Magnetic and optical resonance of two-level quantum systems in modulated fields. II. Floquet Hamiltonian approach

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It is shown that, in modulated magnetic resonance on spin-$\frac{1}{2}$ systems, the time-averaged absorption signal of the optical pump beam can be obtained directly from the dressed-atom energy eigenvalues of the Floquet Hamiltonian for arbitrary orientations of the optical pump beam, namely, by a straightforward generalization of Shirley's well-known result [Phys. Rev. **138**, B979 (1965)]. Analytical and numerical results derived from this generalized diagonalization technique are compared with the vanishing damping solution of the Bloch equations from the preceding paper [W. M. Ruyten, Phys. Rev. A **42**, 4226 (1990)] and are shown to be in complete agreement with the latter.

I. INTRODUCTION

In the preceding paper, to be referred to as Ref. 1, we have studied in detail the resonance behavior of a two-level system driven by either amplitude-modulated or frequency-modulated optical fields, or by a combination of static and oscillating magnetic fields. In particular, for the magnetic-resonance geometry, we have investigated how the absorption of the circularly polarized optical pump-beam depends, in general, on the orientation of the pump beam with respect to the magnetic fields. Although the analysis in Ref. 1 is based on solution of the Bloch equations, namely in terms of matrix continued fractions, we have frequently referred to the concept of level crossings in a dressed-atom energy-level diagram. This concept arises not in the Bloch equation formulation, but rather in formulations where the eigenvalues of the field-atom system (the dressed atom) are determined, for example, by diagonalizing the Floquet Hamiltonian. Specifically, a well-known result, first obtained by Shirley, states that the time-averaged transition probability P_{ab} between two states a and b in such a system can be obtained from the dressed-atom energy eigenvalues q by the simple expression[2]

$$P_{ab}=\frac{1}{2}\left[1-4\left[\frac{\partial q}{\partial\omega_0}\right]^2\right], \tag{1.1}$$

where ω_0 is the level spacing of the spin-$\frac{1}{2}$ system in the absence of the oscillating field (that is, for the case of magnetic resonance, $\omega_0=\gamma H_0$, where γ is the gyromagnetic ratio of the spin-$\frac{1}{2}$ system, and H_0 is the magnitude of the static magnetic field). But since the energy eigenvalues q do not depend on the pump-beam orientation (they *do* depend on the relative orientation of the static and oscillating magnetic fields), the question arises how Eq. (1.1) can be modified to take into account the pump-beam dependence of the resonance behavior of the system. It is the purpose of this paper to show that this generalization can, in fact, be arrived at in a very simple and straightforward manner, namely by the substitution

$$\frac{\partial q}{\partial\omega_0}\rightarrow\mathbf{e}_p\cdot\frac{\partial}{\partial\underline{\omega}_0}q(\underline{\omega}_0) \tag{1.2}$$

in Eq. (1.1), where $\mathbf{e}_p$ is a unit vector along the pump-beam, and $\underline{\omega}_0=\gamma\mathbf{H}_0$ is the Rabi frequency *vector* associated with the static magnetic field $\mathbf{H}_0$. After formally establishing this result in Sec. II, some numerical calculations are performed in Sec. III to show that, indeed, the results obtained with Eqs. (1.1) and (1.2) are in perfect agreement with the results obtained in Ref. 1 using the vanishing damping continued-fraction (VDCF) solution to the Bloch equations. Specifically, it is shown that the special field geometries that lead to pump-beam-independent resonances can be easily inferred from the three-dimensional dressed-atom energy-level diagram of the system. In Sec. IV, the generalized diagonalization results from Eqs. (1.1) and (1.2) are used to confirm some results from the VDCF calculations analytically, namely the shifts of the multiple quantum resonances from the weak-field values $\omega_0=m\omega$ (where m is an integer and ω is the modulation frequency of the oscillating magnetic field), and, also in the weak-field limit, the expression for the transition probability itself. Section V presents a brief conclusion.

II. GENERALIZATION OF THE DIAGONALIZATION TECHNIQUE

Solving the Bloch equations for a magnetic- or optical-resonance problem, as is done in Ref. 1, is only one possible approach to studying the resonance behavior of a modulated interaction. Other approaches that have been used, and are mentioned in Sec. III of Ref. 1, are those based on a resolvent formalism,[3–5] continued-fraction perturbation theory,[6] and unitary transformations of coordinate frames.[7] Here we consider Shirley's method of diagonalization of the Floquet Hamiltonian.[2] Specifically, we show how this method can be generalized to account for an arbitrary orientation of the optical pump-beam in a magnetic-resonance geometry as shown in Fig. 1.

Figure 1(a) depicts the same geometry as that of Fig. 1

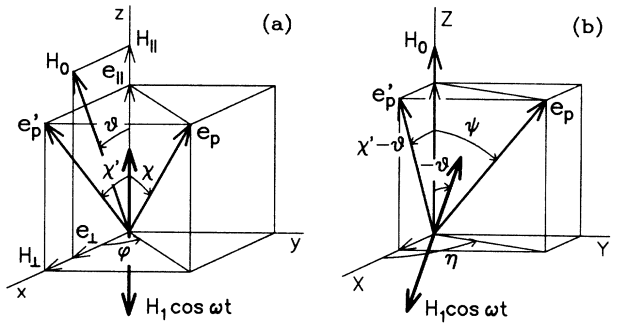

FIG. 1. General magnetic-resonance geometry defined in two different coordinate systems, namely with the oscillating field $\mathbf{H}_1\cos(\omega t)$ along the z axis (a) and the static field $\mathbf{H}_0$ along the Z axis (b), respectively. In (a), the components of the static field and of the pump-beam vector $\mathbf{e}_p$ along and transverse to the oscillating field are denoted by $H_\parallel$ and $e_\parallel$ and by $H_\perp$ and $e_\perp$, respectively. In both cases, $\mathbf{e}_p'$ is the projection of the pump-beam vector on the plane defined by the two magnetic fields, at angles χ' with the z axis and $\chi'-\vartheta$ with the Z axis, respectively.

in Ref. 1. That is, the oscillating radio-frequency (rf) magnetic field is along the z axis in a Cartesian coordinate system, and is denoted by $\mathbf{H}_1\cos(\omega t)$, where ω is the modulation frequency. The static magnetic field $\mathbf{H}_0$ is in the xz plane, making an angle ϑ with the rf field. Finally, the propagation direction of the optical pump beam is denoted by the unit vector $\mathbf{e}_p$, and is specified by the polar angles χ and φ with respect to the xyz system.

