PHYSICAL REVIEW B VOLUME 19, NUMBER 1 1 JANUARY 1979

Theory of ESR dipolar splitting of a spin pair in the presence of avalanche phonons

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(Received 6 July 1978)

The ESR spectrum of a pair of paramagnetic spins $S = \frac{1}{2}$, interacting via dipolar magnetic forces, is calculated in the presence of an incoherent phonon avalanche, which is coupled to the spin pair by linear spin-phonon interaction. The problem is approximately solved by a Green's-function approach. Two cases are discussed, the first for an infinitely narrow phonon spectrum, and the second for finite bandwidth of the avalanche phonons. When the ESR frequency falls out of the avalanche band, the spectrum of the pair is shown to consist of two lines shifted from the no-avalanche resonant field and more closely spaced than in the no-avalanche situation. When the ESR frequency is within the avalanche band, a sharp decrease of the ESR intensity is expected. These results are interpreted in terms of ac Stark shift and of screening of dipolar interaction by the avalanche phonons. Their relationships to electromagnetic power narrowing of ESR lines and to the "magic angle" effect in NMR are discussed. Finally the theoretical results are examined in connection with phonon-avalanche experiments on Ce^{3+}-doped lanthanum magnesium nitrate.

I. INTRODUCTION

Phonons in solids, under suitable conditions, are able to cause screening of electrical charges, and in fact one of the most striking effects of this screening is superconductivity in metals. One may inquire if phonons could also screen dipolar interactions and look for physical effects which are likely to be related to this screening. One of the most common forms of interacting dipoles in physics is a set of magnetic atoms embedded in a solid matrix; usually the magnetic dipolar interaction is not the only source of interaction among the atoms, at least if their magnetism is of electronic origin, but if exchange and lattice strains are small it has good chances of being a dominant mechanism at ordinary temperature. On the other hand, it is well known that magnetic dipoles of electronic origin can interact with lattice vibrations in a crystal via the so-called spin-phonon interaction; so one may expect that, owing to absorption and emission of phonons, an indirect interaction sets up between different magnetic dipoles which might interfere with the ordinary coupling, manifesting itself as a screening of the magnetic dipolar interaction. In fact a coupling due to absorption and emission of virtual phonons has been previously shown to be effective between the atoms of a paramagnetic pair[1]; this coupling is likely to show up at low temperature. We have recently started an investigation of the screening of dipolar interaction in a paramagnetic system by very-high-temperature phonons, such as those which might be emitted in a relatively narrow band during a phonon avalanche.[2] We found the expected effect which turned out to be describable in terms of an effective dipolar coupling constant and of an effective gyromagnetic factor[3]; the unitary transformation used to this aim, however, limited the investigation to the case of an off-resonance phonon avalanche, because of resonant divergences appearing in the unitary transforming operators. The aim of the present paper is to remove these difficulties from the calculation of the ESR spectrum of the system, and to present a more comprehensive account of the phenomenon of reduction of dipolar interaction by a phonon avalanche.

We consider a pair of neighboring spins, in a static magnetic field along z, which are coupled to the lattice vibrations of a host crystal by a direct spin-phonon Hamiltonian in the rotating-wave approximation, and to each other by the truncated dipolar interaction. This system is described by the total Hamiltonian ($\hbar=1$)

$$\mathcal{H} = \omega_0(S_z^1 + S_z^2) + \sum_k \omega_k b_k^\dagger b_k + \tfrac{1}{2}\sum_k \epsilon_k [b_k(S_+^1 + S_+^2) + b_k^\dagger (S_-^1 + S_-^2)] + \mathcal{H}_d \ ,$$

$$\mathcal{H}_d = \alpha [S_z^1 S_z^2 - \tfrac{1}{4}(S_+^1 S_-^2 + S_-^1 S_+^2)] \ , \qquad (1.1)$$

where we have neglected the phase difference between the two spins, under the assumption that $\vec{k}\cdot(\vec{r}_1 - \vec{r}_2) << 1$. The Larmor frequency of the spins located at r_1 and r_2 in the lattice is denoted by ω_0 in (1.1) and the summation over k runs over a narrow band of lattice modes of frequencies ω_k whose average is near ω_0, but does not necessarily coincide with it. The spin-phonon coupling constant to the kth mode is denoted by ϵ_k, and α is the coefficient of the secular part of the dipolar interaction, depending as usual on $|r_1 - r_2|$. Neglecting in (1.1) lattice modes out of the avalanche band is justified because we assume that they are scarcely populated: during a phonon avalanche the temperature of the phonons in the hot band may rise to 10^3–10^4 °K.[4] Moreover we feel entitled to adopt the rotating-wave approximation (RWA) because we assume the average frequency of the hot band never to depart very much from the Larmor frequency ω_0. Available experimental data on avalanche-phonon bandwidth seem to support this assumption.[5] Finally, we base the adoption of the truncated Hamiltonian on our previous off-resonance results,[3] which show the negligible influence of the energy-nonconserving terms in the present situation.

In order to pursue our investigation on the screening we shall develop a theory of ESR of system (1.1). In other words, we add to (1.1) a term describing a small rotating electromagnetic field of amplitude h and frequency Ω as a time-dependent perturbation

$$V(t) = g\mu_B h[(S_+^1 + S_+^2)e^{i\Omega t} + (S_-^1 + S_-^2)e^{-i\Omega t}] \qquad (1.2)$$

and look for the response of the system as a function of ω_0. In particular, the imaginary part χ'' of the susceptibility is proportional to the microwave power at frequency Ω absorbed by the atoms. This power is what is recorded in a typical ESR experiment. In the absence of spin-phonon interaction ($\epsilon_k = 0$) two lines should appear at $\omega_0 = \Omega \pm \frac{3}{4}\alpha$. Our aim is to see how this situation is modified by coupling to avalanche phonons.

We calculate the susceptibility by the well-known Green's-function technique.[6] In fact $\chi'' = -\mathrm{Im}\chi$ and[7]

$$\chi_- \propto \lim_{\eta\to 0} \langle\langle (S_-^1 + S_-^2);(S_+^1 + S_+^2)\rangle\rangle_{E=\Omega+i\eta} \ , \qquad (1.3)$$

where $\chi_- = \chi' - i\chi''$. We define the Green's function of two operators A and B as[8]

$$\langle\langle A;B\rangle\rangle_E = \frac{1}{2\pi Z_0}\sum_{m,m'}(e^{-(1/KT)E_m} - e^{-(1/KT)E_{m'}}) \times \frac{\langle m|A|m'\rangle\langle m'|B|m\rangle}{E + E_m - E_{m'}} \ . \qquad (1.4)$$

In (1.4) E is a complex variable, while $|m\rangle$, $|m'\rangle$ are eigenfunctions of the total Hamiltonian of the system, and $E_m, E_{m'}$ the corresponding eigenvalues. Moreover

$$Z_0 = \sum_m e^{-(1/KT)E_m}$$

is the partition function of the coupled system. Any Green's function is characterized by a set of poles on the real E axis, given by all possible differences $E_m - E_{m'}$ of eigenvalue pairs. It is easy to convince oneself from (1.4) and (1.3) that the poles of

$$\langle\langle (S_-^1 + S_-^2);(S_+^1 + S_+^2)\rangle\rangle_\Omega$$

for real Ω and as a function of ω_0 correspond to the position of lines in the ESR spectrum of the system. Since eigenfunctions and eigenvalues of the complete system are usually unknown, it is not possible in general to calculate the relevant Green's functions directly from (1.4). It is a rigorous property of any Green's function (1.4), however, that

$$E\langle\langle A;B\rangle\rangle_E = \frac{1}{2\pi}\langle [A,B]\rangle + \langle\langle [A,\mathcal{H}];B\rangle\rangle_E \ , \qquad (1.5)$$

where the single brackets $\langle\ \rangle$ denote thermal average. Equation (1.5) can be supplemented by another equation of the same form for $\langle\langle [A,\mathcal{H}];B\rangle\rangle_E$, and iterating this procedure gives rise to an infinite chain of coupled equations. This may not seem very helpful, but if one is able to approximate a few of the Green's functions in terms of the preceding ones in the hierarchy, it is possible to obtain a finite number of linear equations with the same number of Green's functions as unknowns. The above approximation is known as "decoupling procedure," and it is usually a source of conceptual difficulties, because there is no established simple rule to make approximations like

$$\langle\langle [A,\mathcal{H}];B\rangle\rangle_E \simeq \lambda(E)\langle\langle A;B\rangle\rangle_E \ .$$

In order to have a guidance as to the type of decoupling suitable to our needs, we shall first discuss a model simpler than that described by (1.1), where we are able to adopt a satisfactory decoupling procedure which was devised for a different problem.[8] This is a model with just one phonon mode and a spin pair. We shall successively apply this decoupling procedure to the multimode case in Sec. III. From now on, we shall drop suffix E from the Green's-function symbol, except where it can be useful as a reminder of the proper functional dependence.

