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Rare-Earth Ultrasonic Attenuation in Applied Fields

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I show that dynamic screening of phonons by spin-polarized conduction electrons is an important mechanism for anomalous field-dependent acoustical absorption in heavy rare-earth metals. The absorption coefficient is larger by a factor $\{\partial[N(E_F)]^{-1}/\partial E\}^2$ than that arising from the presenlty accepted mechanism of exchange modulation. Here $N(E_F)$ is the density of states at the Fermi energy. The expected behavior in degenerate magnetic semiconductors is also discussed and interesting predictions for experiment are made.

Acoustical absorption in the paramagnetic region of heavy rare-earth metals increases as H^2 in low fields.[1,2] This is in contrast to the expectations of the conventional theory[3] in which phonons scatter from spin fluctuations, since these fluctuations are suppressed by the applied field. Tachiki and Maekawa[4] recognized the importance of off-resonance response of the local $4f$ spins to sound vibrations, and proposed that absorption arises through phonon modulation of the indirect-exchange interaction (volume magnetostriction). Recently, Kim[5] has demonstrated the importance of screening by conduction electrons, in explaining anomalous, *zero-field* absorption in the paramagnetic phase of itinerant magnets. In these materials the electron-density response is strongly exchange-enhanced, increasing sharply as the Curie temperature, T_C, is approached from above. Electron density polarization, coupled to the lattice motion through the electron-phonon interaction, becomes easier as a result of exchange enhancement, so that a softening of the longitudinal phonons and enhanced sound attenuation are predicted.

In rare-earth metals, exchange enhancement among the $5d$-$6s$ conduction electrons is much weaker and itinerant magnetism is not observed. There is no dramatic increase in the electronic response near T_C, below which the well-localized $4f$ moments order magnetically, and in zero field I do not expect anomalous attenuation from a dynamical screening mechanism. A magnetic field, however, induces long-range order among the $4f$ spins, and the conduction spin sub-bands split in the strong c-f (i.e., s-f or d-f) exchange field. Repopulation in the spin sub-bands produces a spin polarization in the electronic density at the Fermi surface.

In this Letter I show that dynamic screening of phonons by spin-polarized conduction electrons is an important mechanism for explaining anomalous field-dependent absorption in the rare-earth metals: The spin-polarized electrons move to screen the electric perturbation and induce an energy-dissipating off-resonance response among the $4f$ electrons, via the c-f exchange interaction. The effect becomes especially important near T_C, where the magnetic response of the $4f$ moments becomes large.

The ionic charge-density oscillations, created by a longitudinal ultrasonic wave of amplitude u_0, wave vector $\vec{q}$, and frequency Ω, that is propagating along the c axis (z direction) of an hcp metal may be written as

$$\delta\rho(\vec{r}, t) = -iz_0 e n_0 u_0 q e^{i(qz-\Omega t)},$$

where $n_0 = N_0/V$ is the number of unit cells per unit volume and z_0 is the ionic valency. In the random-phase approximation (RPA), the linear magnetic response is given by[6]

$$\delta M_{\vec{q},\Omega} = \frac{e^{-1}hV_{\vec{q}}\chi_{\vec{q},\Omega}^{-}\chi_{\vec{q},\Omega}^{H}\delta\rho_{\vec{q}}}{1+V_{\vec{q}}\chi_{\vec{q},\Omega}^{+}-h^2N_0\chi_{\vec{q},\Omega}^{H}(\chi_{\vec{q},\Omega}^{+}+4V_{\vec{q}}\chi_{\vec{q},\Omega}^{0\uparrow}\chi_{\vec{q},\Omega}^{0\downarrow})}, \tag{1}$$