Following Yabuzaki *et al.*'s treatment,[4] it is convenient to define the Hamiltonian for this problem in the alternative coordinate system shown in Fig. 1(b), namely, one in which the static magnetic field is along the Z axis, and the oscillating magnetic field is in the XZ plane, namely, at an angle $-\vartheta$ with respect to the static field (in Yabuzaki *et al.*, the oscillating field is oriented at an angle $+\vartheta$ with respect to the static field;[4] however, the positions and the shifts of the various multiple quantum resonances are independent of the sign of ϑ). In the XYZ system, the orientation of the pump-beam unit vector $\mathbf{e}_p$ is now specified by the polar angles ψ and η, and the Hamiltonian for the interaction can be written as (taking $\hbar=1$)

$$\hat{\mathcal{H}}=\hat{\mathcal{H}}_0+\hat{\mathcal{V}} , \tag{2.1}$$

where $\hat{\mathcal{H}}_0$ and $\hat{\mathcal{V}}$ are the zero-oscillating field and the oscillating field interaction Hamiltonians, given by

$$\hat{\mathcal{H}}_0=\omega_0\hat{J}_Z+\omega\hat{a}^\dagger\hat{a} , \tag{2.2}$$

and

$$\hat{\mathcal{V}}=\lambda\hat{\mathbf{J}}\cdot(\hat{a}\mathbf{e}_1+\hat{a}^\dagger\mathbf{e}_1^*) , \tag{2.3}$$

respectively, where $\omega_0=\gamma H_0$ is the static field Rabi frequency, $\hat{a}$ and $\hat{a}^\dagger$ are creation and annihilation operators of the radio-frequency photons, λ is the coupling coefficient (given, for large average photon numbers $\bar{N}$, by $\lambda=\omega_1/2\bar{N}^{1/2}$—where $\omega_1=\gamma H_1$ is the oscillating field Rabi frequency), and $\mathbf{e}_1=(-\sin\vartheta,0,\cos\vartheta)$ is a unit vector in the direction of the radio-frequency field. Lastly, in Eqs. (2.2) and (2.3), $\hat{\mathbf{J}}$ and $\hat{J}_Z$ are the total spin operator of the spin-$\frac{1}{2}$ atom, and its projection on the static magnetic field $\mathbf{H}_0$. The eigenstates of the Hamiltonian $\hat{\mathcal{H}}_0$ from Eq. (2.2) are denoted by the product states $|\pm,n\rangle=|\pm\rangle\otimes|n\rangle$, where the states $|+\rangle$ and $|-\rangle$ are the eigenstates of the atom in the static magnetic field, and the states $|n\rangle$ indicate photon states of the oscillating magnetic field. These eigenstates and their associated eigenvalues are given by[4]

$$\hat{\mathcal{H}}_0|\pm,n\rangle=(n\omega\pm\tfrac{1}{2}\omega_0)|\pm,n\rangle , \tag{2.4}$$

where it has been assumed that the energy eigenvalues of the unperturbed atom are given by $\pm\frac{1}{2}\omega_0$. In terms of these eigenstates, the nonvanishing matrix elements of the interaction Hamiltonian $\hat{\mathcal{V}}$ from Eq. (2.3) are given by

$$\langle\pm,n|\hat{\mathcal{V}}|\pm,n-1\rangle=\langle\pm,n-1|\hat{\mathcal{V}}|\pm,n\rangle=\pm V\cos\vartheta , \tag{2.5a}$$

and

$$\langle\pm,n|\hat{\mathcal{V}}|\mp,n-1\rangle=\langle\mp,n-1|\hat{\mathcal{V}}|\pm,n\rangle=-V\sin\vartheta , \tag{2.5b}$$

where, for large average photon numbers $\bar{N}$, $V=\frac{1}{4}\omega_1$. With Eqs. (2.4) and (2.5), the Floquet Hamiltonian, relative to the dressed-atom eigenstates $|\pm,n\rangle$, becomes [see Eq. (41) in Ref. 4]

$$\mathcal{H}_F=\begin{array}{c} \\ \vdots \\ |+,n-1\rangle \\ |-,n-1\rangle \\ |+,n\rangle \\ |-,n\rangle \\ |+,n+1\rangle \\ |-,n+1\rangle \\ \vdots \end{array}\begin{array}{c} \begin{array}{ccccccccc} \cdots & |+,n-1\rangle & |-,n-1\rangle & |+,n\rangle & |-,n\rangle & |+,n+1\rangle & |-,n+1\rangle & \cdots \end{array} \\ \begin{bmatrix} \ddots & \vdots & \vdots & \vdots & \vdots & \vdots & \vdots & \vdots \\ \cdots & \frac{1}{2}\omega_0-\omega & 0 & B & A & 0 & 0 & \cdots \\ \cdots & 0 & -\frac{1}{2}\omega_0-\omega & A & -B & 0 & 0 & \cdots \\ \cdots & B & A & \frac{1}{2}\omega_0 & 0 & B & A & \cdots \\ \cdots & A & -B & 0 & -\frac{1}{2}\omega_0 & A & -B & \cdots \\ \cdots & 0 & 0 & B & A & \frac{1}{2}\omega_0+\omega & 0 & \cdots \\ \cdots & 0 & 0 & A & -B & 0 & -\frac{1}{2}\omega_0+\omega & \cdots \\ \cdots & \vdots & \vdots & \vdots & \vdots & \vdots & \vdots & \ddots \end{bmatrix} \end{array} , \tag{2.6}$$

where $A=-V\sin\vartheta$ and $B=V\cos\vartheta$. (Because of their slightly different definition of the angle ϑ, $A=+V\sin\vartheta$ in Yabuzaki *et al.*'s work.[4])

Diagonalization of the infinite-dimensional Floquet Hamiltonian from Eq. (2.6) yields two infinite sets of dressed-atom energy eigenvalues $q_\alpha^{(k)}$ and $q_\beta^{(k)}$. Since the sets are periodic (that is, $q_{\alpha,\beta}^{(k+l)}=q_{\alpha,\beta}^{(k)}+l\omega$) and complementary to each other (that is, $q_\alpha^{(k)}+q_\beta^{(k)}=0$), it suffices to consider only one branch of eigenvalues, say, those with values between 0 and $\frac{1}{2}$, denoted by q:

$$q=q_\alpha^{(k)}-k\omega=-q_\beta^{(k)}-k\omega\ . \tag{2.7}$$

The q's depend on the interaction strength ω_1 (or V), the static Rabi frequency ω_0, and the angle ϑ between the static and the oscillating magnetic fields. For example, Fig. 1 in Ref. 8 displays three-dimensional plots of q for several fixed interaction strengths ω_1 as a function of the static field Rabi frequency components $\omega_\parallel=\omega_0\cos\vartheta$ and $\omega_\perp=\omega_0\sin\vartheta$, that is, the static field Rabi frequency components along and perpendicular to the oscillating field, respectively [see Fig. 1(a)].

