Impurity center in the field of a strong light wave

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Oscillations of the populations of resonance electron levels of an impurity center in the field of a light wave give rise to oscillations of the equilibrium positions of the lattice atoms.. The oscillations, in turn, shift the level positions. Consequently, the oscillations of the populations become nonlinear. Their spectrum is manifested by the appearance of lines in the Raman scattering. The effect should occur for an impurity center as well as for a single molecule. The resultant features in self-induced transparency are discussed.

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Investigations of the optical properties of an impurity center the interaction of an electron with light is normally allowed for using perturbation theory. However, this approach is inappropriate if the light wave is fairly strong. On the other hand, there is a well-known Rabi solution[1] in which the interaction with light is allowed for exactly, but only in the case of a pure electron system which does not interact with phonons. We shall obtain a similar solution (see below in §1) describing the evolution of an impurity electron under the influence of light, but with allowance for its interaction with the phonon subsystem. We shall show that this gives rise to nonlinear oscillations of the populations and that the Raman scattering spectrum (§4) exhibits lines at frequencies characteristic of nonlinear oscillations. In §2 we shall consider the question of self-induced transparency in the case when an impurity electron interacts not only with light but also with phonons. In §3 we shall justify the self-consistent approach employed in §1.

We shall begin with the usual Hamiltonian

$$H=\varepsilon_1 a_1^+a_1+\varepsilon_2 a_2^+a_2+\sum_{\mathbf{k}\lambda}\hbar\omega_{\mathbf{k}\lambda}a_2^+a_2(u_{\mathbf{k}\lambda}^*b_{\mathbf{k}\lambda}+u_{\mathbf{k}\lambda}b_{\mathbf{k}\lambda}^+)$$
$$+\sum_{\mathbf{k}\lambda}\hbar\omega_{\mathbf{k}\lambda}b_{\mathbf{k}\lambda}^+b_{\mathbf{k}\lambda}-(a_1^+a_2+a_2^+a_1)\mathbf{d}_{12}\mathbf{E}_0\cos\Omega t, \qquad (1)$$

where a_i and $b_{\mathbf{k}\lambda}$ are, respectively, the electron and phonon annihilation operators; $\mathbf{d}_{12}$ is the electron dipole moment of a transition; $\mathbf{E}_0$ is the field amplitude; Ω is the field frequency. We shall consider the usual case when the frequencies of phonons obey $\omega_{\mathbf{k}\lambda}\ll\Omega\approx(\varepsilon_2-\varepsilon_1)/\hbar$. States in a resonant electron–phonon system obeying the condition $\omega_{\mathbf{k}\lambda}\equiv\omega_0\approx(\varepsilon_2-\varepsilon_1)/\hbar$ are considered in Ref. 2.

§1. NONLINEAR OSCILLATIONS

We shall begin with a simple and illustrative description of the situation. We shall allow for the interaction between an electron and the phonon subsystem in a self-consistent manner. We note that the phonon subsystem is characterized by $a_2^+a_2-\langle a_2^+a_2\rangle\equiv n_2$. Here and later we shall use $\langle\cdots\rangle$ to denote quantum-mechanical averaging over the exact state. We shall assume that $\dot{n}_2/n_2\ll\omega_0$, where ω_0 is the Debye frequency (this assumption is justified by the final results). In this case we can describe the lattice assuming that n_2 is constant and independent of time. The phonon Hamiltonian can be reduced to

$$H_p=\sum_{\mathbf{k}\lambda}\hbar\omega_{\mathbf{k}\lambda}\bar{b}_{\mathbf{k}\lambda}^+\bar{b}_{\mathbf{k}\lambda}-\sum_{\mathbf{k}\lambda}n_2^2|u_{\mathbf{k}\lambda}|^2\hbar\omega_{\mathbf{k}\lambda},$$

where $\bar{b}_{\mathbf{k}\lambda}=b_{\mathbf{k}\lambda}+n_2u_{\mathbf{k}\lambda}$. Thus, the effect of an electron on the lattice is to shift the equilibrium positions which vary slowly with time. When describing the electron motion, we shall use the substitution

$$u_{\mathbf{k}\lambda}^*b_{\mathbf{k}\lambda}+u_{\mathbf{k}\lambda}b_{\mathbf{k}\lambda}^+\to\langle u_{\mathbf{k}\lambda}^*b_{\mathbf{k}\lambda}+u_{\mathbf{k}\lambda}b_{\mathbf{k}\lambda}^+\rangle=-2n_2|u_{\mathbf{k}\lambda}|^2.$$