where $h = I/(g\mu_B N_0)$, I being the c-f exchange constant, g, the gyromagnetic ratio, and μ_B, the Bohr magneton. The quantity $V_{\vec{q}} = 4\pi e^2/(\kappa_0 q^2)$ is the "bare" Coulomb energy, where the *background* dielectric constant, κ_0, due to core-electron and lattice polarizability, is of order 1 for the rare-earth metals. The quantity $\chi_{\vec{q},\Omega}^{H}$ is the Heisenberg susceptibility of the local spins in the *absence* of conduction electrons and $\chi_{\vec{q},\Omega}^{\pm} \equiv \chi_{\vec{q},\Omega}^{0\uparrow} \pm \chi_{\vec{q},\Omega}^{0\downarrow}$, where $\chi_{\vec{q},\Omega}^{0\sigma}$ are the spin-dependent Lindhard functions.[6] One should note that when the magnetization, σ_0, vanishes (e.g., in the paramagnetic regime with $\vec{H}=0$), $\chi_{\vec{q},\Omega}^{-}=0$ and the "cross coupling" disappears.

For a very-long-wavelength ultrasonic perturbation it is appropriate to use the extreme limit of the Thomas-Fermi approximation: $\chi_{\vec{q},\Omega}^{0\sigma} \to \frac{1}{2}N(E_{F\sigma})$ and $N(E_{F\sigma})V_{\vec{q}} \gg 1$. (Here I neglect $5d$ exchange-correlation effects in the electron gas, which will enhance the value of $\chi^{0\sigma}$ in this limit to some extent.[5]) The quantity $N(E_{F\sigma})$ is the density of states at the Fermi energy of the σ sub-band, where the energies are referred to the bottom of the sub-bands: $E_{F\sigma} = \pm\frac{1}{2}IS\sigma_0 + E_F$. The quantity E_F is the paramagnetic Fermi energy and S is the ground-state spin quantum number of the $4f$ shell. Since $IS\sigma_0 \ll E_F$ for rare-earth metals, I expand the quantity $\chi_{\vec{q},\Omega}^{-} \approx \frac{1}{2}N(E_{F\sigma}) - N(E_{F\sigma})$ to lowest order, and Eq. (1) becomes

$$\delta M_{\vec{q},\Omega} = \chi_{\vec{q},\Omega}^{H}[1-h^2N_0\chi_{\vec{q},\Omega}^{H}N(E_F)]^{-1}\delta H_{\vec{q}}, \tag{2}$$

where $\delta H_{\vec{q}} = [2eg\mu_B N_0 N(E_F)]^{-1} I^2 S\sigma_0(\partial N/\partial E)_{E=E_F}\delta\rho_{\vec{q}}$. The quantity $\delta H_{\vec{q}}$ is an effective field generated by the polarized-conduction-electron charge density, which is screening the phonon mode, $\delta\rho_{\vec{q}}$. The factor multiplying $\delta H_{\vec{q}}$ is the c-f exchange-enhanced Heisenberg susceptibility [see, for example, Ref. 6, Eq. (41)]. Since this RPA expression for the magnetic response is not very reliable in cases where spin-flip processes play an important role,[6] it is better to treat the response function phenomenologically and write $\delta M_{\vec{q},\Omega} \equiv \chi_{\vec{q},\Omega}^{\mathrm{loc}}\delta H_{\vec{q}}$. A good fit to inelastic neutron scattering data[7] in Tb is obtained with a relaxation function of the form $R(\vec{q},t) = \chi_{\vec{q}}\exp(-\Gamma_T t)$, $\chi_{\vec{q}}$ being the wave-vector–dependent susceptibility, and Γ_T, the relaxation frequency of spin fluctuations perpendicular to the crystal c axis.[8] Using the Kubo formula[9] I obtain the generalized susceptibility from $R(\vec{q},t)$:

$$\chi_{\vec{q},\Omega}^{\mathrm{loc}} = \chi_{\vec{q}}\Gamma_T^2(\Gamma_T^2+\Omega^2)^{-1}(1-i\Omega/\Gamma_T).$$