From Shirley's well-known formula, given in Eq. (1.1), the time-averaged transition probability between two states a and b can be found directly from the dressed-atom energy eigenvalues. In the work of Yabuzaki and co-workers,[4,8] which was carried out with an optical pump beam perpendicular to the oscillating field (but in the plane defined by $\mathbf{H}_0$ and $\mathbf{H}_1$), the two states a and b are associated with the atomic eigenstates $|+\rangle$ and $|-\rangle$, and, although this is not mentioned or discussed explicitly in their work, the differentiation with respect to ω_0 in Eq. (1.1) is replaced by differentiation with respect to the static Rabi frequency component $\omega_\perp$ perpendicular to the oscillating rf field (calculation shows that only in this case are Yabuzaki and co-workers' numerical results reproduced correctly).

Up until this point, it may be observed, the orientation of the optical pump beam has not been taken into account. However, as shown in Ref. 1, the resonance behavior of the spin-$\frac{1}{2}$ system depends significantly on the pump-beam orientation. Specifically, the absorption of the optical pump beam is proportional to the projection of the magnetization of the spin-$\frac{1}{2}$ system along the pump-beam propagation direction $\mathbf{e}_p$ [see Eq. (2.5) in Ref. 1]. This means that the appropriate eigenstates for this problem are *not* the states $|+\rangle$ and $|-\rangle$, which are eigenstates of the projection operator $\hat{J}_Z$ along the static magnetic field, but of the states $|+'\rangle$ and $|-'\rangle$, which are eigenstates of the projection operator $\hat{J}_p$, defined along the pump-beam propagation direction. With respect to these latter eigenstates, the Floquet Hamiltonian becomes, in terms of the Hamiltonian from Eq. (2.6),

$$\mathcal{H}'_F=\mathbf{R}^\dagger\mathcal{H}_F\mathbf{R}\ , \tag{2.8}$$

where the rotation matrix $\mathbf{R}$ is given, in terms of the polar angles ψ and η from Fig. 1(b), by[9]

$$\mathbf{R}=\begin{bmatrix}\cos\frac{1}{2}\psi e^{-i\eta/2} & -\sin\frac{1}{2}\psi e^{-i\eta/2}\\ \sin\frac{1}{2}\psi e^{+i\eta/2} & \cos\frac{1}{2}\psi e^{+i\eta/2}\end{bmatrix}\ . \tag{2.9}$$

[Actually, $\mathbf{R}$ is an infinite-dimensional matrix, whose only nonvanishing elements are two-by-two blocks along the diagonal as given in Eq. (2.9).] In writing Eq. (2.9), we have conformed to Messiah's notation, substituting for the Euler angles α, β, and γ in Ref. 9 the values η, ψ, and 0, respectively. Since the rotation matrix from Eq. (2.9) operates on the atomic eigenstates $|\pm\rangle$ but not on the rf photon states $|n\rangle$, it follows that the matrix $\mathcal{H}_F$ from Eq. (2.6) transforms in two-by-two blocks. Namely, the nonvanishing off-diagonal and diagonal blocks transform under the rotation $\mathbf{R}$ as

$$\begin{bmatrix}B & A\\ A & -B\end{bmatrix}\to\begin{bmatrix}B\cos\psi+A\sin\psi\cos\eta & -B\sin\psi+A(\cos\psi\cos\eta+i\sin\eta)\\ -B\sin\psi+A(\cos\psi\cos\eta-i\sin\eta) & -B\cos\psi-A\sin\psi\cos\eta\end{bmatrix}\ , \tag{2.10a}$$

and

$$\begin{bmatrix}\frac{1}{2}\omega_0+n\omega & 0\\ 0 & -\frac{1}{2}\omega_0+n\omega\end{bmatrix}\to\begin{bmatrix}\frac{1}{2}\omega'_\parallel+n\omega & -\frac{1}{2}\omega'_\perp\\ -\frac{1}{2}\omega'_\perp & -\frac{1}{2}\omega'_\parallel+n\omega\end{bmatrix}\ , \tag{2.10b}$$

respectively, where the new parallel and perpendicular projections of the static field Rabi frequency components are defined as $\omega'_\parallel=\omega_0\cos\psi$ and $\omega'_\perp=\omega_0\sin\psi$ (recall that the original components $\omega_\parallel$ and $\omega_\perp$ constitute projections of the static field Rabi frequency vector $\underline{\omega}_0$ with respect to the oscillating rf field, not the optical pump beam).

Having obtained the Floquet Hamiltonian with respect to the new dressed-atom basis states $|\pm',n\rangle$, the important observation is that Shirley's method of diagonalizing the Floquet Hamiltonian and subsequently computing the time-averaged transition probability between two atomic states (here, $|+',n\rangle$ and $|-',n\rangle$) by differentiation of the dressed-atom energy eigenvalues remains essentially unmodified. Indeed, the crucial step in the derivation of Eq. (1.1) is that there exists a variable y for which the operator $\partial\mathcal{H}'_F/\partial y$ is diagonal with respect to the basis in which $\mathcal{H}'_F$ is expressed.[2] Indeed, from Eqs. (2.10), it follows that the new parallel Rabi frequency $\omega'_\parallel$ satisfies this property, namely,

$$\frac{\partial\mathcal{H}'_F}{\partial\omega'_\parallel}|\pm',n\rangle=\pm\tfrac{1}{2}|\pm',n\rangle\ . \tag{2.11}$$

Thus, following the identical steps that led to Eq. (1.1), the time-averaged transition probability between the two states $a'=|+'\rangle$ and $b'=|-'\rangle$ (or vice versa), representing the absorption of the optical pump beam on propagation through the spin-$\frac{1}{2}$ system, is given by

$$P_{a'b'}=\frac{1}{2}\left[1-4\left[\frac{\partial q'}{\partial\omega'_{\parallel}}\right]^2\right], \tag{2.12}$$

where the q's are the eigenvalues of the Floquet Hamiltonian given by Eqs. (2.10) (actually, as discussed at the end of Sec. III, the absorption signal is proportional to the "no-transition probability" $P_{a'a'}=1-P_{a'b'}$).

From Eqs. (2.10) and (2.12), it is possible to calculate the absorption of the optical pump beam for arbitrary orientations of the magnetic fields and of the optical pump beam. However, we now show that this absorption signal can be obtained directly from Yabuzaki *et al.*'s original Floquet Hamiltonian, given in Eq. (2.6), even though this Hamiltonian does not depend on the orientation of the pump beam. To this end, we observe that the eigenvalues q from Eq. (2.6) and q' from Eq. (2.10) must be identical, since the rotation matrix $\mathbf{R}$ from Eq. (2.9) is unitary. Furthermore, we observe that, if some function f is defined in a (three-dimensional) Cartesian coordinate space, the scalar product $\mathbf{a}\cdot\nabla f$, where $\mathbf{a}$ is some vector in this coordinate space, is independent of the choice of coordinates:

$$\mathbf{a}\cdot\nabla f(\mathbf{x})=\mathbf{a}'\cdot\nabla' f(\mathbf{x}'), \tag{2.13}$$

