Thermal magnetoacoustic resonance

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A thermal magnetoacoustic resonance is predicted and observed in chromium sulfide, $Cr_5 S_6$. This is a resonant dependence of the propagation velocity and attenuation of ultrasound on the temperature in helicoidal magnetic materials. It arises at a resonant temperature T_0 , at which the spin-wave velocity, which depends on the temperature, becomes equal to the velocity of an elastic wave. This resonance is unrelated to the frequency magnetoacoustic resonance, which does not occur, since at ultrasonic frequencies ω both elastic and spin waves in a helicoidal magnetic material have a linear dispersion law. The temperature T_0 is thus independent of ω .

1. A periodic helicoidal magnetic structure has substantial effects on the elastic properties of magnetic materials. One mechanism for these effects is quasistatic. Specifically, the appearance of a helicoidal magnetic structure is accompanied by a change in the nature of the quasistatic elastic tensor. The components of this tensor become periodic functions of the coordinates. The number of independent components also increases.^{1,2}

The first of these circumstances gives rise to two regions (bands) of forbidden frequencies in the elastic-wave spectrum and to a region of a repulsion of spectral branches. The second circumstance has the consequence that the polarization of the elastic waves which are propagating along the helicoidal axis and one of the principal crystallographic axes are summed. The polarization of each elastic wave has a longitudinal component, a right-handed transverse component, and a left-handed transverse component simultaneously. These effects are seen in turn in the appearance of structural features when ultrasound reflects from either a semi-infinite magnetic material³ or a plate⁴ and also in the transmission of ultrasound through a plate.⁴ For example, when linearly polarized transverse ultrasound is incident, the reflected ultrasound and the transmitted ultrasound have an elliptical polarization. The major axis of the ellipse of this polarization is rotated with respect to the polarization plane of the incident ultrasound. Longitudinal ultrasound may also arise. Some of these effects are of a resonant nature near the forbidden frequency bands, where the wave vectors of the helicoid, q, and of the ultrasound, k, are comparable.

Another mechanism is a dynamic interaction between the elastic and magnetic subsystems, namely, elastic and spin waves which arise in helicoidal magnetic materials. This interaction may lead to a distinctive thermal magnetoacoustic resonance. The nature of this resonance can be summarized as follows: In the ultrasonic frequency range, with $k \leqslant q$, the dispersion relations $\omega(k)$ for both spin waves^{5,6} and elastic waves are linear, and a frequency resonance is not possible. The velocity of a spin wave, however, depends on q, which in turn may depend strongly on the temperature. At a certain resonant temperature T_0 (at any frequency) at which the propagation velocities of the spin and elastic waves become equal, a thermal magnetoacoustic resonance is thus possible.

2. Let us take a look at the theory of the thermal magnetoacoustic resonance. As an example we consider magnetoelastic waves in crystals of hexagonal symmetry in which there is a helicoidal magnetic structure.⁶ For our purposes it is sufficient to write the free-energy density in the form

$$F = \frac{1}{2} \hat{\beta} M_{z}^{2} + \frac{1}{2} \alpha_{ij} \frac{\partial \mathbf{M}}{\partial x_{i}} \frac{\partial \mathbf{M}}{\partial x_{j}} + \frac{1}{2} \gamma_{ij} \frac{\partial^{2} \mathbf{M}}{\partial x_{i}^{2}} \frac{\partial^{2} \mathbf{M}}{\partial x_{j}^{2}} + b_{ijkl} M_{i} M_{j} u_{kl} + \lambda_{ijkl} \frac{\partial \mathbf{M}}{\partial x_{i}} \frac{\partial \mathbf{M}}{\partial x_{j}} u_{kl} + \frac{1}{2} c_{ijkl}^{M} u_{ij} u_{kl}, u_{ij} = \frac{1}{2} \left(\frac{\partial u_{i}}{\partial x_{j}} + \frac{\partial u_{j}}{\partial x_{i}} \right),$$
(1)

where **M** is the magnetization, u_{ij} is the strain tensor, $\beta = 2K_1/M^2$, and K_1 is the constant of the crystallographic magnetic anisotropy. The tensors α_{ij} and γ_{ij} characterize the nonuniform exchange energies; the tensors b_{ijkl} and λ_{ijkl} describe the uniform and nonuniform magnetostriction, respectively; and the elastic tensors c_{ijkl}^M describe the elastic energy in the case in which the magnetic state of the magnetic material remains unchanged. For brevity we will omit the superscript *M* below.

