Perturbation theory for the quasienergy spectrum of atoms in a strong monochromatic field

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The quasienergy spectrum of atoms is calculated, including higher-order perturbation-theory terms in the interaction between the atom and the field which, among other things, allows an analysis to be made of the range of validity of the first nonvanishing order theory in calculations of resonance and nonresonance perturbations of atomic levels in strong laser fields. The case of an isolated level in a nonresonance elliptically polarized field is investigated, and results are reported of calculations of polarizability and hyperpolarizability for alkali-metal and noble-gas atoms at the frequencies of pulsed solid-state lasers. Frequencies that are small in comparison with the ionization potential or comparable with it are considered, as well as the transition to the constant field. A calculation is given of the quasienergy spectrum in a resonance field, and of the corrections to the multiphoton excitation probability. It is shown that, both in the resonance and the nonresonance cases, the contribution of the higher-order terms becomes appreciable when the field is much smaller than the characteristic intra-atomic field. A separate analysis is given of the quasienergy spectrum of the hydrogen atom which has a number of specific features connected with the presence of a permanent dipole moment in excited states.

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1. INTRODUCTION

The method of quasienergies[1] provides us with a convenient formalism for calculating the spectrum of quasistationary atomic states in strong fields. Substantial progress has been made in recent years in the experimental investigation of this subject.[2] Since calculations of the quasienergy spectrum require the solution of the time-dependent Schrödinger equation, perturbation theory in the interaction between the atom and the field is at present the only method suitable for quantitative calculations involving real atoms. The usual procedure is to confine the analysis to the first nonvanishing order of perturbation theory. In contrast to the constant field, where the perturbation theory series are asymptotic, the question of the convergence of series in a time-dependent field has not as yet been investigated which, among other things, is due to the fact that the terms in the series have an appreciable dependence on the frequency and polarization of radiation. However, it is clear that the inclusion of the higher-order terms in the case of the time-dependent field may turn out to be much more important than for the constant field: The dynamic polarizability determines the quadratic level shift and is zero for a number of frequencies, whereas the hyperpolarizability is not zero at these frequencies.

In a sufficiently weak field (for which perturbation theory calculations are possible), one can define three qualitatively different cases that may occur, depending on the structure of the spectrum of the particular atom:

(1) isolated level in a field of nonresonance frequency;

(2) resonance case when the energy difference between the levels $|i\rangle$ and $|j\rangle$ and the energy k of the photons satisfy the condition $|E_i-E_j|\approx k\hbar\omega$; and

(3) a group of closely lying levels in a nonresonance field (degenerate states of hydrogen, isolated level with $J\neq 0$ in an elliptically polarized field; closely spaced levels of a multiplet for which the matrix element of the interaction with the field is of the order of the multiplet splitting).

Various variants of the perturbation theory can be used to calculate the quasienergy spectrum in such cases,[3–6] and the most satisfactory of them is that proposed in[6]. The latter is a natural generalization of the time-independent perturbation theory to the case of fields that are periodic functions of time. The quasienergy wave function for the Schrödinger equation with a periodic perturbation $V(r,t)$ is

$$\Psi_{\mathscr{E}}(\mathbf{r},t)=\exp\{-i\mathscr{E}t\}U_{\mathscr{E}}(\mathbf{r},t),\quad U(t+2\pi/\omega)=U(t).$$

In this expression, $\mathscr{E}$ is the quasienergy and U satisfies the condition

$$\hat{\mathscr{H}}(t)U_{\mathscr{E}}(\mathbf{r},t)=\left\{H^{(0)}(\mathbf{r})+V(\mathbf{r},t)-i\frac{\partial}{\partial t}\right\}U_{\mathscr{E}}(\mathbf{r},t)=\mathscr{E}U_{\mathscr{E}}(\mathbf{r},t),\qquad(1)$$

which is formally identical with the time-independent equation with the Hamiltonian $\hat{\mathscr{H}}(t)$.[1)] The functions U are defined in the Hilbert space $R\oplus T$, where R is the usual space of stationary function $f(\mathbf{r})$ and T is the space of periodic functions of time with period $2\pi/\omega$. This fact was used in[6] to put forward a method for solving (1) that was analogous to the time-independent case except that the integration with respect to $\mathbf{r}$ was augmented by averaging over the field period T during calculation of the probabilities and averages. It is clear that the complete set of solutions of (1) with $V(\mathbf{r},t)\equiv 0$ (basis functions) is

$$\mathscr{E}_{nq}^{(0)}=E_n^{(0)}+q\omega,\qquad U_{nq}^{(0)}(\mathbf{r},t)=\varphi_n(\mathbf{r})e^{iq\omega t},\quad q=0,\pm 1,\ldots,\qquad(2)$$

where φ_n are the eigenfunctions of the operator $H^{(0)}$ with energies $E_n^{(0)}$. Perturbation theory on the unperturbed basis (2) enables us to consider all the physically interesting cases from a unified point of view. In particular, cases (2) and (3) given above correspond to degeneracy (or the presence of closely spaced levels) in the unperturbed spectrum $\mathscr{E}_{nq}^{(0)}$ and, therefore, the correct functions in the "zero-order approximation" are combinations of the functions $U_{nq}^{(0)}$ corresponding to close levels $\mathscr{E}_{nq}^{(0)}$.

In this paper, we calculate the quasienergy spectrum (QES) of atoms for the typical cases indicated above, taking into account higher-order terms of perturbation theory which, among other things, enables us to establish the limits of applicability of the first-nonvanishing-order perturbation theory in calculations of the perturbation of atomic states in a strong field. Numerical calculations have been carried out for alkali metals, noble gases, and hydrogen because they are of particular interest for experimental studies. To calculate the matrix elements, we used the Green function for the optical electron of an atom in the approximation of the model potential,[7] which enabled us to obtain an analytic expression for the various quantities in which we are interested in the form of rapidly converging hypergeometric series.

2. QUASIENERGY OF AN ISOLATED LEVEL IN A NONRESONANCE FIELD

Consider an isolated level of energy $E_n^{(0)}$ in a field of optical frequency $\omega < E_n^{(0)}$, which corresponds to the ground and low-lying excited states of atoms. If the total angular momentum of the atom is $J \neq 0$, then degeneracy in the magnetic quantum number is removed in an elliptically polarized field, and the splitting of the $2J+1$ magnetic sublevels of the level $E_n^{(0)}$ must be analyzed by perturbation theory in the presence of degeneracy. The quadratic Stark effect was investigated for this case by Zon.[8] For the sake of simplicity in the calculation of the corrections $\sim F^4$, we shall confine our attention to the case $J=0$.

