We have investigated the elementary excitation spectrum of a system of interacting spins and phonons in a solid. A dispersion law for spin-lattice waves has been obtained, and possible methods of studying it experimentally are discussed.

The development of methods of generating and detecting ultrasonic waves at microwave frequencies has made possible the direct investigation of the interaction of lattice oscillations with the spin system. The experimental studies carried out have been concerned, essentially, with two effects: 1) the influence of ultrasound on paramagnetic resonance [1], and 2) the influence of paramagnetic ions on the propagation of ultrasonic waves [2]. The semiclassical approximation was used in [3] to make a theoretical study of the latter effect in a linear crystal. In the present work we carry out a quantum-mechanical analysis of this problem for a three-dimensional medium. This problem can hardly be solved in its most general formulation. However, the solution is considerably simplified if we consider a system at sufficiently low temperatures, when the excited states are close to the ground state. Furthermore, the effects mentioned above are usually observed at low temperatures, since otherwise the spectral lines of the acoustic and electron resonance are greatly broadened. Thus our problem is to find the spectrum of elementary excitations of a system of interacting spins and phonons.

The condition of weak thermal excitation of the system permits us to solve the problem by the method of approximate second quantization (ASQ). The Hamiltonian of the system has the form

\[ \mathcal{H} = \mathcal{H}_s + \mathcal{H}_f + \mathcal{H}_{sf}, \]

where \( \mathcal{H}_s \) is the Hamiltonian of the spin system, \( \mathcal{H}_f \) is the phonon field, and \( \mathcal{H}_{sf} \) is the spin-phonon interaction, respectively; for the case \( S > 1/2 \) and a crystal with cubic symmetry [4] we can write \( \mathcal{H}_{sf} \) as

\[ \mathcal{H}_{sf} = \sum_{k, \sigma} \hbar \omega_k \left( b_k^{\dagger} b_k + \frac{1}{2} \right) \sum_{j} \left( \frac{G_{ij} - G_{ij}}{2} \times \left[ S^z_j - \frac{S(S + 1)}{3} \right] (2e_{xx} - e_{yy}) + \right. \]

\[ + (S^x_j - S^y_j)(e_{xx} - e_{yy}) + 2G_{ij}(S^z_j S^z_j + S^x_j S^y_j + S^y_j S^x_j) e_{xx} + \]

\[ + (S^x_j S^y_j + S^y_j S^x_j) e_{xy} + (S^z_j S^z_j + S^y_j S^x_j) e_{xy} \right] \]  

where \( \omega_k \) is the Larmor frequency; \( S^z_j \) is the z-component of the j-th spin vector operator; \( \omega_k \) is the phonon frequency; \( b_k^{\dagger} \) and \( b_k \) are the creation and annihilation operators of a phonon with wave vector \( k \) and polarization \( \sigma \); the \( G \) constants are the coefficients of the spin-phonon interaction; and \( e_{xx}, e_{xy} \) are the components of the deformation tensor. The magnetic field \( H \parallel z \).

In agreement with the ASQ procedure, we use the Holstein-Primakoff representation (see [5], for example)

\[ S^+ = (2S)^{1/2} \varphi(n) a, \]

\[ S^- = (2S)^{1/2} a^+ \varphi(n), \]

where

\[ S^\pm = S^x \pm i S^y, \quad \varphi(n) = \left( 1 - \frac{n}{2S} \right)^{1/2}, \quad n = a^+ a, \]

and \( a, a^+ \) are Bose operators.

To find the elementary excitation spectrum of the system, it is sufficient to consider only those terms in the Hamiltonian which are quadratic in the Bose operators. Expanding the components of the deformation tensor in the normal modes, and transforming to the second-quantized representation, we obtain:

\[ \mathcal{H} = \mathcal{H}_s + \mathcal{H}_f + \mathcal{H}_o = -N \hbar \omega_0 + \frac{1}{2} \sum_{k, \sigma} \hbar \omega_k + \]

\[ + \frac{1}{V N} \sum_{k, \sigma, j} \left( b_{k\sigma} a_j - b_{k\sigma}^* a_j^* \right) \left( b_{k\sigma} e^{i \mathbf{r}_j} - b_{k\sigma}^* e^{-i \mathbf{r}_j} \right), \]

where

\[ \mathbf{r}_j = \mathbf{k} + \mathbf{V}_j + l \mathbf{k}, \quad \mathbf{V}_j = \mathbf{V} + l \mathbf{k}, \]

\( N \) is the number of particles, \( m \) is the mass of one particle, \( k \) is the wave vector, and \( V \) is the polarization vector of the phonon. It is obvious that our problem consists of the diagonalization of Eq. (5). Performing the transformation
we obtain
\[
\mathcal{H}_1 = \sum_k \hbar \omega_k a_k^+ a_k + \sum_{k,\sigma} B_{-\sigma \sigma} b_{-\sigma}^+ b_{\sigma} + \sum_{k,\sigma} B_{\sigma \sigma} a_{\sigma} b_{\sigma}^+ - \sum_{k,\sigma} B_{\sigma \sigma} a_{\sigma}^+ b_{\sigma} + \sum_k \hbar \omega_k b_{\sigma}^+ b_{\sigma}.
\] (7)

This quadratic form is diagonalized by the canonical transformation \[6\]
\[
a_{\sigma} = \sum \left[ \tilde{\xi}_1 (k) u_{\rho} (k) + \tilde{\xi}_2 (k) \varphi_{\rho}^* (-k) \right],
\]
\[
b_{\sigma} = \sum \left[ \tilde{\xi}_1 (k) u_{\rho} (k) + \tilde{\xi}_2 (k) \varphi_{\rho}^* (-k) \right].
\] (8)