Under the conditions $\alpha_{11} = \alpha_{22} > 0$, $\alpha_{33} < 0$, $\gamma_{33} > 0$, and $\beta > 0$, a helical magnetic structure can arise in a magnetic material. This structure or helicoid is described by

$$\boldsymbol{M}_{0}^{\pm} = \boldsymbol{M}_{0} \exp(\pm i q z), \quad \boldsymbol{M}^{\pm} = \boldsymbol{M}_{x} \pm i \boldsymbol{M}_{y}. \tag{2}$$

The equilibrium value of the wave vector q,

$$q^2 = -\alpha_{ss}/2\gamma_{ss} \tag{3}$$

is found by minimizing the free energy $\Phi = (1/V) \int F dv$.

To analyze the natural magnetoelastic waves which are superposed on the basic state, we must jointly solve the equations of motion of the theory of elasticity and of magnetization. As the latter we use the equation of motion in Hilbert form. We take the damping in the elastic subsystem into account by assuming that the elastic moduli c_{ijkl} are complex: $c_{ijkl} = c'_{ijkl} - ic''_{ijkl}$.

The magnetization and the strain accompanying the magnetoelastic waves as they propagate along the helicoid axis are written in the form

$$\mathbf{M} = \mathbf{M}_{0} + \mathbf{m} \exp\left[-i(\omega t - kz)\right], \quad u_{ij} = u_{ij}^{0} + u_{ij} \exp\left[-i(\omega t - kz)\right], \quad (4)$$

where ω and k are respectively the frequency and wave vec-

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tor of the magnetoelastic waves.

Substituting (4) into the equations of motion of elastic theory and of magnetization, and linearizing these equations near the equilibrium position, we find a coupled system of equations. As in Ref. 6, we consider the interaction of the spin waves with only the longitudinal elastic wave (we ignore the interaction of the spin waves with the transverse elastic waves). The former interaction is due to the nonuniform exchange magnetostriction (λ_{ijkl}), and the latter to the uniform magnetostriction (b_{ijkl}), which is of a magnetic nature.

The dispersion relation for the system is

$$\left(\omega^{2}-\frac{c_{3333}}{\rho}k^{2}\right)\left[\omega^{2}-(\omega_{1}-i\alpha_{1}\omega)(\omega_{2}-i\alpha_{1}\omega)\right]$$
$$-\frac{4gq^{2}k^{4}M_{\rho}^{2}}{\rho}\lambda_{3333}^{2}(\omega_{2}-i\alpha_{1}\omega)=0,$$
(5)

where

$$\omega_{1} = g M_{0} k^{2} \gamma_{33} (k^{2} + 4q^{2}), \quad \omega_{2} = g M_{0} \gamma_{33} \left[(q^{2} - k^{2})^{2} + \frac{\beta}{\gamma_{33}} \right],$$
(6)

 ρ is the density, g is the gyromagnetic ratio, and α_1 is a dimensionless constant, which is a measure of the attenuation in the equation of motion for the magnetization in Hilbert form. Equation (5) can be rewritten as

$$\left(\frac{\omega^{2}}{(s_{l}^{0})^{2}}-k^{2}\right)\left(\frac{\omega^{2}}{(s_{sp}^{0})^{2}}-k^{2}\right)=\xi k^{4},$$
(7)

where ξ is the magnetoelastic coupling coefficient, given by

$$\xi = \frac{M_0^2 \lambda_{3333}^2}{c_{3333} \gamma_{33}} \frac{4q^2}{k^2 + 4q^2} \,. \tag{8}$$

