[9] start with the same sinusoidal perturbation, corresponding to $\delta = 0$ in Eq. (1), Shirley[1] does a further averaging over initial times which is equivalent to our phase averaging. This averaging, however, occurs as a formal step which makes Shirley's method of solution tractable and this phase-averaging step has apparently been overlooked or omitted in some of the more recent treatments[9–11] of the problem

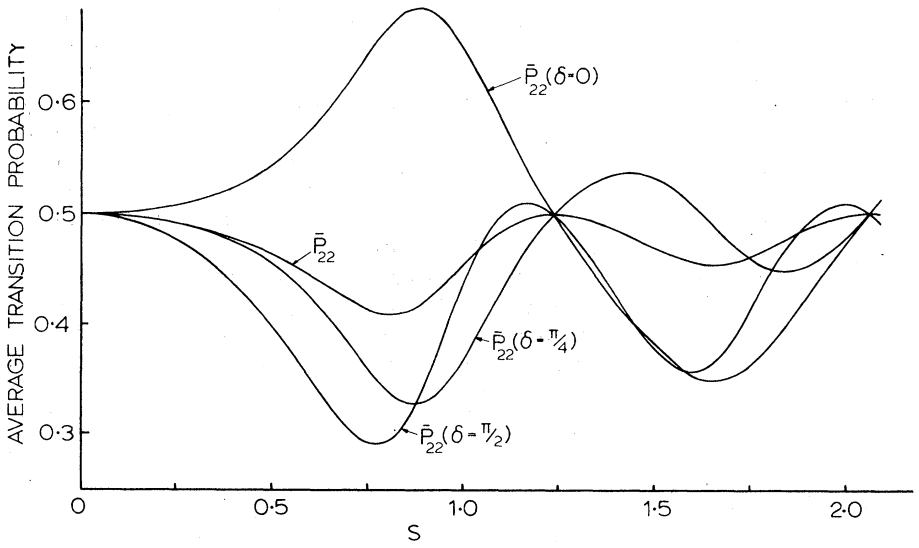

FIG. 1. Phase-averaged steady-state induced transition probability, $\bar{P}_{22}$, and the phase-dependent steady-state induced transition probability $\bar{P}_{22}(\delta)$, $\delta = 0$, $\pi/4$, $\pi/2$, as a function of $s = -\gamma B/(2\omega_0)$ for $\omega = \omega_0$. The initial condition corresponds to $a_1(0) = 1$.

(see however, for example, Refs. 2 and 16). The results in Fig. 1 for $\overline{P}_{22}(\pi/2)$ and $\overline{P}_{22}(\pi/4)$, together with $\overline{P}_{22}(0)$, show the strong phase dependence of the time-averaged induced transition probability and illustrates graphically how $\overline{P}_{22}(\delta)$ can average, as a function of δ, to $\overline{P}_{22} \leq 0.5$ in agreement with Shirley.[1] The points at which $\overline{P}_{22}$ equals 0.5, as a function of s, correspond to successive n-photon resonance peaks crossing the line $\omega = \omega_0$ in the frequency domain [$s \sim 0$, $n = 1$; $s \sim 1.23$–1.24, $n = 3$; $s \sim 2.07$, $n = 5$; and so on].

III. CHARACTERISTIC EXPONENTS

The behavior of the Floquet characteristic exponents in the vicinity of the single-photon transition will be analyzed briefly in this section. The implications of these results for higher photon transitions will become clear in the discussion that follows. It is a straightforward matter to show that the characteristic exponents satisfy the following relations[17]:

$$E_1 + E_2 = -\omega(\Delta_1 + \Delta_2 + m), \quad m = 0, \pm 1, \pm 2, \ldots \tag{6}$$

$$\Delta_j' = \Delta_j + n, \quad n = 0, \pm 1, \pm 2, \ldots . \tag{7}$$

The first relation connects the characteristic exponents with the energy reference[18] $E_r = -E_0 = -(E_1 + E_2)/2$, while the second equation follows from the definition of the characteristic exponents in terms of multibranched inverse trigonometric functions and indicates that there are infinitely many solutions for the Δ_j that are connected in a periodic manner.[1,7,15,19] The original interpretation of the plots of the characteristic exponents versus ω or ω_0 was made by Besset, Horowitz, Messiah, and Winter[20] in the rotating field approximation (which is not applicable for intense fields) and the usefulness of these plots, especially for the Zeeman-tuning experiment, has also been discussed by several authors.[1,4-6] The positions of the anticrossings in the plots correspond to the locations of the resonance maxima (and determine the half-widths of the resonance profiles) while in the presence of a transverse static magnetic field, "forbidden" transitions occur at the positions of the level crossings.

