[Appendix (a)] it has been demonstrated that these terms are essentially equivalent to the inclusion of higher order multipole interactions, and that it is plausible to infer that their influence is most marked for the long-wavelength acoustic modes, whose frequencies depend only on the elastic constants. It follows that both frequency distributions and dispersion curves should be almost unaffected, except in the low-frequency regions, and it would be interesting to test these assertions experimentally.

ACKNOWLEDGMENTS

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Interaction of Ultrasonic Waves with Electron Spins

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We discuss the interaction between paramagnetic atoms and elastic waves at microwave frequencies by means of a total Hamiltonian comprising sound field, interaction, and spins. From this Hamiltonian and the Heisenberg commutation rules we obtain a set of coupled equations of motion. The condition of compatability leads in the usual way to a secular determinant, the solution of which is a dispersion relation exhibiting the familiar anomalous change in velocity and absorption of waves near resonance.

I. INTRODUCTION

With the advent of methods for generating and detecting ultrasonic waves at microwave frequencies, it has become possible to study the interaction between lattice vibrations and electron spin systems directly. Such studies have been carried out by observing the effects of ultrasonic waves on paramagnetic resonance[1,2] and, conversely, by noting the effects of paramagnetic ions on the propagation of ultrasonic waves.[3–6] It is the purpose of this paper to discuss the latter phenomenon, and, in particular, to develop a theory of elastic wave propagation in a solid containing resonant spins.

II. DISPERSION OF SOUND BY RESONANT SPIN SYSTEMS

The experimentally observed change in the velocity of sound propagation[6] when the ultrasonic frequency approaches the resonant frequency of allowed spin transitions closely parallels the behavior of electromagnetic waves propagating in a medium containing resonant atoms. The latter phenomenon of electromagnetic dispersion is well known and easily described by Maxwell's equations for the electromagnetic field and the dynamical equations for the atomic system. When the atomic system is represented by a harmonic oscillator, the problem is particularly simple and readily formulated in terms of Maxwell's equations and Newton's equations of motion for the oscillator, these same ideas being extendable to purely quantum-mechanical systems by means of time-dependent perturbation theory. To treat the dispersion of sound, we employ a model analogous to that of the harmonic oscillator used in elementary treatments of electromagnetic dispersion and derive a set of equations of motion for the composite sound field and spin system, a simultaneous solution of which yields a dispersion relation. We expect the scheme to be extendable to spin systems obeying purely quantum laws of motion by the use of quantum theory. As we shall see, such a program can be carried out subject to the assumption that the spins are uniformly distributed and that there are many spins per sonic wavelength. The system of spin $S=1/2$ is the counterpart of the harmonic oscillator in the optical case.

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[1] E. H. Jacobsen, N. S. Shiren, and E. B. Tucker, Phys. Rev. Letters **3**, 81 (1959).

[2] N. S. Shiren and E. B. Tucker, Phys. Rev. Letters **6**, 105 (1961).

[3] E. B. Tucker, Phys. Rev. Letters **6**, 183 (1961).

[4] N. S. Shiren, Phys. Rev. Letters **6**, 168 (1961).

[5] E. B. Tucker, Phys. Rev. Letters **6**, 547 (1961).

[6] N. S. Shiren, Phys. Rev. **128**, 2103 (1962).

Semiclassical Treatment of $S=1/2$

We start with the Hamiltonian of the complete system (sound+interaction+spin) and derive from it a set of coupled linear equations of motion, the simultaneous solution of which yields a dispersion relation between wave velocity and frequency. For simplicity we consider compressional sound waves propagating along the x direction. We take the Hamiltonian to be

$$\mathcal{H}=\sum_n \left\{\frac{P_n^2}{m}+\frac{K}{2}(U_n-U_{n+1})^2 +\hbar\epsilon(U_{n+1}-U_{n-1})S_x^{(n)}+g\beta H S_z^{(n)}\right\}, \quad (1)$$

where we assume one atom of mass m and spin 1/2 per unit cell. U_n is the displacement and P_n is the momentum of atom n along x. K is the restoring force between nearest neighbors and H is the dc magnetic field along the z direction. $S_x^{(n)}$ and $S_z^{(n)}$ are, respectively, the x and z components of the spin for the unpaired electron on atom n, and ϵ is the coupling constant between the strain at position n and the spin components $S_x^{(n)}$. The spin-lattice coupling is chosen so that a component in $U_{n+1}-U_{n-1}$ at the resonance frequency of the spin is able to induce spin transitions. We do not inquire into the origin of this coupling, except to point out that by Kramers' theorem, ϵ vanishes with H. The simplest assumption is that the lattice oscillations modulate the xz components of the g tensors, in which case ϵ varies linearly with H. From the commutators we obtain the Heisenberg equations of motion for the operators P_n, U_n, and $S^{(n)}$, which are

$$\dot{P}_n=\frac{1}{i\hbar}[P_n,\mathcal{H}]=K(U_{n+1}+U_{n-1}-2U_n) +\hbar\epsilon(S_x^{(n+1)}-S_x^{(n-1)}),$$

$$\dot{U}_n=\frac{1}{i\hbar}[U_n,\mathcal{H}]=\frac{P_n}{m},$$

$$\dot{S}_x^{(n)}=\frac{1}{i\hbar}[S_x^{(n)},\mathcal{H}]=-g\frac{\beta H}{\hbar}S_y^{(n)},$$

$$\dot{S}_y^{(n)}=\frac{1}{i\hbar}[S_y^{(n)},\mathcal{H}]=-\epsilon(U_{n+1}-U_{n-1})S_z^{(n)}+g\frac{\beta H}{\hbar}S_x^{(n)},$$

$$\dot{S}_z^{(n)}=\frac{1}{i\hbar}[S_z^{(n)},\mathcal{H}]=\epsilon(U_{n+1}-U_{n-1})S_y^{(n)},$$

where $(S_x^{(n)},S_y^{(n)})=iS_z^{(n)}$, etc., $\beta=eh/2mc$ so that S_x, S_y, S_z do not contain h. We assume for the moment that the spin-spin relaxation time (to be designated by τ) is infinite, a constraint which is removed when discussing attenuation.