Consequently, the fate of an electron can be described by the Hamiltonian

$$H_e=\varepsilon_1a_1^+a_1+\Big(\varepsilon_2-2n_2\sum_{\mathbf{k}\lambda}\hbar\omega_{\mathbf{k}\lambda}|u_{\mathbf{k}\lambda}|^2\Big)a_2^+a_2-(a_2^+a_1+a_1^+a_2)\mathbf{d}_{12}\mathbf{E}_0\cos\Omega t.$$

The coefficients of the expansion of the electron wave function for the states 1 and 2 can be described by the secular Schrödinger equation (see, for example, Ref. 2). We can also use the equation for the density matrix.[3] In either case, using the variables

$$n(t)=\langle a_2^+a_2\rangle-\langle a_1^+a_1\rangle,$$
$$p(t)=\langle a_1^+a_2\rangle+\langle a_2^+a_1\rangle,$$
$$r(t)=-i\langle a_1^+a_2\rangle+i\langle a_2^+a_1\rangle$$

and bearing in mind that $\langle a_1^+a_1\rangle+\langle a_2^+a_2\rangle=1$, we obtain

$$\dot{n}=-2sr,\quad \dot{p}=(\varepsilon+v)r,\quad \dot{r}=-(\varepsilon+v)p+2sn,\quad v=-f(n+1). \qquad (2)$$

The following notation is used in Eq. (2): $\varepsilon=(\varepsilon_2-\varepsilon_1)/\hbar$, $s=s_0\cos\Omega t$,

$$s_0=-\mathbf{d}_{12}\mathbf{E}_0/\hbar>0,\quad f=\sum_{\mathbf{k}\lambda}\omega_{\mathbf{k}\lambda}|u_{\mathbf{k}\lambda}|^2.$$

Since the variable v is slow and $v\ll\varepsilon$, we can exclude r from Eq. (2). We shall seek the solution of the resultant system in the form

$$p=p_1\cos\Omega t+p_2\sin\Omega t.$$

The variables n, p_1, and p_2 are assumed to be slow compared with the frequency Ω. For these slow variables, we obtain

$$\left.\begin{aligned}\dot{n}&=-s_0p_2,\\ \dot{p}_1&=-(\Delta+fn)p_2,\\ \dot{p}_2&=(\Delta+fn)p_1+s_0n,\end{aligned}\right\} \qquad (3)$$

where $\Delta=\Omega-\varepsilon+f$. It follows from the first and second equations of the system (3) that

$$p_1=s_0^{-1}(\Delta n+fn^2/2+c).$$

The integration constant c can be found from the condi-

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tion that in the state 1 we have $n=-1$, $p_1=0$. This gives $c=\Delta-f/2$. Substituting the expressions for p_1, p_2 in terms of n and $\dot{n}$ in the third equation of the system (3), we obtain

$$-\ddot{n}=f^2n^3/2+3\Delta fn^2/2+(\Delta^2+fc+s_0^2)n+ac,$$

and then

$$\dot{n}^2+f^2n^4/4+\Delta fn^3+(\Delta^2+fc+s_0^2)n^2+2\Delta cn=c_1, \qquad (4)$$

which is identical with the equation of motion of a classical particle in a potential relief described by a polynomial of n. In the absence of the interaction with phonons ($f=0$), the potential is a parabolic well, so that the motion (dependence of n on t) is in the form of oscillations of frequency $(\Delta^2+s_0^2)^{1/2}$. Such oscillations represent the Rabi solution.

