Quadratic Field Dependence of Ultrasonic Attenuation Coefficient in Paramagnetic Holmium*

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The ultrasonic attenuation coefficient α_k has been measured in the paramagnetic phase of Ho in fields from 0 to 50 kG. α_k increased as the square of the field and decreased as the temperature increased. A model has been developed which ascribes this increase in α_k to the interaction of the longitudinal phonons, via modulation of the exchange interaction, with the spin fluctuations associated with the virtual phase transition at the paramagnetic Curie temperature. This theory also agrees with the velocity data obtained by Moran and Lüthi.

We wish to report experimental results which show that the ultrasonic attenuation coefficient in the paramagnetic state of Ho increases as the square of the applied magnetic field. We shall propose a mechanism for this increase in attenuation which shows that it is due to the interaction of the phonons with fluctuations associated with the virtual ferromagnetic phase transition. In fact, the applied field appears to extract the zero-wave-number Fourier component of the fluctuations. Thus, this mechanism presents a new method for determining the anisotropy and temperature dependence of Γ_k, the decay time of the spin-correlation function in the paramagnetic state.

Longitudinal waves in the frequency range 15–225 MHz were propagated along the c axis of a Ho single crystal which was approximately cylindrically shaped, 0.80 cm long and 0.58 cm in diameter. A 15-MHz fundamental X-cut quartz transducer was bonded with epoxy to the end face of the Ho cylinder. The transducer was used as both transmitter and receiver. A Matec ultrasonic rig was used for both production and detection of the ultrasonic echoes. Temperature was controlled with a Sommers[1]-type regulator and measured with a Cu-Constantan thermocouple. Magnetic fields of up to 60 kG were obtained in a Nb-alloy superconducting magnet. In Fig. 1(a) we have plotted the attenuation coefficient for the longitudinal waves as a function of the square of the external magnetic field. We have also drawn straight lines from the origin through the experimental points. A quadratic dependence appears to fit all the data. The attenuation coefficient was set equal to zero at $H=0$. These data were obtained at 148°K, which is about 16°K above the Néel temperature of Ho.

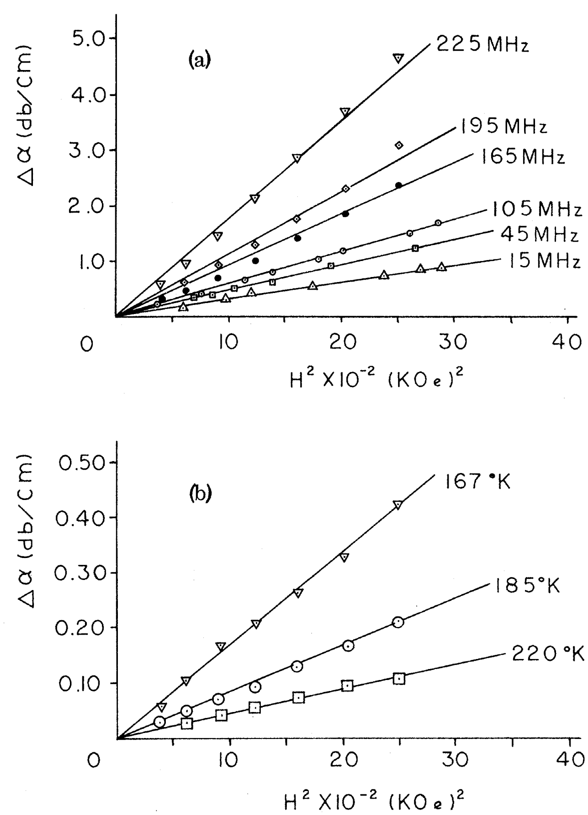

FIG. 1. Attenuation as a function of H^2 for (a) various frequencies at $T=148$°K, (b) various temperatures at $f=15$ MHz.