The behavior of the Δ_j's for the Zeeman tuning of spin-$\frac{1}{2}$ states has been discussed in detail in the literature,[1,4-6] although some confusion has arisen recently with regard to their physical significance[21] and, in particular, to their "singular" behavior in the transition region.[9] The latter point has cast some doubt on the validity of Shirley's derivative formula [Eq. (26) of Ref. 1] for the average induced transition probability due to an apparent discontinuity[9] in the characteristic exponents in this region. Our investigations show that the discontinuity, which is nonphysical, can be made to occur at random in the Zeeman-tuning domain by an artificial adjustment[22] in the energy reference E_r and can be removed from the exactly computed characteristic exponents by choosing $E_r = 0$ in agreement with Shirley.[1] In particular, there is no discontinuity at $\omega = \omega_0$ as implied[23] by Ahmad.[9] The physical significance of the characteristic exponents becomes obvious when $E_r = 0$ as the quantities $\omega\Delta_j$ can be shown to coincide precisely with the "dressed-atom" energies numerically computed by Yabusaki, Murakami, and Ogawa.[5] This coincidence is not surprising when one considers that an intense classical field corresponds to a high photon density in the quantized field resulting in a classical behavior by the latter as discussed, for example, by Shirley.[1] Also the choice of energy reference $E_r = 0$ occurs naturally in the fully quantized approach.

The behavior of the Δ_j's for the frequency sweep case is also important and has not previously been discussed in detail. Unlike the Zeeman-tuning case, the frequency-sweep Δ_j's undergo a radical change of behavior as a function of E_r. In their analysis of the problem, which involved a continued-fraction technique, Autler and Townes[19] observed a very complicated behavior for the Δ_j's as a function of ω which they ascribed to the jumping from one solution to another of the infinitely many solutions for the Δ_j's. Characteristic exponent plots for the frequency-sweep experiment are shown[24] in Figs. 2 and 3 for $E_r = 0$, 0.9, and 1.5 (keeping $E_2 - E_1 = -\gamma B_0 = \omega_0 = 1$). Figure 2 shows how these complications can be eliminated by choosing $E_r = 0$ while Fig. 3 shows that when $E_r \neq 0$ these frequency-dependent char-

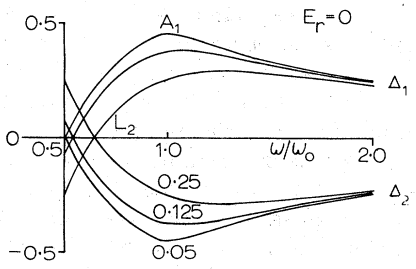

FIG. 2. Frequency-dependent characteristic exponents in the neighborhood of the single-photon transition for the choice of energy reference $E_r = 0$. The curves are labeled by the relevant values of the coupling parameter $s = -\gamma B/(2\omega_0)$. No discontinuity (interchange of identity) occurs at any point in the frequency domain and such plots will also retain their symmetry at all higher photon transition frequencies (Ref. 15). The single-photon resonance peak occurs at the anticrossing A_1 while the level crossing L_2 signifies the position of the "forbidden" two-photon transition.