II. SINGLE-MODEL CASE

We consider a model of a pair of spins interacting via their dipole fields and coupled to a highly populated phonon mode. This model can also be solved by less exotic techniques, but it shall permit us to develop a decoupling procedure which we shall suitably extend to the more complicated multiphonon case. The Hamiltonian of the system is

$$\mathcal{H} = \omega_0(S_z^1 + S_z^2) + \omega b^\dagger b + \tfrac{1}{2}\epsilon[b(S_+^1 + S_+^2) + b^\dagger(S_-^1 + S_-^2)] + \alpha[S_z^1 S_z^2 - \tfrac{1}{4}(S_+^1 S_-^2 + S_-^1 S_+^2)] \; . \tag{2.1}$$

In order to calculate the ESR spectrum obtaiinable by the small field (1.2) the relevant Green's-functions equations are

$$\begin{aligned}
&(E-\omega_0)\langle\langle (S_-^1+S_-^2);(S_+^1+S_+^2)\rangle\rangle \\
&\qquad = -(1/\pi)\langle (S_z^1+S_z^2)\rangle - \epsilon\langle\langle b(S_z^1+S_z^2);(S_+^1+S_+^2)\rangle\rangle + \langle\langle [(S_-^1+S_-^2),\mathcal{H}_d];(S_+^1+S_+^2)\rangle\rangle \; , \\
&(E-\omega)\langle\langle b(S_z^1+S_z^2);(S_+^1+S_+^2)\rangle\rangle \\
&\qquad = (1/2\pi)\langle b(S_+^1+S_+^2)\rangle + \tfrac{1}{2}\epsilon[\langle\langle b^2(S_+^1+S_+^2);(S_+^1+S_+^2)\rangle\rangle - \langle\langle b^\dagger b(S_-^1+S_-^2);(S_+^1+S_+^2)\rangle\rangle \\
&\qquad\quad + \langle\langle (S_z^1+S_z^2)(S_-^1+S_-^2);(S_+^1+S_+^2)\rangle\rangle] \; , \\
&(E+\omega_0-2\omega)\langle\langle b^2(S_+^1+S_+^2);(S_+^1+S_+^2)\rangle\rangle \\
&\qquad = \epsilon[\langle\langle b^\dagger b^2(S_z^1+S_z^2);(S_+^1+S_+^2)\rangle\rangle + \langle\langle b(S_+^1+S_+^2)(S_-^1+S_-^2);(S_+^1+S_+^2)\rangle\rangle] + \langle\langle [b^2(S_+^1+S_+^2),\mathcal{H}_d];(S_+^1+S_+^2)\rangle\rangle \; .
\end{aligned} \tag{2.2}$$

Neglecting for the moment terms in $\mathcal{H}_d$, this set of three equations can be approximately closed by noting that the strength at the poles of a given Green's function depends on the number of b operators appearing on its left-hand side. Consequently we shall neglect

$$\langle\langle b(S_+^1+S_+^2)(S_-^1+S_-^2);(S_+^1+S_+^2)\rangle\rangle$$

in the third member of (2.2) and

$$\langle\langle (S_z^1+S_z^2)(S_-^1+S_-^2);(S_+^1+S_+^2)\rangle\rangle$$

in the second member. Moreover we make a first decoupling by changing operator $b^\dagger b$ into c-number n, the occupation number of the phonon mode,[9] wherever it occurs in (2.2). With these approximations, and in the absence of dipolar coupling, Eqs. (2.2) yield the closed system

$$\begin{aligned}
&(E-\omega_0)G_1 + \frac{1}{\sqrt{2}}\epsilon\sqrt{n}\,G_2 = -\frac{1}{\pi}\langle (S_z^1+S_z^2)\rangle \; , \\
&\frac{1}{\sqrt{2}}\epsilon\sqrt{n}\,G_1 + (E-\omega)G_2 + \frac{1}{\sqrt{2}}\epsilon\sqrt{n}\,G_3 = \frac{1}{2\pi}\left(\frac{2}{n}\right)^{1/2}\langle b(S_+^1+S_+^2)\rangle \; , \\
&\frac{1}{\sqrt{2}}\epsilon\sqrt{n}\,G_2 + (E+\omega_0-2\omega)G_3 = 0 \; ,
\end{aligned} \tag{2.3}$$

where

$$\begin{aligned}
G_1 &= \langle\langle (S_-^1+S_-^2);(S_+^1+S_+^2)\rangle\rangle \; , \\
G_2 &= \left(\frac{2}{n}\right)^{1/2}\langle\langle b(S_z^1+S_z^2);(S_+^1+S_+^2)\rangle\rangle \; , \\
G_3 &= -\frac{1}{n}\langle\langle b^2(S_z^1+S_z^2);(S_+^1+S_+^2)\rangle\rangle \; .
\end{aligned} \tag{2.4}$$

The poles of the various G_i appearing in (2.3) are given by the roots of

$$\begin{vmatrix} E-\omega_0 & (1/\sqrt{2})\epsilon\sqrt{n} & 0 \\ (1/\sqrt{2})\epsilon\sqrt{n} & E-\omega & (1/\sqrt{2})\epsilon\sqrt{n} \\ 0 & (1/\sqrt{2})\epsilon\sqrt{n} & E+\omega_0-2\omega \end{vmatrix} = 0 \tag{2.5}$$

which can be treated as an eigenvalue equation, yielding

$$E_+ = \omega + A \; , \quad E_0 = \omega \; , \quad E_- = \omega - A \; ,$$
$$A = [(\omega_0-\omega)^2 + \epsilon^2 n]^{1/2} \tag{2.6}$$

as eigenvalues, with associated eigenvectors

$$\underline{a_+}=\begin{vmatrix}\frac{1}{2}(1+\cos\theta)\\ -(1/\sqrt{2})\sin\theta\\ \frac{1}{2}(1-\cos\theta)\end{vmatrix}\ ,$$

$$\underline{a_0}=\begin{vmatrix}(1/\sqrt{2})\sin\theta\\ \cos\theta\\ -(1/\sqrt{2})\sin\theta\end{vmatrix}\ ,$$

$$\underline{a_-}=\begin{vmatrix}\frac{1}{2}(1-\cos\theta)\\ (1/\sqrt{2})\sin\theta\\ \frac{1}{2}(1+\cos\theta)\end{vmatrix}\ , \qquad (2.7)$$

where

$$\sin\theta=\epsilon\sqrt{n}/A\ ,\quad \cos\theta=(\omega_0-\omega)/A\ . \qquad (2.8)$$

