Quasienergy states of a two-level atom in a strong lowfrequency electromagnetic field

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The behavior of a two-level atomic system placed in a strong low-frequency electromagnetic field is studied in the framework of the adiabatic approximation. The field strength is not assumed to be small in comparison with the separation of the atomic levels, as was the case in earlier papers on this subject. The region of manyphoton resonances is considered, and also the behavior of the populations and quasienergies of the states in the intervals between resonances. The results are compared with the available exact solutions of the problem, obtained with a computer for particular values of the field parameters. It is found that good agreement between the adiabatic approximation and the exact solutions is already attained beginning with the region of three-photon resonance. The results of an existing experiment in such fields are discussed. Solutions in the adiabatic approximation are constructed both for adiabatic and for instantaneous turning-on of the field.

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1. INTRODUCTION

The adiabatic approximation has previously been applied' to the case of a two-level atom placed in a lowfrequency electromagnetic field with its frequency ω in the neighborhood of a many-photon resonance. By means of an appropriate generalization the same method can be applied to an arbitrary case, including nonresonance conditions, since the adiabatic approximation requires only that the frequency ω be small in comparison with the separation ω_{ba} between the levels. Furthermore the nonresonance condition can be due either to an increase of the detuning from resonance or to an increase of the intensity of the external field, so that the amplitude of the field can be comparable with the distance ω_{ba} between the atomic levels in question. Hitherto the approach to the problem in the nonresonance case has been based exclusively on numerical calculations for particular values of the parameters.^{2,3} There is also an analytic solution⁴ in the nonresonance case when the external field is weak and turned on adiabatically.

The adiabatic approximation makes it possible to deal analytically with the effects of antiresonance terms in the perturbation, which play a very important role for strong fields or nonresonance frequencies.

It is well known that for numerical reasons the quasiclassical approximation gives good agreement even for small quantum numbers, including even the ground state of the system. Analogously, the adiabatic approximation, being an analog of the quasiclassical approximation with coordinates replaced by the time and momenta by the energy, can be applied even when the frequency ω of the external field is not much smaller than the level separation ω_{ba} , but of the same order of magnitude. Important deviations begin to occur only when ω is larger than ω_{ba} . As we shall see, this is well confirmed by comparison of the analytic calculations in the adiabatic approximation with exact calculations made with a computer.

In Sec. 2 we examine the quasienergy states of a two-level atom in a low-frequency strong electromag-

netic field on the basis of the adiabatic approximation. In Sec. 3 the probabilities of level populations are determined both for adiabatic and for sudden turning on of the perturbation. The Conclusion gives a comparison of the analytic and exact calculations, and also an explanation, on the basis of this theory, of experimental data on the populations of levels under the conditions considered here.

2. CALCULATIONS OF QUASIENERGIES IN THE ADIABATIC APPROXIMATION

In the energy-spin representation the wave function of a two-level atom in a monochromatic field V(t)= $V_0 \sin(\omega t + \varphi)$ satisfies the equation (1)

$$i\partial u/\partial t = \hat{H}u, \quad \hat{H} = -i/_2 \omega_{ba} \sigma_3 + V(t) \sigma_1.$$
 (1)

Here ω_{ba} is the separation between the levels, V_0 is the amplitude of the transition matrix element, and σ_i are the Pauli matrices.

The Hamiltonian \hat{H} is invariant under the "screw" transformation

$$\hat{T}_{\pi} = \sigma_{3} \exp\left(\pi \frac{\partial}{\partial \tau}\right),$$

which is a translation by the amount π in the variable $\tau = \omega t + \varphi$ and a simultaneous rotation through the angle π around the third axis in the energy-spin space. Therefore the solutions of Eq. (1) can be chosen so that they are eigenfunctions of the operator \hat{T}_{τ} :

$$\widehat{T}_{\pi}u(\tau) = \sigma_{3}u(\tau+\pi) = \exp(i\pi\varepsilon/\omega)u(\tau).$$
(2)

Since applying the transformation \hat{T}_{τ} twice gives a translation by a whole period, these solutions describe states with a definite quasienergy ϵ (Ref. 5).