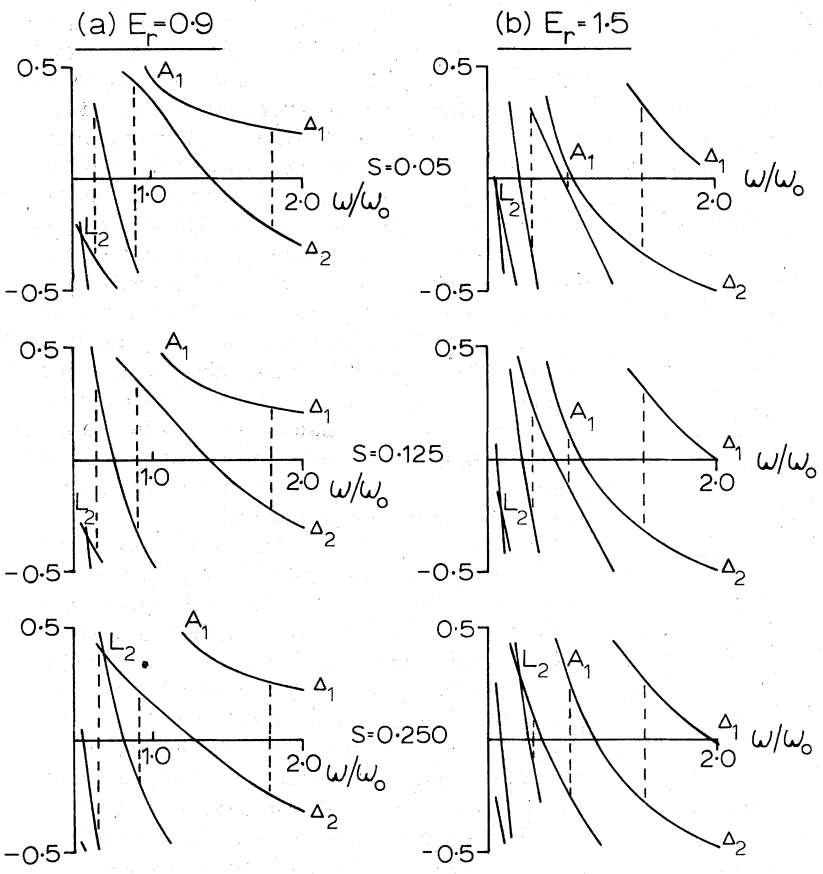

FIG. 3. Frequency-dependent characteristic exponents corresponding to (a) $E_r = 0.9$ and (b) $E_r = 1.5$ over the same frequency domain as in Fig. 2 and for the same values of the coupling parameter s. Moving the energy reference point E_r out of the branch $[-0.5, 0.5]$ has caused a dramatic change in the behavior of the Δ_j. A discontinuity (interchange of identity) in Δ_1 and Δ_2 occurs at the dashed lines. Such characteristic exponent plots will become increasingly difficult to analyze in the vicinity of successive higher photon transitions (Ref. 15). In (b) an interchange of identity occurs at $\omega = \omega_0$, which is not the position of A_1. For all the frequency-sweep plots of Figs. 2 and 3, the positions of the anticrossing A_1 and the level crossing L_2 and the complete spectra are independent of the choice of E_r.

acteristic exponents lose much of their interpretive value and exhibit the same sort of behavior as described in Ref.19.

In general, the interchange[22] of identity between Δ_1 and Δ_2 in both the Zeeman-tuning and frequency-sweep experiments has no effect on the calculated physical observables for the system. It is clear from these results that there is no physically important discontinuity in the characteristic exponent plots for any choice of energy reference. However, the less symmetric the plots of the Δ_j, the less useful they are for locating the positions of the anticrossings and hence the less useful they are for predicting the various characteristics of the spectra assocated with the frequency-sweep experiment. In both experiments the choice of $E_r = 0$ eliminates all problems associated with the interchange of the identities of the Δ_j and with the symmetry of the plots.

The behavior of the characteristic exponents Δ_j in the neighborhood of higher photon resonances will also depend critically on the choice of $E_1 + E_2$. As long as this choice is confined[15] to the branch $[-0.5, 0.5]$ plots of the Δ_j's will remain informative and will provide accurate frequency shifts and half-widths for various n-photon resonances. This is particularly important for $n > 1$ since the shifts are large and the widths are much narrower than for the single photon case. Thus, it is important to have a tractable method for the prediction of the positions of the resonance peaks for multiphoton spectra that does not require a laborious search involving the direct calculation of substantial portions of the spectra themselves. The results presented here for the two-level system also have important implications for analogous studies on multilevel systems. It is clear that the phase of the oscillating field will also play an important role in such studies for intense fields. The nature of the two-level characteristic exponents also suggests that their multilevel analogs will remain well behaved and informative as long as the energy sum $\sum_{i=1}^{N} E_j$ is confined to the branch $[-0.5, 0.5]$. An extension of this study to higher photon resonances and multilevel systems will be the subject of future publications.

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[17]The characteristic exponents used here are related to those of Shirley[1] by the relation $q = -\omega\Delta$. The factor ω must be included in many of the physical interpretations involving the Δ_j, although it is convenient in many applications to use the Δ_j themselves. For example in the Zeeman-tuning experiment q is often plotted in units of ω as a function of ω_0 while in frequency-sweep experiments, involving a wide frequency domain, it is more convenient to plot the Δ_j's rather than the $\omega\Delta_j$'s.
[18]The characteristic exponents for the Rabi (rotating field) solution, in the Schrödinger representation, are given (Ref. 15) by $\Delta^R = -(E_1+E_2)/2 \pm [(\omega-\omega_0)^2+|\gamma B|^2]^{1/2}/2$. The energy reference E_r occurs naturally in this solution which is the weak-field limit of the exact result in the vicinity of $\omega = \omega_0$.
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[22]For $E_r \neq 0$, Δ_1 and Δ_2 can change identity as a function of ω_0. This occurs if $\omega\Delta_1 = \omega\Delta_2$ or $|\omega\Delta_1| = |\omega\Delta_2|$ corresponding to a real or a pseudo degeneracy in the energy levels of the spin $-\frac{1}{2}$—applied-field system. The characteristic exponent plots for $E_r \neq 0$ can be made to agree with those for $E_r = 0$ by using Eq. (7), see also, for example, Fig. 1 of Ref. 1.
[23]Part of the source of this discrepancy may lie in the implied conclusion in Ref. 9 that Shirley constructed characteristic exponent plots by using a singular expansion for the q_j in terms of $\omega - \omega_0$. It is apparent that Shirley's plots were computed for $E_r = 0$ by employing a numerical solution of the time-dependent Schrödinger equation and hence agree with our results. The origin of the singular expansion for the characteristic exponents has been discussed in detail recently by W. A. McClean and S. Swain, J. Phys. B 9, 1673 (1976).
[24]In the examples reported here the Δ_j, $j = 1, 2$, are confined (by choice) to the branch bounded by the branch lines $\pm\frac{1}{2}$ [corresponding to $n = m = 0$ in Eqs. (6) and (7)]. Continuous plots of the Δ_j, as a function of ω_0 or ω, can be obtained by using Eq. (7) with $n \neq 0$.