Eigenvectors (2.7) can be used to assemble new Green's functions

$$\Gamma_+=\tfrac{1}{2}(1+\cos\theta)G_1-\frac{1}{\sqrt{2}}\sin\theta G_2+\tfrac{1}{2}(1-\cos\theta)G_3\ ,$$

$$\Gamma_0=\frac{1}{\sqrt{2}}\sin\theta G_1+\cos\theta G_2-\frac{1}{\sqrt{2}}\sin\theta G_3\ ,$$

$$\Gamma_-=\tfrac{1}{2}(1-\cos\theta)G_1+\frac{1}{\sqrt{2}}\sin\theta G_2+\tfrac{1}{2}(1+\cos\theta)G_3\ , \qquad (2.9)$$

which may be conveniently written

$$\Gamma_+=\langle\langle A_+;(S_+^1+S_+^2)\rangle\rangle\ ,$$

$$\Gamma_0=\langle\langle A_0;(S_+^1+S_+^2)\rangle\rangle\ ,$$

$$\Gamma_-=\langle\langle A_-;(S_+^1+S_+^2)\rangle\rangle\ , \qquad (2.10)$$

where

$$\begin{aligned}A_+=&\tfrac{1}{2}(1+\cos\theta)(S_-^1+S_-^2)\\ &-\sin\theta(1/\sqrt{n})b(S_z^1+S_z^2)\\ &-\tfrac{1}{2}(1-\cos\theta)(1/n)b^2(S_+^1+S_+^2)\ ,\\ A_0=&(1/\sqrt{2})\sin\theta(S_-^1+S_-^2)\\ &+\cos\theta(2/n)^{1/2}b(S_z^1+S_z^2)\\ &+(1/\sqrt{2})\sin\theta(1/n)b^2(S_+^1+S_+^2)\ ,\\ A_-=&\tfrac{1}{2}(1-\cos\theta)(S_-^1+S_-^2)\\ &+\sin\theta(1/\sqrt{n})b(S_z^1+S_z^2)\\ &-\tfrac{1}{2}(1+\cos\theta)(1/n)b^2(S_+^1+S_+^2)\ .\end{aligned} \qquad (2.11)$$

Operators (2.11) have the very useful property

$$[A_i,\mathcal{H}]=E_iA_i+[A_i,\mathcal{H}_d] \qquad (2.12)$$

which can be used to write directly equations for Γ_i in the presence of dipolar coupling as

$$\begin{aligned}(E-E_+)\Gamma_+ &=\frac{1}{2\pi}\langle-(1+\cos\theta)(S_z^1+S_z^2)-\frac{1}{\sqrt{n}}\sin\theta b(S_+^1+S_+^2)\rangle\\ &\quad+\langle\langle[A_+,\mathcal{H}_d];(S_+^1+S_+^2)\rangle\rangle\ ,\\ (E-E_0)\Gamma_0 &=\frac{1}{\sqrt{2}\pi}\langle-\sin\theta(S_z^1+S_z^2)+\frac{1}{\sqrt{n}}\cos\theta b(S_+^1+S_+^2)\rangle\\ &\quad+\langle\langle[A_0,\mathcal{H}_d];(S_+^1+S_+^2)\rangle\rangle\ ,\\ (E-E_-)\Gamma_- &=\frac{1}{2\pi}\langle-(1-\cos\theta)(S_z^1+S_z^2)+\frac{1}{\sqrt{n}}\sin\theta b(S_+^1+S_+^2)\rangle\\ &\quad+\langle\langle[A_-,\mathcal{H}_d];(S_+^1+S_+^2)\rangle\rangle\ .\end{aligned} \qquad (2.13)$$

A most delicate step is the decoupling procedure which is necessary to express Green's functions

$$\langle\langle[A_i,\mathcal{H}_d];(S_+^1+S_+^2)\rangle\rangle$$

in terms of the Γ_i's in order to close system (2.13). We use a procedure developed in a previous paper,[8] and begin by considering explicitly

$$\langle\langle[A_+,\mathcal{H}_d];(S_+^1+S_+^2)\rangle\rangle =\frac{1}{2\pi Z_0}\sum_{m,m'}\left((e^{-(1/KT)E_m}-e^{-(1/KT)E_{m'}})\frac{\langle m|A_+\mathcal{H}_d|m'\rangle\langle m'|(S_+^1+S_+^2)]m\rangle-\langle m|\mathcal{H}_dA_+|m'\rangle\langle m'|(S_+^1+S_+^2)|m\rangle}{E+E_m-E_{m'}}\right) \qquad (2.14)$$

which we try to relate to

$$\langle\langle A_+;(S_+^1+S_+^2)\rangle\rangle =\frac{1}{2\pi Z_0}\sum_{m,m'}(e^{-(1/KT)E_m}-e^{-(1/KT)E_{m'}}) \times \frac{\langle m|A_+|m'\rangle\langle m'|(S_+^1+S_+^2)|m\rangle}{E+E_m-E_{m'}} \quad (2.15)$$

in the simplest possible way as

$$\langle\langle [A_+,\mathcal{H}_d];(S_+^1+S_+^2)\rangle\rangle \simeq \lambda(E)\langle\langle A_+;(S_+^1+S_+^2)\rangle\rangle \ . \quad (2.16)$$

Here $\lambda(E)$ is a c function of E which we wish to determine approximately. We treat $\mathcal{H}_d$ in (2.14) at the lowest possible order in α, by keeping its diagonal matrix elements only. Consequently we approximate

$$\langle\langle [A_+,\mathcal{H}_d];(S_+^1+S_+^2)\rangle\rangle \simeq \frac{1}{2\pi Z_0}\sum_{m,m'}(e^{-(1/KT)E_m}-e^{-(1/KT)E_{m'}})(\mathcal{H}_d^{m'm'}-\mathcal{H}_d^{mm}) \times\frac{\langle m|A_+|m'\rangle\langle m'|(S_+^1+S_+^2)m\rangle}{E+E_m-E_{m'}} \ , \quad (2.17)$$

where $\mathcal{H}_d^{mm}$ are diagonal matrix elements of $\mathcal{H}_d$ on the eigenstates of $\mathcal{H}-\mathcal{H}_d$. In order to relate (2.17) to (2.15) as in (2.16), we have to require that (2.16) be a good approximation mainly in the neighborhood of $E=E_+$, which is the only pole of Γ_+ in the absence of dipolar coupling, and presumably its most important pole in the presence of α. In fact in the neighborhood of this pole we have $E_{m'}-E_m=E_+$; consequently, even if we do not know eigenstates $|n,+\rangle$, $|n,0\rangle$, and $|n,-\rangle$ of $\mathcal{H}-\mathcal{H}_d$, corresponding to eigenvalues $n\omega+E_+$, $n\omega+E_0$, and $n\omega+E_-$, we do know that if $|m\rangle=|n,0\rangle$ then $|m'\rangle=|n+1,+\rangle$, while if $|m\rangle=|n,-\rangle$ then $|m'\rangle=|n+1,0\rangle$, because only these two pairs of states satisfy $E_{m'}-E_m=E_+$. On the other hand, the matrix elements of $\mathcal{H}_d$ on eigenstates $|n,i\rangle$ $(i=+,0,-)$ are easily found by transforming $\mathcal{H}_d$ as

$$T^{-1}\mathcal{H}_dT=\alpha\begin{vmatrix}\frac{1}{8}(3\cos^2\theta-1) & (3/4\sqrt{2})\sin\theta\cos\theta & \frac{3}{8}\sin^2\theta\\ (3/4\sqrt{2})\sin\theta\cos\theta & -\frac{1}{4}(3\cos^2\theta-1) & -(3/4\sqrt{2})\sin\theta\cos\theta\\ \frac{3}{8}\sin^2\theta & -(3/4\sqrt{2})\sin\theta\cos\theta & \frac{1}{8}(3\cos^2\theta-1)\end{vmatrix} \quad (2.18)$$

by the matrix

$$T=\begin{vmatrix}a_+^1 & a_0^1 & a_-^1\\ a_+^2 & a_0^2 & a_-^2\\ a_+^3 & a_0^3 & a_-^3\end{vmatrix} \ , \quad (2.19)$$

obtainable from (2.7). We therefore conclude that there are the following two possibilities:

