ULTRASONIC ATTENUATION IN THE HELICAL SPIN STATE OF THE RARE-EARTH METALS*

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Theory and experiment are presented for the ultrasonic attenuation of the rare-earth metals. In the helical spin state of the metals, the sound energy transferred to the spin system is dissipated to the lattice by the spin rotation around the c axis, arising from the spin-lattice relaxation mechanism, which may explain the large increases of the attenuation observed on Dy and Ho single crystals below their Néel temperatures.

It is the purpose of this Letter to propose a mechanism for ultrasonic attenuation which is characteristic of the helical spin structure and to show ultrasonic attenuation experimental results obtained by Dy and Ho. It is well known that, between their two transition temperatures, the heavy rare-earth elements such as Tb, Dy, and Ho have a helical spin structure propagating in the c direction.[1] This helical spin structure results from the long-range nature of the indirect exchange through the conduction electrons and the s-f interaction.[2] If the x and y axes are taken perpendicular to the c axis (the z axis) and the helical spin arrangement is expressed as $S_{ix} = S\cos(\vec{q}\cdot\vec{r}_i)$ and $S_{iy} = S\sin(\vec{q}\cdot\vec{r}_i)$, the exchange energy may be written as

$$E_{\text{ex}} = NS^2 J(\vec{q}), \quad J(\vec{q}) = \sum_j J_{i,j}\cos(\vec{q}\cdot\vec{r}_i), \tag{1}$$

where $J_{i,j}$ is the indirect exchange integral between spins i and j, and N is the total number of spins. The stable helical spin structure is determined in such a way that $J(\vec{q})$ is minimized with respect to the wave vector $\vec{q}$.

Let us consider a portion of the sample, whose length l is longer than the exchange-interaction range but shorter than the ultrasonic wavelength. Then the strain induced by the wave is considered to be uniform in this portion, and if the longitudinal wave propagating along the c axis is expressed as $u = u_0\sin(kz-\omega t)$, the strain becomes

$$\epsilon_{zz} = ku_0\cos\omega t. \tag{2}$$

If a static stress is abruptly applied to the crystal, the conduction band is distorted and the conduction electrons assume a new stable configuration in the conduction band which has been altered by the strain. The relaxation time for this rearrangement is usually 10^{-11}-10^{-14} sec and is shorter than the inverse of the usual ultrasonic frequency. Therefore, during passage of the wave, the stable configuration of the conduction electrons follows the strain (2) adiabatically. The Fourier component of the exchange integral corresponding to the electron configuration under strain $J(q, \epsilon_{zz})$ is expanded as a power series of ϵ_{zz} and Δq, where Δq is the deviation of q from its equilibrium value in the absence of strain. The term linear in Δq vanishes because of the stability requirement for the helical spin structure. The term linear in strain gives the static strain below the Néel temperature (the exchange striction) and may be eliminated if strains are measured on the lattice that has already been distorted by the exchange striction. Thus, the exchange energy may be written, neglecting a constant term and terms higher than second order with respect to ϵ_{zz} and Δq, as

$$E_{\text{ex}} = NS^2\left[\frac{1}{2}\frac{\partial^2 J}{\partial q^2}(\Delta q)^2 + \frac{\partial^2 J}{\partial q\,\partial\epsilon_{zz}}(\Delta q)\epsilon_{zz} + \frac{1}{2}\frac{\partial^2 J}{\partial\epsilon_{zz}^2}\epsilon_{zz}^2\right]. \tag{3}$$

If q is fixed, the exchange energy (3) is always increased by the strain since $\partial^2 J/\partial\epsilon_{zz}^2$ is positive. Thus in order to decrease the exchange energy, q is changed so that the sum of the energy from the first and second terms in the bracket of (3) is minimized. The spin rotation around the c axis, which corresponds to this change of q, may be caused by the spin-lattice relaxation mechanism. Thus we assume that Δq obeys the equation

$$\frac{d\Delta q}{dt} = -\frac{\Delta q - (\Delta q)_{\text{static}}}{\tau}, \tag{4}$$

where τ is the spin-lattice relaxation time, and

$(\Delta q)_{\text{static}}$ is Δq for a static strain and is calculated from the condition $\partial E_{\text{ex}}/\partial(\Delta q)=0$, using (2) and (3). The energy dissipation of the spin system or the energy dissipation from the sound wave is calculated from the equation

$$\left\langle \frac{dW}{dt} \right\rangle = -\left\langle \frac{\partial E_{\text{ex}}}{\partial q}\frac{dq}{dt} \right\rangle, \tag{5}$$

the angular brackets indicating time averages. Using (2)-(5), the ultrasonic-attenuation constant α_l is obtained as

$$\alpha_l = \left\langle \frac{dW/dt}{v_l U_l} \right\rangle = \frac{nS^2}{\rho v_l^3}\frac{(\partial^2 J/\partial q \partial \epsilon_{zz})^2}{\partial^2 J/\partial q^2}\frac{\tau\omega^2}{1+\tau^2\omega^2}, \tag{6}$$

where n is the number of spins per unit volume, and v_l and U_l are the velocity and energy density of the longitudinal wave, respectively. v_l is obtained from (3) as $\{[c_{11}+(\partial^2 J/\partial \epsilon_{zz}^2)S^2]/\rho\}^{1/2}$ and U_l is given by $\frac{1}{2}\rho\omega^2 u_0^2$, where ρ is the density of the sample and c_{11} is the elastic-stiffness constant.

Since it has been shown by neutron diffraction experiments that the helical-turn angle depends considerably on the hydrostatic pressure, we can expect a large contribution from this mechanism to the ultrasonic attenuation in the helical spin states of the rare-earth metals.[3] This ultrasonic-attenuation mechanism is characteristic of the helical spin structure and does not work in ferromagnets and antiferromagnets where the spin structures are stable for a small change of the exchange interaction ($\partial^2 J/\partial q \partial \epsilon_{zz} = 0$).