The preceding equations can be rearranged to give

$$m\ddot{U}_n=K(U_{n+1}+U_{n-1}-2U_n) +\hbar\epsilon(S_x^{(n+1)}-S_y^{(n-1)}), \quad (2)$$

$$d^2S_x^{(n)}/dt^2=\omega_0\epsilon(U_{n+1}-U_{n-1})S_z^{(n)}-\omega_0^2S_x^{(n)}. \quad (3)$$

They would be linear were it not for the term $(U_{n+1}-U_{n-1})S_z^{(n)}$. Noting that the sonic wavelength is long compared with the atomic spacing "a" and that the rate of change of S_z with time is of order ϵ^2, a reasonable physical approximation is to replace S_z by its average value per unit volume. A solution is now readily found by assuming that both U_n and $S_x^{(n)}$ vary as $e^{i(\omega t-kna)}$ where "na" is the position along x of the nth atom. k and ω are then related by a typical dispersion relation,

$$(m\omega^2-Kk^2a^2)(\omega^2-\omega_0^2)+4\epsilon^2\hbar\omega_0\langle S_z\rangle k^2a^2=0, \quad (4)$$

where we have taken the long wave limit and replaced $\sin ka$ by ka and set $g\beta H=\hbar\omega_0$. With the definition $v_0^2=Ka^2/m$, the square of the phase velocity in zero magnetic field, we obtain finally the relation for the sonic index of refraction as a function of frequency, where $\langle S_z\rangle$ is the mean value of S_z per unit volume.

$$\left(\frac{v_0}{v}\right)^2=\left[1+\frac{4\epsilon^2 g\beta H\langle S_z\rangle/K}{\omega_0^2-\omega^2}\right]^{-1} =1-\frac{4\epsilon^2 g\beta H\langle S_z\rangle/K}{\omega_0^2+4\epsilon^2 g\beta H\langle S_z\rangle/K-\omega^2}. \quad (5)$$

It is interesting to compare the above equation with that obtained in the case of optical dispersion near a single resonant frequency ω_0,[7]

$$\left(\frac{c}{v}\right)^2=1+\frac{4\pi Ne^2/m}{\omega_0^2-\omega^2}.$$

Both equations take the same form when $\langle S_z\rangle$ is negative, i.e., when we have a normal population. The small difference between the two expressions results from the fact that in our model the spins are coupled to the elastic *strain* rather than to the *amplitude* of the atomic displacement. A plot of $(v_0/v)^2$ appears in Fig. 1(a) for negative values of $\langle S_z\rangle$. A positive value of $\langle S_z\rangle$ corresponds to an inverted population, and we shall return to a consideration of this point presently. Finally, we would emphasize that since we are dealing with coupled systems [Eq. (2)], the disturbance which propagates is a mixture of sound and transverse spin waves (i.e., waves in S_x and S_y, but not S_z). For small coupling ϵ, most of the wave energy is contained in the elastic strain field and so propagates as a nearly pure sound wave. As ϵ increases, and particularly near resonance,

[7] W. K. H. Panofsky and M. Phillips, *Classical Electricity and Magnetism* (Addison-Wesley Publishing Company, Inc., Reading, Massachusetts, 1955), Chap. 21.

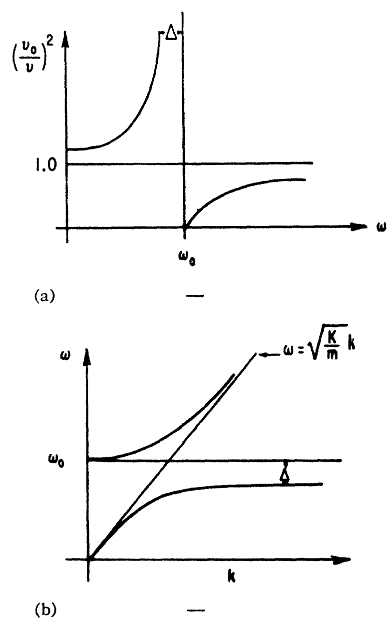

FIG. 1. (a) Sonic index of refraction (v_0/v) as a function of elastic wave frequency ω, where v_0 is the phase velocity in the absence of spins, v is the phase velocity with spins, ω_0 is the spin resonance frequency, and $\langle S_z\rangle<0$ (normal population). $\Delta^2=4\epsilon^2 g\beta H\langle S_z\rangle/K$; see text. (b) Dispersion relation between elastic wave frequency ω and wave vector $k=2\pi/\lambda$. $\Delta=[4\epsilon^2 g\beta H\langle S_z\rangle/K]^{1/2}$ is a stop band near the spin resonance frequency ω_0; see text. $\langle S_z\rangle<0$ (normal population). In the case of a ferromagnetic spin system, the exchange coupling between spins makes possible the propagation of spin waves entirely apart from the phonon field. Hence, the straight line at ω_0 is replaced by a parabolic dispersion law, discussed by Kittel.[8]

more energy is propagated in the companion spin wave with the result that the disturbance is no longer purely sonic and is propagated at a modified velocity. (Similar ideas apply to phonon-magnon dispersion in a ferromagnet.)[8] An alternative description of a dispersive medium is via the relation $\omega=f(k)$, which can be obtained from the secular equation (4), a plot of which appears in Fig. 1(b). These relations show that the group velocity $\partial\omega/\partial k$ changes radically as ω approaches the resonant frequency ω_0. For negative $\langle S_z\rangle$ (thermal equilibrium case) a wave packet slows down in the neighborhood of ω_0, as reported earlier.[6] Moreover, a stop band exists, between ω_0 and