Here s_l^0 and s_{sp}^0 are the phase velocities of the elastic and spin waves, respectively, in the absence of a magnetoelastic interaction ($\xi = 0$), and

$$(s_{i}^{0})^{2} = (s_{i}^{0})_{0}^{2} (1 - i\alpha_{i}),$$

$$(s_{sp}^{0})^{2} = (s_{sp}^{0})_{0}^{2} \frac{1 - i\alpha_{1}\omega/\omega_{2}}{1 + \alpha_{1}^{2} + i\alpha_{1}\omega_{2}/\omega},$$
(9)

are the velocities of elastic and spin waves, respectively, when the interaction of these waves and the attenuation are ignored. These quantities are given by

$$(s_{l}^{0})_{0}^{2} = c_{3333}^{\prime}/\rho, \quad \alpha_{l} = c_{3333}^{\prime\prime}/c_{3333}^{\prime}, \quad (s_{sp}^{0})_{0}^{2} = \omega_{1}\omega_{2}/k^{2}.$$

(10)

It follows from (7) that if the phase velocities of the elastic and spin waves, s_l^0 and s_{sp}^0 , are nearly the same (or if the corresponding wavelengths are equal), a magnetoelastic resonance may arise.

In general, the dispersion relation for a spin wave, $\omega(k)$, is nonlinear,^{5,6} according to (10) and (6), in contrast with that for an elastic wave. There could thus be a frequency magnetoelastic resonance at frequencies such that the dispersion branches, i.e., the curves of ω versus k for the spin and elastic waves (if the interaction between these waves is ignored), intersect. However, the wave vector of the helicoid, **q**, is usually much larger than the wave vector of the ultrasound, $\mathbf{k}: |\mathbf{q}| \ge |\mathbf{k}|$. Accordingly, at frequencies in the ultrasonic range, both $(s_{sp}^0)_0$ and ξ become independent of k according to (10), (6), and (8). Specifically, we find

$$(s_{\bullet p}^{\bullet})_{\bullet}^{2} = (2gM_{\bullet})^{2} \gamma_{\bullet \bullet}^{2} q^{2} (q^{\bullet} + \beta/\gamma_{\bullet \bullet}), \quad \xi = M_{\bullet}^{\bullet} \lambda_{3333}^{2} / C_{3333} \gamma_{33},$$
(11)

in other words, the dispersion relations for both the elastic and spin waves become linear. The dispersion branches do not intersect when the frequency changes, so in this case a frequency magnetoelastic resonance should not arise. The wave vector of the helicoid, however, depends on the temperature T. As T is varied, a distinctive thermal magnetoelastic (or magnetoacoustic) resonance can occur at a certain temperature T_0 , at which the velocity of a spin wave, which depends on q, becomes equal to the velocity of an elastic wave. Let us examine this resonance.

If s_{sp}^0 and ξ are independent of k, the solutions of Eq. (7) for $s_{1,2} = \omega/k_{1,2}$ are

$$\frac{1}{s_{1,2}} = \frac{1}{2(1-\xi)^{\frac{1}{2}}} \left\{ \left[\left(\frac{1}{s_{l}^{0}} + \frac{1}{s_{sp}^{0}} \right)^{2} - \frac{2(-(1-\xi)^{\frac{1}{2}})}{s_{l}^{0}s_{sp}^{0}} \right]^{\frac{1}{2}} \\ \pm \left[\left(\frac{1}{s_{l}^{0}} - \frac{1}{s_{sp}^{0}} \right)^{2} + \frac{2(1-(1-\xi)^{\frac{1}{2}})}{s_{l}^{0}s_{sp}^{0}} \right]^{\frac{1}{2}} \right\}.$$
(12)

These solutions describe two coupled magnetoelastic waves. Two cases are possible here, depending on the value of the parameter A, which is given by

$$A = \frac{2[1 - (1 - \xi)^{\frac{1}{2}}]s_{l}^{0}s_{sp}^{0}}{(s_{l}^{0} - s_{sp}^{0})^{2}}.$$
 (13)

For A > 0 we are dealing with the case of strong coupling. As T is varied, and we move out of the region with $(s_{sp}^0)_0 > (s_l^0)_0$ into the region with $(s_{sp}^0)_0 < (s_l^0)_0$, the magnetoelastic wave described by the root s_1 converts from an elastic-like wave (by which we mean a wave which carries primarily elastic energy) into a spin-like wave (one which carries primarily exchange energy). The inverse conversion occurs for the wave described by the root s_2 . The dispersion branches do not intersect in this case.