where ∇ and ∇' represent differentiation with respect to the coordinates $\mathbf{x}$ and $\mathbf{x}'$, respectively, and $\mathbf{a}$ and $\mathbf{a}'$ represent the same vector, expressed in either coordinate system. Associating the eigenvalues q and q' with the function f, associating, furthermore, the coordinates $\mathbf{x}$ and $\mathbf{x}'$ with projections of the static magnetic field $\mathbf{H}_0$ on three mutually orthogonal axes [namely, those in the original coordinate system from Fig. 1(a) and the rotated system in which Eqs. (2.10) and (2.12) are expressed, respectively], and, lastly, associating the vectors $\mathbf{a}$ and $\mathbf{a}'$ from Eq. (2.13) with the pump-beam vector $\mathbf{e}_p$, we can write

$$\mathbf{x}=(\omega_{\parallel},\omega_{\perp},\omega_3),\quad \mathbf{a}=\mathbf{e}_p=(\mathbf{e}_{\parallel},\mathbf{e}_{\perp},0), \tag{2.14a}$$

and

$$\mathbf{x}'=(\omega'_{\parallel},\omega'_{\perp},\omega'_3),\quad \mathbf{a}'=\mathbf{e}_p=(1,0,0), \tag{2.14b}$$

respectively, where $e_{\parallel}=e_z$ and $e_{\perp}=e_x$ are the projections of the pump-beam vector $\mathbf{e}_p$ along and perpendicular to the oscillating magnetic field $\mathbf{H}_1$ in Fig. 1(a). From Eqs. (2.13) and (2.14), we then conclude that

$$\frac{\partial q'}{\partial\omega'_{\parallel}}=\mathbf{e}'_p\cdot\nabla' q'=\mathbf{e}_p\cdot\nabla q=\frac{\partial q}{\partial\omega_{\parallel}}e_{\parallel}+\frac{\partial q}{\partial\omega_{\perp}}e_{\perp}. \tag{2.15}$$

This is exactly the generalization of Shirley's result, constituted by Eqs. (1.1) and (1.2), that we set out to prove. That is, for arbitrary pump-beam orientations $\mathbf{e}_p$, the time-averaged transition probability associated with the absorption of the optical pump beam on propagation through the spin-$\frac{1}{2}$ system is given by

$$P_{a'b'}=\tfrac{1}{2}\left[1-4\left[\frac{\partial q}{\partial\omega_{\parallel}}e_{\parallel}+\frac{\partial q}{\partial\omega_{\perp}}\omega_{\perp}\right]^2\right]. \tag{2.16}$$

It should be noted that the choice of coordinate system in which the Rabi frequency vector $\underline{\omega}_0$ is expressed is completely arbitrary, and the choice on the right-hand sides of Eqs. (2.15) and (2.16) are for convenience only [much of the discussion on the resonance behavior of this system has been in terms of the static field Rabi frequency components $\omega_{\parallel}$ and $\omega_{\perp}$, associated with the projections $H_{\parallel}$ and $H_{\perp}$ of the static field with respect to the oscillating field in Fig. 1(a)]. Note that, by Eq. (2.16), the term $\partial q/\omega_0$ in Shirley's formula from Eq. (1.1) may be replaced, for the much studied magnetic-resonance geometries with the pump beam along and perpendicular to the oscillating magnetic field, by $\partial q/\partial\omega_{\parallel}$ and $\partial q/\partial\omega_{\perp}$, respectively, thereby justifying Yabuzaki *et al.*'s choice ($\partial q/\partial\omega_{\perp}$) in Refs. 4 and 8.

III. NUMERICAL RESULTS

In this section we discuss some numerical results, obtained with the generalized diagonalization technique, and compare the results to those obtained with the vanishing damping continued-fraction solution of the Bloch equations from Ref. 1.

First, Fig. 2 shows a typical three-dimensional energy-level diagram, similar to those in Fig. 1 of Ref. 8, in which the dressed-atom eigenvalues q are displayed, for a fixed interaction strength $\omega_1=2\omega$, as a function of the static Rabi frequency components $\omega_{\parallel}$ and $\omega_{\perp}$. The displayed values of q belong to one of the two central branches of eigenvalues, obtained by truncating and diagonalizing the Floquet Hamiltonian from Eq. (2.6) at $n=\pm 10$ terms. By Eq. (2.16), the time-averaged transition probability $P_{a'b'}$ is independent of the orientation of the pump beam if the condition

$$\frac{\partial q}{\partial\omega_{\parallel}}=\frac{\partial q}{\partial\omega_{\perp}}=0 \tag{3.1}$$

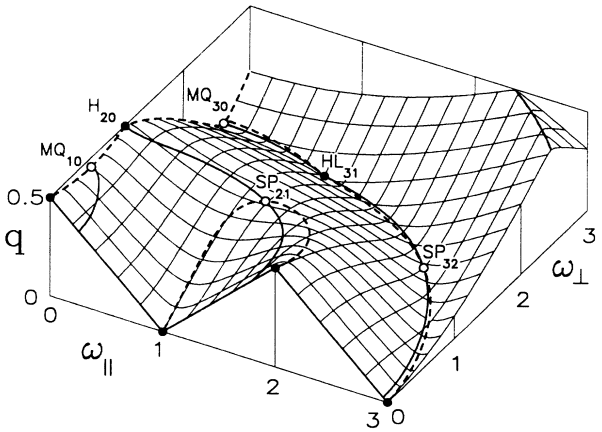

FIG. 2. Three-dimensional rendering of one branch of the dressed-atom energy eigenvalues q as a function of the two static field Rabi frequency components $\omega_{\parallel}$ and $\omega_{\perp}$ (in units of the modulation frequency ω), for an interaction strength $\omega_1=4\omega$. Dashed and solid curves indicate points for which $\partial q/\partial\omega_{\parallel}=0$ and $\partial q/\partial\omega_{\perp}=0$, respectively. Closed and open symbols indicate level crossings and saddle points, respectively, and are labeled in the same way as the corresponding points in Fig. 7(a) in Ref. 1.