$$\omega_0-[4\epsilon^2 g\beta H|\langle S_z\rangle|/K]^{1/2}$$

as indicated on the graph. Within the stop band, k is imaginary so that normal wave propagation is not possible. Thus, such a wave impinging on the boundary of a medium containing resonant spins would be reflected as light is reflected in the case of "frustrated internal reflection" or as x rays are reflected when the Bragg law is satisfied. As $L\rightarrow\infty$, the reflection at surface X_0 approaches 100%. This behavior is outlined in Fig. 2. Thus, insofar as we may neglect losses and nonlinear effects in the derivation of (4), we expect a slowing down and distortion of a wave packet of ultrasonic energy as it travels through a resonant medium. When the sound frequency approaches ω_0, the dispersion becomes severe, and finally, the wave is highly reflected when ω falls within the stop band Δ. k is then purely imaginary within the resonant medium. Although our resonant spin system is not periodic, it has much in common with systems which are, such as electrical delay lines and crystals. Because of the slow propagation of sound these effects are readily observed in ultrasonic pulse experiments, as reported earlier, and are manifestations of dispersion, a common property of periodic and resonant systems. In nature any resonant system will have a finite Q factor, or linewidth, which implies a finite loss for wave propagation in the resonant medium. In our model of $S=1/2$ it is convenient to treat this linewidth phenomenologically in terms of a relaxation time τ. We can do so by adding the quantities S_x/τ and S_y/τ to the left side of the Bloch equations for S_x and S_y. By so doing we interpret τ as the spin-spin or transverse relaxation time. However, even in the absence of this interaction, a finite linewidth would still exist, in which case τ would represent the effect of spontaneous emission to the phonon field. In practice we should expect τ to represent both of these level-broadening effects although in our discussion here we attribute τ entirely to spin-spin interaction since we assume a fairly concentrated spin system. The equations of motion are then

$$\begin{aligned} m\ddot{U}_n&=K(U_{n-1}+U_{n+1}-2U_n)\\ &\quad+\hbar\epsilon(S_x^{(n+1)}-S_x^{(n-1)}),\\ \dot{S}_x^{(n)}+S_x^{(n)}/\tau&=-\omega_0 S_y^{(n)},\\ \dot{S}_y^{(n)}+S_y^{(n)}/\tau&=-\epsilon(U_{n+1}-U_{n-1})S_z+\omega_0 S_x^{(n)}, \end{aligned} \tag{6}$$

which contract to the coupled wave equations (7).

$$\begin{aligned} m\ddot{U}_n&=K(U_{n-1}+U_{n+1}-2U_n)+\hbar\epsilon(S_x^{(n+1)}-S_x^{(n-1)}),\\ \ddot{S}_x^{(n)}&+2\dot{S}_x^{(n)}/\tau+(1/\tau^2+\omega_0^2)S_x^{(n)}\\ &=\epsilon\omega_0\langle S_z\rangle(U_{n+1}-U_{n-1}). \end{aligned} \tag{7}$$

Again assuming traveling wave solutions of the form $e^{i(\omega t-kna)}$ for U_n and $S_x^{(n)}$ we arrive at the more general dispersion relation (8).

$$\frac{Kk^2a^2}{m\omega^2}=\left(\frac{v_0}{v}\right)^2 =1-\frac{4\epsilon^2 g\beta H\langle S_z\rangle/K}{[\omega_0^2+1/\tau^2+4\epsilon^2 g\beta H\langle S_z\rangle/K]-\omega^2+2i\omega/\tau}. \tag{8}$$

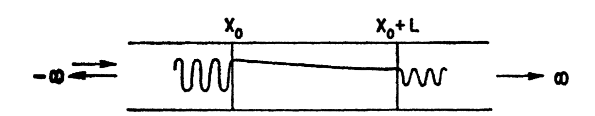

FIG. 2. Behavior of ultrasonic oscillations within a resonant medium, of infinite Q, when the wave frequency falls within the stop band. The exponential decay $e^{-\alpha x}$ within the medium for $X_0\leqslant x\leqslant X_0+L$ signifies reflection of the incident wave at X_0, which becomes total reflection as $L\rightarrow\infty$.

[8] C. Kittel, Phys. Rev. 110, 836 (1958).

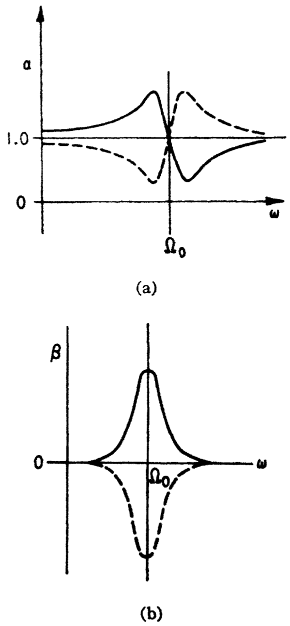

FIG. 3. (a) The real part (dispersion) and (b) the imaginary part (absorption) as a function of elastic wave frequency ω; $\langle S_z\rangle>0$ (normal population) solid line, $\langle S_z\rangle>0$ (inverted population) dashed line. See text.

For convenience, let us assume $|(4\epsilon^2 g\beta H\langle S_z\rangle\tau/2\omega K|\ll 1$ and rewrite (8) in terms of real and imaginary parts. Then

$$\left(\frac{v_0}{v}\right)=(\alpha-i\beta)=1-\frac{2\epsilon^2 g\beta H\langle S_z\rangle}{K}\times\left(\frac{\Omega_0^2-\omega^2-2i\omega/\tau}{(\Omega_0^2-\omega^2)^2+(2\omega/\tau)^2}\right), \quad (9)$$