The attenuation coefficient is given by $\alpha_L \equiv P/(c_s U_p)$, where $P = \frac{1}{2}\Omega\,\mathrm{Im}\chi_{\vec{q},\Omega}^{\mathrm{loc}}|\delta H_{\vec{q}}|^2$ and c_s is the speed of sound. The elastic energy density $U_p = \frac{1}{2}Mn_0c_s^2u_0^2q^2$, where M is the unit-cell mass. Using the mean-field expression $\chi_{\vec{q}} = N_0(g\mu_B)^2S(S+1)[3k_B(T-T_{\vec{q}})]^{-1}$, and taking $\vec{H}$ perpendicular to the c axis, I obtain

$$\alpha_L = \frac{2(g\mu_B)^2J^3(J+1)^3\Gamma_T\Omega^2H^2}{27k_B^3Mc_s^3(\Gamma_T^2+\Omega^2)(T-T_0)^2(T-T_{\vec{q}})}\left\{\frac{z_0(\lambda-1)^2I^2}{2\sqrt{2}\,N(E_F)}\left(\frac{\partial N}{\partial E}\right)_{E=E_F}\right\}^2. \tag{3}$$

Here I have made the usual projection $\vec{S}\to(\lambda-1)\vec{J}$, where $\vec{J}$ is the total-angular-momentum operator and λ is the Landé factor.

The quantity $T_{\vec{q}}$ is the ordering temperature for a helical spin arrangement in the hexagonal plane of wave vector $\vec{q}$, and is given explicitly by the usual expression $T_{\vec{q}} = 2S(S+1)(3k_B)^{-1}J_{\vec{q}}$, where $J_{\vec{q}}$ is the Fourier transform of the Ruderman-Kittel-Kasuya-Yosida (RKKY) indirect-exchange energy. For ultrasonic wavelengths, $T_{\vec{q}} \approx T_0$, the paramagnetic Curie temperature. Equation (3) is valid for low fields and yields the H^2 dependence, observed in Ho and Tb.[1,2] This H^2 dependence of $\alpha_L \sim |\delta H_{\vec{q}}|^2$ arises from the replacement $\sigma_0 \to \chi_0 H \sim H(T-T_0)^{-1}$ in the expression for $\delta H_{\vec{q}}$ below Eq. (2) and is valid only for low fields. At higher fields, local spins saturate and are no longer capable of responding to the spin-polarized wave of screening charge. The absorption is expected to drop rapidly to zero, producing a peak structure. Such peaks are observed in the isotherms of $\alpha_L(\vec{H},T)$ in Tb,[2] the peak position corresponding roughly to fields at which the magnetization be-

gins to saturate.[10]

Equation (3) was written in a form that can be easily compared with the attenuation, expected from the exchange-modulation mechanism[1,2,4] [see Eq. (8) of Ref. 1]. The identical temperature and field dependence of these low-field formulas can be attributed to the form of the relaxation function, assumed in both derivations, and to the fact that in both cases the "driving field", $\delta H_{\vec{q}}$, is proportional to σ_0. In the exchange-modulation theory, however, this field is proportional to the magnetostrictive energy, $E_{MS}=\sum_i z_{ij}J_{ij}/z_i$, whereas in the present model it is proportional to the factor enclosed in curly brackets in Eq. (3). This factor represents, essentially, the energy of the *spin*-density wave, which is screening the phonon mode in the field of the local moments. Using the usual expression[11] for J_{ij}, the RKKY indirect-exchange interaction, one finds that $E_{MS} \sim (\lambda-1)^2 I^2 N(E_F)$, which is the energy of the corresponding *charge*-density wave in the field of the local moments. Comparing these expressions, one sees that the present mechanism yields an attenuation coefficient which is larger by the *square* of a factor $\Delta \equiv d[N(E_F)]^{-1}/de$. Since E_F lies within a sharp spike in the density of states,[12,13] Δ is quite large for the heavy rare-earth metals.