In analyzing the $f\neq 0$ case we have to find the roots of the polynomial in Eq. (4). The problem simplifies greatly if the frequency of light Ω is equal to the frequency of a zero-phonon transition $\varepsilon-f$. Then, $\Delta=0$ and the polynomial becomes biquadratic:

$$\dot{n}^2+\frac{f^2}{4}\left(n^4+2\frac{-f^2+2s_0^2}{f^2}n^2-c_1\right)=0. \qquad (5)$$

The nature of the dependence of the expression in parentheses on n depends strongly on the relationship between the parameters f^2 and s_0^2, and it is shown in Figs. 1(a), 1(b), and 1(c). The constant c_1 is selected so that the smallest root

$$n=\pm\left[\frac{f^2-2s_0^2}{f^2}\pm\left(\left(\frac{f^2-2s_0^2}{f^2}\right)^2-c_1\right)^{1/2}\right]^{1/2}$$

is -1, since we are dealing with the case when at $t=0$ the system is in the state 1 ($n=-1$). This gives $c_1=(f^2-4s_0^2)/f^2$. Consequently,

$$n=\pm\left[\frac{f^2-2s_0^2}{f^2}\pm\left(1-\frac{f^2-2s_0^2}{f^2}\right)\right]^{1/2}.$$

We can see that there are always real roots $n=\pm 1$. Moreover, if $f^2\geq 4s_0^2$, new real roots [Fig. 1(c)] are obtained:

$$n=\pm[(f^2-4s_0^2)/f^2]^{1/2}.$$

It is clear from Fig. 1 that if $f^2<4s_0^2$, the influence of the lattice is not so important. If at $t=0$ an electron is in the state with $n=-1$, then after a time it goes over entirely to an excited state which corresponds to $n=1$ if $\Delta=0$. The situation changes if $f^2\geq 4s_0^2$. An electron then oscillates within the limits

$$-1\leq n\leq-[(f^2-4s_0^2)/f^2]^{1/2}.$$

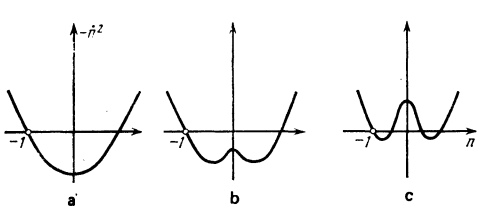

FIG. 1. Effective potential in the cases of $f^2<2s_0^2$ (a), $2s_0^2<f^2<4s_0^2$ (b), and $f^2>4s_0^2$ (c).

If $f^2-4s_0^2+0$, the oscillation period $T\to\infty$. If $f^2=4s_0^2$, the function $n(t)$ rises monotonically, beginning from $n(0)=-1$ and we find that $n(t)\to 0$ when $t\to\infty$.

It is quite easy to write down the solution of Eq. (5). It has all the features described qualitatively above. In the case when the interaction with the lattice is important ($f^2\geq 4s_0^2$), the solution is (see, for example, Ref. 2)

$$n^2(t)=1-\frac{4s_0^2}{f^2}\,\mathrm{sn}^2\frac{ft}{2},$$

where sn x is an elliptic sine which depends on the parameter $\lambda=2s_0/f$. The quantity f corresponds to the Huang-Reiss factor known from the theory of impurity centers and it can exceed considerably the Rabi frequency. In this case, we have sn $x=\sin x$ and then

$$n(t)=-1+\frac{2s_0^2}{f^2}\sin^2\frac{ft}{2}. \qquad (6)$$

An impurity is weakly excited and the oscillation amplitude is $s_0^2/f^2\ll 1$. The dependence (6) corresponds to harmonic oscillations of a classical particle near the left-hand minimum of the potential shown in Fig. 1(c). The condition $\dot{n}_2/n_2\ll\omega_0$ is satisfied if f, $s_0\ll\omega_0$.

§2. ROLE OF THE ELECTRON-PHONON INTERACTION IN SELF-INDUCED TRANSPARENCY

The evolution of an electron in the field of a soliton is again described by the system (2) but $s=-\mathbf{d}_{12}\mathbf{E}/\hbar$ is no longer a given function but a dynamic variable defined by the wave equation

$$\frac{\partial^2 s}{\partial t^2}-c^2\frac{\partial^2 s}{\partial z^2}=4\pi\alpha\frac{\partial^2 p}{\partial t^2}. \qquad (7)$$

Here and later, c denotes the velocity of light in a medium, $\alpha=Nd_{12}^2/\varkappa^2\hbar$, $\varkappa$ is a nonresonant part of the refractive index, and N is the impurity concentration (cm^{-3}).