The interaction operator $V(\mathbf{r}, t)$ in (1) in the case of an elliptically polarized wave propagating along the z axis has the form (in the nonrelativistic dipole approximation)

$$V(\mathbf{r}, t) = \mathbf{F}(t)\mathbf{r} = -\mathbf{A}(t)\mathbf{P} + {}^1/_2\mathbf{A}^2(t), \tag{3}$$

where

$$\mathbf{F}(t) = -\frac{\partial \mathbf{A}}{\partial t}, \quad \mathbf{A}(t) = \left\{\frac{F}{\omega}\sin\omega t, -\varepsilon\frac{F}{\omega}\cos\omega t, 0\right\},$$

and $-1 \leqslant \varepsilon \leqslant 1$ is the elliptic polarization such that $\varepsilon = 0$ correspond to linear and $\varepsilon = +1$ (-1) to the right-handed (left-handed) circular polarization. Although the two forms of (3) are equivalent in the dipole approximation,[9] one of the two may turn out to be technically more convenient in particular cases.

The perturbation theory series for the quasienergy

$$\mathscr{E}_n = E_n^{(0)} + \mathscr{E}_n^{(2)} + \mathscr{E}_n^{(4)} + \ldots$$

is analogous to the series for the energy in time-independent perturbation theory. The expression for the correction $\mathscr{E}_n^{(k)}$ has a form analogous to that for $E_n^{(k)}$, but summation in the intermediate states is carried out over the unperturbed basis (excluding the state $U_{n,0}^{(0)}$) and the matrix elements are averaged over the period of the field. All that remains is to note that the presence of the two terms in the operator

$$V(\mathbf{r}, t) = V(\mathbf{r})e^{i\omega t} + V^+(\mathbf{r})e^{-i\omega t}$$

leads to a substantial increase in the number of matrix elements in the series for $\mathscr{E}_n$ and U_n, as compared with the time-independent case. For example,

$$\mathscr{E}_n^{(2)} = \sum_{k,r}{}' \frac{\langle\langle U_{n,0}^{(0)} | V(\mathbf{r}, t) | U_{k,r}^{(0)}\rangle\rangle \langle\langle U_{k,r}^{(0)} | V(\mathbf{r}, t) | U_{n,0}^{(0)}\rangle\rangle}{E_n^{(0)} - \mathscr{E}_{k,r}^{(0)}}$$
$$= F^2 \frac{1+\varepsilon^2}{4} \sum_k{}' \frac{2(E_n^{(0)} - E_k^{(0)}) |\langle\varphi_n | z | \varphi_k\rangle|^2}{(E_n^{(0)} - E_k^{(0)})^2 - \omega^2} = -\frac{1+\varepsilon^2}{4}\alpha_n(\omega)F^2, \tag{4}$$

where α_n is the dynamic polarizability. Similarly, the quantity $\mathscr{E}_n^{(4)}$ can be written in the form

$$\mathscr{E}_n^{(4)} = -\frac{1}{4!}\gamma_n(\omega, \varepsilon)F^4,$$

where γ_n is the dynamic hyperpolarizability which can be expressed in terms of the components of the nonlinear susceptibility tensor of the atom,[2)] χ_{ijkl}:

$$\gamma_n(\omega, \varepsilon) = {}^9/_4\{(1-\varepsilon^2)\chi_{xxxx} + 8\varepsilon^2\chi_{xxyy}\}. \tag{5}$$

The formulas for χ include a combination of fourth-order matrix elements.[10] They are rather unwieldy and will not be reproduced here.

The components of the tensor χ are normalized as follows:

$$\chi_{xxxx}(-\omega; \omega, \omega, -\omega)|_{\omega=0} = 3\chi_{xxyy}(-\omega; \omega, \omega, -\omega)|_{\omega=0} = \chi_0,$$

where $\chi_0 = 222.1875$ for hydrogen. Moreover,

$$\gamma(\omega, \varepsilon=0)|_{\omega=0} = {}^3/_8\gamma_0, \quad \gamma(\omega, \varepsilon=\pm1)|_{\omega=0} = \gamma_0,$$

where γ_0 is the static hyperpolarizability which determines the fourth-order correction to the energy in a constant electric field $\mathbf{F}$. The factor $\frac{3}{8}$ for $\varepsilon = 0$ corresponds to the value of $\mathbf{F}^4(t)$ averaged over the period. It follows from (4) and (5) that $\mathscr{E}_n^4$ is more frequency-dependent than $\mathscr{E}_n^{(2)}$. In particular, in addition to single-photon poles of intermediate p-states, which are also present in $\alpha_n(\omega)$, the quantity γ_n has two-photon resonance singularities near which there is a considerable increase in $\mathscr{E}_n^{(4)}$. Moreover, when $|\varepsilon| \neq 1$, the quantity γ_n has two-photon resonances on virtual S states that are absent in the case of circular polarization, which shows that $\mathscr{E}_n^{(4)}$ depends on the degree of elliptical polarization.

Table 1 shows the results of a numerical calculation of $\alpha(\omega)$, $\chi_{\parallel} = \chi_{xxxx}$ and $\chi_{\perp} = \chi_{xxyy}$ for the ground states of hydrogen, alkali metals, and noble gases at the frequencies of the ruby ($\omega_R = 0.0656$) and neodymium ($\omega_N = 0.043$) lasers, and for $\omega = 0$. The ratio of $\mathscr{E}_n^{(2)}$ to $\mathscr{E}_n^{(4)}$

TABLE 1. Polarizabilities and components of the nonlinear susceptibility tensor of rank four for the ground states of atoms.

Atom	$\omega=0$		$\omega_N=0.043$			$\omega_R=0.0656$		
	α	χ_0	α	$\chi_\perp$	$\chi_\parallel$	α	$\chi_\perp$	$\chi_\parallel$
H	4.5	222.1875	4.55	82.967	251.577	4.618	85.78	264.62
He	1.49	9.5	1.49	3.18	9.55	1.49	3.24	9.76
Ne	5.2	51.0	5.25	16.9	51.1	5.27	17.1	51.9
Ar	13.1	390	13.2	130	408	13.3	140	442
Kr	18.2	800	18.3	292	898	18.5	338	1065
Xe	27.1	2020	27.4	705	2150	27.9	800	2460
Li	166	2.7(5)	277	−8.8(4)	2.4(5)	2.46(3)	−5.5(8)	−1.7(9)
Na	162	1.04(6)	234	2.5(5)	1.0(6)	575	9.7(7)	1.1(8)
K	290	3.48(6)	607	1.3(7)	2.6(7)	−1320	7.8(7)	1.5(8)
Rb	318	5.42(6)	692	1.1(8)	1.7(8)	−1206	6.2(7)	1.3(8)
Cs	396	1.18(7)	1117	−6.1(7)	−4.4(8)	−775	–	–

Note. $\chi_0=\chi_\parallel(\omega=0)$. Numbers of parenthesis show powers of ten: $(n)=10^n$. Values of $\chi_\perp$ and $\chi_\parallel$ for Cs at $\omega=0.0656$ are not given because of the presence of the two-photon resonance $6S$–$9D$.