In the case of extremely strong coupling, in which we can ignore the wave attenuation $(\alpha_1 = 0, \alpha_l = 0)$, we should replace the complex quantities s_l^0 and s_{sp}^0 in (10) by the real quantities $(s_l^0)_0$ and $(s_{sp}^0)_0$. It follows from (12) that the dispersion branches $\omega(k)$ and therefore the frequency-independent velocities s_1 and s_2 move close together with increasing T (if q decreases in the process), as the resonant temperature T_0 is approached from the side $T < T_0$. At $T = T_0$, the distance between the branches is at a minimum and is given by

$$s_1^{-1} - s_2^{-1} = \left[2 \left(1 - (1 - \xi)^{\frac{1}{2}} \right) / (1 - \xi) \left(s_1^{\circ} \right)_0^2 \right]^{\frac{1}{2}},$$

$$(s_{\bullet p}^{\circ})_{\bullet} = (s_1^{\circ})_0.$$

Then for $T > T_0$ the branches move away from each other. This difference does not depend on ω .

In the weak-coupling case, with $A \leq 1$, we can take square roots approximately. In this case the roots s_1 and s_2 determine the velocities of an elastic-like wave, s_1 , and a spin-like wave, s_{sp} , respectively:

$$\frac{1}{s_{l}} = \frac{1}{s_{l}^{0}(1-\xi)^{\prime l_{0}}} \left[1 - \frac{(s_{l}^{0})^{2}(1-(1-\xi)^{\prime l_{0}})}{(s_{l}^{0})^{2} - (s_{sp}^{0})^{2}} \right],$$

$$\frac{1}{s_{sp}} = \frac{1}{s_{sp}^{0}(1-\xi)^{\prime l_{0}}} \left[1 + \frac{(s_{sp}^{0})^{2}(1-(1-\xi)^{\prime l_{0}})}{(s_{l}^{0})^{2} - (s_{sp}^{0})^{2}} \right].$$
(14)

Using (9), (10), and (14), we find the following relations for $s_l = s'_l - is''_l$ and $s_{sp} = s'_{sp} - is''_{sp}$ under the conditions $\alpha_l \ll 1, \alpha_1 \ll 1, \xi \ll 1$, and $\omega/\omega_2 \ll 1$:

$$\frac{s_{l}'}{(s_{l}^{0})_{0}} = 1 + \frac{\xi}{2} \frac{x^{2}(1-x^{2})}{(1-x^{2})^{2}+\alpha^{2}},$$

$$\frac{s_{l}''}{(s_{l}^{0})_{0}} = \frac{\alpha_{l}}{2} + \frac{\xi}{2} \frac{\alpha x^{2}}{(1-x^{2})^{2}+\alpha^{2}},$$

$$s_{sp}' = (s_{sp}^{0})_{0} \left[1 - \frac{\xi}{2} \frac{1-x^{2}+(\alpha^{*})^{2}}{(1-x^{2})^{2}+\alpha^{2}} \right],$$
(15)

$$s_{sp}'' = (s_{sp}^{0})'' - \frac{\xi}{2} \frac{\alpha x^2}{(1-x^2)^2 + \alpha^2} (s_{sp}^{0})_0, \qquad (16)$$

where

$$x = \frac{(s_{sp}^{0})_{0}\delta}{(s_{l}^{0})_{0}}, \quad \delta = \frac{1}{(1+\alpha_{sp}\alpha_{l})},$$
$$\alpha_{sp} = \frac{\alpha_{1}\omega_{2}}{\omega}, \quad \alpha^{*} = \alpha_{sp} - \alpha_{l}, \ \alpha = \alpha^{*}\delta.$$
(17)

The parameter α_{sp} is a measure of the attenuation of the spin wave in the approximation $\omega/\omega_2 \ll 1$, and α_1 serves a corresponding role for an elastic wave. The ω dependence of α_{sp} is determined by (17) in the case of the Hilbert equation. Its ω dependence, however, can also be found from experimental data on the attenuation of spin waves. Expressions (15) and (16) hold in the case $A \ll 1$, in which we have

$$A = 2\xi/(\alpha^*)^2.$$
(18)