We shall seek the solution of the system (2), (7) in the form[3]

$$s=W\cos\Phi,$$
$$p=Q\cos\Phi+R\sin\Phi,$$
$$\Phi=kz-\omega t+\varphi,$$

where W, Q, R, and φ are assumed to be slow, compared with Ω, function of the variable $\xi=t-z/u$. In the case of slow variables, we obtain as usual

$$\left.\begin{aligned}&\dot{n}=\omega WR/\varepsilon, \quad \dot{W}=\lambda\varepsilon^2R, \quad \dot{\varphi}=\lambda\varepsilon^2Q/W+\Omega_0,\\ &\dot{R}-\dot{\varphi}Q=(\varepsilon+v-\omega)Q-(\varepsilon+v)nW/\omega,\\ &\dot{Q}+\dot{\varphi}R=-(\varepsilon+v-\omega)R, \quad v=-f(n+1),\end{aligned}\right\} \qquad (8)$$

where

$$\lambda=\frac{2\pi\alpha\omega^2}{(c^2ku^{-1}-\omega)\varepsilon^2}\ll 1, \quad \Omega_0=-\frac{\omega^2-c^2k^2}{2(c^2ku^{-1}-\omega)}.$$

The derivatives in the system (8) are with respect to ξ. In the first three equations the correction v to ε is omitted since $\lambda\ll 1$ and $v\ll\varepsilon$. The difference from the situation described earlier is that the equations now have a new variable v which gives rise to an additional nonlinearity in the system (8). The first and second equations of the system yield

$$n=-1+\omega W^2/2\lambda\varepsilon^2.$$

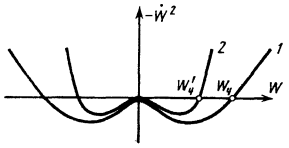

FIG. 2. Effective potential for $g=0$ (curve 1) and $g^2\lambda \gg 1$ (curve 2); $W_4 = 2\lambda^{1/2}\varepsilon$, $W_4' = 2\lambda^{1/4}g^{-1/2}\varepsilon$.

Therefore, we find that

$$v = -gW^2/\varepsilon, \qquad g = f\omega/2\lambda\varepsilon^3.$$

All the variables in the system (8) can be expressed in terms of W, and then W can be described by an expression which, for the frequency and wave vector selected so that $\omega = ck = \varepsilon$, is

$$W^2 - [\lambda\varepsilon^2 W^2 - {}^1/_4(1+2\lambda g)W^4 + \varepsilon^{-2}(-g^2/16 + g/6)W^6] = 0. \tag{9}$$

If $g=0$, the polynomial in the brackets in the above expression has the form represented by curve 1 in Fig. 2. Its roots are $W_{1,2}=0$ and $W_{3,4}=\pm 2\lambda^{1/2}\varepsilon$.

Since $v \ll \varepsilon$ for all cases when $W^2 \leq W_{3,4}^2$, it follows that $2g\lambda \ll 1$. Thus, the condition $g \neq 0$ leaves unaffected the variables $\sim W^2$ and $\sim W^4$ in Eq. (9) and only gives rise to a term $\sim W^6$. On increase in g, it begins to play an important role if at the point $W^2 = 4\lambda\varepsilon^2$ it becomes comparable with the first two quantities:

$$\frac{g^2}{16\varepsilon^2} \cdot 64\lambda^3\varepsilon^6 \geq 4\lambda^2\varepsilon^4.$$

Consequently, the electron-phonon interaction discussed here is important if $g^2\lambda \gtrsim 1$ ($f \gtrsim 2\lambda^{1/2}\varepsilon$). This corresponds exactly to the condition $f \geq 2s_0$ obtained earlier and it means that the maximum shift of a level $|v| = 4g\lambda\varepsilon$ becomes comparable with the field (expressed in suitable units) $W \approx 2\lambda^{1/2}\varepsilon$. However, if $g^2\lambda \gg 1$, the term $\sim W^6$ dominates Eq. (9). The polynomial has the form represented by curve 2 in Fig. 2. The term $\sim W^4$ can be ignored altogether and the soliton envelope is then given by the equation

$$W^2 - W^2(\lambda\varepsilon^2 - g^2W^4/16\varepsilon^2) = 0.$$

Its solution can be written in the form

$$W = (\lambda/g^2)^{1/4} 2\varepsilon \{\mathrm{ch}[2\lambda^{1/2}\varepsilon(t - z/u)]\}^{-1/2}.$$

For comparison, we recall that if $g=0$, the envelope has the form[3]

$$W = 2\lambda^{1/2}\varepsilon/\mathrm{ch}[\lambda^{1/2}\varepsilon(t - z/u)].$$

The wave amplitude decreases, compared with the amplitude when $g=0$, by a factor $g^{1/2}\lambda^{1/4}$. The condition $\dot{n}/n \ll \omega_0$ is satisfied if $\lambda \ll \omega_0/\varepsilon$.