Hereafter we shall consider the case of a low-frequency field, $\omega \ll \omega_{ba}$, for which we can use the adiabatic solution of Eq. (1)

$$u_{1}(\tau) = \begin{pmatrix} \cos^{4}/_{2}\theta \\ -\sin^{4}/_{2}\theta \end{pmatrix} \exp\left(i\int\Omega d\tau\right),$$

$$u_{2}(\tau) = \begin{pmatrix} \sin^{4}/_{2}\theta \\ \cos^{4}/_{2}\theta \end{pmatrix} \exp\left(-i\int\Omega d\tau\right).$$
(3)

Here the notation is

$$tg \theta = q \sin \tau, \quad \Omega = \frac{\omega_{ba}}{2\omega} (1 + q^2 \sin^2 \tau)^{\frac{1}{2}}, \quad q = \frac{2V_a}{\omega_{ba}}.$$

The function Ω has the complex branch points

 $\tau_n = \pi n \pm i \operatorname{arsh}(1/q).$

Let us write the general solution in the interval $(0, \pi)$ as a superposition of the linearly independent basic solutions shown in Eq. (3):

$$u(\tau) = C_1 u_1(\tau) + C_2 u_2(\tau).$$
 (4)

We continue this solution analytically along the real axis into the next interval $(\pi, 2\pi)$, avoiding the branch points τ_1 (for the function u_1) and τ_1^* (for u_2). Then, using the conditions (2) we get the following equations for the coefficients C_1 and C_2 :

$$C_{1}((1-R^{2})^{\prime_{h}}e^{is}-e^{i\pi\epsilon/*})+C_{2}Re^{-is}=0,$$

-C_{2}((1-R^{2})^{\prime_{h}}e^{-is}+e^{i\pi\epsilon/*})+C_{4}Re^{is}=0. (5)

Here the new notation is

$$H = \exp\left\{2i\int_{0}^{\infty}\Omega d\tau\right\}$$
$$= \exp\left\{-\frac{\omega_{ba}}{\omega}\frac{D\left[\left(1+q^{2}\right)^{-\frac{1}{2}}\right]}{(1+q^{2})^{\frac{1}{2}}}\right\},$$
$$S = \int_{0}^{\pi}\Omega d\tau = \frac{\omega_{ba}}{\omega}\left(1+q^{2}\right)^{\frac{1}{2}}E\left(\frac{q}{(1+q^{2})^{\frac{1}{2}}}\right),$$
(6)

and E and D are the complete elliptic integrals of the second and third kinds, respectively.

The quantity S is a quasiclassical phase taken on the interval $(0, \pi)$, and R is analogous to the quasiclassical coefficient of above-barrier reflection.⁶ In the low-frequency range considered $(\omega \ll \omega_{ba})$ the phase S is large, and $R \ll 1$ (with $q \ll \omega_{ba}/\omega$, but either $q \gtrless 1$).

Equating the determinant of the system (5) to zero, we get the following equation for the quasienergy:

$$\sin(\pi \varepsilon/\omega) = (1-R^2)^{\frac{1}{2}} \sin S, \tag{7}$$

which has two solutions, ε and $\varepsilon' = \omega - \varepsilon$. Far from resonance (i.e., when S is not close to an odd multiple of $\pi/2$) we can neglect the small quantity R, and the quasienergy is given by⁷

$$\varepsilon \approx \frac{\omega}{\pi} S = \frac{\omega_{ba}}{\pi} (1+q^2)^{\frac{1}{2}} E\left(\frac{q}{(1+q^2)^{\frac{1}{2}}}\right) .$$
(8)

In a weak field $(q \ll 1)$ the first terms of the expansion in powers of q are identical with the result given by Meli-kyan⁸ (see also Ref. 9)

$$\epsilon \approx 1/2 \omega_{ba} (1+q^2/4-3q^4/64)$$

In the neighborhood of a many-photon resonance the phase is

$$S = \frac{i}{2\pi} (2K+1) + \Delta S, \quad \Delta S \rightarrow 0$$

and the quasienergy is of the form¹

$$\varepsilon = \frac{\omega}{2} \cdot (2K+1) \pm \frac{\omega}{\pi} \left((\Delta S)^2 + R^2 \right)^{\frac{1}{2}}.$$
 (9)