in the case of hydrogen and the noble gases is not very different from that in the case of a constant field. This is due to the fact that ω is small in comparison with the energy difference between the ground and the first excited states. In the alkali metals, the difference is substantial and corrections $\sim F^4$ become appreciable for fields $F \gtrsim 10^6$ V/cm, where, depending on ε, the quantity γ_n may change sign. This was, in fact, the case, for example, in the lithium atom at the frequencies ω_N and $2\omega_N$. Consequently, departures from the first-nonvanishing-order perturbation theory can be seen even in fields $F \gtrsim F_{\text{crit}}$ much smaller than the "interatomic" field F_{at}, and for the alkali metals F_{crit} is less than in the case when $\omega=0$. This is connected with two factors: firstly, with the large magnitudes of the matrix elements of order N for atomic potentials with long-range Coulomb asymptotics, which are much greater than the corresponding values for rapidly decreasing potentials[3)] (the statistical hyperpolarizability of hydrogen exceeds the hyperpolarizability of a particle in a short-range potential by a factor of 40[12]), and this difference increases with increasing N. Secondly, at frequencies ω that are comparable with the frequencies of atomic transitions, the ratio γ_n/α_n is found to increase quite substantially (and, correspondingly, there is a reduction in F_{crit}) in comparison with the static case because the expression for $\gamma_n(\omega)$ includes the product of three energy denominators of the form $E_k^{(0)} - E_k^{(0)} \pm \omega$, the magnitude of which decreases with increasing ω.

Thus, it is impossible to formulate a combined criterion for estimating the contribution of higher-order perturbation theory terms in a time-dependent field because $\mathscr{E}_n$ depends appreciably on both ω and ε. For example, in the region of normal dispersion ($\omega < \omega_{in} = E_i^{(0)} - E_n^{(0)}$, where $E_i^{(0)}$ is the energy of the state nearest to $|n\rangle$ state, the transition to which is allowed by selection rules), the quantities α and γ can be expanded into series in ω^2 which converge for $\omega < \omega_{in}$ and $\omega < \omega_{in}/2$, respectively. In that case, the first $N \approx \omega_{in}/\omega$ terms of the perturbation-theory series give the asymptotic expansion of $\mathscr{E}_n$ in the parameter F^2 to within the N-th term, inclusively. At high frequencies, the asymptotic character of the series is violated. (See the analogy with the level shift in a short-range potential.[12])

Let us now consider the frequency region $\omega \gtrsim I_n$, when single-photon ionization of the state under consideration is possible. This situation is characteristic for highly excited states in the light field. For sufficiently large n, the electron is weakly bound to the atom, and the unperturbed value of the quasienergy must include the mean oscillatory energy in the wave field $\mathscr{E}_{\text{osc}} = \mathbf{F}^2(t)/2\omega^2$, which may be comparable with I_n. To calculate $\mathscr{E}_n$ in this case, it is convenient to use the operator $V(\mathbf{r}, t)$ in (3), written in terms of the vector potential. The term $\frac{1}{2}\mathbf{A}^2$ contains the terms $\frac{1}{2}(1+\varepsilon^2)F^2 = \mathscr{E}_{\text{osc}}$, and we therefore included it in $H^{(0)}(\mathbf{r})$. This corresponds to the replacement of $\mathscr{E}_{nq}^{(0)}$ in (2) by $\mathscr{E}_{nq}^{(0)\prime} = \mathscr{E}_{nq}^{(0)} + \mathscr{E}_{\text{osc}}$, and the remaining part of V is discussed in terms of perturbation theory. In the second order, we have (for the sake of simplicity, we confine ourselves to the case $\varepsilon=0$)

$$\mathscr{E}_n^{(2)\prime} = \frac{F^2}{\omega^2} \sum_k{}' \frac{2\omega_{kn} |\langle n|p_z|k\rangle|^2}{\omega_{kn}{}^2 - \omega^2} \equiv -\frac{1}{4}\alpha_n{}'(\omega) F^2.$$

It is important to note that the formulas for the higher-order corrections have the same form as for the interaction $\mathbf{F}(t)\mathbf{r}$, provided we make the replacement $Fz \to Fp_z/\omega$. Hence, the term $\frac{1}{2}\mathbf{A}^2(t)$ provides a contribution only in the second order and determines the specific dependence of $\mathscr{E}_n$ on ω at high frequencies. Using the sum rule, we can readily show that

$$\mathscr{E}_n^{(2)\prime} = -F^2/2\omega^2 + \mathscr{E}_n^{(2)},$$

where $\mathscr{E}_n^{(2)}$ is given by (4) and, consequently, in practical calculations for small ω ($\omega \ll I_n$), the expression given by (4) is convenient. For $\omega \gg I_n$, the quantity $\mathscr{E}_n^{(2)}$ decreases rapidly with increasing n. In particular, for the nS states of hydrogen,[3]

$$\mathscr{E}_n^{(2)\prime} = \mathscr{E}_{\text{osc}} \frac{4}{3n^3\omega^2}. \tag{6}$$

Similarly, it can be shown that $\mathscr{E}_n^{(4)} \approx F^4/n^5\omega^6$. Thus, in contrast to the constant field, where the level shift increases rapidly with increasing n, the quasienergy spectrum in a time-dependent field reaches the asymptotic value $\mathscr{E}_{n,0}^{(0)\prime}$ as n increases. Moreover, when $\omega > I_n$, the quantity $\mathscr{E}_n^{(2)\prime}$ has an imaginary part which determines the level width and is related to the single-photon ionization cross section σ_n. It follows from the optical theorem that

$$\operatorname{Im} \mathscr{E}_n^{(2)\prime} = \frac{\alpha\omega}{8\pi} \sigma_n F^2,$$

where α is the fine-structure constant, When $\omega > \frac{1}{2} I_n$, $\operatorname{Im} \mathscr{E}_n^{(4)}$ gives the two-photon level width, and so on.

The case $\omega \sim I_n$ is of particular interest, especially in the case of resonance multiphoton ionization, when the asymptotic approximation of the form given by (6) is invalid for $\mathscr{E}_n^{(2)\prime}$. Table 2 shows numerical values of the "renormalized" polarizabilities α_n' for a series of xenon levels in the field produced by the neodymium laser (it includes only states with zero angular momentum components along the polarization vector). Despite the complexity of the xenon spectrum, which results in a

TABLE 2. Polarizabilities α_n' of excited states of Xe at neodymium-laser frequency.