where $\Omega_0^2\equiv(\omega_0^2+1/\tau^2+4\epsilon^2 g\beta H\langle S_z\rangle/K)$. The real and imaginary parts are plotted against ω in Figs. 3(a) and 3(b). As before when $\omega\rightarrow\Omega_0$ the sound wave experiences anomalous dispersion in the vicinity of resonance, but with now a concomitant rise in attenuation.[9] Moreover, with finite value of τ a stop band no longer exists; wave propagation at frequencies within the resonance bandwidth is still possible although the physical meaning of group velocity is not clear if the dispersion is pronounced. However, an energy velocity can always be defined. The problem of wave propagation at frequencies within the region of anomalous dispersion, particularly as it applies to the propagation of pulses, is an interesting and delicate matter which we do not take up here, but which is dealt with at some length by Brillouin.[10] We point out, however, that a pulse of sound incident on the surface X_0 in Fig. 2 will be distorted for three reasons. First, net absorption of energy may occur, leading to changes in $\langle S_z\rangle$. Secondly, the various side bands will be differentially reflected at X_0 since the mechanical impedance of the resonant medium will be frequency dependent, as seen from Fig. 3(a). Thirdly, the frequency components within the linewidth will each have different phase velocities as well as suffering varying degrees of attenuation. The relative dispersion and attenuation will depend, of course, upon the magnitudes of coupling constant ϵ and loss factor $1/\tau$.

The case of an inverted population, $\langle S_z\rangle>0$, merits further comment. First, our analysis has assumed that $\langle S_z\rangle$ is constant in space and time, and this is now unlikely to be correct except possibly for very short time intervals. Nonetheless, a literal interpretation of Eq. (9) for positive $\langle S_z\rangle$ suggests that as $\omega\rightarrow\Omega_0$ the measured velocity of pulses will increase because of the resulting inverted dispersion [Fig. 3(a)]. However, the analysis of Sommerfeld[11] and Brillouin[10] indicate that, to the contrary, it is not possible to propagate signals at velocities greater than $v_0(\omega\rightarrow\infty)$. Thus for an elastic continuum, where v_0 is independent of frequency, we would not expect to observe an increase in the pulse velocity with an inverted population. In contrast to the behavior of dispersion (in so far as the velocity of pulses is concerned), the absorption is directly related to the sign and magnitude of $\langle S_z\rangle$, as given by Eq. (8) and portrayed in Fig. 3(b). In this case the sign and magnitude of $\langle S_z\rangle$ represents the potential for amplifying or attenuating the sonic wave train. Thus an inverted spin system, $\langle S_z\rangle>0$, predicts negative absorption (amplification) by stimulated emission.

An experiment to demonstrate the amplification of sound pulses by stimulated emission from an inverted population in ruby was suggested by one us (E.H.J.) to Tucker and successfully carried out by the latter, in accordance with the behavior implied by Eq. (9).[5,12] It is to be noted that the amplification must not be too large (i.e., $\epsilon^2\langle S_z\rangle$ must not be too large) in pulse experiments if distortion caused by large dispersion is to be avoided. On the other hand, for cw narrow band amplification (Fig. 4), the product $\epsilon^2\langle S_z\rangle$ can be increased accordingly. However, some means is needed in this case to prevent reflected waves within the resonant medium so as to avoid the buildup of self-sustaining standing waves, i.e., feed back must be eliminated in order to suppress self-sustained oscillations, as with any amplifier. In contrast to amplification, the production of self-sustained oscillation, if desired, should

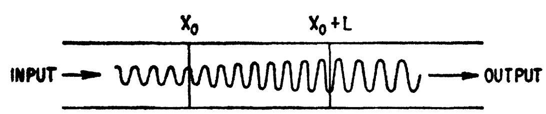

FIG. 4. Amplification of elastic wave by inverted spin system within the resonant medium $X_0\leqslant x\leqslant X_0+L$.

[9] By attenuation we mean dissipation of energy from the ultrasonic wave unless otherwise stated.

[10] L. Brillouin, *Wave Propagation and Group Velocity* (Academic Press Inc., New York, 1960).

[11] A. Sommerfeld, Ann. Physik 44, 177 (1914); we are also indebted to N. S. Shiren for discussion of this point.

[12] See also the discussion on phonon masers by C. H. Townes and N. Bloembergen, in *Quantum Electronics*, edited by C. H. Townes (Columbia University Press, New York, 1960), p. 405–9; C. Kittel, Phys. Rev. Letters 6, 449 (1961).

be easily realizable from an inverted spin system under proper conditions of reflection or feedback. Such an oscillator, at microwave sound frequencies, would involve essentially the same features encountered in the optical maser since the wavelengths are comparable. In particular, we would expect to observe a series of modes excited within the natural spin-resonance linewidth, the spacing of which depends upon the ratio of sonic wavelength of crystal length. Further development of quantum methods of sonic amplification will doubtless continue and may afford the opportunity for detailed exploration of ultrasonic phenomena at frequencies well beyond the present microwave range.

Our model of $S=1/2$ coupled to a longitudinal wave through the x-z component on the g tensor is somewhat idealized and, though illustrative in all the essential ideas of dispersion and attenuation, is not a common example of what we find in nature. A typical spin system involves a more complicated Hamiltonian containing terms arising from the action of the crystal field on the spin through the spin-orbit interaction. As it turns out, these terms are usually much more sensitive to lattice distortion than are the components of the g tensor and so provide the main coupling between sound field and spins. An example of such a system is Fe^{++} in MgO which is described by an effective $S=1$, and we now address ourselves to the mathematical treatment of this more general problem.