The dependence of the quasienergy on the field, which is determined by the expression (7), is shown in Fig. 1. The solid lines show the branch of the spectrum corresponding to the state that arises from the lower state of the atom when the field is turned on adiabatically. The breaks in this curve at certain values of the amplitude q of the external field correspond to many-photon resonances.





3. CALCULATION OF POPULATIONS OF THE LEVELS

Corresponding to the two values of the quasienergy, ε and ε' , there are two sets of coefficients C_1 and C_2 :

$$C_{i}^{\pm} = [\frac{1}{2} \{ 1 \pm [(1 - R^{2}) (1 + R^{2} \operatorname{tg}^{2} S)^{-1}]^{\frac{1}{2}} \}]^{\frac{1}{2}},$$
(10)

$$C_{2}^{\pm} = \pm \operatorname{sign} \cos Se^{iS} \left[\frac{1}{2} \left\{ 1 \mp \left[(1 - R^{2}) (1 + R^{2} \operatorname{tg}^{2} S)^{-1} \right]^{\frac{1}{2}} \right\} \right]^{\frac{1}{2}},$$

with the choices of signs corresponding to Eq. (7).

When the interaction is turned off adiabatically $(q \rightarrow 0, R \rightarrow 0, \Omega \rightarrow \omega_{ba}/2\omega)$ the solution $u^{\dagger} = C_1^{\dagger}u_1 + C_2^{\dagger}u_2$ goes over into the wave function

$$\begin{pmatrix} 1\\ 0 \end{pmatrix} \exp\left(i\frac{\omega_{ba}}{2}t\right),$$

which describes an atom which is in the lower state.

It suffices to define the wave function of a quasienergy state on the interval $(0, \pi)$ only, since it can be constructed easily on other intervals by means of the condition (2).

The probability of transition from the lower state to the upper one during adiabatic turning on of the perturbation is determined by the square of the lower component of the spinor $u^*(\tau)$ and is a periodic function of τ with period π :

$$W(\tau) = \frac{1}{2} \left\{ 1 - \left(\frac{1 - R^2}{1 + R^2 \log^2 S} \right)^{\frac{1}{12}} \cos \theta - \sin \theta \frac{R}{(1 + R^2 \log^2 S)^{\frac{1}{12}} \cos S} \cos \left(S - 2 \int_0^{\tau} \Omega \, d\tau \right) \right\} \quad (0 \le \tau \le \pi).$$
(11)

The last term describes rapid oscillations with frequency $\sim \omega_{ba}$, with small amplitude except in the neighborhood of a many-photon resonance, where these oscillations are simply modulated by the function $\sin\theta(\tau)$. The time appears in the transition probabilities (11) only in combination with the phase constant of the field $(\tau = \omega t + \varphi)$ so that averaging over the phase constant of the field (which is equivalent to averaging over time with a fixed phase constant) leads to a transition probability which is independent of the time:

$$W = \frac{1}{2}(1 - \mathcal{P}), \tag{12}$$

where \mathcal{P} is given by the expression

$$\mathscr{P} = \frac{2}{\pi} \left[\frac{1 - R^2}{1 + R^2 \operatorname{tg}^2 S} \right]^{\frac{1}{2}} \frac{1}{(1 + q^2)^{\frac{1}{2}}} K\left(\frac{q}{(1 + q^2)^{\frac{1}{2}}}\right).$$
(13)

Here K is the complete elliptic integral of the first kind. At a resonance point $\mathscr{P} = 0$ and $W = \frac{1}{2}$. That the populations are equal at the resonance point is a fact not related to the adiabatic approximation, and is true also for the exact solution of the problem.