(i) $|m\rangle=|n,0\rangle$ and $|m'\rangle=|n+1,+\rangle$; then

$$\mathcal{H}_d^{mm}=-\tfrac{1}{4}\alpha(3\cos^2\theta-1) \ ,$$

$$\mathcal{H}_d^{m'm'}=\tfrac{1}{8}\alpha(3\cos^2\theta-1) \ ,$$

$$\mathcal{H}_d^{m'm'}-\mathcal{H}_d^{mm}=\tfrac{3}{8}\alpha(3\cos^2\theta-1)\equiv+\lambda \ ; \quad (2.20a)$$

(ii) $|m\rangle=|n,-\rangle$ and $|m'\rangle=|n+1,0\rangle$; then

$$\mathcal{H}_d^{mm}=\tfrac{1}{8}\alpha(3\cos^2\theta-1) \ ,$$

$$\mathcal{H}_d^{m'm'}=-\tfrac{1}{4}\alpha(3\cos^2\theta-1) \ ,$$

$$\mathcal{H}_d^{m'm'}-\mathcal{H}_d^{mm}=-\tfrac{3}{8}\alpha(3\cos^2\theta-1)\equiv-\lambda \ . \quad (2.20b)$$

Since $\mathcal{H}_d$ has been treated at the lowest possible order in α, it should not cause mixing of states $|n,i\rangle$, and it should only shift their energies. These shifts are likely to cause different values for $E_{m'}-E_m$ in cases (i) and (ii) above, and hence two slightly displaced poles of (2.17) in the neighborhood of $E=E_+$ instead of one. Near one of them, vanishing of energy denominators in (2.17) should emphasize the role of terms with $|m\rangle$ and $|m'\rangle$ as in (2.20a); the opposite should be true near the other pole. A little thought yields the following decoupling

$$\langle\langle [A_+,\mathcal{H}_d];(S_+^1+S_+^2)\rangle\rangle$$
$$\sim \lambda_+(E)\,\langle\langle A_+;(S_+^1+S_+^2)\rangle\rangle = \lambda_+(E)\Gamma_+ \;, \quad (2.21)$$

where

$$\lambda_+(E)=\begin{cases}+|\lambda| & (E\gtrsim E_+)\\ -|\lambda| & (E\lesssim E_+)\end{cases} \;. \quad (2.22)$$

Within the same approximations, an analogous procedure yields the following decoupling:

$$\langle\langle [A_0,\mathcal{H}_d];(S_+^1+S_+^2)\rangle\rangle \sim 0 \;,$$
$$\langle\langle [A_-,\mathcal{H}_d];(S_+^1+S_+^2)\rangle\rangle$$
$$\sim \lambda_-(E)\,\langle\langle A_-;(S_+^1+S_+^2)\rangle\rangle = \lambda_-(E)\Gamma_- \;, \quad (2.23)$$

where

$$\lambda_-(E)=\begin{cases}+|\lambda| & (E\gtrsim E_-)\\ -|\lambda| & (E\lesssim E_-)\end{cases} \;. \quad (2.24)$$

Substituting (2.21) and (2.23) into (2.13) we find

$$[E-E_+-\lambda_+(E)]\Gamma_+$$
$$=\frac{1}{2\pi}\langle -(1+\cos\theta)(S_z^1+S_z^2)-\frac{1}{\sqrt{n}}\sin\theta\, b(S_+^1+S_+^2)\rangle$$
$$\equiv \langle R_+\rangle \;,$$

$$(E-E_0)\Gamma_0$$
$$=\frac{1}{\sqrt{2}\pi}\langle -\sin\theta(S_z^1+S_z^2)+\frac{1}{\sqrt{n}}\cos\theta\, b(S_+^1+S_+^2)\rangle$$
$$\equiv \langle R_0\rangle \;,$$

$$[E-E_--\lambda_-(E)]\Gamma_-$$
$$=\frac{1}{2\pi}\langle -(1-\cos\theta)(S_z^1+S_z^2)+\frac{1}{\sqrt{n}}\sin\theta\, b(S_+^1+S_+^2)\rangle$$
$$\equiv \langle R_-\rangle \;. \quad (2.25)$$

In Fig. 1 the Γ's obtained from (2.25) are represented as functions of E for fixed ω_0 and ω. The location of their poles correspond to approximate eigenvalues of the complete Hamiltonian $\mathcal{H}$. Inverting Eq. (2.9), we obtain

$$\langle\langle (S_-^1+S_-^2);(S_+^1+S_+^2)\rangle\rangle \equiv G_1=\tfrac{1}{2}(1+\cos\theta)\Gamma_+ +(1/\sqrt{2})\sin\theta\,\Gamma_0+\tfrac{1}{2}(1-\cos\theta)\Gamma_-$$
$$=\frac{1}{4\pi}\Bigg\{\frac{\langle -(1+\cos\theta)^2(S_z^1+S_z^2)-(1/\sqrt{n})(1+\cos\theta)\sin\theta\, b(S_+^1+S_+^2)\rangle}{E-E_+-\lambda_+(E)}$$
$$+\frac{\langle -\sin^2\theta(S_z^1+S_z^2)+(1/\sqrt{n})\sin\theta\cos\theta\, b(S_+^1+S_+^2)\rangle}{E-E_0}$$
$$+\frac{\langle -(1-\cos\theta)^2(S_z^1+S_z^2)+(1/\sqrt{n})(1-\cos\theta)\sin\theta\, b(S_+^1+S_+^2)\rangle}{E-E_--\lambda_-(E)}\Bigg\} \;. \quad (2.26)$$

Next we consider $G_1(E=\Omega+i\eta)$, and assume $\Omega>\omega$. It is easily seen that the only important term on the right-hand side of (2.26) is the first, whose denominator as $\eta\to 0$ takes the form

$$(\Omega-\omega)-[(\omega_0-\omega)^2+\epsilon^2 n]^{1/2}-\lambda_+(\Omega) \;. \quad (2.27)$$

This denominator in fact may vanish as ω_0 is varied like in a typical ESR experiment. Since λ_+, as defined by (2.22) and (2.20), is also a function of $\omega_0-\omega$, the zeros of (2.27) cannot in general be found analytically. We shall assume that α is small enough, however, and put

$$(\omega_0-\omega)^2 \simeq (\Omega-\omega)^2-\epsilon^2 n$$

in the expression for λ_+. The zeros of (2.27) are thus given by

$$(\omega_0-\omega)^2$$
$$=(\Omega-\omega-\lambda_+)^2-\epsilon^2 n$$
$$\simeq \left\{\Omega-\omega\mp\frac{3\alpha}{8}\frac{|2(\Omega-\omega)^2-3\epsilon^2 n|}{(\Omega-\omega)^2}\right\}^2-\epsilon^2 n \;. \quad (2.28)$$

We remark that, for small α, these zeros are real only for

$$(\Omega-\omega)^2>\epsilon^2 n \;. \quad (2.29)$$

Moreover they coincide for

$$(\Omega-\omega)^2=\tfrac{3}{2}\epsilon^2 n \;. \quad (2.30)$$

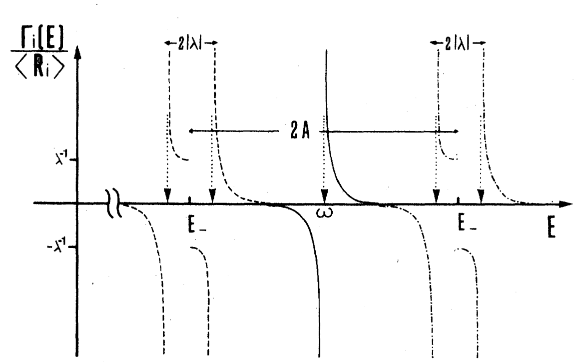

FIG. 1. Diagonal Green's functions along real E axis. Poles are located at approximate eigenvalues of Hamiltonian, and their position is indicated by dotted vertical arrows. – – – $i=-$; —— $i=0$; – · – · – · $i=+$.