§3. EXACT EQUATIONS OF MOTION

The complete orthonormalized basis of the wave functions in the problem under consideration is the set

$$\Psi_1\{n_{k\lambda}\} = a_1^+ \prod_{k\lambda} \frac{(b_{k\lambda}^+)^{n_{k\lambda}}}{(n_{k\lambda}!)^{1/2}} |0\rangle \exp\left(-i\left(\frac{\varepsilon_1}{\hbar} + \sum_{k\lambda} n_{k\lambda}\omega_{k\lambda}\right)t\right),$$

$$\Psi_2\{n_{k\lambda}\} = a_2^+ \prod_{k\lambda} \frac{(b_{k\lambda}^+)^{n_{k\lambda}}}{(n_{k\lambda}!)^{1/2}} |0\rangle \exp\left(-i\left(\frac{\varepsilon_2}{\hbar} + \sum_{k\lambda} n_{k\lambda}\omega_{k\lambda}\right)t\right).$$

The wave function of any state can be represented in the form

$$\Psi = \sum_{\{n_{k\lambda}\}} [c_1\{n_{k\lambda}\}\Psi_1\{n_{k\lambda}\} + c_2\{n_{k\lambda}\}\Psi_2\{n_{k\lambda}\}],$$

where the summation is carried out over all possible sets of the occupation numbers $\{n_{k\lambda}\}$.

The Schrödinger equations for the expansion coefficients are

$$\begin{aligned} i\dot{c}_1\{n_{k\lambda}\} &= e^{-ist}sc_2\{n_{k\lambda}\}, \\ i\dot{c}_2\{n_{k\lambda}\} &= e^{ist}sc_1\{n_{k\lambda}\} + \sum_{k\lambda}[u_{k\lambda}^*\omega_{k\lambda}e^{-i\omega_{k\lambda}t}\cdot\sqrt{n_{k\lambda}+1}\,c_2\{n_{k\lambda}+1\} \\ &\quad + u_{k\lambda}\omega_{k\lambda}e^{i\omega_{k\lambda}t}\sqrt{n_{k\lambda}}c_2\{n_{k\lambda}-1\}]. \end{aligned} \tag{10}$$

Summation with respect to $k\lambda$ in the second of the above equations relates the coefficient $c_2\{n_{k\lambda}\}$ to all the coefficients $c_2\{n_{k\lambda}\pm 1\}$ in which one of the occupation numbers differs by unity. By definition, the coefficient $c_2\{n_{k\lambda}\}$ vanishes if at least one of the numbers obeys $n_{k\lambda} < 0$.

It is easily shown that, in the $s \equiv 0$ case, the wave function

$$\Psi = a_2^+ \prod_{k\lambda} \exp(-u_{k\lambda}b_{k\lambda}^+ + u_{k\lambda}^* b_{k\lambda})|0\rangle \tag{11}$$

is the eigenfunction for the Hamiltonian of the problem. In fact, if we use

$$b_{k\lambda}\Psi = -u_{k\lambda}\Psi,$$

we obtain

$$(u_{k\lambda}^* b_{k\lambda} + u_{k\lambda}b_{k\lambda}^+ + b_{k\lambda}^+ b_{k\lambda})\Psi = -|u_{k\lambda}|^2\Psi.$$

Therefore,

$$H\Psi = \left(\varepsilon_2 - \sum_{k\lambda}\hbar\omega_{k\lambda}|u_{k\lambda}|^2\right)\Psi. \tag{12}$$

The wave function (11) describes the shift of the equilibrium positions of the lattice atoms caused by the electron-phonon interaction. It is used also widely in the polaron theory. The correction to the energy (12) is the polaron shift denoted above by $\hbar f$.