Far from resonances the quantity R can be neglected and

$$W = \frac{1}{2} \left\{ 1 - \frac{2}{\pi} \frac{1}{(1+q^2)^{\frac{1}{h}}} K\left(\frac{q}{(1+q^2)^{\frac{1}{h}}}\right) \right\} .$$

For small q this expression goes over into the perturbation-theory result,⁴ as could be expected.

As is well known, by differentiating the quasienergy with respect to parameters one can obtain the average values of various physical quantities.⁷ For example, the average over a period of the probability of finding the system in the upper state is given by

$$W = \frac{1}{2} \left(1 - \frac{\partial \varepsilon}{\partial (1/2\omega_{ba})} \right) ,$$

which, when Eqs. (6) and (7) are used, leads to the expression (12), as it should.

Let us now turn to the case of instantaneous turningon of the perturbation. In this case the solution is a superposition of the quasienergy states u^* and u^- . The coefficients of this superposition are found from the condition that at a certain initial time the system is in the lower state.

Omitting the intermediate steps, we find that the average, taken over the phase constant φ and the time the observation is made, of the probability that the upper level is occupied is

$$W = \frac{1}{2}(1 - \mathcal{P}^2),$$
 (14)

where the value of \mathcal{P} is given by Eq. (13).

Figures 2 and 3 show the probability that the upper level is occupied, calculated by means of Eqs. (14) and (13) (solid curve) and the results of calculations with a computer (from Ref. 2) (dashed curve). Figure 2 corresponds to a field of moderate intensity, with q= 0.5, while Fig. 3 corresponds to a rather strong field, with q = 1.54.

At points where the condition

$$\frac{\omega_{ba}}{\omega} (1+q^2)^{\nu_b} E\left(-\frac{q}{(1+q^2)^{\nu_b}}\right) = \frac{2k+1}{2} \pi,$$
(15)



FIG. 2. Probability of occupation of the upper level, averaged over the phase of the field and the time of observation, as a function of ω/ω_{ba} for a relatively weak field, q=0.5. The solid curve shows the analytic solution found according to Eqs. (13) and (14) in the adiabatic approximation, while the dashed curve represents the exact solution obtained with a computer in Ref. 2. The peaks correspond to the one-photon resonance, the three-photon resonance, and so on.



FIG. 3. The same as in Fig. 2, but for a very strong field, q=1.54.

holds (with k an integer), the probability of occupation for each level is $\frac{1}{2}$. The condition (15) gives the positions of many-photon resonances in the case of a strong field and does not depend on the way the external field is turned on.

4. DISCUSSION OF THE RESULTS

Comparing the numerical and analytic solutions in Figs. 2 and 3, we see that beginning with even the three-photon resonance the adiabatic approximation agrees very well with the exact solution. Furthermore it is hard to continue the exact solution into the range of very small frequencies because of the strong oscillations of the probability as a function of the time.

We note that in the resonance region the agreement between the approximate and exact solutions is better than in the nonresonance region. As could be expected, the agreement becomes worse with increasing frequency ω . It is not bad, however, even in the neighborhood of the fundamental harmonic k = 0, especially when the strength q of the perturbation is not too large. At small frequencies, i.e., actually behond the region of three-photon resonance, the analytical formulas (14), (13) agree well with the exact solution over the entire range of frequencies, including the nonresonance intervals, in which the transitions between the upper and lower states are given not by the many-photon matrix elements, as in the neighborhoods of resonances, but by the ordinary nonresonance matrix elements, which do not require that there be energy conservation with the number of absorbed photons taken into account. For example, in the case of weak fields, the populations in the nonresonance intervals are determined by the perturbation-theory term of first order in q. It corresponds to the expansion of the elliptic integral K in powers of q with $q \ll 1$.

As can be seen from Figs. 2 and 3, with increasing multiplicity the resonances rapidly become extremely narrow, if only the perturbation q is not too large. As q increases, there is a gradual raising of the curve of W toward the horizontal asymptotic line $W = \frac{1}{2}$, as could be expected.