From these results it is possible to predict the following features of the ESR spectrum with $\Omega > \omega$ and small α.

(i) If detuning $\Omega - \omega$ is large enough to satisfy (2.29), two lines should appear, almost symmetric about the field

$$\omega_0 = \omega + [(\Omega - \omega)^2 - \epsilon^2 n]^{1/2} \quad . \tag{2.31}$$

This can be described as a shift towards smaller values of the static magnetic field away from $\omega_0 = \Omega$, which is the value of the resonant field for $\epsilon = 0$. There is also a "ghost" doublet at

$$\omega_0 = \omega - [(\Omega - \omega)^2 - \epsilon^2 n]^{1/2} \quad ,$$

but for this doublet $\omega_0 < \omega$ and $\cos\theta < 0$ from (2.8). Consequently it should be much fainter than the first (or "main") doublet because of intensity factor $(1 + \cos\theta)^2$ in the first term of (2.26) and we shall neglect it.

(ii) The splitting of the two components of the main doublet is approximately given by

$$\frac{3\alpha}{4} \frac{|2(\Omega - \omega)^2 - 3\epsilon^2 n|}{|\Omega - \omega| [(\Omega - \omega)^2 - \epsilon^2 n]^{1/2}} \tag{2.32}$$

and it vanishes when (2.30) is satisfied. This effect can be attributed to screening of the dipolar interaction by phonons.

(iii) If detuning $\Omega - \omega$ is small so that (2.29) is not satisfied, the zeros of (2.27) move into the complex ω_0 plane out of the real axis and the ESR spectrum does not display poles, as a function of ω_0. This effect can be qualitatively understood in terms of the single-spin–single-mode model of Fig. 2. The uncoupled ($\epsilon = 0$) energies of the $S =$ (spin $\frac{1}{2}$) + (ω mode) system are represented by dashed lines as functions of Larmor frequency ω_0 of the spin in the static magnetic field along z. If the ESR frequency Ω is slightly larger than ω, resonance occurs for $\omega_0 = \Omega$, as shown in the figure for the n-phonon subspace. When the spin-phonon coupling is turned on ($\epsilon \neq 0$), levels are changed as shown by the continuous lines, and forbidden gaps of amplitude $\sim \epsilon\sqrt{n}$ are created. It is easy to realize that, unless (2.29) is satisfied, the system is out of tune with Ω and cannot absorb this frequency for any value of ω_0. This explains the disappearance of the ESR lines predicted by the present theory, at least within our approximations.

For $\Omega < \omega$ the important term on the right-hand side of (2.26) is the third, whose denominator takes the form

$$(\Omega - \omega) + [(\omega_0 - \omega)^2 + \epsilon^2 n]^{1/2} - \lambda_-(\Omega) \quad .$$

The argument goes through as previously, except that

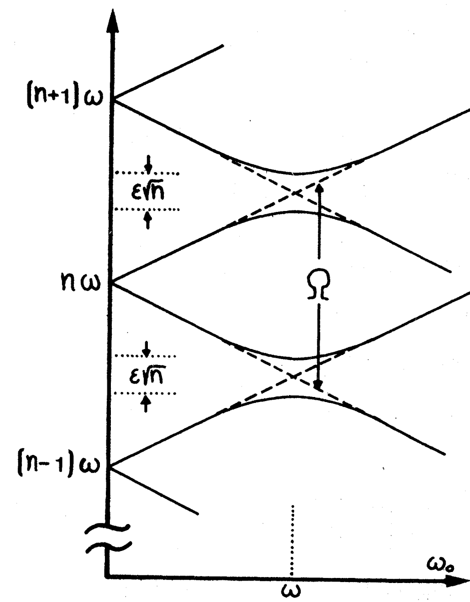

FIG. 2. Energy levels of single-spin–single-mode (ω) system for $\epsilon = 0$ (dashed lines) and $\epsilon \neq 0$ (continuous lines) as functions of static magnetic field ω_0. Energy $\Omega > \omega$ of monitoring photons falls within "forbidden" gaps of amplitude $\epsilon\sqrt{n}$.

now the main doublet is centered at

$$\omega_0 = \omega - [(\Omega - \omega)^2 - \epsilon^2 n]^{1/2} \quad .$$

The splitting of the two lines of this doublet is again given by (2.32), and the same considerations as before can account for its disappearance when (2.29) is not satisfied.

III. MULTIMODE CASE

We are now ready to tackle the multimode problem of (1.1) which we write in the form

$$\mathcal{H} = \mathcal{H}_0 + \mathcal{H}_1 \quad ,$$

$$\mathcal{H}_0 = \omega_0 (S_z^1 + S_z^2) + \omega \sum_k b_k^\dagger b_k + \tfrac{1}{2}\epsilon \sum_k [b_k (S_+^1 + S_+^2) + b_k^\dagger (S_-^1 + S_-^2)] \quad , \tag{3.1}$$

$$\mathcal{H}_1 = \sum_k (\omega_k - \omega) b_k^\dagger b_k + \mathcal{H}_d \quad ,$$

where we have approximated by neglecting the small dependence of ϵ_k from k within the narrow hot band. The coupled Green's-function equations relevant to our purpose are

$$(E-\omega_0)\langle\langle(S_-^1+S_-^2);(S_+^1+S_+^2)\rangle\rangle = -\langle(S_z^1+S_z^2)\rangle - \epsilon\left\langle\!\!\left\langle\left[\sum_k b_k\right](S_z^1+S_z^2);(S_+^1+S_+^2)\right\rangle\!\!\right\rangle + \langle\langle[(S_-^1+S_-^2),\mathcal{H}_1];(S_+^1+S_+^2)\rangle\rangle \;,$$

$$(E-\omega)\left[\sum_k b_k\right](S_z^1+S_z^2);(S_+^1+S_+^2)\rangle\!\rangle = \frac{1}{2\pi}\left\langle\left[\sum_k b_k\right](S_+^1+S_+^2)\right\rangle + \tfrac{1}{2}\epsilon\Bigg[\left\langle\!\!\left\langle\left[\sum_k b_k\right]^2(S_+^1+S_+^2);(S_+^1+S_+^2)\right\rangle\!\!\right\rangle - \left\langle\!\!\left\langle\left[\sum_k b_k^\dagger\right]\left[\sum_k b_k\right](S_-^1+S_-^2);(S_+^1+S_+^2)\right\rangle\!\!\right\rangle + \mathfrak{N}\langle\langle(S_z^1+S_z^2)(S_-^1+S_-^2);(S_+^1+S_+^2)\rangle\rangle\Bigg] + \left\langle\!\!\left\langle\left[\left[\sum_k b_k\right](S_z^1+S_z^2),\mathcal{H}_1\right];(S_+^1+S_+^2)\right\rangle\!\!\right\rangle \;,$$

$$(E+\omega_0-2\omega)\left\langle\!\!\left\langle\left[\sum_k b_k\right]^2(S_+^1+S_+^2);(S_+^1+S_+^2)\right\rangle\!\!\right\rangle = \epsilon\Bigg[\left\langle\!\!\left\langle\left[\sum_k b_k^\dagger\right]\left[\sum_k b_k\right]^2(S_z^1+S_z^2);(S_+^1+S_+^2)\right\rangle\!\!\right\rangle + \mathfrak{N}\left\langle\!\!\left\langle\left[\sum_k b_k\right](S_+^1+S_+^2)(S_-^1+S_-^2);(S_+^1+S_+^2)\right\rangle\!\!\right\rangle\Bigg] + \left\langle\!\!\left\langle\left[\left[\sum_k b_k\right]^2(S_+^1+S_+^2),\mathcal{H}_1\right];(S_+^1+S_+^2)\right\rangle\!\!\right\rangle \;, \quad (3.2)$$