We shall represent the wave function (11) in the form of an expansion in respect of the selected basis:

$$\Psi \exp[-i(\varepsilon_2/\hbar - f)t] = \sum_{\{n_{k\lambda}\}} c^{(0)}\{n_{k\lambda}\}\Psi_2\{n_{k\lambda}\}.$$

The coefficients of the expansion $c^{(0)}\{n_{k\lambda}\}$ are readily found if we multiply both sides of the above equality by $\Psi_2\{n_{k\lambda}\}$:

$$c^{(0)}\{n_{k\lambda}\} = \exp i\left(f + \sum_{k\lambda} n_{k\lambda}\omega_{k\lambda}\right)t \prod_{k\lambda} \frac{(-u_{k\lambda})^{n_{k\lambda}}}{(n_{k\lambda}!)^{1/2}} \exp\left(-\frac{1}{2}|u_{k\lambda}|^2\right). \tag{13}$$

Substitution of Eq. (13) in Eq. (10) demonstrates that $c_1\{n_{k\lambda}\} = 0$ and $c_2\{n_{k\lambda}\} = c^{(0)}\{n_{k\lambda}\}$ represent the solution of the system in the $s=0$ case.

We shall seek the solution of the system (10) with $s \neq 0$ in the form $c_1\{n_{k\lambda}\} = c_1 c\{n_{k\lambda}\}$, $c_2\{n_{k\lambda}\} = c_2 c\{n_{k\lambda}\}$. Substituting these expressions in Eq. (10), we obtain

$$\begin{aligned} ic_1\dot{c}\{n_{k\lambda}\} + ic_1\dot{c}\{n_{k\lambda}\} &= e^{-ist}sc_2 c\{n_{k\lambda}\}, \\ ic_2\dot{c}\{n_{k\lambda}\} + ic_2\dot{c}\{n_{k\lambda}\} &= e^{ist}sc_1 c\{n_{k\lambda}\} \\ + c_2\sum_{k\lambda}[u_{k\lambda}^*\omega_{k\lambda}e^{-i\omega_{k\lambda}t}(n_{k\lambda}+1)^{1/2}c\{n_{k\lambda}+1\} &+ u_{k\lambda}\omega_{k\lambda}e^{i\omega_{k\lambda}t}\sqrt{n_{k\lambda}}c\{n_{k\lambda}-1\}]. \end{aligned} \tag{14}$$

Substituting in the system (14) the expression for $c\{n_{k\lambda}\}$ which differs from $c^{(0)}\{n_{k\lambda}\}$ of Eq. (13) by the substitution $u_{k\lambda} \to |c_2|^2 u_{k\lambda} \equiv n_2 u_{k\lambda}$ $(f \to n_2^2 f)$. We shall multiply by $c^*\{n_{k\lambda}\}$ and sum over $\{n_{k\lambda}\}$, which yields the following simple equations for c_1 and c_2:

$$i\dot{c}_1 - 2c_1 f n_2^2 = e^{-ist} s c_2,$$
$$i\dot{c}_2 - 2c_2 f(n_2^2 - n_2) = e^{ist} s c_1.$$

Introducing new variables

$$\alpha_1 = c_1 \exp \int_0^t 2if n_2^2\, dt, \quad \alpha_2 = c_2 \exp \int_0^t 2if(n_2^2 - n_2)\, dt,$$

we find that

$$\left.\begin{aligned} i\dot{\alpha}_1 &= s\alpha_2 \exp\left[-i\int_0^t (\varepsilon - 2fn_2)\, dt\right], \\ i\dot{\alpha}_2 &= s\alpha_1 \exp i\int_0^t (\varepsilon - 2fn_2)\, dt, \end{aligned}\right\} \tag{15}$$

which is equivalent to the system (2) obtained in §1. In fact, the dipole moment of an atom

$$\mathbf{p}(t) = \mathbf{d}_{12}(c_1 c_2^* e^{ist} + c_2 c_1^* e^{-ist}) = \mathbf{d}_{12} p$$

can be expressed in terms of the quantity

$$p = \alpha_1 \alpha_2^* \exp i\int_0^t (\varepsilon - 2fn_2)\, dt + \alpha_1^* \alpha_2 \exp\left[-i\int_0^t (\varepsilon - 2fn_2)\, dt\right].$$