A similar calculation gives the attenuation constant of a transverse wave propagating along the c axis:

$$\alpha_t = \frac{nS^2 U_t}{2\rho^2 v_t^5}\frac{(\partial^3 J/\partial q \partial \epsilon_{xz}^2)}{\partial^2 J/\partial q^2}\frac{\tau\omega^2}{1+4\tau^2\omega^2}, \tag{7}$$

where v_t and U_t are the velocity and energy density of the transverse wave, respectively. The magnitude of α_t is much smaller than that of α_l because Δq is coupled with the shear strain only to second order. α_l is independent of the longitudinal sound energy, but α_t is proportional to the transverse sound energy. The denominator of the frequency dependent part of α_t is different from that of α_l.

In addition to the present mechanism, there is a contribution to the attenuation from spin fluctuations (these include both spin fluctuations that are described by means of spin waves at low temperatures and critical fluctuations of spins near the Néel temperature). The sharp peak in attenuation arising from critical fluctuations may be expected to be not so different from that found in ferromagnetic and antiferromagnetic states. These have already been observed in MnF_2,[4] MnO,[5] MnTe,[6] and Gd.[7]

Applying a longitudinal wave propagating along the c axis, we have measured the attenuation of sound in Ho and Dy single crystals. Typical temperature dependences of the attenuation are shown in Fig. 1 for Ho and in Fig. 2 for Dy. The maximum in the attenuation coefficient was chosen as the Néel temperature. The sharp peak at the Néel temperature comes from the critical fluctuations of spins as mentioned above. The attenuation below the Néel temperature is much larger than that above the Néel temperature, and increases as temperature decreases, as expected from (6). Since the attenuation in ferromagnets and antiferromagnets has been observed to decrease as temperature decreases, we believe that the anomalous increase of the attenuation below the Néel temperatures in Ho and Dy originates, at least in part, from the present mechanism.[8] The attenuation obtained experimentally is independent of the applied sound energy. This is consistent with the present theory. The frequen-

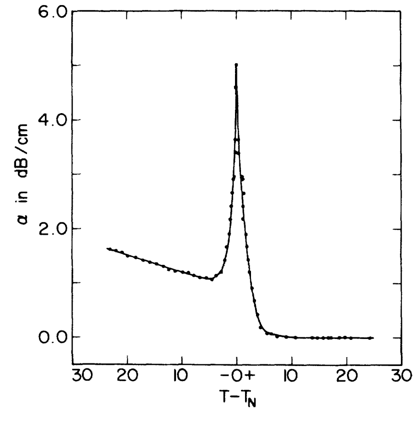

FIG. 1. Ultrasonic-attenuation coefficient as a function of temperature for holmium near the paramagnetic-antiferromagnetic phase transition. Data obtained with longitudinal waves at 45 Mc/sec propagating along the c axis are shown. The Néel temperature was chosen as the temperature at which the attenuation is a maximum.

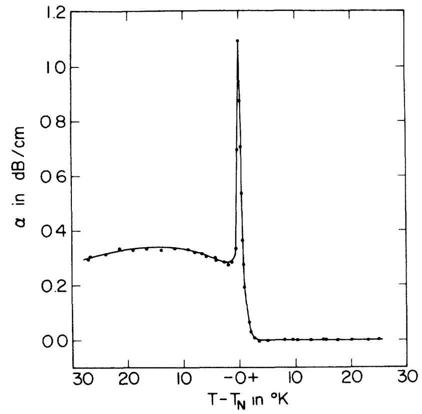

FIG. 2. Ultrasonic-attenuation coefficient of 15-Mc/sec longitudinal waves in dysprosium near T_N.

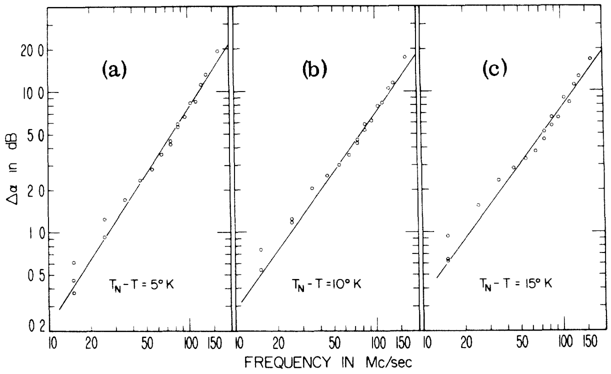

FIG. 3. Frequency dependence of the change in attenuation coefficient (see text) at three temperatures for holmium. (a) Slope of line = 1.5. (b) Slope of line = 1.4. (c) Slope of line = 1.3.

cy dependence of $\Delta\alpha_l$ for Ho is shown in Fig. 3. In this figure, $\Delta\alpha_l$ is defined as the difference in attenuation between the attenuation immediately above the Néel temperature, where the attenuation is almost constant as a function of temperature, and the attenuation at the desired temperature below the Néel temperature. As seen from this figure, $\Delta\alpha_l$ is proportional to the 1.3-1.5 powers of ω, depending on the temperature. $\Delta\alpha_l$ for Dy is almost proportional to ω. If τ is independent of the frequency of sound, it is very difficult to explain the frequency dependences of the attenuation from the present phenomenological theory. Thus, we are investigating the microscopic mechanism of the spin-lattice relaxation of the helical spins.

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[8] Recently, Evans and Daniel have observed large ultrasonic attenuation independent of temperature below the Néel temperature in MnO and CoO. This attenuation may originate from domain-wall motion and its mechanism is considered to be similar to the present mechanism.