State	ν_n	α_n'	State	ν_n	α_n'
$6s[1^1/_2]_2$	1.89	2070	$7d[1^1/_2]_1$	4.63	$-275+i160$
$6p[2^1/_2]_3$	2.38	-220	$5f[1^1/_2]_1$	4.96	$-80+i49.3$
$5d[1^1/_2]_2$	2.52	2080	$9s[1^1/_2]_2$	4.97	$87.6+i0.53$
$7s[1^1/_2]_2$	2.94	354	$9p[2^1/_2]_3$	5.45	$480+i27$
$7p[2^1/_2]_3$	3.42	$815+i97$	$8d[1^1/_2]_1$	5.90	$-13+i52$
$6d[1^1/_2]_2$	3.75	$-630+i240$	$6f[1^1/_2]_1$	5.95	$8.8+i29.7$
$8s[1^1/_2]_2$	3.95	$238+i6.2$	$10s[1^1/_2]_2$	5.98	$92.6+i0.22$
$4f[1^1/_2]_1$	3.96	$-1150+i85$	$7f[1^1/_2]_1$	6.95	$154+i19$
$8p[2^1/_2]_3$	4.44	$430+i57$	$11s[1^1/_2]_2$	6.98	$94.3+i0.12$

considerable difference in the values of α_n' for sufficiently closely lying energy levels, the quantity α_n' rapidly decreases with increasing "principal quantum number" $\nu_n = (-2E_n^{(0)})^{-1/2}$, as expected. The individual deviations are due to the approach to resonance with the lower-lying level. However, the conditions of single-photon resonance $V_{nn'} \gtrsim |E_n^{(0)} - E_{n'}^{(0)} \pm \omega|$ are not satisfied for any of these states for field strengths up to $F \approx 5\times10^7$ V/cm.

Eleven-photon ionization of xenon with resonance at $\omega = 9450$ cm^{-1} was observed in[13] for $F \approx 5\times10^7$ V/cm and laser linewidth ~2 cm^{-1}. It follows from Table 2 that the minimum detuning corresponding to the two-photon resonance on the field-shifted levels $5p^{6}\,{}^1S_0$ and $9p[2\frac{1}{2}]_3$ is then ~130 cm^{-1}.

For frequencies in the neighborhood of which $\alpha_n(\omega) \sim 0$, and for $\omega \approx |E_n^{(0)} - E_k^{(0)}|$, where $|k\rangle$ are virtual states with the same parity as $|n\rangle$, corrections to $\mathscr{E}_n^{(2)}$ that are connected with the inclusion of quadrupole terms in $V(\mathbf{r}, t)$, and are of the order of $(\alpha\omega)^2$, may turn out to be appreciable. Numerical calculations show that the contribution of nondipole terms is appreciable when the frequency differs by more than 0.01–0.1 cm^{-1} from the above frequencies, i.e., in fields ~10^6 V/cm the contribution of terms ~F^4 is more important than corrections due to retardation.

The above results are not valid at low frequencies when

$$\omega \lessdot |W_{nq;nq'}| = \left| {\sum_{k,r}}' \frac{\langle\langle U_{n,q}^{(0)}|V|U_{k,r}^{(0)}\rangle\rangle\langle\langle U_{k,r}^{(0)}|V|U_{n,q'}^{(0)}\rangle\rangle}{\mathscr{E}_{nq}^{(0)} - \mathscr{E}_{kr}^{(0)}} \right|, \tag{7}$$

where W is the composite matrix element for the transition between the harmonics of the level $E_n^{(0)}$. In particular, when $\omega \to 0$, the expressions given by (4) and (5) do not become identical with the formula for the static Stark effect because of the dependence on the degree of elliptical polarization. Similarly to the case of closely-lying levels in the time-independent perturbation theory, the correct QES function corresponding to the "zero-order approximation" must have the form of a superposition of harmonics when the inequality given by (7) is satisfied:

$$U_n(\mathbf{r}, t) = \sum_q c_q U_{nq}^{(0)}(\mathbf{r}, t). \tag{8}$$

The matrix element $W_{nq,nq'}$ is nonzero for $q' = q$, $q\pm2$, where

$$W_{nq,nq} = (1+\varepsilon^2)W_n, \qquad W_{nq,nq\pm2} = (1-\varepsilon^2)W_n,$$

$$W_n(\pm\omega) = \frac{F^2}{4}\sum_k \frac{|\langle\varphi_n|z|\varphi_k\rangle|^2}{E_n^{(0)} - E_k^{(0)} \pm \omega}.$$

For small ω, the dependence of W_n on ω can be neglected, and the expression for U_n turns out to be

$$U_n(\mathbf{r}, t) = \exp\left\{i(1-\varepsilon^2)\frac{W_n}{\omega}\sin 2\omega t\right\}\varphi_n(\mathbf{r}). \tag{9}$$

Under these conditions, $\mathscr{E}_n = E_n^{(0)} - 2(1+\varepsilon^2)W_n$, which is identical with (4) for $\omega \to 0$, and the weights of the harmonics are determined by the Bessel functions $c_q = J_q[(1-\varepsilon^2)W_n/\omega]$. It is readily seen that the time-dependent argument of the exponential in the function $\Psi_{\mathscr{E}_n}(\mathbf{r}, t)$ becomes $E_n^{(0)} - \frac{1}{2}\alpha_n(0)F^2$ when $\omega \to 0$, and corresponds to the inclusion of the Stark shift in a constant field $\mathbf{F}$. The expression for the correction $\mathscr{E}_n^{(4)}$ can be obtained in a similar way when (7) is satisfied. In a circularly polarized field, the harmonics of a given state are "nondegenerate" in the sense that $W_{nq,nq\pm2} = 0$. The various features which arise for $\omega \lesssim W_n$ and, in particular, the large weight of the harmonics $U_{nq}^{(0)}$ with $q \pm 0$, are determined by the "linearly polarized component" of the wave ($c_q = \delta_{q0}$ for $\varepsilon = \pm1$) which is connected with the fact that an atomic electron can absorb an arbitrary number of linearly polarized photons without a change in the quantum numbers l and m.

An interesting feature of the behavior of the level with $J \neq 0$ in an elliptically polarized field is the coalescence of some of the split $(2J+1)$ magnetic sublevels at low frequencies. In particular, $U_n(\mathbf{r}, t)$ is a superposition of $(2J+1)$ harmonics $\exp(iq\omega t)$ with $q = 0, \pm1, \ldots, \pm J$ in the case of circular polarization and provided (7) is satisfied. The secular determinant splits into the product of determinants of order J and $J+1$, which correspond to the two independent sets of magnetic sublevels with even and odd m. Thus, U_n is a superposition of states with different m and, when $\omega \ll W_n$, the conserved quantum number is the component of the angular momentum along the direction of $\mathbf{F}$. It is readily verified that the $2J+1$ roots of the determinant then coincide in pairs, which corresponds to degeneracy in the constant field with respect to the sign of the component along the vector $\mathbf{F}$. At low frequencies, the separation between the coalescing levels is ~ω^2.

3. PERTURBATION THEORY IN THE PRESENCE OF RESONANCES

The two-level resonance approximation[14] (the generalization to the case of resonance between two multiplets is given in[4]) is usually employed in the presence of resonances and is equivalent to the zero-order approximation of the perturbation theory for the QES.[6] However, in fields of the order of 10^6–10^7 V/cm, the corrections representing the presence of nonresonance levels become very important and this leads to a modification of both the quasienergy spectrum and the weights of the quasienergy harmonics of the resonance level.