Semiclassical Treatment of $S=1$

We start by considering the total Hamiltonian for $S=1$, in a cubic crystalline field, with a compressional sound wave propagating along the [100] direction:

$$\mathcal{H}=\sum_n\left\{\frac{P_n^2}{2m}+\frac{K}{2}(U_n-U_{n-1})^2+g\beta\mathbf{H}\cdot\mathbf{S}^{(n)}+\frac{\mathfrak{D}\hbar}{2}[(S_x^{(n)})^2-\tfrac{2}{3}](U_{n+1}-U_{n-1})\right\}.$$

As in the previous example of $S=1/2$, we assume linear restoring forces between nearest neighbors only, and that each atom has a spin. However, in contrast with $S=1/2$ we here introduce coupling to the lattice via the $\mathfrak{D}$ term instead of through a component of the g tensor. Moreover, we may expect that for a general spin Hamiltonian a quantum-mechanical treatment will be required similar to that used in describing optical dispersion in a real atomic system, in contrast to the harmonic oscillator model. To this end we describe presently a quantum-mechanical approach which is applicable to a general spin system. We describe first, however, a method analogous to choosing normal modes in a many-body linear system obeying classical laws. This method was pointed out to one of us (E.H.J.) in some detail by M. H. L. Pryce of the University of Bristol for the case of $S=1$ and H_{dc} perpendicular to the direction of sound propagation. We have found that this method can be applied to a more complicated spin Hamiltonian with $S=1$, although extension to systems with $S>1$ is rather difficult owing to, apparently, the algebraic properties of the spin operators for such systems.

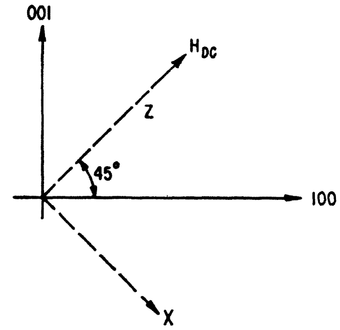

FIG. 5. Rotation of x, z axes relative to cubic axes. H_{dc} parallel to z axis. Compressional elastic wave propagating along [100], see text.

In describing this essentially "classical" method we are concerned with deriving and linearizing equations of motion for operators like U_n, S_x^2, S_y^2, $S_xS_y+S_yS_x$, etc. For convenience let us take the case pertaining to Fe^{++} in MgO with H_{dc} 45° to [100] and with atomic displacements along [100] only. The Hamiltonian is then

$$\mathcal{H}=\sum_n\left\{\frac{P_n^2}{2m}+\frac{K}{2}(U_n-U_{n-1})^2+\frac{\sqrt{2}}{2}g\beta H(S_{[001]}^{(n)}+S_{[100]}^{(n)})+\frac{\mathfrak{D}\hbar}{2}[(S_{[100]}^{(n)})^2-\tfrac{2}{3}](U_{n+1}-U_{n-1})\right\}. \quad (11)$$

The algebra is simplified if we take a new set of axes with z parallel to H_{dc}, as shown in Fig. 5, and transform the spin operators $S_{[100]}$ and $S_{[001]}$ into the new x, z frame. Since these operators transform as components of a pseudovector, we have that

$$S_{[100]}=(\sqrt{2}/2)(S_z+S_x);\quad S_{[001]}=(\sqrt{2}/2)(S_z-S_x),$$
$$S_{[100]}^2=\tfrac{1}{2}(S_z^2+S_x^2+S_xS_z+S_zS_x), \quad (12)$$

so that

$$\mathcal{H}=\sum_n\left\{\frac{P_n^2}{2m}+\frac{K}{2}(U_n-U_{n-1})^2+g\beta HS_z^{(n)}+\frac{\mathfrak{D}\hbar}{4}[(S_z^{(n)})^2+(S_x^{(n)})^2+S_x^{(n)}S_z^{(n)}+S_z^{(n)}S_x^{(n)}-\tfrac{4}{3}](U_{n+1}-U_{n-1})\right\}, \quad (13)$$

where U_n still denotes the displacement of the nth atom from its equilibrium position along the [100] direction.

As before, using the commutation rules for the time dependence of an operator, we derive a set of simultaneous equations of the motion. The equation for U_n becomes

$$m\ddot{U}_n=K(U_{n+1}+U_{n-1}-2U_n)+\sum_{r=n-1}^{r=n+1}\frac{\mathfrak{D}\hbar}{4}(S_z^2+S_x^2+S_xS_z+S_zS_x-\tfrac{4}{3})^{(r)}\times(\delta_{r-1,n}-\delta_{r+1,n}). \quad (14)$$

Equations (13) and (14) suggest that we may need equations of motion for products of operators like S_x^2, $S_xS_y+S_yS_x$, $S_xS_z+S_zS_x$, etc. Some experimentation shows that the following set of equations are sufficient and lead to the correct dispersion relation, which can be derived by an alternate method.

$$\begin{aligned}
d(S_x^{(n)})^2/dt&=-\omega_0(S_xS_y+S_yS_x)^{(n)}-\tfrac{1}{4}\mathfrak{D}(U_{n+1}-U_{n-1})S_y^{(n)},\\
d(S_y^{(n)})^2/dt&=\omega_0(S_xS_y+S_yS_x)^{(n)},\\
d(S_z^{(n)})^2/dt&=\tfrac{1}{4}\mathfrak{D}(U_{n+1}-U_{n-1})S_y^{(n)},\\
d(S_xS_y+S_yS_x)^{(n)}/dt&=2\omega_0(2S_x^2+S_z^2-2)^{(n)}-\tfrac{1}{4}\mathfrak{D}(U_{n+1}-U_{n-1})\times(S_z^{(n)}+S_x^{(n)}),\\
d(S_yS_z+S_zS_y)^{(n)}/dt&=\omega_0(S_yS_z+S_zS_y)^{(n)}-\tfrac{1}{4}\mathfrak{D}(U_{n+1}-U_{n-1})\times(S_z^{(n)}-S_x^{(n)}),\\
d(S_xS_z+S_zS_x)^{(n)}/dt&=-\omega_0(S_yS_z+S_zS_y)^{(n)}.
\end{aligned} \quad (15)$$