where $\mathfrak{N}$ is the number of modes in the hot band. We remark that in the absence of $\mathcal{H}_1$ the structure of system (3.2) is similar to that of system (2.2) in the same limit. Moreover, under the conditions of phonon avalanche, we expect[2] $n/\mathfrak{N} \gg 1$, where n is the *total* number of phonons in the hot band. Consequently we may neglect the $\mathfrak{N}$-dependent terms in (3.2). We also wish to approximate

$$\left\langle\!\!\left\langle\left[\sum_k b_k^\dagger\right]\left[\sum_k b_k\right]\hat{O};(S_+^1+S_+^2)\right\rangle\!\!\right\rangle \sim n\,\langle\langle\hat{O};(S_+^1+S_+^2)\rangle\rangle \;, \quad (3.3)$$

where $\hat{O}$ is an operator in (3.2). This is not a trivial approximation, since it involves neglecting all terms containing

$$\frac{\langle m|b_k^\dagger b_{k'}\hat{O}|m'\rangle\langle m'|(S_+^1+S_+^2)|m\rangle}{E+E_m-E_{m'}} \quad (k\neq k')$$

as a factor in the expression of the Green's functions. In fact, processes of absorption and emission of photons in different modes might interfere and contribute fairly substantially to the residue of the exact Green's functions at their poles, as thoroughly discussed by Swain[10] in connection with the problem of resonance fluorescence. We remark, however, that these terms should not change the location of the poles of our Green's functions, as long as the effects of $\mathcal{H}_1$ are assumed negligible, since all modes have the same frequency in $\mathcal{H}_0$. Moreover the contribution of these processes to the residues must disappear in the limit of an infinitely narrow avalanche band; so we expect this contribution to be negligible if the hot band is narrow enough, which should always be the case for a phonon avalanche.[4,5] Consequently we feel entitled to use approximation (3.3) in (3.2) which, upon neglect of $\mathcal{H}_1$, takes the form

$$(E-\omega_0)G_1+(1/\sqrt{2})\epsilon\sqrt{n}\,G_2 = -(1/\pi)\langle(S_z^1+S_z^2)\rangle \;,$$
$$(1/\sqrt{2})\epsilon\sqrt{n}\,G_1+(E-\omega)G_2+(1/\sqrt{2})\epsilon\sqrt{n}\,G_3 = \frac{1}{2\pi}\left(\frac{2}{n}\right)^{1/2}\left\langle\left[\sum_k b_k\right](S_+^1+S_+^2)\right\rangle \;, \quad (3.4)$$
$$(1/\sqrt{2})\epsilon\sqrt{n}\,G_2+(E+\omega_0-2\omega)G_3 = 0 \;,$$

where

$$G_1 = \langle\langle(S_-^1+S_-^2);(S_+^1+S_+^2)\rangle\rangle \;,$$
$$G_2 = \left(\frac{2}{n}\right)^{1/2}\left\langle\!\!\left\langle\left[\sum_k b_k\right](S_z^1+S_z^2);(S_+^1+S_+^2)\right\rangle\!\!\right\rangle \;, \quad (3.5)$$
$$G_3 = -\frac{1}{n}\left\langle\!\!\left\langle\left[\sum_k b_k\right]^2(S_z^1+S_z^2);(S_+^1+S_+^2)\right\rangle\!\!\right\rangle \;.$$

System (3.4) is essentially the same as (2.3), and it can be solved by the same technique used in the previous section. We thus arrive at the "diagonal" Green's functions

$$(E-E_+)\Gamma_+=(1/2\pi)\langle -(1+\cos\theta)(S_z^1+S_z^2) - \frac{1}{\sqrt{n}}\sin\theta\left(\sum_k b_k\right)(S_+^1+S_+^2)\rangle + \langle\langle [A_+,\mathcal{H}_1];(S_+^1+S_+^2)\rangle\rangle ,$$

$$(E-E_0)\Gamma_0=(1/\sqrt{2}\pi)\langle -\sin\theta(S_z^1+S_z^2) + \frac{1}{\sqrt{n}}\cos\theta\left(\sum_k b_k\right)(S_+^1+S_+^2)\rangle + \langle\langle [A_0,\mathcal{H}_1];(S_+^1+S_+^2)\rangle\rangle , \quad (3.6)$$

$$(E-E_-)\Gamma_-=(1/2\pi)\langle -(1-\cos\theta)(S_z^1+S_z^2) + \frac{1}{\sqrt{n}}\sin\theta\left(\sum_k b_k\right)(S_+^1+S_+^2)\rangle + \langle\langle [A_-,\mathcal{H}_1];(S_+^1+S_+^2)\rangle\rangle ,$$

where the E_i's are still given by (2.6), but with a different meaning for ω and n as discussed above, and where $\sin\theta$ and $\cos\theta$ have been chosen to be formally the same as (2.8). Also the Γ_i's are formally the same as in (2.10), but

$$A_+=\tfrac{1}{2}(1+\cos\theta)(S_-^1+S_-^2) - \sin\theta\frac{1}{\sqrt{n}}\left(\sum_k b_k\right)(S_z^1+S_z^2) - \tfrac{1}{2}(1-\cos\theta)\frac{1}{n}\left(\sum_k b_k\right)^2(S_+^1+S_+^2) ,$$

$$A_0=(1/\sqrt{2})\sin\theta(S_-^1+S_-^2) + \cos\theta\left(\frac{2}{n}\right)^{1/2}\left(\sum_k b_k\right)(S_z^1+S_z^2) + \frac{1}{\sqrt{2}}\sin\theta\frac{1}{n}\left(\sum_k b_k\right)^2(S_+^1+S_+^2) , \quad (3.7)$$

$$A_-=\tfrac{1}{2}(1-\cos\theta)(S_-^1+S_-^2) + \sin\theta\frac{1}{\sqrt{n}}\left(\sum_k b_k\right)(S_z^1+S_z^2) - \tfrac{1}{2}(1+\cos\theta)\frac{1}{n}\left(\sum_k b_k\right)^2(S_+^1+S_+^2) .$$

It is easy to show that unitary operator

$$T=\exp\left(\frac{\theta}{2\sqrt{n}}\sum_k[b_k(S_+^1+S_+^2)-b_k^\dagger(S_-^1+S_-^2)]\right) \quad (3.8)$$

can be used to obtain (3.7) as

$$T^{-1}\begin{vmatrix} (S_-^1+S_-^2) \\ \left(\frac{2}{n}\right)^{1/2}\left(\sum_k b_k\right)(S_z^1+S_z^2) \\ -\frac{1}{n}\left(\sum_k b_k\right)^2(S_+^1+S_+^2)\end{vmatrix} T \simeq \begin{vmatrix} A_+ \\ A_0 \\ A_- \end{vmatrix} \quad (3.7a)$$

within the same approximations as used in decoupling from (3.2) to (3.4). Since we have

$$[A_i,H_0]\simeq E_iA_i$$

as in (2.12), we may use T to express eigenfunctions $|m\rangle$ of $\mathcal{H}_0$ in the Green's functions in terms of the free-field eigenfunctions as

$$\begin{vmatrix}\{|n,+\rangle\} \\ \{|n,0\rangle\} \\ \{|n,-\rangle\}\end{vmatrix} \simeq T\begin{vmatrix}\{|n,\uparrow\uparrow\rangle\} \\ \{|n+1,\rightleftarrows\rangle\} \\ \{|n+2,\downarrow\downarrow\rangle\}\end{vmatrix} , \quad (3.9)$$

where $\{|n,\uparrow\uparrow\rangle\}$ denotes the ensemble of all degenerate eigenstates of the free-field Hamiltonian with n phonons in the hot band and both spins up. Within the mentioned approximations, operator T in (3.9) transforms this ensemble into that of the eigenfunctions of $\mathcal{H}_0$ relative to eigenvalue $n\omega+E_+$, which we indicate by $\{|n,+\rangle\}$. Analogously, $\{|n+1,\rightleftarrows\rangle\}$ represents the ensemble of eigenstates with $n+1$ phonons and

$$\frac{1}{\sqrt{2}}(|\uparrow\downarrow\rangle+|\downarrow\uparrow\rangle)$$

as a spinor part, while $\{|n+2,\downarrow\downarrow\rangle\}$ is relative to free-field eigenstates with $n+2$ phonons and both spins down. These two ensembles are transformed by T into $\{|n,0\rangle\}$ and $\{|n,-\rangle\}$ respectively, which belong to eigenvalues $n\omega+E_0$ and $n\omega+E_-$ of $\mathcal{H}_0$.

