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INFLUENCE OF MAGNON-PHONON COUPLING ON THE LOW-TEMPERATURE MAGNETIC PROPERTIES OF AN ANTIFERROMAGNET*

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There have been several experiments[1-4] bearing on the temperature dependence of the sublattice magnetization, parallel susceptibility, and nuclear relaxation rates of antiferromagnets. The surprising result is that, with the possible exception of MnF_2,[3] none of the experiments show any evidence for the expected energy gap[5] in the spin-wave spectrum. Furthermore, the experiments do not agree with the predictions of the usual spin-wave theory[6,7] even when the anisotropy is negligible and no energy gap is expected. The measurements[2,3] of the longitudinal nuclear relaxation time T_1 are also in disagreement with the theoretical predictions.[8] We give here a theory which is able to explain most of these results, but in terms of one coupling constant which is not too well known independently. The central idea is that the magnetostrictive terms in the Hamiltonian produce a magnon component in the thermal phonon spectrum, so that the phonons participate directly in magnetic processes.

We define the temperature T_{AE} corresponding to the energy gap by $k_B T_{AE} = \hbar\gamma(2H_e H_A)^{1/2}$, where γ is the gyromagnetic ratio, H_e the exchange field, and H_A the anisotropy field. For $T \ll T_{AE}$ the magnetic thermodynamic properties will be mainly determined by the admixture of spin-wave character into the acoustic phonons. The effective density of thermally excited magnons will then not exhibit the exponential temperature variation predicted by ordinary spin-wave theory.[5]

For the bilinear coupling between the magnons and phonons we take

$$\mathcal{H}' = G\sum_i (S_i^x S_i^z \epsilon_i^{xz} + S_i^y S_i^z \epsilon_i^{yz}), \qquad (1)$$

where G is the coupling constant and $\epsilon_i^{\mu\nu}$ the strain tensor. In a forthcoming paper[9] the transformations to the mixed magnon-phonon modes are given in detail. We have calculated the magnetic properties arising from the low-lying quasi-phonons for $T \ll T_{AE}$. The deviation of the magnetization is

$$[M(0) - M(T)]/M(0) = \frac{3(6\pi^2)^{2/3} G^2 S^2}{2(\gamma H_A)^2 k_B \Theta M a^2} (T/\Theta)^4 \Gamma(4)\zeta(4), \qquad (2)$$

where M is the ionic mass; a is the lattice constant; $\Gamma(x)$ and $\zeta(x)$ are, respectively, the gamma function and Riemann zeta function; and Θ is the Debye temperature. For $\Theta = 110°K$, $a = 10^{-7}$ cm, $H_A = 10^3$ oe, and $M = 10^{-22}$ g, (2) agrees with the T^4 dependence of the sublattice magnetization observed by Poulis and Hardeman[1] in $CuCl_2 \cdot 2H_2O$ if we set $G \cong 10^{-13}$ erg. In MnF_2, Jaccarino and Walker[3] find that the observed sublattice magnetization does not seem to obey a unique power law, but (2) gives the observed order of magnitude when $G \cong 10^{-13}$ erg. In MnF_2, $H_A = 10^4$ oe, $\Theta = 450°K$, and $a \cong 3 \times 10^{-8}$ cm; the other parameters are as for $CuCl_2 \cdot 2H_2O$. This contribution to the magnetization deviation is comparable with the contribution arising from the ordinary magnon branches of the spectrum in MnF_2.

For $T \ll T_{AE}$ the parallel susceptibility is proportional to the deviation of the sublattice magnetization and is given by

$$\chi_{\parallel} = (4\gamma\hbar/H_e a^3)[M(0) - M(T)]/M(0). \qquad (3)$$

The parallel susceptibility also has a T^4 temperature dependence. Such a variation has been observed by Stout and Matarrese[4] for $T \ll T_{AE}$ in CoF_2 and FeF_2; here $G \cong 10^{-12}$ erg fits the data quantitatively.

The low-lying quasi-phonon branch of the spectrum also plays an important role for the longitudinal nuclear relaxation time T_1. We calculate the direct relaxation rate,

$$1/T_1 \cong \frac{3\pi(6\pi^2)^{2/3}\hbar A^4 S^6 G^2}{2Ma^2(k_B\Theta)^4(\hbar\gamma H_A)^2}(T/\Theta), \qquad (4)$$

where A is the hyperfine coupling constant. In MnF_2, where $A \cong 10^{-18}$ erg, a relaxation rate proportional to T has been observed[3] and agrees in order of magnitude with (4) when the parameters above are used. Thus the nuclear relaxation and magnetization data find a consistent explanation. The Raman relaxation rate has a T^7 temperature dependence and agrees again with the observations[2] in $CuCl_2 \cdot 2H_2O$ where $G \cong 10^{-13}$ erg. The expression for the Raman relaxation rate is

$$1/T_1 \cong \frac{27\pi^3(6\pi^2)^{1/3}A^2S^6G^4}{16\hbar(\gamma H_A)^4M^2a^4(k_B\Theta)^3}(T/\Theta)^7 P_6, \qquad (5)$$

where A is the strength of the dipolar coupling and P_6 is given by

$$P_6 = \int_0^{x_{\max}} \frac{x^6 e^x}{(e^x-1)^2}dx; \quad x_{\max} = \hbar v k_{\max}/k_B T. \qquad (6)$$

Thus by the use of the magnon-phonon interaction we are able to explain several experiments. We require, however, somewhat larger coupling constants than have been reported in more symmetrical crystals.

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RELATION BETWEEN THERMAL CONDUCTIVITY DEFECT IN SODIUM CHLORIDE AND ACOUSTIC RELAXATION EFFECTS AT LOW TEMPERATURES

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Recently an anomalously large reduction in thermal conductivity at low temperatures has been measured[1] for NaCl single crystals grown by the Harshaw process. No such reduction occurs for crystals grown free from atmospheric contamination, and evidence is presented that the reduction in thermal conductivity is associated with an impurity ion in the crystal in which oxygen is one of the components. The thermal conductivity varies nearly as the square of the temperature from 1°K to 20°K. Evidence is presented that the mean free path satisfies an equation of the type

$$\lambda = (3.3\times10^{-5}\,\omega/V)^{-1}\ \text{cm}, \qquad (1)$$

where V is the average sound velocity in the Debye sense and $\omega = 2\pi$ times the frequency of the phonon. Several mechanisms for this effect were considered but no conclusive evidence was found for any of them.

One possibility was that a relaxation process might be involved. Recently a mechanical relaxation effect has been measured at low temperatures which is associated with the same set of impurity ions, and it is the purpose of this note to show that these relaxations are in agreement with the loss in thermal conductivity.

The crystals were obtained from Harshaw and from Isomet and were provided with oriented, parallel, flat faces. X-cut and AC-cut quartz plates were bonded to the specimens with 4-methyl-1-pentene. The temperature was controlled by