Equation (8) and the equations of motion for the operators \(a_k\) and \(b_k\) give us a system of homogeneous equations to be solved to determine the coefficients of the transformation:

\[
[\hbar \omega_0 - E_1 (k) ] \varphi_{\rho} (-k) + B_1 [\varphi_{\rho} (k) - \varphi_{\rho} (-k) ] = 0,
\]
\[
[\hbar \omega_0 - E_1 (k) ] u_{\rho} (k) - B_1 [u_{\rho} (k) - u_{\rho} (-k) ] = 0,
\]
\[
[\hbar \omega_0 + E_1 (k) ] \varphi_{\rho} (k) + B_1 [u_{\rho} (k) + u_{\rho} (-k) ] = 0,
\]
\[
[\hbar \omega_0 + E_1 (k) ] u_{\rho} (k) + B_1 [\varphi_{\rho} (k) + \varphi_{\rho} (-k) ] = 0.
\] (9)

We neglected the interactions between lattice oscillations of different polarization, which is possible if the frequency branches of the lattice oscillations are not degenerate, and have used

\[
B_1 = B_{-1}, \quad \varphi_{\rho} = \varphi_{-\rho}.
\]

The solvability condition of Eq. (9) gives the dispersion equation

\[
E_1^2 (k) - \hbar^2 (w_0^2 + w_0^2) E_1^2 (k) + \hbar^4 (w_0^2 + w_0^2 - \frac{4}{B_1^2} |B_0| w_0 w_0) = 0.
\] (10)

Noting that the energy of an elementary excitation is positive, and setting \(E = \hbar \omega_0\), we obtain

\[
\pm \sqrt{\left( w_0^2 - w_0^2 \right)^2 + \frac{16}{B_1^2} |B_0|^2 w_0 w_0}.
\] (11)

The dispersion curves are pictured in Fig. 1. In the long-wave region, branch \(\Omega_1 (k)\) describes spin oscillations with the same frequency as free oscillations, while branch \(\Omega_2 (k)\) in this region corresponds to acoustic waves with an altered velocity of sound. In the shortwave region \(\Omega_1\) corresponds to acoustic waves with the original velocity, and \(\Omega_2\) to spin oscillations with an altered frequency. The meaning of the solution we have obtained is that, in the presence of the spin-phonon interaction, mixed oscillations appear which, generally speaking, are neither spin-type nor lattice-type. The greatest "mixing" occurs, naturally, in the region of the resonance frequencies. This circumstance is characteristic of paramagnetic crystals, since at paramagnetic centers the lower group of energy levels always lies in the zone of the quasi-continuous spectrum of lattice oscillations. It is obvious that \(\mathcal{H}_1\) in the \(\xi, \xi^\dagger\) operators will have the following form:

\[
\mathcal{H}_1 = \Delta E + \sum_{k,\sigma} E_1 (k) \xi_\sigma^* (k) \xi_\sigma (k),
\] (12)

where

\[
\Delta E = - \sum_{k,\sigma} E_1 (k) |a_{\sigma} (k)|^2.
\]

From the normalization condition and Eq. (9) we find:

\[
|\varphi_{\rho} |^2 = \frac{|B_0|^2 \hbar \omega_0 (\hbar \omega_0 - E_1 (k))^2}{E_1 (k) \left( (\hbar \omega_0^2 - E_1^2 (k))^2 + 4 |B_0|^2 \hbar^2 \omega_0 \omega_0 \right)}
\]
\[
|\varphi_{\rho} |^2 = \frac{|B_0|^2 \hbar \omega_0 (\hbar \omega_0 + E_1 (k))^2}{E_1 (k) \left( (\hbar \omega_0^2 - E_1^2 (k))^2 + 4 |B_0|^2 \hbar^2 \omega_0 \omega_0 \right)}
\]
\[
|u_{\rho} |^2 = \frac{\left( E_1 (k) + \hbar \omega_0 \right) \left( \hbar \omega_0^2 - E_1^2 (k) \right)^2}{4 \hbar \omega_0 E_1 (k) \left( (\hbar \omega_0^2 - E_1^2 (k))^2 + 4 |B_0|^2 \hbar^2 \omega_0 \omega_0 \right)}
\]
\[
|u_{\rho} |^2 = \frac{\left( E_1 (k) - \hbar \omega_0 \right) \left( \hbar \omega_0^2 - E_1^2 (k) \right)^2}{4 \hbar \omega_0 E_1 (k) \left( (\hbar \omega_0^2 - E_1^2 (k))^2 + 4 |B_0|^2 \hbar^2 \omega_0 \omega_0 \right)}
\] (13)

Equations (4), (11), (12), and (13) now give the solution of the problem.

The explicit expressions obtained above for the coefficients of transformation (8) permit us to determine the contribution from lattice and spin oscillations in each of the branches \(\Omega_1 (k)\) and \(\Omega_2 (k)\). These coefficients are also needed in the study of the various elementary-excitation scattering processes. It would be of interest to test experimentally the results obtained here. In particular, the spatial dispersion of spin oscillations due to the interaction with phonons can be studied by means of neutron scattering from a paramagnetic crystal. It would be even more interesting to extend the experiments on the propagation of sound in paramagnetic crystals \[2\]. In high-quality samples, we might expect to be able to observe splitting of the mechanical resonance line, if the sound frequency is close to the frequency of the spin oscillations. This effect could also be observed indirectly, in the study of the Rayleigh scattering of light (for more details on this, see \[6\]).

REFERENCES