There is a relatively simple expression for the probability W(t) of the upper level being occupied, averaged over the phase at which the field is turned on and over small-scale fluctuations as a function of the time of observation t (of the order $1/\omega$), at an exact resonance of arbitrary multiplicity:

$$W(t) = \frac{1}{2} - \frac{2}{\pi^2 (1+q^2)} K^2 \left(\frac{q}{(1+q^2)^{\frac{1}{2}}} \right) \cos\left(\frac{2\omega t}{\pi} \arcsin R \right).$$
(16)

From this it can be seen that for instantaneous turning on the probability undergoes large-scale fluctuations, which are the analog of the Rabi fluctuations for the case of a strong field. The amplitude of these oscillations is given by

$$\mathscr{A} = \frac{2}{\pi^2 (1+q^2)} K^2 \left(\frac{q}{(1+q^2)^{1/2}} \right) .$$

For example, for q = 1.54 we find that the amplitude of the oscillations is $\mathcal{A} = 0.26$. We note that in the adiabatic approximation the amplitude of the oscillations does not depend on the mulitplicity of the resonance, i.e., on the frequency ω . Their frequency is given by

$\Omega = 2\omega \pi^{-1} \arcsin R.$

It does, of course, depend strongly on the multiplicity of the resonance. In particular, for q = 1.54 and a fivephoton resonance it is $\Omega = 0.115\omega$. We see that it is small compared with ω , and still smaller compared with $\omega_{ba} = 5\omega$, as it should be.

Table I shows the values of the Stark shifts calculated from Eq. (15); these are the shifts of the positions of the maximum values $W = \frac{1}{2}$ relative to the unperturbed positions $\omega = \omega_{ba}/k$ (k an odd integer). They are compared with the exact values of the Stark shifts as found in Ref. 2 by means of a computer. The results given are for the case of a strong field, with q = 1.54. In this case, by calculating the elliptic integral we get from Eq. (15) a simple formula for determining the Stark shift of a resonance of arbitrary multiplicity k: $\Delta \omega = 0.44 \omega_{ba}/k$. We see that, except for the case k = 1, all the other Stark shifts agree well with the exact values calculated with the computer. The differences between the approximate and the exact values of the Stark shift are proportional to $1/k^2$, which is quite natural, since the phases of the adiabatic functions are determined with fractional accuracy $\sim 1/k = \omega/\omega_{ba}$. We emphasize that in the case in question the Stark shifts are comparable with the distance between the unperturbed levels.

An experiment relating to our present subject is described in Ref. 10. An excited level of the sodium atom was split with a constant magnetic field (the Zeeman effect). A state with definite magnetic quantum number m was populated by the absorption of polarized light of optical frequency. An alternating magnetic field of the radio frequency between the states m and m+1 was applied. It played the part of the strong monochromatic field of our present case. Measurements were made on the spontaneous emission of light of definite polarization, corresponding to transition from the state m + 1. The constant magnetic field was varied, so as to have various values of the distance ω_{ba} between the levels of the two-level system in question. The resulting curves for the intensity of the spontaneous radiation as a function of the constant magnetic field are very similar to the curves shown in Figs. 2 and 3. A quantitative comparison between theory and experiment is not possible, however, since in this experiment the steady magnetic field was only approximately constant, and the alternating field produced res-

TABLE I. Stark shifts of many-photon resonances in the strong-field case (q=1.54).

Multiplicity	Adiabatic approximation	Exact	Multiplicity	Adiabatic	Exact
of resonance		solution	of resonance	approximation	solution
1	0.44	0.15	5	0.090	0.094
3	0.150	0.160	7	0.063	0.065

onances of the even harmonics.

Accordingly, we can conclude that the adiabatic approximation correctly describes the behavior of a twolevel system in a strong electromagnetic field, with high quantitative accuracy achieved already in the neighborhood of the three-photon resonance.