The derivation of these equations makes explicit use of the algebra for the spin operators associated with $S=1$. For example, products of the form $S_z^2S_y+S_yS_z^2$ and $S_x^2S_y+S_yS_x^2$ reduce simply to S_y. To carry the analysis further it is necessary at this point to linearize the above equations by dropping out all second-order terms such as $U_nS_y^{(n)}$ and by assuming that $S_z^{(n)}$ is in a definite state S_z, constant in space and time when it appears on the right-hand side of Eqs. (15). These equations taken together with that for U_n yield the following set of linear simultaneous equations:

$$\begin{aligned}
&-m\ddot{U}_n+K(U_{n+1}+U_{n-1}-2U_n)+\frac{\mathfrak{D}\hbar}{4}\sum_{r=n-1}^{n+1}\{S_x^2+\langle S_z\rangle^2+S_zS_x+S_xS_z-\tfrac{4}{3}\}^{(r)}\times(\delta_{r-1,n}-\delta_{r+1,n})=0,\\
&\frac{d^2}{dt^2}(S_x^{(n)})^2-\frac{\mathfrak{D}}{4}\omega_0\langle S_z\rangle(U_{n+1}-U_{n-1})+2\omega_0^2[2(S_x^{(n)})^2+\langle S_z\rangle^2-2]=0,\\
&\frac{d^2}{dt^2}(S_zS_x-S_xS_z)^{(n)}+\omega_0^2(S_zS_x+S_xS_z)^{(n)}+\frac{\mathfrak{D}}{4}\omega_0\langle S_z\rangle(U_{n+1}-U_{n-1})=0.
\end{aligned} \quad (16)$$

By a slight change in the quantity $\{S_x^2+S_z^2+S_zS_x+S_xS_z-\frac{4}{3}\}$ to read $\{S_x^2+\frac{1}{2}S_z^2-1+S_zS_x+S_xS_z\}$ and assuming traveling wave solutions of the form $e^{i(\omega t-kna)}$ where "na" measures the position of the nth atom along the [100] direction and "a" is the atomic spacing, we easily arrive at the following secular equation, where we have already taken the long-wavelength limit by replacing finite differences with derivatives.

$$\begin{vmatrix}(Ka^2k^2-m\omega^2)/\hbar & ik(\mathfrak{D}a/4) & ik(\mathfrak{D}a/4)\\ ik(\mathfrak{D}a/4) & (4\omega_0^2-\omega^2)/\omega_0\langle S_z\rangle & 0\\ ik(\mathfrak{D}a/4) & 0 & (\omega_0^2-\omega^2)/\omega_0\langle S_z\rangle\end{vmatrix}=0. \quad (17)$$

This secular determinant yields the following dispersion relation[13]:

$$\frac{k^2a^2K}{m\omega^2}=\left(\frac{v_0}{v}\right)^2=\left[1+\left(\frac{\mathfrak{D}}{4}\right)^2\frac{g\beta H\langle S_z\rangle}{K}\left(\frac{1}{\omega_0^2-\omega^2}+\frac{1}{4\omega_0^2-\omega^2}\right)\right]^{-1}. \quad (18)$$

A plot of Eq. (18) in Fig. 6 shows the familiar anomalous dispersion near ω_0 and $2\omega_0$ corresponding to transitions between the spin levels -1, 0, 1 in Fig. 7 for $S=1$. Equation (18) can be inverted and solved for ω vs k the plot of which appears in Fig. 8. The above equations and graphs show a variety of dispersion phenomena having to do with the interaction of sound waves with resonant systems, and similar in almost all detail to

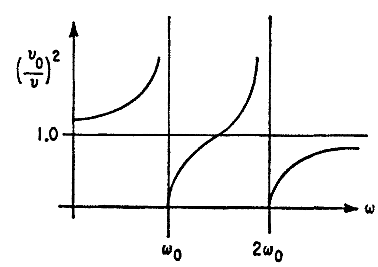

FIG. 6. Sonic index of refraction (v_0/v) vs elastic wave frequency ω for $S=1$ system. Two resonances appear, one for transitions between $S_z=0$ and $+1$ or -1, and the other between $S_z=+1$ and $S_z=-1$, for a compressional elastic wave propagating along the [100] cubic axis (see Fig. 5).

[13] The constant $(D/4)^2g\beta H\langle S_z\rangle/K$ appearing in Eq. (18) can be made to agree with the corresponding constant in Shiren's dispersion relation (see reference 6) by making the following substitutions in Eq. (13): Let $(Da\hbar/4)(\text{spins})(U_{n+1}-U_{n-1})/a=G(\text{spins})(U_{n+1}-U_{n-1})/a$ from which $D/4=G/\hbar a$.

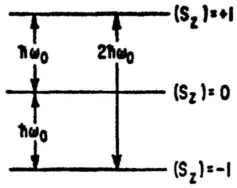

FIG. 7. Spin resonance energy levels for $S=1$ system.

that encountered in the optical spectrum of electromagnetic radiation. Because of the comparative slowness of sound and the ease of changing level populations at microwave frequencies, it is possible to study experimentally the dispersion surfaces in rather great detail. The above relations were derived with the assumption of no loss or level broadening. We know from our earlier study of $S=1/2$ system that the general behavior is not greatly modified when a loss term or relaxation time is introduced, the main effect being to eliminate the stop bands and provide either absorption or amplification for waves propagating near the resonant frequencies, instead of causing complete reflection at a boundary as depicted in Fig. 2.

Treatment of General Spin System via Contact Transformation

As remarked earlier the dispersion relation for $S=1$ was derived by a semiclassical approach which is difficult to carry out for general spin systems. Thus, for these cases we turn finally to a purely quantum mechanical method.