is satisfied. In Fig. 2, the points for which $\partial q/\partial\omega_{\parallel}=0$ and $\partial q/\partial\omega_{\perp}=0$ are indicated by dashed and solid curves, respectively. The intersections between these curves then give the locations of the points $(\omega_{\parallel},\omega_{\perp})$ for which the condition from Eq. (3.1) holds. These intersections fall into two categories, namely, those that correspond to level crossings in the dressed-atom energy-level diagram (the solid symbols in Fig. 2, for which $q=0$ or $q=\frac{1}{2}$), and those that correspond to level anticrossings, but are located at saddle points in the same diagram (open symbols in Fig. 2—actually, it is not obvious that these points correspond to saddle points, rather than local extrema; since all our calculations do, however, show these points to be saddle points, we shall refer to them as such without further justification). The level crossings are given by the well-known parametric resonances for $\omega_{\perp}=0$ and $\omega_{\parallel}=p\omega$ (with p a nonzero integer), the Haroche resonances (namely, at $\omega_{\parallel}=0$), and the Haroche-like resonances (namely, at $\omega_{\parallel}=p\omega$, with $\omega_{\perp}\neq 0$). Indeed, these level crossings have been observed, at the same locations, for pump-beam orientations both along[10] and perpendicular[4,8] to the oscillating magnetic field. Like the level crossings, the saddle points also occur for both zero and nonzero values of $\omega_{\parallel}$. The former are given by the well-known multiple quantum resonances, and the latter, termed simply saddle-point resonances, were first discussed in Ref. 1. Numerical calculation shows that the locations of these various special points, namely at which the condition from Eq. (3.1) holds, are in exact agreement with the values of $\omega_{\parallel}$ and $\omega_{\perp}$ at which the pump-beam-independent resonances were found using the VDCF technique from Ref. 1 (compare the special points in Fig. 2 with those in Fig. 7 in Ref. 1). Also, the dashed and solid curves in Fig. 2 are in exact agreement with the similarly coded curves in Fig. 7 in Ref. 1 which correspond to pump-beam orientations with $\mathbf{e}_p\|\mathbf{H}_1$ and $\mathbf{e}_p\perp\mathbf{H}_1$, respectively.

Of course, not only the positions of the various resonances can be found from Eq. (2.16), but also the transition probabilities themselves. The results of such a calculation are shown in Fig. 3, which displays the quantity $P_{a'b'}$ from Eq. (2.16) as a function of the parallel Rabi frequency $\omega_{\parallel}$ for fixed values $\omega_{\perp}$ and ω_1, and for various pump-beam angles χ' [as in Ref. 1, χ' is the angle between the static magnetic field $\mathbf{H}_0$ and the projection of the pump-beam vector $\mathbf{e}_p$ on the plane defined by the two magnetic fields, that is, the xz plane in Fig. 1(a)]. This calculation, carried out by truncating the Floquet matrix from Eq. (2.6) at $n=\pm 10$ terms and evaluating the derivatives in Eq. (2.16) as finite differences (with a step size of $10^{-3}\omega$), is found to be in agreement with the VDCF solution from Ref. 1 to better than one part in one thousand for all points in Fig. 3. That is, by numerical calculation, the time-averaged transition probability $P_{a'b'}$ from the generalized diagonalization technique is found to be related to the vanishing damping, time-averaged absorption signal $(S^{(0)})^{\mathrm{VD}}$ as

$$P_{a'b'}=\tfrac{1}{2}[1-(S^{(0)})^{\mathrm{VD}}],\quad \gamma_{xz}=\gamma_{yz}=1 \tag{3.2}$$

where, according to Eqs. (4.12)–(4.20) in Ref. 1, the VDCF absorption signal is given by

$$(S^{(0)})^{\mathrm{VD}}=\frac{(\omega_{\perp}e_x+\alpha e_z)^2}{\alpha^2+\omega_1^2\beta_{21}}\ , \tag{3.3}$$

with

$$\alpha=\omega_{\parallel}+\omega_1(\mathbf{X}_1^{(0)})_{11}\ , \tag{3.4}$$

$$\beta_{21}=\gamma_{xz}+2(\mathbf{X}_1^{(1)})_{21}\ , \tag{3.5}$$

and the matrix continued fractions $\mathbf{X}_1^{(0)}$ and $\mathbf{X}_1^{(1)}$ follow by backward iteration of

$$\mathbf{X}_k^{(0)}=\left[\frac{2}{\omega_1}\begin{bmatrix}-\omega_{\parallel} & k-\omega_{\perp}^2/k\\ k & -\omega_{\parallel}\end{bmatrix}-\mathbf{X}_{k+1}^{(0)}\right]^{-1}\ , \tag{3.6}$$

and

$$\mathbf{X}_k^{(1)}=\mathbf{X}_k^{(0)}\left[\begin{bmatrix}0 & \gamma_{yz}+\omega_{\perp}^2/k^2\\ \gamma_{xz} & 0\end{bmatrix}+\mathbf{X}_{k+1}^{(1)}\right]\mathbf{X}_k^{(0)}\ . \tag{3.7}$$

However, as indicated in Eq. (3.2), the two methods are found to be in general agreement only for unit damping ratios γ_{xz} and γ_{yz} in the VDCF solution. This result, although not trivial, is quite plausible since, in contrast to the VDCF method, no damping terms appear in the derivation of the generalized diagonalization results. Figure 4 shows explicitly that, although the positions of the resonances (the points for which $P_{a'b'}=\frac{1}{2}$) are independent of these damping ratios, the results from the VDCF

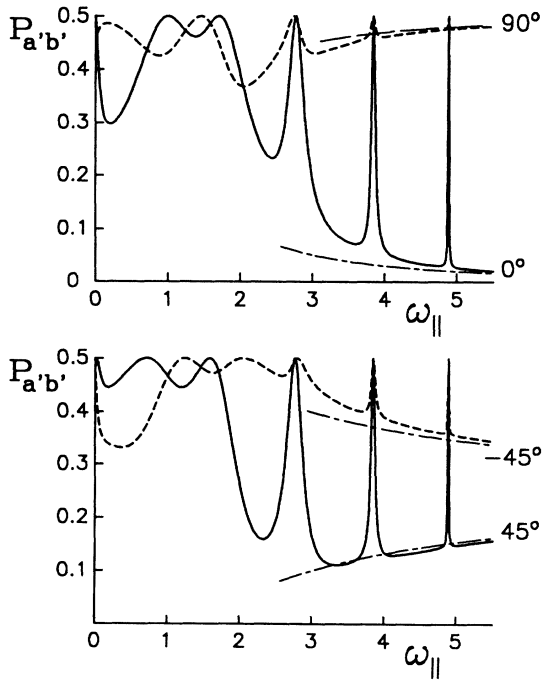

FIG. 3. Time-averaged transition probabilities for fixed values $\omega_1=2\omega$ and $\omega_{\perp}=\omega$ as a function of the parallel Rabi frequency $\omega_{\parallel}$ (in units of ω), for various pump-beam angles χ'. The thin, dot-dashed curves show the approximations given in Eq. (4.12). The solid curves are found both with the generalized diagonalization technique and the VDCF technique from Ref. 1.

solution do, in general, depend on the values of γ_{xz} and γ_{yz}. Note that, since the absorption of the circularly polarized pump beam takes place only from the excited state level that was originally populated by the pump beam, the absorption signal should be proportional to the probability $P_{a'a'}=1-P_{a'b'}$, that is, the probability that the atom remain in its original state, as is indeed found in Eq. (2.3). The factor $\frac{1}{2}$ in Eq. (2.3) then is due merely to the normalization of the respective quantities in the two methods.

Finally, it should be mentioned that, although the two methods give identical results, the VDCF solution is much less computationally intensive than the diagonalization method, and that, furthermore, it allows damping to be taken into account more easily, namely, by solving the Bloch equations in terms of continued fractions without taking the vanishing damping limit.