The attenuation coefficient was also measured as a function of magnetic field at three higher temperatures, 167, 184, and 219°K, at 15 MHz. These data are shown in Fig. 1(b). Again, α_k is seen to depend on H.[2] Thus, the important feature of our data is the increase of α_k as the square of the magnetic field, and the decrease in $\alpha_k(H)$ as the temperature is increased.

The spin-phonon interaction responsible for the attenuation of longitudinal sound in the rare-earth metals is assumed to arise from the strain modulation of the exchange interaction. Via this interaction, the spin thermal fluctuations cause random forces to act on the acoustic normal modes. According to Tani and Mori,[3] the sound attenuation coefficient α_k is expressed as the time-correlation function of these random forces, and is proportional to the four-spin correlation function, which is approximated by a product of the two-spin correlation functions at high temperatures. The two-spin correlation function is divided into the thermal-fluctuation part and the static spin-polarization part induced by an external magnetic field. The field dependence of α_k originates from the cross product of these two terms.

By introducing the Fourier amplitude of spin, $\vec{S}_{\vec{q}} = (1/\sqrt{N})\sum_i \vec{S}_i \exp(-i\vec{q}\cdot\vec{R}_i)$, Tani and Mori's formulas may be written as

$$\alpha_k = (NMV_l k^2)^{-1}\left[\sum_{\vec{q}}\sum_{\vec{q}'} g_{\vec{q}}^* g_{\vec{q}'} \int_0^\infty (\vec{S}_{\vec{q}}(t)\cdot\vec{S}_{-\vec{q}-\vec{k}}(t), \vec{S}_{-\vec{q}'}(0)\cdot\vec{S}_{\vec{q}'+\vec{k}}(0)) \exp(-i\omega_k t)dt\right], \tag{1}$$

$$g_{\vec{q}} = \sum_j \exp(i\vec{q}\cdot\vec{R}_{ji})[\exp(i\vec{k}\cdot\vec{R}_{ji}) - 1](k^{-1}\vec{k}\cdot\partial J_{ij}/\partial\vec{R}_i), \tag{2}$$

where $\vec{k}$ is the wave number of the longitudinal sound and ω_k its angular frequency, N the total number of spins, M the mass of the unit cell, V_l the longitudinal sound speed, J_{ij} the exchange integral between spins at positions i and j, and $\vec{R}_{ij} = \vec{R}_i - \vec{R}_j$ the separation between positions i and j. In (1) the relaxation function is defined by $(A, B) = \int_0^\beta d\lambda \langle e^{\lambda H} A e^{-\lambda H} B\rangle - \beta\langle A\rangle\langle B\rangle$, where $\langle C\rangle = \mathrm{Tr}(e^{-\beta H}C)/\mathrm{Tr}e^{-\beta H}$. For rare-earth metal ions the total angular momentum is the good quantum number. Therefore, $\vec{S}$ in this paper stands for the total angular momentum. The total angular momentum of a Ho ion is very large ($S = 8$) and, thus, behaves like classical angular momentum. Also, the data were obtained at temperatures that were rather high with respect to the transition temperature. Therefore, to a good approximation $(\vec{S}_{\vec{q}}(t)\cdot\vec{S}_{-\vec{q}-\vec{k}}(t), \vec{S}_{-\vec{q}'}(0)\cdot\vec{S}_{\vec{q}'+\vec{k}}(0))$ in Eq. (1) is replaced by $\beta\langle\vec{S}_{\vec{q}}(t)\cdot\vec{S}_{-\vec{q}-\vec{k}}(t), \vec{S}_{-\vec{q}'}(0) \cdot S_{\vec{q}'+\vec{k}}(0)\rangle$. Now since the temperature is high, the four-spin correlation function may be decoupled, and the random-phase approximation may be used. Once this is done Eq. (1) is reduced to

$$\alpha_k = (2NMV_l k^2 k_B T)^{-1}\,\mathrm{Re}\sum_{\vec{q}} (g_{\vec{q}}^* g_{\vec{q}} + g_{\vec{q}}^* g_{-\vec{q}-\vec{k}}) \sum_\nu \int_0^\infty \langle S_{\vec{q}}^\nu(t) S_{-\vec{q}}^\nu(0)\rangle \times\langle S_{-\vec{q}-\vec{k}}^\nu(t) S_{\vec{q}+\vec{k}}^\nu(0)\rangle \exp(-i\omega_k t)\,dt. \tag{3}$$