Following the same lines as in Sec. II, for $i=+,0,-$ we approximate:

$$\langle\langle [A_i,\mathcal{H}_1];(S_+^1+S_+^2)\rangle\rangle \simeq \frac{1}{2\pi Z_0}\sum_{m,m'}(e^{-\beta E_m}-e^{-\beta E_{m'}})(\mathcal{H}_{\rm I}^{m'm'}-\mathcal{H}_{\rm I}^{mm}) \times \frac{\langle m|A_i|m'\rangle\langle m'|(S_+^1+S_+^2)|m\rangle}{E+E_m-E_{m'}} \tag{3.10}$$

in the neighborhood of $E\sim E_{m'}-E_m\sim E_i$, where

$$|m\rangle\subset\{|n,0\rangle\} \text{ and } |m'\rangle\subset\{|n+1,+\rangle\} \text{ or } |m\rangle\subset\{|n,-\rangle\} \text{ and } |m'\rangle\subset\{|n+1,0\rangle\} \quad (i=+);$$

$$|m\rangle\subset\{|n,+\rangle\} \text{ and } |m'\rangle\subset\{|n+1,+\rangle\}, \text{ or } |m\rangle\subset\{|n,0\rangle\} \text{ and } |m'\rangle\subset\{|n+1,0\rangle\}\ ,$$

or (3.11)

$$|m\rangle\subset\{|n,-\rangle\} \text{ and } |m'\rangle\subset\{|n+1,-\rangle\} \quad (i=0)\ ;$$

$$|m\rangle\subset\{|n,+\rangle\} \text{ and } |m'\rangle\subset\{|n+1,0\rangle\} \text{ or } |m\rangle\subset\{|n,0\rangle\} \text{ and } |m'\rangle\subset\{|n+1,-\rangle\} \quad (i=-)\ .$$

We consider separately the two contributions to $\mathcal{H}_{\rm I}^{m'm'}-\mathcal{H}_{\rm I}^{mm}$. As for the phonon part, it is evident from (3.11) that the average number of phonons in states $|m'\rangle$ is always larger than in states $|m\rangle$ by one. Therefore we should expect always

$$\langle m'|\sum_k(\omega_k-\omega)b_k^\dagger b_k|m'\rangle-\langle m|\sum_k(\omega_k-\omega)b_k^\dagger b_k|m\rangle\sim\omega_{k'}-\omega\ , \tag{3.12}$$

where k' is the mode selected by a particular pair m,m'. The average of (3.12) over all possible pairs should vanish, however, since presumably the various pairs yield in turn as many positive as negative values of $\omega_{k'}-\omega$. On this basis, we shall neglect contributions of the phonon part of $\mathcal{H}_1$ to $\mathcal{H}_{\rm I}^{m'm'}-\mathcal{H}_{\rm I}^{mm}$. The problem is thus reduced to evaluating $\mathcal{H}_d^{m'm'}$ and $\mathcal{H}_d^{mm}$. Using (3.9), we find the following results relative to the cases in (3.11): for $i=+$,

$$\mathcal{H}_{\rm I}^{m'm'}=\{\langle n+1,\uparrow\uparrow|\}T^{-1}\mathcal{H}_dT\{|n+1,\uparrow\uparrow\rangle\},\quad \mathcal{H}_{\rm I}^{mm}=\{\langle n+1,\rightleftarrows|\}T^{-1}\mathcal{H}_dT\{|n+1,\rightleftarrows\rangle\}$$

or

$$\mathcal{H}_{\rm I}^{m'm'}=\{\langle n+2,\rightleftarrows|\}T^{-1}\mathcal{H}_dT\{|n+2,\rightleftarrows\rangle\},\quad \mathcal{H}_{\rm I}^{mm}=\{\langle n+2,\downarrow\downarrow|\}T^{-1}\mathcal{H}_dT\{|n+2,\downarrow\downarrow\rangle\}\ ;$$

for $i=0$,

$$\mathcal{H}_{\rm I}^{m'm'}=\{\langle n+1,\uparrow\uparrow|\}T^{-1}\mathcal{H}_dT\{|n+1,\uparrow\uparrow\rangle\},\quad \mathcal{H}_{\rm I}^{mm}=\{\langle n,\uparrow\uparrow|\}T^{-1}\mathcal{H}_dT\{|n,\uparrow\uparrow\rangle\}$$

or

$$\mathcal{H}_{\rm I}^{m'm'}=\{\langle n+2,\rightleftarrows|\}T^{-1}\mathcal{H}_dT\{|n+2,\rightleftarrows\rangle\},\quad \mathcal{H}_{\rm I}^{mm}=\{\langle n+1,\rightleftarrows|\}T^{-1}\mathcal{H}_dT\{|n+1,\rightleftarrows\rangle\}$$

or

$$\mathcal{H}_{\rm I}^{m'm'}=\{\langle n+3,\downarrow\downarrow|\}T^{-1}\mathcal{H}_dT\{|n+3,\downarrow\downarrow\rangle\},\quad \mathcal{H}_{\rm I}^{mm}=\{\langle n+2,\downarrow\downarrow|\}T^{-1}\mathcal{H}_dT\{|n+2,\downarrow\downarrow\rangle\}\ ;$$

for $i=-$,

$$\mathcal{H}_{\rm I}^{m'm'}=\{\langle n+2,\rightleftarrows|\}T^{-1}\mathcal{H}_dT\{|n+2,\rightleftarrows\rangle\},\quad \mathcal{H}_{\rm I}^{mm}=\{\langle n,\uparrow\uparrow|\}T^{-1}\mathcal{H}_dT\{|n,\uparrow\uparrow\rangle\}$$

or

$$\mathcal{H}_{\rm I}^{m'm'}=\{\langle n+3,\downarrow\downarrow|\}T^{-1}\mathcal{H}_dT\{|n+3,\downarrow\downarrow\rangle\},\quad \mathcal{H}_{\rm I}^{mm}=\{\langle n+1,\rightleftarrows|\}T^{-1}\mathcal{H}_dT\{|n+1,\rightleftarrows\rangle\}\ .$$

(3.13)

Transforming the dipolar Hamiltonian by unitary operator (3.8) with the usual approximations and neglecting out of diagonal operators yields

$$T^{-1}\mathcal{H}_d T \simeq \tfrac{1}{2}(3\cos^2\theta - 1)\mathcal{H}_d \tag{3.14}$$

and (3.13) become

$$\mathcal{H}_1^{m'm'} = \tfrac{1}{3}\lambda \text{ and } \mathcal{H}_1^{mm} = -\tfrac{2}{3}\lambda$$

or

$$\mathcal{H}_1^{m'm'} = -\tfrac{2}{3}\lambda \text{ and } \mathcal{H}_1^{mm} = \tfrac{1}{3}\lambda \quad (i=+) ,$$

$$\mathcal{H}_1^{m'm'} = \mathcal{H}_1^{mm} = \tfrac{1}{3}\lambda \text{ or } -\tfrac{2}{3}\lambda \quad (i=0) , \tag{3.15}$$

$$\mathcal{H}_1^{m'm'} = -\tfrac{2}{3}\lambda \text{ and } \mathcal{H}_1^{mm} = \tfrac{1}{3}\lambda$$

or

$$\mathcal{H}_1^{m'm'} = \tfrac{1}{3}\lambda \text{ and } \mathcal{H}_1^{mm} = -\tfrac{2}{3}\lambda \quad (i=-) ,$$

where λ is formally the same as in (2.20), although θ is different since we are dealing with the multimode case. Upon substitution of (3.15) in (3.10) we find the following decoupling:

$$\langle\langle [A_i, \mathcal{H}_1]; (S_+^1 + S_+^2)\rangle\rangle \simeq \lambda_i(E)\Gamma_i , \tag{3.16}$$

where $\lambda_i(E)$ is given by (2.22) and (2.24) for $i=+,-$, while $\lambda_0(E)=0$. Thus we see that, within the present approximations, the results for the multimode case are not formally different from those obtained for the single-mode case.