Since the unperturbed states $|n\rangle$ and $|n'\rangle$ with energies satisfying the relation $E_{n'}^{(0)} - E_n^{(0)} \approx k\omega$, $k = 0, 1, 2, \ldots,$

correspond to degenerate states of the operator $\mathscr{H}(t)$ in (1),[4)] the QES function is a superposition of degenerate states in (2) in the zero-order approximation:

$$\Psi_{\mathscr{E}}(\mathbf{r},t)=e^{-i\mathscr{E}t}\{c_n\varphi_n(\mathbf{r})+c_{n'}\varphi_{n'}e^{ik\omega t}\}, \tag{10}$$

where $\mathscr{E}$ is the root of the secular equation

$$\|V_{ij}+(\mathscr{E}_i^{(0)}-\mathscr{E})\delta_{ij}\|=0 \quad i,j=n,n'. \tag{11}$$

In this expression, V_{ii} is the correction to the quasienergy of the i-th isolated level, where, however, the states $|n\rangle$ and $|n'\rangle$ are absent from the sums over the intermediate states. The nondiagonal element $V_{nn'}$ can also be written as a series in F, beginning with the term F^k. Since the shift of the quasienergy of an individual level is quadratic in F, and resonance effects are important only when terms $\sim F^k$ are included, one is in practice interested in cases with $k\leqslant 2$ when the weights of the harmonics $c_{n,n'}$ in (10) can be comparable in magnitude.

A. Single-photon resonance

We shall now take into account in V_{ij} terms up to the second order in F, inclusive, and will write the solution of (11) in the form

$$\begin{gathered}\mathscr{E}=\mathscr{E}_n^{(0)}+\Delta_\pm;\\ 2\Delta_\pm=-{}^1/_4F^2(\alpha_n+\alpha_{n'})+\delta\pm[a^2F^2+(\delta+{}^1/_4\alpha_nF^2-{}^1/_4\alpha_{n'}F^2)^2]^{1/2},\\ \delta=\mathscr{E}_{n'}^{(0)}-\mathscr{E}_n^{(0)},\quad a=\langle n|z|n'\rangle,\quad \acute{\alpha}_i=\sum_{p\neq n,n'}\frac{2\omega_{p_i}|\langle i|z|p\rangle|^2}{\omega_{p_i}^2-\omega^2}.\end{gathered} \tag{12}$$

It follows from (12) that the quantities $\Delta_\pm$ are linear functions of F when $\delta\ll aF$, and the shift of the quasienergy corresponds to the shift of the isolated levels $|n\rangle$ and $|n'\rangle$ when $\delta\gg aF$. Inclusion of second-order corrections is unimportant when $a\gg(\alpha_n-\alpha_{n'})$ and, in the opposite case, the formulas of the two-level approximation are invalid.

As an example, we have calculated the parameters of the resonance between the $3d_3$ and $5V$ levels of the Xe atom (in the Paschen notation) for which the detuning at the frequency of the neodymium laser is $\delta=74$ cm^{-1}:

$$a=1.37,\quad \alpha_{3d}=740,\quad \alpha_{5V}=-1470+i\cdot 39.$$

The relative deviation $\{\Delta_\pm-\Delta_\pm(\alpha_i\equiv 0)\}/\Delta_\pm$ is ~ 1 in a field $F\approx 10^7$ V/cm, and calculations based on perturbation theory are valid in this case for $F\lesssim 5\times 10^6$ V/cm.

The quantity $|V_{nn'}|^2$ in (11) is proportional to the parity of the K-photon excitation of the level $|n'\rangle$ in a weak field. It is clear that, when higher-order corrections are included, $V_{nn'}$ can be written in the form

$$V_{nn'}=A^{(K)}F^K+B^{(K)}F^{K+2}+\ldots,$$

where $B^{(K)}$ is a combination of matrix elements of order $(K+2)$, which corresponds to the inclusion of virtual processes in the re-emission of the photon. The following table lists the calculated values of $A^{(1)}$ and $B^{(1)}$ for the transition between the ground state $nS_{1/2}$ and the first excited state $nP_{3/2}$ of alkali-metal atoms:

Atom:	Li	Na	K	Rb	Cs
$A^{(1)}$:	0.35	0.37	0.43	0.44	0.47
$B^{(1)}\cdot 10^{-4}$:	−1.7	−1.7	−0.83	−1.6	−1.5

The negative value of B leads to a slower increase in the probability with increasing F, as compared with the results of the first-nonvanishing-order perturbation theory, and this corresponds to the "smoothing" of the resonance when the nonresonance levels are taken into account. Zaretskiĭ and Kraĭnov[15] have obtained in the adiabatic approximation the expression for the probability of K-photon excitation in the two-level system:

$$W_K\sim F^{2K}\exp(-\gamma KF^2),$$

where $\gamma=\frac{1}{2}$. The considerable increase in the parameter γ in atoms ($\gamma=2B/A\approx 10^4$) and the associated reduction in the critical field F_{crit}, for which the first nonvanishing order of perturbation theory is valid, is of the same origin as the reduction in F_{crit} in comparison with F_{at} for the level shift, which was discussed in Sec. 2.

We note that generalization of the results of Zaretskiĭ and Kraĭnov[15] to the case of real atoms[21] leads to a negative value for γ (thus, for the $1S\to 2S$ transition in hydrogen, $\gamma=-446$[21]) although the numerical values of the pre-exponential factor in W_K are in reasonable agreement with perturbation-theory calculations. The negative value of γ shows that the precision of the method used in[21] is insufficient for calculations of the deviation from perturbation theory in resonance transitions in atoms. This may be due to the fact that transitions through different intermediate states [see[14], Eq. (53.10) and thereafter], which compete with the direct $1\to 2$ transition, and whose contribution increases with increasing F, were neglected in[21].

B. Two-photon resonance

This case is formally different from the foregoing only by the fact that $V_{nn'}$ is a composite second-order matrix element. The functional dependence $\Delta(F)$ remains quadratic in form. However, in contrast to the isolated level, the "polarizability" of this quasienergy state is given by

$$\begin{aligned}\alpha_+&=-4(\Delta_+-\delta)/F^2,\\ \alpha_-&=-4\Delta_-/F^2,\end{aligned}$$

and is a function of the field (for fixed detuning $\delta=E_{n'}^{(0)}-E_n^{(0)}-2\omega$). When $F^2\ll\delta$, $\alpha_+\to\alpha_{n'}(\omega)$, $\alpha_-\to\alpha_n(\omega)$:

$$\begin{gathered}\alpha_\pm={}^1/_2\left(\alpha_n'(\omega)+\alpha_{n'}'(\omega)\mp\frac{4\delta}{F^2}\right)\\ \pm\left[{}^1/_4\left(\alpha_n'-\alpha_{n'}'-\frac{4\delta}{F^2}\right)^2\right.\\ \left.+|W_{nn'}|^2\right]^{1/2},\\ \alpha_i'(\omega)=\sum_k\frac{\omega_{ki}|\langle i|z|k\rangle|^2}{\omega_{ki}^2-\omega^2},\\ W_{nn'}=\sum_{k\neq n,n'}\frac{\langle n|z|k\rangle\langle k|z|n'\rangle}{\omega_{kn'}+\omega}.\end{gathered} \tag{13}$$

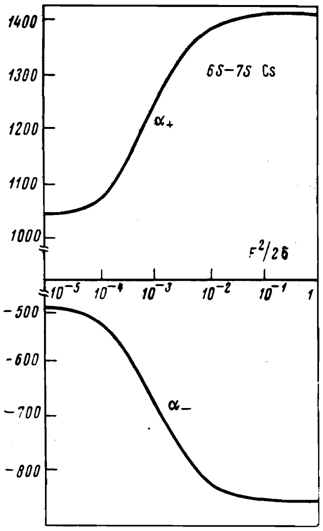

FIG. 1. "Polarizabilities" $\alpha_\pm$ (in atomic units) as functions of $F^2/2\delta$ for the two-photon resonance between the $6S_{1/2}$ and $7S_{1/2}$ levels of Cs.