From these equations we can distinguish the following basic features of the thermal magnetoacoustic resonance.

a) A necessary condition for the occurrence of this resonance is that the difference between the phase velocities for the propagation of spin and elastic waves vanish at some resonant temperature T_0 .

b) The condition for the occurrence of a resonance and the value of the resonant temperature T_0 are independent of ω in a first approximation.

c) The conditions under which the strong-coupling and weak-coupling cases prevail are determined by the ratio of the magnetoelastic coupling coefficient ξ to the square of the constant $\alpha^* = \alpha_{sp} - \alpha_l$, which is a measure of the attenuation of the spin waves (α_{sp}) and of the elastic waves (α_l) . These conditions depend on ω , because of the strong ω dependence of α_{sp} .

d) The values of the resonant extrema of the real and imaginary (quality factor) terms of the propagation velocity and also the widths of the resonant extrema, $s_i^{"}$ and $s_{sp}^{"}$, depend on ω in the case in which the weak-coupling condition holds, because of the ω dependence of the constant α . Since we have $\alpha = (\alpha_{sp} - \alpha_l)\delta$, at $T = T_0$ and under the condition $\alpha_{sp} > \alpha_l$ we should see a maximum of $s_l^{"}$, while under the condition $\alpha_{sp} < \alpha_l$ we should see a minimum. We should find

the opposite picture for s_{sp}'' .

e) In cases in which the attenuation of the spin wave is very strong (and the spin wave does not propagate), the existence of the spin wave nevertheless affects the propagation velocity and attenuation of the elastic wave.

From this discussion we find the conditions which must be satisfied by magnetic materials with a helicoidal structure if a thermal magnetoacoustic resonance is to be observed in them.

First, the condition $s_{sp} > s_l$ must hold in a certain temperature interval. This condition can be satisfied in magnetic materials which have large values of q and of the constant γ_{33} , which is determined by exchange forces, in this temperature interval.

Second, q must depend strongly on T. This condition would apparently hold in magnetic materials in which a magnetostructural transition from ferromagnetism to a helicoidal magnetism is observed at a certain temperature T_1 . At T_1 , the constant α_{33} changes sign, so q vanishes according to (3).

It turns out that these conditions hold for chromium sulfide, Cr_5S_6 .

3. Chromium sulfide, $\operatorname{Cr}_5 S_6$ (space group \mathscr{D}_{3d}^2), has two magnetic phase transitions, at $T_1 \simeq 165$ and $T_c \simeq 305$ K. As the temperature is lowered, the following sequence of magnetic structures is observed in this material: paramagnetism (at $T > T_c$), a collinear ferrimagnetic structure $(T_1 < T < T_c)$, and an unusual helicoidal structure $(T < T_1)$.⁷

For a study of the elastic properties, we used polycrystalline chromium sulfide, prepared by the procedure described in Ref. 8. An x-ray diffraction analysis and measurements of the temperature dependence of the magnetization revealed that the alloy which resulted corresponded to the $Cr_5 S_6$ phase.

For the acoustic measurements we used a resonance method involving a composite longitudinal-vibration piezoelectric vibrator working at a frequency of about 87 kHz. The measurement procedure and equations for determining the velocity of ultrasonic waves and the internal friction in the material are given in Ref. 9. A quartz piezoelectric vibrator used to excite the longitudinal vibrations had dimensions of $3 \times 3 \times 28$ mm; the test sample had dimensions of $3 \times 3 \times 14$ mm. The measurements were carried out during heating and cooling. The sample temperature changed by less than 12 deg/h.

Figure 1 shows the temperature dependence of the velocity of longitudinal ultrasound (more precisely, of an elastic-like wave) in Cr_5S_6 . Near the Curie temperature $(T_c \simeq 305 \text{ K})$, we see only a small anomaly on the $s_l(T)$ curve. The curves of $s_l(T)$ measured during the heating and cooling cycles coincide, near T_1 showing no thermal hysteresis even in the transition region.