IV. DISCUSSION AND CONCLUSIONS

On the basis of the results of the treatment outlined in Sec. III, we may conclude that the features of the ESR spectrum should be approximately the same as those discussed at the end of Sec. II, except that ω and ϵ in (2.31) and (2.32) should be interpreted as average phonon frequency and average coupling constant, respectively, taken over all modes in the hot band, and n as the total number of avalanche phonons. Naturally we expect that the two lines in the spectrum, which are split as in (2.32), should become broadened when the finite width of the avalanche band is taken into account in a better approximation. Moreover, we remark that the present treatment might run into trouble if the width of the hot band becomes so large as to include the ESR frequency Ω. We feel confident enough that our model represents the main features of the problem, however, to expect that a sharp decrease in the intensity of the ESR line should take place when the avalanche band includes the ESR frequency Ω, as discussed at the end of Sec. II for the single-mode case. We remark that the physical origin of this effect appears to be of a different nature than ordinary saturation broadening, and more similar to the classical splitting of the response peak of two coupled oscillators of the same frequency. For large detunings, we adapt (2.31) to the multimode case as discussed above by putting

$$\omega = \langle\omega_k\rangle, \quad \epsilon = \langle\epsilon_k\rangle, \quad n = \sum_k n_k . \tag{4.1}$$

Furthermore, we develop (2.31) in powers of ϵ, obtaining

$$\Omega \sim \omega_0 - \langle\epsilon_k\rangle^2 n / 2(\langle\omega_k\rangle - \omega_0) , \tag{4.2}$$

which is a result analogous to expression (7) of our previous paper,[3] when counter-rotating terms are neglected, and which can be taken as the effective Larmor frequency of the spins. The decrease in the effective dipolar interaction, as obtained by (2.32) upon substitution of (4.1), should be a most prominent feature of the ESR spectrum for

$$\Omega - \langle\omega_k\rangle = \left[\tfrac{3}{2}\langle\epsilon_k\rangle^2 \left(\sum_k n_k\right)\right]^{1/2} . \tag{4.3}$$

In this case, in fact, it should manifest itself experimentally as a drastic reduction of the linewidth in the presence of the phonon avalanche. The shift of the resonance field is likely to be of the same nature as the ac Stark effect predicted and observed in optical spectroscopy.[11] The reduction of dipolar broadening may be attributed to screening of dipolar interaction due to absorption and reemission of phonons in the avalanche band. Result (4.3) is very much remindful of the "magic angle" concept, which was introduced not long ago to explain the increase of relaxation time T_2 in the presence of a strong coherent electromagnetic field in NMR[12] and ESR[13] experiments. We remark however that in our case the effect is due to an incoherent phonon avalanche, and in fact a substantially different, and perhaps more sophisticated, approach has been necessary in order to arrive at the present conclusions.

The experimental results which may be correlated to the present theory are rather scanty since no experiment, to the best of our knowledge, has yet been performed specifically to the purpose of investigating this problem. Phonon avalanche generated by initially inverted populations of magnetic levels of Ce impurities in lanthanum magnesium nitrate was obtained by Brya and Wagner,[2] and its time evolution was monitored by looking at the ESR lineshape of the Ce atoms. Since the ESR was performed by suddenly shifting the Larmor frequency of the Ce atoms out of the hot band of average frequency $\langle\omega_k\rangle$ through monitoring frequency Ω ($\Omega < \langle\omega_k\rangle$), on the basis of our theory a displacement of the line towards low fields and a broadening given by (2.32) should have been observed. Using the values of the various physical parameters given by Brya and Wagner, and calculating in the Debye approximation

$$\epsilon \sim \hbar[2\pi/g(\omega_0)T_1]^{1/2} ,$$
$$g(\omega_0) = 3\omega_0^2/2\pi^2 c^3 , \quad (4.4)$$

we predict, under the assumption of a detuning $\omega - \Omega = 10$ G, a shift of $\sim$2.7 G and a width reduction factor of $\sim$0.42 for $n = 10^{18}$ phonons in the hot band, or a shift of $\sim$0.2 G and a width reduction factor of $\sim$0.95 for $n = 10^{17}$. We are inclined to favor the latter situation as more realistic, in spite of the fairly long lifetime of avalanche phonons experimentally measured,[5] because of partial initial inversion of the paramagnetic atoms in the sample and other likely losses. If our expectations are correct, the shift was too small to be observed in Brya and Wagner's experiment, since it should have been masked by larger shifts which have been experimentally observed and attributed to demagnetization effects.[2] A line narrowing should have been observed, however, even if the dipolar interaction is only one of the contributions to the experimental width, and indeed it cannot be excluded on the basis of the experimental line profiles displayed in Fig. 13 of Brya and Wagner's paper,[2] especially towards the end of the avalanche. Two criticisms of our approach should be explicitly made at this point. First, our calculations are valid for small α, and in particular for $\alpha < \epsilon\sqrt{n}$ at small detuning; in the experimental setup discussed above, however, detuning $\omega - \Omega$ is likely to be large enough to warrant applicability of the theory. Second and more important, the Green's-function technique applies to quasiequilibrium situations, while in Brya and Wagner's experiment the system is far from equilibrium for most of the avalanche process; consequently one should be cautious in using our theory in such an experimental context. Towards the end of the avalanche however, the spins may reach a state not too far from equilibrium with the hot-band phonons which have not yet disappeared because of finite phonon lifetime, and we may hope to extend the approximate validity of our results to this region of time. With the above warnings, we wish to mention also the spin-echo results on the same paramagnetic material by Mims and Taylor,[5] who measured a T_2 relaxation time of 4 μsec during a phonon avalanche. This may be contrasted with a previous measurement in the absence of any avalanche by Cowan and Kaplan,[14] who found $T_2 \sim 1$ μsec. This increase of spin-spin relaxation time during a phonon avalanche may not be very meaningful, however, due to experimental difficulties inherent in this kind of measurement.[5]

In conclusion, we have studied, by a Green's-function technique, the effects of a strong phonon avalanche on the ESR spectrum of a pair of paramagnetic atoms with dipolar interaction and spin-phonon coupling. When the monitoring ESR frequency Ω falls out of the hot band of phonons, our theory predicts a shift of the ESR line and its narrowing. The latter effect is attributed to screening of the spin-spin interaction, and its magnitude depends on the physical parameters of the system; in ideal circumstances (2.32) yields a "magic" number of avalanche phonons which gives zero dipolar broadening. For small detuning, the details of the ESR spectrum should be calculated numerically; also the applicability of our theory might become doubtful if the monitoring frequency falls within the hot band, although we expect a reduction of intensity on the basis of the single-spin—single-mode case and of qualitative arguments. The results of the present theory agree with those previously found for the eigenvalue spectrum of (1.1) in the off-resonance case by a unitary transformation technique.[3] Comparison of our calculation with previous experimental results[2,5,14] does not yield conclusive evidence as to the existence of effects predicted by the present theory. It would be interesting to have a specific investigation of this problem done, since an increase of spin-spin relaxation time T_2 during a phonon avalanche may prove important in lowering the threshold for phonon maser operation.[15]

ACKNOWLEDGMENT

This work was partially supported by Comitato Regionale Ricerche Nucleari e Struttura della Materia (CRRNSM), Palermo, Italy.

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