It follows from (13) that resonance effects do not lead to a change in the polarizability of the states $|n\rangle$ and $|n'\rangle$ when $|W_{nn'}| \ll |\alpha'_n - \alpha'_{n'}|$.

As an example, Fig. 1 shows graphs of $\alpha_+(4\delta/F^2)$ for the resonance $6S_{1/2}$ and $7S_{1/2}$ levels of cesium. Mixing is important for these states and, when $\delta \ll F^2$, one observes the characteristic "repulsion" between closely lying levels in the external field. When $\delta \gg F^2$, we have

$$\alpha_- \to \alpha_{6S}(\omega = {}^1\!/_2\omega_{6S,\,7S}) = 1050,\quad \alpha_+ \to \alpha_{7S}(\omega = {}^1\!/_2\omega_{6S,\,7S}) = -495,$$
$$W_{6S,\,7S} = -838.$$

Nonresonance corrections which, in this case, are comparable with terms $\sim F^2$ in a field $F \approx 5\times 10^6$ V/cm can be included by analogy with the procedure used in the case of the single-photon resonance.

It thus follows from the above calculations, performed for a number of typical cases, that the validity of the resonance perturbation theory is restricted to the same range of field strengths as in the nonresonance case.

4. QUASIENERGY SPECTRUM OF EXCITED STATES OF HYDROGEN

The particular feature of hydrogen is the presence of a permanent dipole moment in excited states, and this leads to a linear Stark effect in a constant field. In a time-dependent field $\mathbf{F}(t)$, effects connected with the presence of the permanent dipole moment are important only for frequencies ω that are comparable with the change in the frequencies of atomic transitions due to the linear Stark effect ($\omega \lesssim V\langle nlm|\mathbf{F}\cdot\mathbf{r}|nl'm'\rangle$) but can be ignored in a rapidly-varying field where the quasienergy spectrum is determined by the interaction between the induced dipole moment $\mathbf{p} \sim \alpha\mathbf{F}(t)$ and the field. We note that the quadratic shift and the level splitting were investigated in[3,16] for $V \ll \omega$, whereas the case $V \gg \omega$ was investigated in[17,20] for linear and circular polarizations, neglecting terms quadratic in F. The analysis given below is valid for an arbitrary relationship between V and ω, which, in particular, enables us to investigate the transition from the linear to the quadratic Stark effect (and vice versa) as a function of the relation between F and ω.

A. Circularly polarized wave

In this case, the problem can be reduced to the time-independent problem by transforming to a system of coordinates that rotates with the frequency of the field:

$$U_{\mathscr{E}}(\mathbf{r}, t) = \exp\{i\varepsilon\omega\hat{L}_z t\}\varphi_{\mathscr{E}}(\mathbf{r}),\qquad \varepsilon = \pm 1,$$

where $\varphi_{\mathscr{E}}$ satisfies the time-independent equation

$$\{H^{(0)}(\mathbf{r}) + Fx + \varepsilon\omega\hat{L}_z - \mathscr{E}\}\varphi_{\mathscr{E}}(\mathbf{r}) = 0. \tag{14}$$

In the rotating system, $E_n^{(0)}$ splits into $(2n-1)$ sublevels with different m: $E_n^{(0)\prime} = E_n^{(0)} + m$ (henceforth, we shall assume that $\varepsilon = +1$). When $V \ll \omega$, sublevels with different m can be considered independently, and the result is that the level $E_n^{(0)}$ splits in the field into n^2 sublevels with definite values of m, the parity, shift and splitting of which are $\sim F^2$. When $V \gtrsim \omega$, the sublevels with different m must be considered in terms of the perturbation theory for close levels, and $U_{\mathscr{E}}$ is a superposition of harmonics with $m = 0, \ldots, \pm(n-1)$.

To simplify the laborious diagonalization of matrices, we shall confine our attention to $n = 2$. The state $|2p0\rangle$ then remains isolated, and we have the quadratic shift:

$$\mathscr{E}_{2p0} = E_n^{(0)} - {}^1\!/_2\alpha_{2p0}(\omega)F^2.$$

The polarizability α is calculated with the aid of the Coulomb Green function:

$$\alpha_{2p0}(\omega) = \omega^{-2}(P(\nu_+) + P(\nu_-) - 1),$$
$$P(\nu) = \frac{2\nu^2}{(2+\nu)^2(3-\nu)}\,{}_2F_1\left(1, -2-\nu; 4-\nu; \left(\frac{2-\nu}{2+\nu}\right)^2\right),\quad \nu_\pm = (|E_n^{(0)}| \mp \omega)^{-1/2}. \tag{15}$$

When $\omega = 0$, we have $\alpha = 156$, which corresponds to the constant in the quadratic Stark effect.[18] (In the parabolic set of coordinates in which $\mathbf{F}$ lies along one of the axes, the level $|2p0\rangle$ corresponds to the state with $n_1 = n_2 = 0$ for which the linear Stark effect is absent.)

The $|2s\rangle$ and $|2p,\pm 1\rangle$ states are mixed by the field and

$$U_{\mathscr{E}}(\mathbf{r}, t) = c_0|2s\rangle + c_1|2p, 1\rangle e^{i\omega t} + c_{-1}|2p, -1\rangle e^{-i\omega t}. \tag{16}$$

The solution of the secular equation for $\mathscr{E}$, when the terms $\sim F^2$ are included, have the form ($\Delta = E_n^{(0)} - \mathscr{E}$)

$$(\Delta = E_n^{(0)} - \mathscr{E})$$
$$\Delta_{1,2} = \pm\beta \mp \frac{\omega}{2\beta}(W_{11} - W_{-1-1}) - \frac{1}{2}(W_{11} + W_{-1-1})$$
$$+ \left(\frac{3F}{2\beta}\right)^2 \{W_{11} + W_{-1-1} + W_{1-1} + W_{-11} - 2W_{00}\},$$
$$\Delta_3 = -W_{00} - \left(\frac{3F}{2\beta}\right)^2 \{W_{11} + W_{-1-1} + W_{1-1} + W_{-11} - 2W_{00}\},\quad \beta = \sqrt{\omega^2 + 9F^2}, \tag{17}$$

where

$$W_{ij} = F^2 \sum_{k\neq i,j} \frac{\langle i|x|k\rangle\langle k|x|j\rangle}{E_j^{(0)\prime} - E_k^{(0)\prime}}$$

are the composite matrix elements between states with $m = 0, \pm 1$ in (16). Similarly, (15) can be expressed in