IV. CALCULATION OF SOME ANALYTICAL RESULTS

Although the exact agreement between the numerical results from the VDCF solution from Ref. 1 and the present, generalized diagonalization technique would seem to establish the correctness of both methods beyond all reasonable doubt, it is of interest to derive also some analytical results from the generalized diagonalization technique, and compare these with the results from the VDCF solution. Thus we derive in this section expressions for the lowest (second) -order shifts of the system's multiple quantum resonances from the weak-field resonance conditions $\omega_0=m\omega$, and for the transition probability $P_{a'b'}$, also in the weak-field limit.

To obtain expressions for the shifts of the multiple quantum resonances, we use here a perturbation method described by Salwen[11] and used by Shirley.[2] In it, the infinite-dimensional Floquet Hamiltonian from Eq. (2.6) can be approximated by a two-by-two matrix $\tilde{\mathbf{H}}$. Specifically, near a resonance between two dressed-atom eigenstates $|\alpha\rangle$ and $|\beta\rangle$ of the unperturbed Hamiltonian $\hat{\mathcal{H}}_0$ from Eq. (2.2), the elements of $\tilde{\mathbf{H}}$ are given, in terms of the Floquet matrix $\mathcal{H}_F$ from Eq. (2.6), by

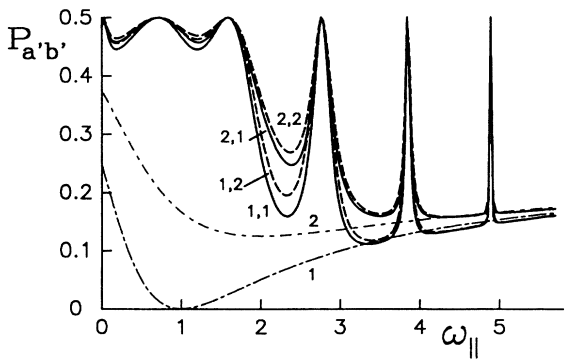

FIG. 4. As Fig. 3 for a pump-beam angle $\chi'=\pi/4$, but for several values of the damping rates γ_{xz},γ_{yz} in the VDCF solution from Eqs. (3.2)–(3.7). The thin, dot-dashed curves show the approximation from Eq. (4.13), and are labeled with the value of γ_{xz}.

$$\tilde{H}_{\mu\nu}=(\mathcal{H}_F)_{\mu\nu}+\sum_{\xi\neq\alpha,\beta}\frac{(\mathcal{H}_F)_{\mu\xi}(\mathcal{H}_F)_{\xi\nu}}{q_0-(\mathcal{H}_F)_{\xi\xi}},\quad \mu,\nu=\alpha,\beta \tag{4.1}$$

where q_0 is an approximation for the exact eigenvalue q near the resonance. Identifying the generic states $|\alpha\rangle$ and $|\beta\rangle$ with the actual states $|+,n\rangle$ (with unperturbed eigenvalue $q_\alpha=\frac{1}{2}\omega_0$) and $|-,n+m\rangle$ (with unperturbed eigenvalue $q_\beta=-\frac{1}{2}\omega_0+m\omega$, where m is an arbitrary integer), respectively, and setting $q_0=q_\alpha=q_\beta=\frac{1}{2}\omega_0$ (which is approximately true near the resonance condition $\omega_0=m\omega$), the elements of $\tilde{\mathbf{H}}$ become, as a function of the photon number m,

$$\tilde{H}^{(m)}_{\alpha\alpha}=\tfrac{1}{2}\omega_0+(A^2/\omega)c_m\ , \tag{4.2a}$$

$$\tilde{H}^{(m)}_{\beta\beta}=-\tfrac{1}{2}\omega_0+m\omega-(A^2/\omega)c_m\ , \tag{4.2b}$$

and

$$\tilde{H}^{(m)}_{\alpha\beta}=\tilde{H}^{(m)}_{\beta\alpha}=A\delta_{|m|,1}-m(AB/\omega)\delta_{|m|,2}\ , \tag{4.2c}$$

where A and B are as defined following Eq. (2.6), and the coefficients c_m are given by

$$c_m=\begin{cases} m/2, & m=\pm1\ , \\ 2m/(m^2-1) & \text{otherwise}\ . \end{cases} \tag{4.3}$$

As an example of the calculations leading to Eqs. (4.2) and (4.3), we observe that, for the single photon resonance with $m=+1$, Eqs. (2.6) and (4.1) yield

$$\begin{aligned}\tilde{H}^{(1)}_{\alpha\alpha}-\tfrac{1}{2}\omega_0&=\frac{B^2}{q_0-\frac{1}{2}\omega_0+\omega}+\frac{A^2}{q_0+\frac{1}{2}\omega_0+\omega}+\frac{B^2}{q_0-\frac{1}{2}\omega_0-\omega}\\&\simeq\frac{B^2}{\omega}+\frac{A^2}{2\omega}-\frac{B^2}{\omega}=\frac{A^2}{2\omega}\ ,\end{aligned} \tag{4.4}$$

where the approximations $q_0\simeq\omega_0\simeq\omega$ have been used. With Eqs. (4.2), the approximate values $\tilde{q}^{(m)}$ of the perturbed eigenvalues of $\tilde{\mathbf{H}}$ are given by

$$\tilde{q}^{(m)}\simeq\tfrac{1}{2}m\omega\pm\tfrac{1}{2}[(\omega_0-m\omega+2c_m A^2/\omega)^2+4\tilde{A}^2]^{1/2}\ , \tag{4.5}$$

where $\tilde{A}$ represents the quantity on the right-hand side of Eq. (4.2c). Using the results

$$\frac{\partial\omega_0}{\partial\omega_{\|}}=\cos\vartheta,\quad \frac{\partial\omega_0}{\partial\omega_{\perp}}=\sin\vartheta\ , \tag{4.6a}$$

and

$$\begin{aligned}\frac{\partial A^2}{\partial\omega_{\|}}&\simeq-\frac{2V^2}{\omega}\sin^2\vartheta\cos\vartheta\ ,\\ \frac{\partial A^2}{\partial\omega_{\perp}}&\simeq\frac{2V^2}{\omega}\sin\vartheta\cos^2\vartheta\ ,\end{aligned} \tag{4.6b}$$

the resonance shifts of the m-quantum resonances now follow, by Eq. (2.16), from the condition $\mathbf{e}_p\cdot\nabla\tilde{q}^{(m)}=0$, or $(\partial\tilde{q}^{(m)}/\partial\omega_{\|})e_{\|}+(\partial\tilde{q}^{(m)}/\partial\omega_{\perp})e_{\perp}=0$. With Eqs. (4.3), (4.5), and (4.6), and substituting $e_{\|}\sim\cos\chi'$ and $e_{\perp}\sim\sin\chi'$, we thus obtain

$$\omega_0 \simeq \pm[\omega - (V^2/\omega)\sin^2\vartheta + (4V^2/\omega)\sin\vartheta\cos\vartheta\tan(\vartheta-\chi')] , \quad (4.7)$$