Figure 2 shows the temperature dependence of the internal friction $Q^{-1} = 2s_i''/s_i'$. We see that there are no substantial structural features on the curve of $Q^{-1}(T)$ at T_c , while there is a rounded maximum at T_1 .

4. Let us discuss the results. In the experiments we measured the temperature dependence of the phase velocity and quality factor. In the theoretical work, these properties have been calculated as functions of the ratio of the velocities of



FIG. 1. Temperature dependence of the velocity of longitudinal ultrasound in chromium sulfide, Cr_5S_6 .

elastic and spin waves, rather than as functions of T. It was thus necessary to convert the experimental results on $s_l(T)$ and $Q^{-1}(T)$ into results on s_l and Q^{-1} as a function of $(s_{sp}^0)_0$. This conversion requires knowledge of the temperature dependence of $(s_{sp}^0)_0$.

A slight complication in efforts to compare a theory derived for the case of a simple ferromagnetic helicoid with experimental data on $Cr_5 S_6$ is that the distribution of magnetic moments of chromium ions in the latter material is more complex, as is the magnetic structure. For this reason, our comparison of the experimental and theoretical behavior is only qualitative, and our estimates of the constants α_{33} , γ_{33} , λ_{3333} , and α_{sp} (about which absolutely nothing yet is known) are merely order-of-magnitude estimates.

We introduce an effective magnetic order, and we consider a helicoid which is formed by the resultant magnetization of the magnetic moments of a magnetic cell. Judging from the neutron-diffraction measurements of Ref. 7, the resultant magnetization of $Cr_5 S_6$ is the sum of two terms, $M = M_1 + M_2$.

The first term arises from ions of types a and c. The magnetic moments of the c ions are parallel to each other. Those of the a ions are oriented antiparallel to the magnetic moments of the c ions. However, the moments do not cancel



FIG. 2. Temperature dependence of the internal friction in Cr_5S_6 .

out completely, since certain sites which would ordinarily have c ions are vacant. The number of vacancies is equal to the number of remaining c ions. For this reason, there is a resultant magnetization \mathbf{M}_1 . This term is nonzero over the entire temperature range $T < T_c$. For $T > T_1$ the magnetization \mathbf{M}_1 is equal to the spontaneous magnetization \mathbf{M}_s found experimentally. In the region $T < T_1$ the temperature dependence of \mathbf{M}_1 can be found by extrapolating the $\mathbf{M}_s(T)$ curve recorded for $T > T_1$ into the region $T < T_1$, under the assumption of a $T^{3/2}$ law.¹⁰

The origin of the term \mathbf{M}_2 is as follows. Magnetic moments of b ions are oriented parallel to each other, while for $T \ge T_1$ the magnetic moments of the ions of type f are oriented antiparallel to those of b ions. For $T < T_1$ the magnetic moments of the f ions begin to rotate with respect to the magnetic moments of the b ions. The angle φ through which they rotate, varies with the temperature, from $\varphi = 180^\circ$ at $T = T_1$ to $\varphi = 129^\circ$ at T = 4.2 K. Judging from the experimental data of Ref. 7, the dependence of $\cos\varphi$ on T can be described by

$$\cos\varphi=0,37\left(\frac{q}{q_0}\right)^2-1,$$

where q_0 is the value of **q** for T = 0.

The resultant-magnetization vector $\mathbf{M} = \mathbf{M}_1 + \mathbf{M}_2$ forms a helicoid in space with a wave vector **q** for $T < T_1$. The axis of this helicoid is oriented along the hexagonal axis. The q(T) dependence is as shown in Fig. 3, according to Ref. 7. We can find some information about the relationship between M and the anisotropy constant K—these quantities determine the T dependence of $(s_{sp}^0)_0$ and ω_2 , according to (11) and (6)—from data in the literature.¹¹ It turns out that $\beta = 2K / M_0^2 = 21 \cdot 10^2$ is essentially independent of T. The value of γ_{33} at $T = T_0$ can be found by requiring $(s_{sp}^0)_0 = (s_1^0)_0$; the result is $\gamma_{33} = 1.2 \cdot 10^{-23}$ cm⁴. We use the approximation that γ_{33} is, like β , independent of the temperature. We can then plot $(s_{sp}^0)_0$ versus T. Knowing the dependence of $(s_{sp}^0)_0$ on T, we reconstructed s_1' and Q^{-1} as functions of $(s_{sp}^0)_0$ (see the experimental points in Figs. 4 and 5). These curves clearly have the resonant shape characteristic