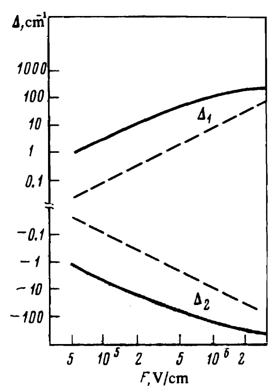

FIG. 2. Quasienergies of $n=2$ states in hydrogen as functions of field F (double log scale). Solid line—$\omega=20\ \text{cm}^{-1}$, broken lines—$\omega=1000\ \text{cm}^{-1}$.

terms of the hypergeometric functions ${}_2F_1$. Figure 2 shows the function $\Delta_{1,2}(F)\mp\omega$ for $\omega=20$ and $1000\ \text{cm}^{-1}$. The change in the slope of the curves for $\omega=20\ \text{cm}^{-1}$ corresponds to the transition from the quadratic to the linear level shift as F increases.

It is clear that $\Delta_{1,2}$ are linear functions of F for $\omega<3F$ and are determined by the interaction between the field and the degenerate states of the level $|n\rangle$. Inclusion of the interaction between the shells gives corrections $\sim F^2$. When $\omega\ll|E_n^{(0)}-E_{n'}^{(0)}|$, the dependence of W_{ij} on frequency can be neglected, and we have the following expression for Δ_i:

$$\begin{aligned}\Delta_{1,2}&=\mp\beta+3F^2\{31-27F^2/\beta^2\}\pm\omega,\\ \Delta_3&=6F^2\{10+27F^2/\beta^2\},\\ \Delta_4&=E_2^{(0)}-\mathscr{E}_{2p0}=78F^2.\end{aligned}\tag{18}$$

When $\omega=0$, (18) leads to the formulas for the static Stark effect.[18] In particular, for the coefficient c_i in (16), which corresponds to the root Δ_1, we have $c_0=1/\sqrt{2}$, $c_{-1}=-c_1=\frac{1}{2}$ (state with parabolic quantum numbers $n_1=m=0$, $n_2=1$, where m is the component along the vector $\mathbf{F}$). Similarly, the root Δ_2 corresponds to the state with $n_1=1$, $n_2=m=0$, and Δ_3 corresponds to the level $n_1=n_2=0$, $m=1$. We note that the levels $\mathscr{E}_{2p0}$ and $\mathscr{E}_3=E_2^{(0)}-\Delta_3$ coalesce for $\omega\to 0$. Thus, some of the split n^2 sublevels coalesce as the frequency is reduced, and when $\omega\ll 3nF/2$, the number of split components is $\frac{1}{2}n(n+1)$, as in the case of the constant field.

In the opposite limit ($F\ll\omega$), the roots Δ_i and the corresponding wave functions have the form

$$\begin{aligned}\Delta_{1,2}&=\pm\omega\pm\frac{9F^2}{2\omega}-W_{\pm1,\pm1},\quad U_{\mathscr{E}}(\mathbf{r},t)=\varphi_{2p,m=\pm1}e^{\pm i\omega t},\\ \Delta_3&=-W_{00},\ U_{\mathscr{E}}(\mathbf{r})=\varphi_{2s}(\mathbf{r}),\end{aligned}$$

which is in agreement with the results in[3,16].

For low frequencies $\omega\ll E_n^{(0)}-E_{n'}^{(0)}$, we can obtain an analytic solution for an arbitrary level $E_n^{(0)}$ if we take into account the analogy between (14) and the Schrödinger equation for an atom in crossed electric and magnetic fields. In particular, to within terms representing the interaction between the field and states with the same n, we have[19,20]

$$\mathscr{E}_{nn'n''}=E_n^{(0)}+(n'+n'')\,\omega\sqrt{1+\left(\frac{3nF}{2\omega}\right)^2},\quad n',n''=-\frac{n-1}{2},\dots,+\frac{n-1}{2}.\tag{19}$$

The quantum numbers n', n'' correspond to the components of the vectors $\mathbf{J}_\pm=\frac{1}{2}(\mathbf{L}\pm\mathbf{A})$ along the directions of $\mathbf{\Omega}_\pm=\omega\mathbf{e}_z\pm 3nF\mathbf{e}_x/2$. In these expressions, $\mathbf{L}$ is the angular momentum and $\mathbf{A}$ is the Runge–Lenz vector in the subspace of states with given n.[19] The number of split levels $(2n-1)$ remains the same as in the case of the linear Stark effect. The characteristic parameter $\gamma^2=(3nF/2\omega)^2$ in (19), which determines the transition from the square shift to the linear shift as F increases, can be written in the form

$$\gamma^2=9\mathscr{E}_{\text{osc}}/4I_n,$$

where $\mathscr{E}_{\text{osc}}=e^2F^2/2m\omega^2$ is the mean energy of an electron in the field and I_n is the ionization potential.5)

In the above approximation, the QES function is given as a superposition of unperturbed functions $\psi_{ki_1i_2}^{(0)}(\mathbf{r})$ in parabolic coordinates with the quantization axis lying along the direction of propagation of the wave (the z axis):

$$\begin{aligned}\Psi_{nn'n''}(\mathbf{r},t)=e^{-i\mathscr{E}_{nn'n''}t}\sum_{i_1,i_2=-k}^{k}D_{n'i_1}^{k}\left(0,-\alpha-\frac{\pi}{2},0\right)\cdot\\ \cdot D_{n''i_2}^{k}\left(0,\alpha-\frac{3\pi}{2},0\right)\psi_{ki_1i_2}^{(0)}e^{i(i_1+i_2)\omega t},\end{aligned}\tag{20}$$

where $k=(n-1)/2$ is the modulus of the vectors $\mathbf{J}_\pm$, i_1, i_2 are their components along the z axis, $\alpha=\operatorname{arc\,cot}\gamma$, and D is the finite rotation matrix.

It follows from (20) that, when $\gamma\sim 1$, the wave function $\Psi_{\mathscr{E}}(\mathbf{r},t)$ includes the superposition of harmonics with $m=0,\dots,\pm(n-1)$, the component of the momentum is not a quantum number of the QES (although each individual harmonic has a definite component $m=i_1+i_2$ along the z axis), and the QES quantum numbers are the components n' and n'' introduced above. It is readily verified that, when $\gamma\gg 1$, the function $\psi_{nn'n''}$ becomes identical with $\psi_{k,n',-n''}^{(0)}$ with fixed value of the component $m=n'-n''$ along the direction of $\mathbf{F}$ (the x axis), and $\mathscr{E}_{nn'n''}$ is given by the usual formula for the linear Stark effect. In the opposite limiting case ($\gamma\ll 1$), we have

$$\Psi_{nn'n''}(\mathbf{r},t)=\exp\left\{-i\left[E_n^{(0)}-\frac{9}{8}\frac{mn^2F^2}{\omega}\right]t\right\}\psi_{k,-n',-n''}^{(0)}(\mathbf{r}),$$

where $m=-n'-n''$ is the component along the z axis.