If I approximate the density-of-states peak by a cutoff Lorentzian of height $N_m(E_m)$ and full width at half-maximum γ, then for E_F within the peak I obtain $\Delta = 8(N_m\gamma^2)^{-1}|E_F - E_m|$. Taking the relativistic augmented-plane-wave bands of Dy as a typical example,[12] $N_m = 25.5$ Ry^{-1} per atom, $|E_F - E_m| \sim 0.01$ Ry, and $\gamma \sim 0.03$ Ry, we obtain $\Delta \cong 3.5$. A similar value is obtained using Tb bands.[13] This rather crude estimate indicates that the present mechanism may well be dominant for the rare-earth metals.

In heavily doped semiconductors (e.g., EuO doped with 2% La), this mechanism should lead to very anomalous absorption. If the spacing between singly ionized nonmagnetic impurities is much less than the ultrasonic wavelength, then a well-defined impurity-charge-density oscillation is set up by the sound wave. For small doping concentrations, n_c, the attenuation coefficient is found to be

$$\alpha_L = (Mn_0 c_s^3)^{-1}\Omega\hbar^2 \mathrm{Im}\chi^{H}_{\vec{q},\Omega}(n_\uparrow - n_\downarrow)^2,$$

where n_σ is the concentration of electrons in the σ-spin sub-band. Since $E_F \approx IS\sigma_0$ for the degenerate magnetic semiconductor, small changes of the magnetization produce radical repopulation effects. Defining $x_\sigma = n_\sigma/n_c$, conservation of conduction electrons yields the relations $x_\uparrow^{2/3} - (1-x_\uparrow)^{2/3} = IS\sigma_0/E_{SB}$ for $IS\sigma_0 \le E_{SB}$, and $x_\uparrow = 1$ for $IS\sigma_0 > E_{SB}$. Here $E_{SB} = \hbar^2(6\pi^2 n_c)^{2/3}/2m^*$ is the Fermi energy of the spin-up sub-band when the spin-down sub-band is completely depopulated (split-band limit).

In EuO, a low-frequency spin-lattice channel,[14,15] as well as a high-frequency spin-spin relaxation[16] channel, must be considered for Ω in the upper MHz range: $10 \lesssim \Omega \lesssim 500$ MHz. Assuming an exponential form for the relaxation function[17] with $\omega_{s1} \ll \Omega \ll \omega_{ss}$ I obtain $\mathrm{Im}\chi^{H}_{\vec{q},\Omega} \cong -\chi_{\vec{q}}[(\omega_{s1}/\Omega)\chi_W + (\Omega/\omega_{ss})(1-\chi_W)]$ for the relaxation of spin fluctuations along the field direction. Here ω_{s1} and ω_{ss} are, respectively, the spin-lattice and spin-spin relaxation frequencies, and $\chi_W \equiv 1 - \chi_s/\chi_0$, where χ_s and χ_0 are, respectively, the adiabatic and isothermal susceptibilities. The weighting factors involving χ_W may be understood as follows: If $\chi_s \approx \chi_0$, then there is little flow of heat to the lattice bath, during one period of sound vibration. In this case, the spin system is well-isolated from the lattice, and only the high-frequency spin-spin relaxation channel is open. Using this expression for $\mathrm{Im}\chi^{H}_{\vec{q},\Omega}$ in the limit of long wavelengths, we obtain for the attenuation

$$\alpha_L = \frac{I^2 f_c^2 (x_\uparrow - x_\downarrow)^2 \chi_0}{Mc_s^3 (g\mu_B)^2 N_0}[\omega_{s1}\chi_W + \omega_{ss}^{-1}(1-\chi_W)\Omega^2], \quad (4)$$

where f_c is the fraction of impurities.

Neutron diffraction studies of EuO and EuS indicate[18] that the criterion[19] $(\kappa_c/q)^2 > 0.08$ is well satisfied to within experimental resolution of temperature near T_0, where κ_c is the spin correlation length. This justifies to some extent[19] the use of the mean-field approximation, $M = N_0 g\mu_B S B_S(\vec{H}, T)$, in the calculations. Here B_S is the Brillouin function and $S = \frac{7}{2}$ for EuO.