The scheme is to use a contact transformation to transform the original total Hamiltonian [Eq. (16)] into a new function which does not contain the spin Hamiltonian but which is time dependent, involving the resonance frequencies in the form $e^{i\omega_{12}t}$. We carry out this transformation by means of an operator $T=\exp(i\mathcal{H}_0t/\hbar)$ which is used to define new variables $\tilde{S}_i=TS_iT^*$, etc., and a new Hamiltonian

$$\mathcal{H}'=\mathcal{H}+i\hbar T^*\partial T/\partial t.$$

The new Hamiltonian $\mathcal{H}'$ is no longer the energy, but still gives the equations of motion.[14] Thus for the total Hamiltonian [Eq. (13)] for $S=1$, we are concerned with the following spin functions:

$$S_z=\exp(-i\mathcal{H}_0t/\hbar)\tilde{S}_z\exp(i\mathcal{H}_0t/\hbar)=\tilde{S}_z,$$
$$S_x=\exp(-i\mathcal{H}_0t/\hbar)\tilde{S}_x\exp(i\mathcal{H}_0t/\hbar)$$
$$=\tfrac{1}{2}\{\tilde{S}_-\exp(i\omega_0t)+\tilde{S}_+\exp-i\omega_0t\},$$

where

$$\tilde{S}_\pm=(\tilde{S}_x\pm i\tilde{S}_y),$$

and

$$\mathcal{H}=g\beta HS_z=\hbar\omega_0S_z.$$

So that $\mathcal{H}'$ becomes

$$\mathcal{H}'=\sum_n\bigg\{\frac{P_n^2}{2m}+\frac{K}{2}(U_n-U_{n-1})^2+\frac{\mathfrak{D}\hbar}{4}(U_{n+1}-U_{n-1})\{\tilde{S}_z^2+\tfrac{1}{4}[\tilde{S}_-^2e^{i2\omega_0t}+\tilde{S}_+^2e^{-i2\omega_0t}+\tilde{S}_-\tilde{S}_++\tilde{S}_+\tilde{S}_-+2(\tilde{S}_-\tilde{S}_z+\tilde{S}_z\tilde{S}_-)e^{i\omega_0t}+2(\tilde{S}_+\tilde{S}_z+\tilde{S}_z\tilde{S}_+)e^{-i\omega_0t}]-\tfrac{4}{3}\}^{(n)}\bigg\}. \quad (19)$$

For convenience we henceforth drop the tilde, and proceed to get equations of motion for the operators U_n, S_+^2, and $S_+S_z+S_{z+}$:

$$m\ddot{U}_n=K(U_{n+1}+U_{n-1}-2U_n)+\frac{\mathfrak{D}\hbar}{16}\sum_{r=n-1}^{n+1}\{4S_z^2+S_-^2e^{i2\omega_0t}+S_+^2e^{-i2\omega_0t}+2(S_-S_z+S_zS_-)e^{i\omega_0t}+2(S_+S_z+S_zS_+)e^{-i\omega_0t}+S_-S_++S_+S_-\}^{(r)}(\delta_{r-1,n}-\delta_{r+1,n}),$$

$$\frac{d}{dt}(S_\pm^{(n)})^2=\mp i\frac{\mathfrak{D}}{4}(U_{n+1}-U_{n-1})\langle S_z\rangle e^{\pm i2\omega_0t}+\text{terms in }(S_\pm^{(n)}),$$

$$\frac{d}{dt}(S_\mp S_z+S_zS_\mp)^{(n)}=\pm i\frac{\mathfrak{D}}{4}(U_{n+1}-U_{n-1})\langle S_z\rangle e^{\mp i\omega_0t}+\text{terms in }(S_\pm^{(n)}),$$

where the spin operators on the right-hand side have been replaced by constants, such as $\langle S_z\rangle$. This implies that the integration to follow is correct only for small time intervals. We integrate the last two equations by assuming $U_n\sim e^{i(\omega t-kna)}$ to yield

$$(S_\pm^{(n)})^2=\mp\frac{\mathfrak{D}}{2}\langle S_z\rangle\sin ka\frac{U_ne^{\pm i2\omega_0t}}{i(\omega\pm2\omega_0)}+\text{const},$$

$$(S_\pm S_z+S_zS_\pm)^{(n)}=\mp\frac{\mathfrak{D}}{2}\langle S_z\rangle\sin ka\frac{U_ne^{\pm i\omega_0t}}{i(\omega\pm\omega_0)}+\text{const},$$

and substitute these results in the equation for U_n

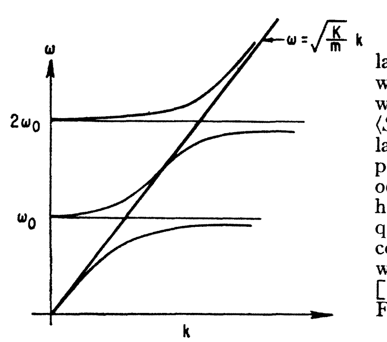

FIG. 8. Dispersion relation between elastic wave frequency ω and wave vector $k=2\pi/\lambda$. $\langle S_z\rangle<0$ (normal population). Anomalous dispersion and stop bands occur in the neighborhood of the resonant frequencies ω_0 and $2\omega_0$ for compressional elastic wave propagation along [100] cubic axis (see Fig. 5).

[14] P. A. M. Dirac, *The Principles of Quantum Mechanics* (Clarendon Press, Oxford, 1947), 3rd ed., Chap. 5, Sec. 44; W. Heitler, *The Quantum Theory of Radiation* (Oxford University Press, New York, 1957), 3rd ed.

which then gives the dispersion relations, Eq. (18), derived earlier. In the method just outlined the "terms in $(S_\pm^{(n)})$" are dropped on the assumption that they vanish at the starting time for the integration. The range of integration must be such that no appreciable change in population occur.