We shall also determine the functions r and n:

$$r = i\alpha_1 \alpha_2^* \exp i\int_0^t (\varepsilon - 2fn_2)\, dt - i\alpha_1^* \alpha_2 \exp\left[-i\int_0^t (\varepsilon - 2fn_2)\, dt\right],$$
$$n = 1 - 2|\alpha_1|^2.$$

Differentiating n, p, and r and using the system (15), we obtain the system (2). We can easily see that the transition from (14) to (15) represents a self-consistent approximation. In fact, if $c\{n_{k\lambda}\}$ is substituted in Eq. (14) we obtain

$$\left.\begin{aligned} &i\dot{c}_1 - c_1 \sum_{k\lambda}\left[|u_{k\lambda}|^2 \omega_{k\lambda} n_2^2 \left(1 + \frac{i\dot{n}_2}{n_2\omega_{k\lambda}}\right) + n_{k\lambda}\omega_{k\lambda}\left(1 - \frac{i\dot{n}_2}{n_2\omega_{k\lambda}}\right)\right] = sc_2 e^{-ist}, \\ &i\dot{c}_2 - c_2 \sum_{k\lambda}\left[|u_{k\lambda}|^2 \omega_{k\lambda} n_2^2 \left(1 - \frac{1}{n_2} + \frac{i\dot{n}_2}{n_2\omega_{k\lambda}}\right)\right. \\ &\left. + n_{k\lambda}\omega_{k\lambda}\left(1 - \frac{1}{n_2} - \frac{i\dot{n}_2}{n_2\omega_{k\lambda}}\right)\right] = sc_1 e^{ist}. \end{aligned}\right\} \tag{16}$$

The summation over $\{n_{k\lambda}\}$ used in the derivation of the system (15) corresponds to the following substitution in the system (16):

$$n_{k\lambda} \to \langle n_{k\lambda}\rangle = \sum_{\{n_{k\lambda}\}} n_{k\lambda} |c\{n_{k\lambda}\}|^2 = n_2^2 |u_{k\lambda}|^2.$$

This substitution is valid if the deviation of $n_{k\lambda}$ from $\langle n_{k\lambda}\rangle$ within the dispersion limits

$$(\langle n_{k\lambda}^2\rangle - \langle n_{k\lambda}\rangle^2)^{1/2} = n_2^2 |u_{k\lambda}|^2$$

gives rise to small corrections in the system (16). This means that the following relationships should be satisfied:

$$\left.\begin{aligned} &|\dot{\alpha}_1/\alpha_1| \gg n_2^2 \sum_{k\lambda} |u_{k\lambda}|^2 \omega_{k\lambda} = n_2^2 f, \\ &|\dot{\alpha}_2/\alpha_2| \gg (n_2^2 - n_2) f, \quad |\dot{n}_2/n_2\omega_0| \ll 1. \end{aligned}\right\} \tag{17}$$

The last condition for the system (17) means that at every moment we can define the concept of dispersion $n_{k\lambda}$ which varies slowly with time. The relationships (17) are obeyed in the case of a weak electron-phonon coupling $(f \ll \omega_0)$, since it follows from the solutions found in §1 that if $s_0 \gg f$, then

$$|\dot{\alpha}_1/\alpha_1|, \quad |\dot{\alpha}_2/\alpha_2| \sim s_0,$$

whereas for $s_0 \ll f$,

$$|\dot{\alpha}_1/\alpha_1|, \quad |\dot{\alpha}_2/\alpha_2| \sim f, \quad n_2 \ll 1.$$

§4. OPTICAL PROPERTIES

The polarization of an atom p can be expressed in terms of n and $\dot{n}$:

$$\left.\begin{aligned} p &= p_1 \cos\Omega t + p_2 \sin\Omega t, \\ p_1 &= s_0^{-1}(\Delta n + fn^2/2 + \Delta - f/2), \\ p_2 &= -s_0^{-1}\dot{n}. \end{aligned}\right\} \tag{18}$$