We have given the adiabatic solutions both for the case of adiabatic turning on of the perturbation and for instantaneous turning on. What are the criteria for the instantaneous or adiabatic nature of the turning on? In the case of the ordinary Rabi problem for fields small in comparison with the distance between the levels, the criteria are¹¹: $\Delta T \ll 1$ or $\Delta T \gg 1$, where T is the duration of the turning on, and Δ is the detuning of the resonance. Therefore, in the immediate neighborhood of a resonance the turning on is instantaneous, and far from all resonances it is of course adiabatic. In the case of a strong field the same criteria hold, but the resonance detuning must be considered with the Stark shift included, because in a very strong field this shift is large and decidedly alters the position of the resonance. This follows from the general statement that the turning on is adiabatic when the quasienergy levels do not approach each other too closely in their quasienergies, so that they do not become intermixed during the turning on of the perturbation.

If the levels have finite width, in particular the natural width, the minimum detuning will be determined by their width, so that in the stated criteria we must replace \triangle with the level width. For radiofrequency fields at resonance the turning on is instantaneous. This statement is confirmed by the observation of the well known Autler-Townes effect in the radiofrequency range.¹² Far from resonance, however, as Δ increases the turning on becomes adiabatic. Although the level width can vary widely, for example if we consider transitions between hfs sublevels or the Zeeman sublevels of the ground state of an atom, from both the experimental and the theoretical points of view we can conclude rather generally that the turning on of the perturbation is instantaneous at resonances and adiabatic away from them.

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On the possibility of measuring the population, orientation, and alignment relaxation times by the photon echo method

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Stimulated photon echo in a resonant gaseous medium is investigated theoretically. It is found that the echo intensity and the polarization depend markedly on the relaxation characteristics repsonsible for the relaxation of the population, magneto-dipole (orientation relaxation) moment, and quadrupole (alignment relaxation) moment of the resonance levels. These dependences make it possible to carry out experimental measurements of these relaxation times by the photon echo method. The observed effect should stimulate the setting up of new experiments on the photon echo in gases with the aim of measurement of the above-mentioned characteristics of the resonance levels.

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A photon echo is formed in a medium after passage of two exciting light pulses separated by a time interval τ_s , and represents spontaneous coherent radiation from a superradiant state created by the first exciting pulse.¹ By increasing τ_s and observing experimentally the attenuation of the intensity of the photon echo, we can effectively measure the relaxation characteristics of the resonance transition by the photon echo method. Thus, in experiments on the photon echo in gases (see, for example, Refs. 2-5), this method was used successfully to find relaxation characteristic $\gamma^{(1)}$ responsible for the damping of the component of the optical coherence matrix, which is proportional to the polarization of the medium. There are also theoretical works,^{6,7} showing the possibility of measurement by the photon echo method of other relaxational characteristics $\gamma^{(\eta)}(\varkappa)$ \neq 1) of the optical coherence matrix.

In the present work, it is shown that we can also measure the relaxation characteristics of the resonance levels themselves by the photon echo method: the relaxation of the population, orientation, and alignment. For this purpose, it is proposed to observe not the ordinary but the so-called stimulated photon echo.

The stimulated echo was predicted and observed by $Hahn^8$ in the radiofrequency range. This echo is cre-

ated at an instant of time approximately equal to $2\tau_1 + \tau_2$ upon successive passage through the medium of three exciting pulses, separated by the respective time intervals τ_1 and τ_2 . We note that the three-pulse method of excitation of resonant media has already progressed at the present time from the radiofrequency to the optical range (see, for example, Trgd. 9–11). In particular, stimulated photon echo in ruby has been observed in the work of Samatsev *et al.*¹¹ Thus, the fact of the possibility of observation of a stimulated echo in the optical range raises no doubts whatever.

In the present work, we have carried out a calculation of the intensity and polarization of the stimulated photon echo produced in a gaseous medium. In this case we have taken into account the degeneracy of the resonance levels of the considered transition and the effect of elastic depolarizing atomic (molecular) collisions on the interaction of the atoms (molecules) of the gas with the resonance electromagnetic field. The calculations that have been carried out show that it is possible to select such experimental conditions under which the damping of the components of the maximum of intensity of the stimulated photon echo in the direction of polarization of the third exciting pulse and in the perpendicular direction will be determined either only by the relaxation times of the population of the resonan-