The above procedure can be applied to a general and more complicated example to yield a general result. Suppose

$$\mathcal{H}=\mathcal{H}(\text{lattice})+\hbar\epsilon(U_{n+1}-U_{n-1})f(S_x^{(n)},S_y^{(n)},S_z^{(n)})+\mathcal{H}_0(\text{spin}),$$

and that the eigenfrequencies $\hbar\omega_{ij}=E_i-E_j$ of $\mathcal{H}_0$ (spin) are known. If now the contact transformation is applied to $\mathcal{H}$ such that $\mathcal{H}'=\mathcal{H}+i\hbar T^*\partial T/\partial t$, the new Hamiltonian function $\mathcal{H}'$ will take the form

$$\mathcal{H}'=\mathcal{H}(\text{lattice})+\sum_n \hbar\epsilon(U_{n+1}-U_{n-1})\{a_{12}^{(n)}e^{i\omega_{12}t}+a_{12}^{*(n)}e^{-i\omega_{12}t}+a_{13}^{(n)}e^{i\omega_{13}t}+a_{13}^{*}e^{-i\omega_{13}t}+a_{23}^{(n)}e^{i\omega_{23}t}+a_{23}^{*(n)}e^{-i\omega_{23}t}+\cdots+\text{products of } a^{(n)}\text{'s}\}, \quad (20)$$

where the $a^{(n)}$'s are functions of the spin operators $S_x^{(n)}$, $S_y^{(n)}$, $S_z^{(n)}$. If we specify that all spins are initially in a definite state "l", we deduce a general dispersion law of the form

$$\left(\frac{v_0}{v}\right)^2=\left[1+\sum_{ij}\frac{A_{ij}}{\omega_{ij}-\omega^2}\right]^{-1}, \quad (21)$$

where $A_{ij}\equiv\langle l|a_{ij}^{(n)},a_{ij}^{*(n)}|l\rangle$ and where the operation $(a_{ij}^{(n)},a_{ij}^{*(n)})$ yields a quantity independent of position such as, for example, $\langle S_z\rangle$ in the case of $S=1/2$. The foregoing discussion has omitted level-broadening effects which could be included in the Hamiltonian or added phenomenologically to the equations of motion as was done in the example for $S=1/2$; in either case the result would produce a more general dispersion law of the form

$$\left(\frac{v_0}{v}\right)^2=\left[1+\sum_{ij}\frac{A_{ij}}{\omega_{ij}^2+(1/\tau_{ij})^2-\omega^2-(2i\omega/\tau_{ij})}\right]^{-1}, \quad (22)$$

where τ_{ij} is a relaxation time associated with the $i\rightarrow j$ transition, and the dynamical behavior would follow closely that already encountered in the $S=1/2$ system. The foregoing ideas apply equally well to a three-dimensional sound field interacting with a general, but not too dilute, spin system, in which case we can expect a variety of sonic phenomena such as Raman scattering, parametric effects, and rotary polarization in addition to dispersion, loss, and amplification.

III. DISCUSSION

In our analysis we have, essentially, assumed that at some definite time, t_0, the spins are in known states and that a lattice wave is being propagated. We have then solved the equations on the assumption that the spins do not change their states significantly. We find that near the resonance frequencies of the spins it is not realistic to think in terms of purely lattice oscillations, for each such disturbance is accompanied by a wave motion in the transverse components of the spin moments. The coupling of the lattice and spin disturbances, which in the uncoupled system would have different velocities, results in their being a change in the apparent velocity of sound. There is an associated attenuation, by which we mean that the amplitude of the disturbance decays with distance due to irreversible degradation of energy. With an infinite spin-spin relaxation time there is no energy dissipation within the resonant medium. With a finite spin-spin relaxation time energy dissipation occurs, and one may ask where this energy goes to. Unfortunately, it has not proved feasible to treat the dipolar interaction between the spins completely, and we have been forced to use the phenomenological description given by τ, the spin-spin relaxation time. There is, however, good experimental evidence to support this phenomenological description. If, then, it is accepted that the introduction of the concept of spin-spin relaxation is a valid one, the energy dissipation occurs because to propagate a wave near resonance a wave motion must be set up in the transverse spin moments. Energy is required to do this. Furthermore, the spin-spin relaxation is constantly trying to destroy such coherences and we must suppose that a continual supply of energy is necessary to maintain the spin wave. It is possible for the destruction of spin coherence to take place by mutual spin flips, so that $\sum S_z$ does not change. That is, no work is done on or by the external field, H. However, this does not mean that the energy of the spin is unaltered, for the dipolar energy may be changed. Thus the energy loss we are considering is a process whereby energy in a sound wave near resonance is transferred to the mutual dipolar energies of the spins, where it is effectively randomized in the sense that if a quantum $\hbar\omega_0$ is taken from a lattice oscillator it is broken down into many smaller units and given to the dipolar interactions. Quite what happens to it then is not entirely clear, but it seems plausible that as every lattice mode is slightly coupled to the spins the energy may be fed back to all the lattice modes through various higher order interactions to establish a lattice temperature. The above process must clearly be distinguished from direct absorption of sound, resulting in there being changes in $\sum_n S_z^{(n)}$. Both processes may occur simultaneously, and it would be necessary to distinguish them in any typical pulse echo experiment.

Finally, a remark seems appropriate on the oft-alluded-to phonon bottleneck. In the theory of the direct process in spin-lattice relaxation the spins exchange energy with lattice modes "on speaking terms," that is, with lattice modes having frequencies close to

ω_0. Orbach[15] has used thermodynamic arguments to show that heating of phonon modes near ω_0 via the direct process and under typical conditions will probably be small. We suggest that the dissipative effect of spin-spin interaction will diminish still further the heating of these modes and so correspondingly reduce the likelihood of a phonon bottleneck.

IV. CONCLUSIONS

We have developed a theory of ultrasonic dispersion resulting from the interaction between transverse spin moments and elastic strain fields oscillating at microwave frequencies. Neglecting nonlinear effects, the theory predicts a reduction in the group velocity and and increase or decrease in the absorption of elastic waves near the spin resonance frequency for normal and inverted spin populations, respectively. The effect of damping is treated phenomenologically in terms of the spin-spin interaction time τ. The change in elastic wave propagation near the spin resonance frequency is known as anomalous dispersion and is the manifestation of two coupled wave fields, one an elastic wave, the other a spin wave in the transverse components of spin, which propagate with the same phase velocity. Finally, we suggest that the dipolar spin-spin interaction may act in a dissipative manner to extract energy irreversibly from the phonon modes near the spin resonance frequency and thereby reduce the likelihood of a phonon bottleneck under typical conditions of magnetic resonance.

[15] R. Orbach, Proc. Roy. Soc. (London) **A264**, 481 (1961).

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