From thermodynamics

$$\chi_W(\vec{H}, T) = 1 - C_M/C_H = T(C_H\chi_0)^{-1}(\partial M/\partial T)_H^2,$$

where $\chi_0 \equiv (\partial M/\partial H)_T$ and the specific heats C_M and C_H are strong functions of $\vec{H}$ and T, although from conditions of thermodynamic stability $0 \le \chi_W \le 1$. Using the general relation

$$(\partial C_H/\partial H)_T = T(\partial^2 M/\partial T^2)_H$$

to calculate the field dependence of C_H, and taking $C_H(\vec{H}=0) \approx 30$ J/mole K at $|T - T_0| = 1.0$ K,[20] I find that $\chi_W(\vec{H})$ rises rapidly from zero as H^2, levels off above 1.5 kOe, and drops very slowly back to zero at infinite field. For fields in the

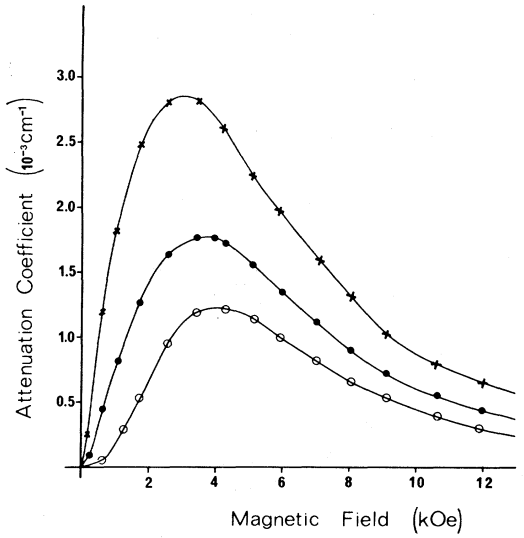

FIG. 1. Predicted attenuation-coefficient isotherms vs applied magnetic field in paramagnetic 2%-La-doped EuO, with $|T-T_0|=1.0$ K, at three sound frequencies: open circles, 10 MHz; solid circles, 300 MHz; crosses, 500 MHz.

range $2.6 \lesssim H \lesssim 20$ kOe, χ_W remains greater than 0.90.

The expected absorption isotherms at $|T-T_0|=1.0$ K for a 2%-La-doped sample are shown in Fig. 1 at three typical frequencies. No adjustable parameters were employed and the experimental values $c_s=4.60\times10^5$ cm/sec,[21] $I=0.12\times10^{-12}$ erg,[22] $\omega_{sl}\approx 1.5\times10^6$ sec^{-1},[15] and $\omega_{ss}\approx 10^{10}$ sec^{-1} [16] were used in Eq. (4). For the relaxation frequencies assumed, one expects frequency-independent attenuation, dominated by the spin-lattice mechanism, for $10 \lesssim \Omega \lesssim 100$ MHz. The lowest curve in Fig. 1 characterizes the absorption profile in this frequency range. Above 100 MHz, the spin-spin mechanism is dominant and the absorption increases as Ω^2. The upper curves in Fig. 1 show the profiles at 300 and 500 MHz.

The value that I used for ω_{ss} is rather rough, since we estimate it from the EPR linewidth, which narrows severely in the neighborhood of T_0 and in strong magnetic fields.[16,23] In fact, an experimental investigation of the high-field structure would yield new information on the field dependence of ω_{ss}, since the field dependence of the other quantities in Eq. (4) can be calculated with some degree of certainty. In this respect, acoustical attenuation has a decided advantage over EPR, since the field at fixed frequency may be varied easily in a continuous manner. Another appealing aspect is that information on *both* relaxation frequencies can be obtained from a single experiment.

The important conclusion of this paper is that, in metals possessing good local moments and large c-f exchange coupling, the dominant mechanism for sound absorption in applied fields is the off-resonance response of the local moments to the conduction-electron density, which is adiabatically screening the lattice wave. In rare-earth metals, where the Fermi energy lies within a large density-of-states peak, it is most probably the response of the local moments to the spin-density wave, rather than to the corresponding charge-density wave, which is of paramount importance for attenuation.

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