Now we use Landau's fluctuation theory[4] to calculate the correlation function $\langle S_{\vec{q}}^\nu(t) S_{-\vec{q}}^\nu(0)\rangle$ in an external magnetic field H. When H is applied along the μ axis, the Zeeman energy $\sqrt{N} g\mu_B S_0^\nu H$ appears in the free energy. To eliminate this term, which is linear in the spin operator, we introduce the following transformation:

$$\sigma_{\vec{q}}^\nu = S_{\vec{q}}^\nu + \frac{\sqrt{N} g\mu_B S(S+1)H}{3k_B(T - T_{\vec{q}}^\nu)}\delta_{q0}\delta_{\nu\mu}, \tag{4}$$

where $T_{\vec{q}}^\nu$ is the ordering temperature for the ν component of spins varying sinusoidally with wave number q. Using this transformation, Landau's free energy in the paramagnetic state may be written as

$$F = -\frac{CH^2}{2(T - T_0^\nu)} + \frac{3}{2}\frac{k_B}{S(S+1)}\sum_{\vec{q}}\sum_\nu (T - T_{\vec{q}}^\nu)\sigma_{-\vec{q}}^\nu \sigma_{\vec{q}}^\nu, \tag{5}$$

C being the Curie constant, $N(g\mu_B)^2 S(S+1)/3k_B$. The first term in Eq. (5) indicates the free energy of the spin system at the thermal-equilibrium point, and the second term expresses the change in the free energy associated with the thermal fluctuations of σ_q^ν. When $\vec{q}$ is small, we assume the hydrodynamic form $\langle\sigma_{\vec{q}}^\nu(t)\sigma_{-\vec{q}}^\nu(0)\rangle = \langle\sigma_{\vec{q}}^\nu\sigma_{-\vec{q}}^\nu\rangle\exp(-\Gamma_{\vec{q}}^\nu t)$. The static correlation function $\langle\sigma_{\vec{q}}^\nu\sigma_{-\vec{q}}^\nu\rangle$ is calculated by using the free energy (5):

$$\langle\sigma_{\vec{q}}^\nu\sigma_{-\vec{q}}^\nu\rangle = \tfrac{1}{3}S(S+1)T/(T - T_{\vec{q}}^\nu). \tag{6}$$

Equation (6) can also be obtained from the well-known result

$$\chi_\nu(q) = \frac{N(g\mu_B)^2}{k_B T}\langle \sigma_{\vec{q}}^{\nu}\sigma_{-\vec{q}}^{\nu}\rangle = \frac{N(g\mu_B)^2 S(S+1)}{3k_B(T-T_{\vec{q}}^{\nu})}.$$

With the use of Eqs. (4) and (6), the spin-correlation function for small $\vec{q}$ is obtained as

$$\langle S_{\vec{q}}^{\nu}(t)S_{-\vec{q}}^{\nu}(0)\rangle = \frac{S(S+1)}{3}\frac{T}{T-T_{\vec{q}}^{\nu}}\exp(-\Gamma_{\vec{q}}^{\nu}t) + \frac{N(g\mu_B)^2S^2(S+1)^2H^2}{9k_B^2(T-T_0^{\nu})^2}\delta_{\vec{q}0}\delta_{\nu\mu}. \quad (7)$$