FIG. 3. Temperature dependence of the wave function of the helicoid in Cr_5S_6 , according to Ref. 7.

of the case in which the weak-coupling condition holds. Further evidence that the weak-coupling condition holds comes from the fact that the measurements by the composite-vibrator method yielded only a single resonance peak, which corresponded to a geometric resonance of the elasticlike wave. Under the strong-coupling condition, in contrast, we should have observed two peaks, corresponding to geometric resonances of two natural magnetoelastic waves.⁹

We can draw conclusions about the T dependence of $(s_{sp}^0)_0$, which we are not showing here, by comparing the scales along the abscissa axis for $(s_{sp}^0)_0$ and T in Figs. 4 and 5.

We can draw a basic qualitative conclusion from the results shown in Figs. 1, 2, 4, and 5: The thermal magnetoacoustic resonance arises at a resonant temperature $T_0 = 132$ K in Cr₅S₆ at $T < T_1$.

That the relation $(s_{sp}^0)_0 > (s_l^0)_0$ holds at $T < T_0$ can be confirmed by an independent estimate based on α_{33} , γ_{33} , and q, along with relations (3) and (11). For an estimate of $|\alpha_{33}|$ in the limit $T \rightarrow 0$, we use the method proposed by Lifshitz¹² for the case of ferromagnets. Specifically, we equate the energy of the antiparallel spins to the thermal energy at the Curie point, T_c . We find

$$\frac{|\alpha_{33}|}{2} = \frac{k_B T_c}{dM_0^2},$$
(19)

where d is the distance between the magnetoactive atoms, and k is the Boltzmann constant. In $\operatorname{Cr}_5 S_6$ we have d = c/4, $c = 11.5 \cdot 10^{-8}$ cm, and $M_0 = 38.3$ G. Using $q(0) = 2 \cdot 10^6$ cm⁻¹, and working from (3) and (11), we find α_{33} $= 2 \cdot 10^{-9}$ cm², $\gamma_{33}(0) = 25 \cdot 10^{-23}$ cm⁴, and $(s_{sp}^0)_0 = 30 \cdot 10^5$ cm/s. Although this is a very crude estimate, it does tell us, at the very least, that the condition $(s_{sp}^0)_0 > (s_I^0)_0$ holds as $T \rightarrow 0$.

By comparing the experimental and theoretical curves, we can estimate the values of the parameters ξ and α , which determine the effect. In general, these parameters may depend on T and thus on $(s_{sp}^0)_0$. Estimates show that we have $\omega_2 \sim 10^{12} \text{ s}^{-1}$ and that the condition $\omega_2/\omega \gg 1$ holds for the frequency of the ultrasound, $\omega \sim 5 \cdot 10^5 \text{ s}^{-1}$. We plotted a theoretical curve of s_i versus $(s_{sp}^0)_0$ (the solid curve in Fig. 4), ignoring in a first approximation the temperature depen-



FIG. 4. Velocity of longitudinal ultrasound versus the spin-wave velocity. Solid line-Theoretical; points-experimental.



FIG. 5. Internal friction as a function of the spin-wave velocity.

dence of λ_{3333} , α_1 , and α_l . The best agreement between the experimental data (the points in Fig. 4) and the theoretical curve was found with the values $\xi = 3.2 \cdot 10^{-2}$, $\alpha_1 = 4.8 \cdot 10^{-7}$, and $\alpha = 1$. The value of α_l was found from the attenuation of the ultrasound away from the resonant region: $\alpha_l = 7 \cdot 10^{-3}$.