The function n is periodic and has a spectrum. It follows from the system (18) that this function is reflected in the dependence $p(t)$ and, consequently, in the spectrum of scattered light. Thus, a spectroscopic analysis of the scattered light can give direct information on oscillations of the populations. In the limiting cases of $f \gg s_0$ and $f \ll s_0$ the function n varies harmonically but the frequencies are different in these two cases. If $f \gg s_0$ and $\Delta = 0$, the oscillation frequency is f, so that the scattered light has components with Ω, $\Omega \pm f$, and $\Omega \pm 2f$. However, if $s_0 \gg f$, then n oscillates at a frequency $\omega_s = (\Delta^2 + s_0^2)^{1/2}$ and the scattered light includes frequencies Ω, $\Omega \pm \omega_s$ and $\Omega \pm 2\omega_s$. We can easily find the intensities of the various lines. If $f \gg s_0$, and $\Delta = 0$, then Eqs. (18) and (6) yield the following expression for the dipole moment of an atom:

$$\mathbf{p} = \mathbf{d}_{12}\left[-\frac{s_0}{f}\left(1 - \frac{3s_0^2}{4f^2}\right)\cos\Omega t + \frac{s_0}{f}\left(1 - \frac{s_0^2}{f^2}\right)\cos(\Omega + f)t\right.$$
$$\left. - \frac{s_0^2}{2f^2}\cos(\Omega - f)t + \frac{s_0^3}{8f^3}\cos(\Omega + 2f)t + \frac{s_0^3}{8f^3}\cos(\Omega - 2f)t\right].$$

Each atom becomes a source of dipole radiation[4] with the following intensities of the various components:

$$\left.\begin{aligned} I(\Omega) &= \frac{s_0^2}{f^2} I_0(\Omega) \sim 10^{-8} \text{ erg/sec}, \\ I(\Omega + f) &= \frac{s_0^2}{f^2} I_0(\Omega + f) \sim 10^{-8} \text{ erg/sec}, \\ I(\Omega - f) &= \frac{s_0^4}{4f^4} I_0(\Omega - f) \sim 10^{-13} \text{ erg/sec}, \\ I(\Omega \pm 2f) &= \frac{s_0^6}{64f^6} I_0(\Omega \pm 2f) \sim 10^{-13} \text{ erg/sec}, \end{aligned}\right\} \tag{19}$$

where $I_0(\Omega) = (d_{12}^2 \Omega^4 / 8\pi c^3) \sin^2\theta\, do$, θ is the angle between $\mathbf{d}_{12}$ and the direction of emission of radiation, and do is a solid-angle element. The above numerical estimates apply to the total radiation in the $s_0/f = 10^{-1}$ case. The components of frequencies Ω and $\Omega + f$ are proportional to the intensity of the scattered light, whereas the components with frequencies $\Omega - f$ and $\Omega \pm 2f$ are proportional to the cube of the intensity. Similar results are obtained also in the case when $f \gg s_0$.

Since we can assume that the field is classical and known, the Hamiltonian (1) does not include the radiative width of the upper level γ_0. We shall consider the terms of the interaction which are linear with respect

to the displacements and, therefore, we shall ignore also the broadening of the level γ_p because of the electron-electron interaction, which is obtained if the quadratic terms are included.[5] However, the effects considered here, like the Rabi oscillations, appear in a real situation only when their characteristic frequencies are considerably greater than the damping $\gamma = \gamma_0 + \gamma_p$ ignored by us. Thus, the inequality $\gamma T \ll 1$ should be satisfied (T is the period of nonlinear oscillations in §1 or the pulse duration in §2).

The quantities γ_0 and γ_p vary within wide limits and for many impurity centers they are well known.[3,5] For example, in the case of ruby exhibiting self-induced transparency at helium temperatures, we have $\gamma_0 \ll \gamma_p$ and $\gamma_p \sim 10^7 - 10^8$ sec^{-1} (Ref. 3). The pulse duration satisfies the condition $\gamma T \ll 1$, as indicated by the fact the phenomenon itself is observed. Nonlinear oscillations in the $f \gg s_0$ case are characterized by the period $T = 2\pi f^{-1}$ so that Eqs. (6) and (19) remain valid if $\gamma/f \ll 1$, which again is easy to satisfy. Investigations of impurity centers in strong electromagnetic fields are particularly promising. The well-developed methods of laser spectroscopy of gases can give unique information. The possibility of spectroscopic analysis within an inhomogeneously broadened line (which corresponds to the intra-Doppler spectroscopy of gases) is particularly attractive. It is interesting to consider also the possibility of investigating impurity centers by a weak probe wave in the presence of a strong saturating wave. To the author's knowledge, such investigations have not yet been made.

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