for the $m=\pm 1$ resonances, and

$$\omega_0 \simeq m\omega - \frac{4m}{m^2-1}V^2\sin^2\vartheta , \quad (4.8)$$

for the m-quantum resonances with $m\neq\pm 1$. Comparison of Eqs. (4.7) and (4.8) with the results from the vanishing damping continued-fraction calculation [Eqs. (5.3)–(5.7) in Ref. 1] shows that, to order V^2, exactly the same shifts are obtained using the generalized diagonalization technique, including the pump-beam-dependent term for the single quantum resonances. Note that it is exactly the first-order off-diagonal element $A=-V\sin\vartheta$ in Eq. (4.2c) that gives rise to the pump-beam-dependent shift for the single quantum resonances. By comparison, although the $m=\pm 2$ resonances also have a nonvanishing matrix element, namely, $\tilde{A}=\pm(V^2/m)\sin 2\vartheta$, this element is of second order in the interaction strength V, and contributes only to the next higher order (fourth-order) shift of the $m=\pm 2$ resonances. Generally, for small interactions, the interaction strength for the m-quantum resonances is proportional to $V^{2|m|}$ [for $|m|>2$, this follows from Eq. (4.1) only in higher-order perturbation theory], and the lowest pump-beam-dependent shift is of order $V^{2|m|}$, as was already established in Sec. IV of Ref. 1.

It deserves mention that Salwen's perturbation technique is very closely related to (but precedes, by a decade or so) the resolvent formalism, developed by Cohen-Tannoudji.[3–5] In this formalism, the resolvent of the Floquet Hamiltonian $\mathcal{H}_F$ from Eq. (2.6), projected onto the same states $|\alpha\rangle$ and $|\beta\rangle$ that are assumed in Eq. (4.1), takes the form

$$\bar{G}(q)=[q-\bar{\mathcal{H}}_0-\bar{R}(q)]^{-1} , \quad (4.9)$$

where the so-called level shift operator $\bar{R}$, as a function of the (eigen)value q, can be expanded as

$$\bar{R}=\bar{\mathcal{V}}+\mathcal{P}\mathcal{V}\frac{\mathcal{Q}}{q-\mathcal{H}_0}\mathcal{V}\mathcal{P}+\cdots , \quad (4.10)$$

and where the projection operators $\mathcal{P}$ and $\mathcal{Q}$ are defined as $\mathcal{P}=|\alpha\rangle\langle\alpha|+|\beta\rangle\langle\beta|$ and $\mathcal{Q}=1-\mathcal{P}$ (in terms of these operators, $\bar{\mathcal{H}}_0=\mathcal{P}\mathcal{H}_0\mathcal{P}$ and $\bar{\mathcal{V}}=\mathcal{P}\mathcal{V}\mathcal{P}$, where $\mathcal{H}_0$ and $\mathcal{V}$ are the operators from Eqs. (2.2) and (2.3)—for convenience we have omitted the carets). Comparison of Eq. (4.10) with Eq. (4.1) shows that the two expressions are almost identical. Indeed, the diagonal elements of $\bar{R}$ are equal to the interaction parts on the right-hand sides of Eqs. (4.2a) and (4.2b), namely, $\bar{R}_{\alpha\alpha}=-\bar{R}_{\beta\beta}=(A^2/\omega)c_m$. However, it is not clear how the resolvent formalism can be used to determine the shifts of the resonances in an absorption experiment with an arbitrary orientation of the optical pump beam relative to the magnetic fields. Specifically, the zero-oscillating field Hamiltonian $\mathcal{H}_0$ from Eq. (2.2) is no longer diagonal with respect to the states $|\pm',n\rangle$ projected along the pump beam, as follows from the transformed two-by-two matrix in Eq. (2.10b). Note that, for Salwen's Eq. (4.1) to be used, the matrix $\mathcal{H}_F$ must be diagonal also. In this case, however, the pump-beam dependence of the resonances follows *after* the eigenvalues of the Floquet Hamiltonian have been determined.

Using Salwen's technique, higher-order corrections to the shifts of the resonances may be obtained as well. To do this, one must first calculate the perturbed states $|\alpha'\rangle$ and $|\beta'\rangle$ (to order V^2) in terms of the unperturbed states $|\alpha\rangle$ and $|\beta\rangle$, and substitute the new, perturbed matrix elements $(\mathcal{H}_F)_{\mu\nu}$ (with $\mu,\nu=\alpha',\beta'$) into the right-hand side of Eq. (4.1). We will not pursue this approach here. However, we do want to show that, for weak interactions, another analytical result can be derived easily, namely for excitation far off resonance. To this end we observe that, for weak interaction, the eigenvalues of the dressed atom are given approximately by

$$q \simeq \pm\tfrac{1}{2}\omega_0 + n\omega . \quad (4.11)$$

From Fig. 2 it follows that this expression is a good approximation for the eigenvalues not only for small values of the interaction strength, but also for relatively large values of the rf Rabi frequency ω_1, as long as $\omega_0 \gtrsim \omega_1$, that is, if the static magnetic field is sufficiently stronger than the oscillating magnetic field. With Eqs. (2.16), (3.2), and (4.11), the time-averaged transition probability and absorption signal in this case become

$$P_{a'b'}=\tfrac{1}{2}\sin^2\psi, \quad S^{(0)}=\cos^2\psi , \quad (4.12)$$

where ψ is the angle between the static magnetic field and the pump beam [see Fig. 1(b)]. (Note that , in terms of the polar angles ϑ, χ, and φ in Fig. 1, $\cos\psi=\cos\vartheta\cos\varphi+\sin\vartheta\sin\chi\cos\varphi$.) Results calculated with Eq. (4.12) are shown in Fig. 3 as the thin dashed curves. It is seen that, indeed, at least for $\omega_0 \gtrsim \omega_1$, and away from the narrow multiple quantum resonances, Eq. (4.12) is a good approximation.

For comparison, the approximation given in Eq. (4.12) can also be obtained from the vanishing damping continued-fraction solution. Namely, if we approximate, to zeroth order in the interaction strength V, $\alpha=\omega_{\parallel}$ in Eq. (3.4) and $\beta_{21}=\gamma_{xz}$ in Eq. (3.5), then the time-averaged absorption signal from Eq. (3.3) becomes

$$(S^{(0)})^{\mathrm{VD}}=\frac{\cos^2\psi}{1-(1-\gamma_{xz})\sin^2\vartheta} . \quad (4.13)$$

This approximation is shown in Fig. 4, where it is seen that, indeed, in the VDCF calculation, the resonance behavior depends on the damping ratios γ_{xz} and (other than in the weak-field limit) γ_{yz}. For a damping ratio $\gamma_{xz}=1$, the VDCF result from Eq. (4.13) is identical to the result from Eq. (4.12), obtained from the generalized diagonalization technique, thereby establishing once more the equivalence of the two approaches, at least for unit damping ratios in the VDCF method.