If we insert (7) into (3), the field-dependent part of α_k for waves propagating along the c axis, with Z being the z component of R, is obtained as

$$\Delta\alpha_k(H) = \frac{2}{27}\frac{(g\mu_B)^2S^3(S+1)^3}{k_B^3MV_l}k^2\left(\sum_i Z_{ij}\frac{\partial J_{ij}}{\partial Z_i}\right)^2\frac{H^2}{(T-T_0^{\mu})^2(T-T_k^{\mu})}\frac{\Gamma_k^{\mu}}{(\Gamma_k^{\mu})^2+\omega_k^2}. \quad (8)$$

Equation (8) shows that the attenuation *increases* as the square of the magnetic field, which is in agreement with the experimental results. Using inelastic scattering of neutrons, Als-Nielsen *et al.*[5] have obtained the decay time of the spin correlation function in paramagnetic Tb, $\Gamma_Q^{\perp}$, where the Q indicates the spiral wave number of the ordered structure and $\perp$ denotes the component perpendicular to the c axis. According to their results, the dynamical slowing down of $\Gamma_Q^{\perp}$ occurs in an extremely narrow temperature region on the order of $(T-T_N)/T_N < 0.02$. Above $(T-T_N)/T_N \sim 0.1$, $\Gamma_Q^{\perp}$ is almost constant independent of temperature. The value of this constant is about 31 GHz in Tb, which is much larger than the acoustic frequencies in our experiments. Krueger and Huber[6] have developed a self-consistent theory for $\Gamma_{\vec{q}}^{\nu}$ and explained the experimental results mentioned above. According to their theory, the component parallel to the C axis, $\Gamma_q^{\parallel}$, vanishes as $q \to 0$ (kinematic slowing down). However, the leading term of $\Gamma_q^{\perp}$ for small q is constant independent of q. In this context the spiral wave number Q is small and the impressed phonon wave number k is certainly small.

Since Tb and Ho are neighboring rare-earth elements in the periodic table and both metals have a helical spin structure below T_N, we may assume that $\Gamma_{\vec{q}}^{\nu}$ for Ho has a character similar to that for Tb. Our data were obtained at rather high temperatures $(T-T_N)/T_N > 0.12$. Therefore, we assume that the value of $\Gamma_k^{\perp}$ for Ho in our measured temperature range is constant independent of temperature. Then, the temperature-dependent part of $\Delta\alpha_k(H)$ may be written as $(T-T_0^{\perp})^{-3}$, if we consider that $T_k^{\perp}$ is nearly equal to $T_0^{\perp}$, since $k \sim 0$. It should be noted that $T_0^{\perp}$ is not the ordering temperature of the helical state T_N, but the paramagnetic Curie temperature θ. The experimental values obtained for the slopes of the straight lines passing through the 15-MHz data in Figs. 1(a) and 1(b) are plotted as a function of $T-T_0^{\perp}$ in Fig. 2, where the experimental value[7] of θ_a (= 88°K) was used for $T_0^{\perp}$. As seen in this figure, the experimental values are almost on a straight line whose gradient is -2.9, which is in reasonable agreement with the theoretical value -3.

Because of the factor $(T-\theta)^{-3}$ in $\Delta\alpha_k(H)$, the largest field effect is expected in ferromagnets, where θ is positive and nearly equal to T_c. Although Ho is an antiferromagnet which has a helical spin structure below 132°K, the θ is positive. This accounts for the large value of $\Delta\alpha_k(H)$ which we observe experimentally in Ho. In a pure antiferromagnet such as MnF_2, which has a negative θ, $\Delta\alpha_k(H)$ may be small.

As seen from (8), $\Delta\alpha_k(H)$ is proportional to ω_k^2, when $\Gamma_k \gg \omega_k$. The experimental results show the ω_k^2 dependence for frequencies higher than 135 MHz at 148°K and at 50 kOe. However, below