The formulas given by (19) and (20) are valid for $\gamma\gtrsim 1$, when we can neglect terms $\sim F^2$ that correspond to the inclusion of the interaction between the shells, and lead to complete removal of the degeneracy of the level $E_n^{(0)}$. The algebraic method used in[19] does not take into account correctly terms that are quadratic in F because, when $\gamma<1$, the parabolic symmetry of the problem is lost and the use of the algebra of the operators $\mathbf{J}_\pm$ is ineffective (the correction $\sim F^2$ obtained by Demkov *et al.*[19] is incorrect because it was erroneously assumed in their derivation that the second-order composite matrix element was diagonal in the parabolic quantum numbers6)).

B. Linearly polarized wave

In a linearly polarized optical-frequency wave ($V\ll\omega$), the level $E_n^{(0)}$ splits into $\frac{1}{2}n(n+1)$ sublevels, the shift of which is quadratic in F, and the wave functions are superpositions of the zero-order harmonics (2) corresponding to the degenerate states of the level $|n\rangle$.[16] When $V \gtrsim \omega$, the QES function in the zero-order approximation should be sought in the form

$$U_n(\mathbf{r}, t) = \sum_{\gamma} \sum_{q} c_{\gamma q} \varphi_{n\gamma}(\mathbf{r}) e^{iq\omega t},$$

which takes into account the "degeneracy" of harmonics with different q in (2). For the $\varphi_{n\gamma}$ we take the hydrogen functions in parabolic coordinates with $z \parallel \mathbf{F}$: $\equiv \{n_1, n_2, m\}$. Since the absorption of a photon of plane-polarized radiation by an atom is not accompanied by a change in the component of the angular momentum along the quantization axis, the secular determinant is infinite in the indices q and q' (in contrast to circular polarization):

$$\det \| [(E_n^{(0)} - \mathscr{E} - q\omega)\delta_{\gamma\gamma'} + W_{\gamma\gamma'}(\omega) + W_{\gamma\gamma'}(-\omega)]\delta_{qq'} + V\delta_{\gamma\gamma'}\delta_{q,q'\pm 1} + W_{\gamma\gamma'}(\omega)\delta_{q,q'-2} + W_{\gamma\gamma'}(-\omega)\delta_{q,q'+2} \| = 0, \tag{21}$$

where $V = \frac{3}{2}nF(n_1 - n_2)$ and $W_{\gamma\gamma'}$ is the composite matrix element of the form given by (7) in which we can neglect the dependence on ω at low frequencies. The matrix element $W_{\gamma\gamma'}$ is not diagonal in γ and γ' (for the state $n=2$, $m=0$, for example, we have $W_{\gamma\gamma} = -84F^2$, $W_{\gamma\gamma'} = -24F^2$), but for $V \gtrsim \omega$, the terms in (21) that are nondiagonal in γ provide a contribution to the order of $VW_{\gamma\gamma'}$ to $\mathscr{E}$, and can be neglected. Thus, in this approximation, the quasienergy spectrum is characterized by parabolic quantum numbers, similarly to the static Stark effect, and we have

$$\Psi_{\mathscr{E}_n}(\mathbf{r}, t) = \exp\{-i\mathscr{E}_n^{(2)} t\}\varphi_{n\gamma}(\mathbf{r}) U(t), \quad \mathscr{E}_n^{(2)} = E_n^{(0)} + 2W_{\gamma\gamma},$$

$$U(t) = \exp i\left\{\frac{V}{\omega}\sin\omega t + \delta\sin 2\omega t\right\} = \sum_{q=-\infty}^{\infty} c_q(F, \omega) e^{iq\omega t} \tag{22}$$

$$c_q = J_0(\delta) J_q\left(\frac{V}{\omega}\right) + \sum_{p=1}^{\infty} J_p(\delta)\left\{J_{q-2p}\left(\frac{V}{\omega}\right) + (-1)^p J_{q+2p}\left(\frac{V}{\omega}\right)\right\},$$

where $\delta = W_{\gamma\gamma}/2\omega$. In particular, when $n=2$, $m=0$, we have $\delta = 28F|V|/\omega$, and for states with $n=2$, $m=\pm 1$, we have $V=0$, $X_{\gamma\gamma} = \frac{1}{4}\alpha_{2p0}F^2$; the quantity α_{2p0} is defined by (15).

It follows from (22) that, as $\omega \to 0$ ($V \gg \omega$), the argument of the exponential in $\Psi_{\mathscr{E}}(\mathbf{r}, t)$ includes both $\mathscr{E}_n^{(2)}$ and the term V which corresponds to the linear Stark effect. When $V \ll \omega$, the weights c_q of harmonics with $q \neq 0$ are small in comparison with $c_0 \sim 1$, and $(\mathscr{E}_n - \mathscr{E}_n^{(0)}) \sim F^2$. As in the case of circular polarization, the role of terms that are linear in F for states with $n_1 \neq n_2$ becomes important for $V \gtrsim \omega$. The inclusion of terms that are quadratic in F leads not only to a shift and a splitting of the QES level $E_n^{(0)}$, but also to a change in the weights c_q of the harmonics. For example, for the state with $n=2$, $n_1=1$, $n_2=0$, the ratio $r_q = c_q/c_q(\delta=0)$ has the following values for $F = 5\times 10^{-3}$, $\omega = 10^{-2}$: $r_0 = 0.997$; $r_1 = 0.92$; $r_{-1} = 1.06$; $r_2 = 1.11$; $r_{-2} = 0.88$. The departure from symmetry in the weights of the positive and negative harmonics is analogous to the change in the symmetry of the shift of levels relative to $E_n^{(0)}$ when quadratic corrections to the static Stark effect is taken into account.

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1)We use the atomic system of units.

2)We note that the quantity γ_n is of independent interest in nonlinear optics of atomic gases because it governs the correction to the refractive index of the medium, which is a function of intensity.

3)An analogous situation obtains in the problem of ionization of an atom where the inclusion of the Coulomb potential results in a considerable increase in ionization probability.[14]

4)$k=0$ corresponds to the presence of close levels for which the matrix element of the interaction (simple or composite) is of the order of the distance between the levels.

5)The parameter nF/ω is identical with the parameter ξ introduced by Ritus.[3]

6)We note that the shift and the splitting of states with $n=2$ in crossed fields ($\mathbf{F} \perp \mathbf{H}$), including terms quadratic in F, are given by (18) if we suppose that $\omega = eH/2mc$ and we omit the terms $\pm\omega$ in $\Delta_{1,2}$.

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