V. CONCLUSION

We have shown how Shirley's well-known result for the time-averaged transition probability of a spin-$\frac{1}{2}$ system under the influence of a sinusoidally driven interac-

tion can be generalized to account for an arbitrary orientation of the optical pump beam in modulated magnetic resonance. Thus the time-averaged absorption signal can, as in Shirley's original method, be obtained directly from the dressed-atom energy eigenvalues of the system, which follow by diagonalization of the Floquet Hamiltonian. The results are in complete agreement with those obtained from the vanishing damping matrix continued-fraction solution of the Bloch equations from the preceding paper.[1] Although the generalized diagonalization technique is more computationally intensive than this latter VDCF solution, and damping is not as easily taken into account, the technique offers the advantage of easy visualization of the time-averaged resonance behavior of the system, namely by inspection of the three-dimensional dressed-atom energy-level diagram. Thus, for example, the various special points for which the resonances are pump-beam independent are immediately found to correspond to level crossings and saddle points in the dressed-atom energy-level diagram. As in the preceding paper, it is shown that the shifts of the various multiple quantum resonances (particularly the lowest-order shift of the single quantum resonance) depend on the orientation of the optical pump beam, unlike what is implied in some previous work on the problem.[4,6,7]

Although, in this paper, our discussion has been primarily in terms of magnetic resonance, we note once more that, as is pointed out in some detail in Refs. 1 and 12, each magnetic-resonance geometry corresponds, upon making the rotating-wave approximation, to an optical-resonance experiment. Thus, using the generalized diagonalization technique, one can obtain the time-averaged fluorescence signal for narrow-band amplitude-modulated (AM) or frequency (phase) -modulated (FM) excitation of an optical two-level system from the dressed-atom energy-level diagram using Eqs. (1.1) and (1.2) after substituting the appropriate optical variables (see Sec. II in Ref. 1). For example, the resonances of the time-averaged fluorescence signal in optical resonance for AM and FM excitation are given by the solid and dashed curves in Fig. 2, respectively, with the quantities on the axes ($\omega_{\parallel}$ and $\omega_{\perp}$) corresponding to Rabi frequency and detuning for AM excitation, and vice versa for the FM case (see Fig. 2 in Ref. 1). Indeed, Floquet techniques have been used to solve a variety of optical-resonance problems, for both two-level and multilevel systems.[13,14] In this regard, we point out that Shirley's approach is not restricted to the case of a two-level system. However, for more than two levels, more complicated expressions for the transition probabilities result, which require not only determination of the eigenvalues of the Floquet Hamiltonian, but of the eigenvectors as well.[2,13,14] In this case, it is no longer obvious how the resonance behavior of the system is determined, say, by the location of level crossings in the dressed-atom energy-level diagram. An exception is the symmetric three-level system, which, as shown by Hermann and Swain, can be treated in essentially the same way as the two-level system, resulting in an expression for the time-averaged transition probability similar to Eq. (1.1).[15] It would be of interest to obtain the appropriate generalization of their result [the same as Eq. (1.2)?] for arbitrary magnetic-resonance geometries and to investigate, as in the present work, the existence and special roles of level-crossing and saddle-point resonances.

Finally, we remark once again that, although the existence of Haroche-like level crossings in the two-level system at integer values of the parallel Rabi frequency $\omega_{\parallel}$ has been established firmly in an empirical sense (Refs. 4, 8, 1, and the present work), a formal proof of this property is still lacking.

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[1]W. M. Ruyten, preceding paper, Phys. Rev. A **42**, 4226 (1990). For some summary results, published earlier, see W. M. Ruyten, in *Coherence and Quantum Optics VI*, edited by L. Mandel and E. Wolf (Plenum, New York, 1990), p. 1035, and W. M. Ruyten, J. Phys. B **23**, L207 (1990).

[2]J. H. Shirley, Phys. Rev. **138**, B979 (1965).

[3]C. Cohen-Tannoudji, in *Cargèse Lectures in Physics*, edited by M. Lévy (Gordon and Breach, New York, 1968), Vol. 2, p. 347; C. Cohen-Tannoudji, J. Dupont-Roc, and C. Fabre, J. Phys. B **6**, L214 (1973); **6**, L218 (1973).

[4]T. Yabuzaki, S. Nakayama, Y. Murakami, and T. Ogawa, Phys. Rev. A **10**, 1955 (1974).

[5]M. Allegrini and E. Arimondo, J. Phys. B **4**, 1008 (1971); E. Arimondo and G. Moruzzi, *ibid.* **9**, 709 (1976); **9**, 723 (1976).

[6]W. A. McClean and S. Swain, J. Phys. B **9**, 1673 (1976); see also S. Swain, J. Phys. A **6**, 192 (1973); Adv. At. Mol. Phys. **22**, 387 (1986); F. T. Hioe and J. H. Eberly, Phys. Rev. A **11**, 1358 (1975); A. D'Andrea, *ibid.* **39**, 5143 (1989).

[7]D. T. Pegg, J. Phys. B **10**, 1027 (1977); see also D. T. Pegg and G. W. Series, *ibid.* **3**, L33 (1970); D. T. Pegg, *ibid.* **6**, 241 (1973); **6**, 246 (1973).

[8]T. Yabuzaki, Y. Murakami, and T. Ogawa, J. Phys. B **9**, 9 (1976).

[9]A. Messiah, *Quantum Mechanics* (North-Holland, Amsterdam, 1963), Vol. II, Eq. (C.74).

[10]N. Tsukada, T. Koyama, and T. Ogawa, Phys. Lett. **44A**, 501 (1973); N. Tsukada, T. Koyama, and T. Ogawa, J. Phys. B **7**, 799 (1974).

[11]H. Salwen, Phys. Rev. **99**, 1274 (1955).

[12]W. M. Ruyten, Phys. Rev. A **39**, 442 (1989).

[13]S.-I. Chu and T.-S. Ho, Israel. J. Chem. **24**, 237 (1984); T.-S. Ho and S.-I. Chu, J. Phys. B **17**, 2101 (1984); S.-I. Chu, in *Advances in Atomic and Molecular Physics*, edited by D. Bates and B. Bederson (Academic, New York, 1985), Vol. 21, p. 197; S.-I. Chu, in *Advances in Multi-Photon Processes and Spectroscopy* (World Scientific, Singapore, 1987), Vol. 2, p.

175.
[14]G. C. Stey and G. Gusman, Phys. Lett. **50A**, 111 (1974); D. R. Dion and J. O. Hirschfelder, Adv. Chem. Phys. **35**, 265 (1976); J. V. Moloney and W. J. Meath, J. Phys. B **11**, 2641 (1978).
[15]J. Hermann and S. Swain, J. Phys. A **9**, 1947 (1976); J. A. Hermann and S. Swain, J. Phys. B **10**, 2111 (1977).