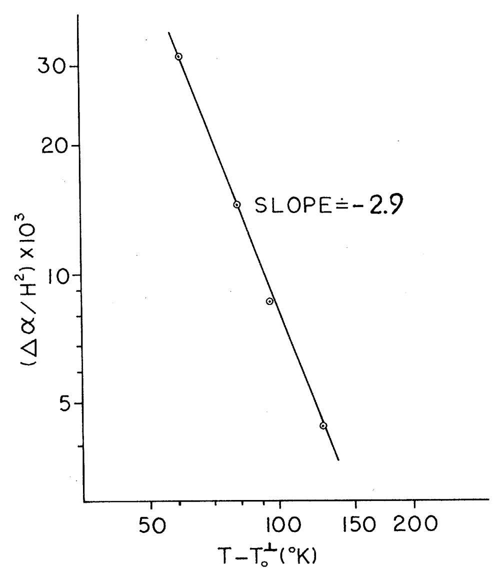

FIG. 2. Slopes of the straight lines passing through the 15-MHz data in Figs. 1(a) and 1(b) as a function of $T-T_0^{\perp}$. $T_0^{\perp}=\theta_a=88$°K.

these frequencies, $\Delta\alpha_k(H)$ depends more weakly on frequency ($\sim\omega_k^{1/2}$).

In the present model, the field dependence of the sound velocity may be obtained in the following way. When a strain is applied to a crystal, the field-dependent part in the free energy [the first term in Eq. (5)] has a change through the change of T_0^μ due to the strain. This causes the change in the elastic constant, and thus a change in the sound velocity, which depends on the strength of the magnetic field. Using this procedure, the change in the velocity for the longitudinal sound wave propagating along the c axis is calculated as

$$\frac{\Delta V_l}{V_l} = -\frac{1}{9}\frac{N(g\mu_B)^2S^2(S+1)^2}{k_B{}^2C_{33}}\left[\frac{2S(S+1)}{3k_B}\left(\sum_i Z_{ij}\frac{\partial J_{ij}}{\partial Z_i}\right)^2\frac{H^2}{(T-T_0{}^\mu)^3} - \frac{1}{2}\left(\sum_i Z_{ij}{}^2\frac{\partial^2 J_{ij}}{\partial Z_i{}^2}\right)\frac{H^2}{(T-T_0{}^\mu)^2}\right], \tag{9}$$

where C_{33} is an elastic constant. This result can also be obtained by using the formulas for the sound velocity given by Kawasaki and Ikushima[8] [Eqs. (4.12) and (4.13) in their paper] and Eq. (7). The velocity change (9) is proportional to H^2, which is in agreement with the experimental results obtained by Moran and Lüthi.[9] As seen from (9), the temperature dependence of ΔV_l is expressed in terms of $(T-T_0{}^\mu)^{-3}$ and $(T-T_0{}^\mu)^{-2}$. Analysis of the experimental results of Moran and Lüthi[9] shows that the decrease of the velocity is proportional to $(T-\theta_a)^{-2.7}$ for Dy and $(T-\theta_a)^{-2.6}$ for Ho, where $\theta_a = 169°$K and $\theta_a = 88°$K were used for Dy and Ho, respectively.

If the second term in the bracket of (9) is assumed to be negligible in the rare-earth metals and in the temperature range we are concerned with, Eqs. (8) and (9) yield the relation[10]

$$\Gamma_k{}^\mu = -\frac{\omega_k{}^2}{V_l}\frac{\Delta V_l(H)/V_l}{\Delta\alpha_k(H)} \quad (\Gamma_k \gg \omega_k). \tag{10}$$

$\Gamma_k{}^\mu$ may be estimated using the following values: $\Delta V_l/V_l = -1.67\times10^{-2}$, $V = 2.95\times10^5$ cm/sec, $\omega_k = 2\pi\times225\times10^6$ Hz, and $\Delta\alpha(H) = 4.4$ dB/cm, which are obtained by extrapolating Moran and Lüthi's data on $\Delta V_l/V$ to $H = 50$ kOe at $T = 147°$K and employing $\Delta\alpha(H)$ in the present paper. A value of 35 GHz is obtained which is comparable to the value for Tb mentioned above. The anisotropy of $\Gamma_k{}^\mu$ may be measured changing the direction of the applied magnetic field.

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