Knowing ξ , we can use (11) to determine the component λ_{3333} of the tensor λ_{ijkl} (which describes the nonuniform exchange magnetostriction) in order of magnitude:

$$\lambda_{3333} = (c'_{3333} \gamma_{33} \xi)^{\frac{1}{2}} / M_0.$$
⁽²⁰⁾

We find $\lambda_{3333} = 4 \cdot 10^{-8} \text{ cm}^2$.

The value which we found for the quantity α_1 , which is a measure of the attenuation in the phenomenological equation of motion for the magnetization, turns out to be considerably smaller than the values which are seen experimentally for α_1 for ferromagnets. We might point out in this connection that there is a corresponding situation in the case of antiferromagnets.¹³ The constant which determines the attenuation of the spin wave, $\alpha_{sp} = \alpha_1 \omega_2 / \omega \sim 1$, turns out to be very large, despite the value $\delta = 1$. At this value of α_{sp} , a spin wave should be attenuated over a distance no longer than the wavelength of the wave. However, despite the strong attenuation in the spin subsystem, the latter may have a strong effect on the behavior of the elastic subsystem under conditions of thermal magnetoacoustic resonance.

The estimate of α_{sp} from the experimental results on $s_e(T)$ is the least reliable estimate. The reason is that the theory was derived for a single crystal, while the experiments were carried out on a polycrystalline sample. We know that in (for example) polycrystalline ferromagnets in which the grains can be assumed to be independent the width of the ferromagnetic-resonance line is determined, in the case of weak attenuation, not by the attenuation but by the spread of the resonance fields for the variously oriented crystallites.¹⁴ This width turns out to be on the order of the crystallographic-anisotropy field. In an analysis of the thermal magnetoacoustic resonance in a polycrystalline sample, we should thus allow for the anisotropy of the propagation velocity of the spin wave and of the coupling coefficient.

With the z axis oriented along the hexagonal axis for each crystallite, we find

$$\omega_{1} = gM_{0}\{(k^{2}-k_{z}^{2}) [\alpha_{11}+\gamma_{11}(k^{2}-k_{z}^{2})] + k_{z}^{2}\gamma_{33}(k_{z}^{2}+4q_{z}^{2})\},\$$

$$\omega_{2} = gM_{0}\{(k^{2}-k_{z}^{2}) [\alpha_{11}+\gamma_{11}(k^{2}-k_{z}^{2})] + (k_{z}^{2}-q_{z}^{2})^{2}\gamma_{33}+\beta\}.$$
(21)

Under the condition $k \leq q$, we have, according to (21) and (10),

$$(s_{sp}^{0})_{0}^{\perp}/(s_{sp}^{0})_{0}^{\parallel}=^{1}/_{2}(\alpha_{11}/\alpha_{33})^{\frac{1}{2}},$$

where $(s_{sp}^0)_0^{\perp}$ and $(s_{sp}^0)_0^{\parallel}$ are the velocities of spin waves propagating respectively perpendicular to and parallel to the helicoid axis.

The coupling of a longitudinal elastic wave with a spin wave, which is determined by the component λ_{3333} and the derivative $\partial M^0_{\tau}/\partial z$, is at a maximum when the elastic wave is propagating along the helicoid axis. In contrast, when a longitudinal elastic wave is propagating perpendicular to the helicoid axis, the coupling with a spin wave is zero, since it is determined by the component λ_{1111} , and we have

$\partial M_x^{\circ}/\partial x = \partial M_y^{\circ}/\partial y = \partial M_z^{\circ}/\partial z = 0.$

A slightly unexpected result was that the magnitude of the increment $(\Delta Q^{-1}) = Q^{-1} - Q_{l}^{-1}$ at resonance found experimentally is an order of magnitude smaller than the maximum value $2s_l''/s_l' - \alpha_l$. Here Q_l^{-1} is the internal friction away from the resonance, which is due to the elastic subsystem.

In summary, it follows from this comparison of experimental data and the theoretical results that a thermal magnetoacoustic resonance is observed in $Cr_5 S_6$. We have found order-of-magnitude estimates of the thermodynamic parameters α_{33} , γ_{33} , and λ_{3333} , about which absolutely nothing was known previously. Our estimates of the relaxation constants α and α_{sp} are problematical.

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