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Acoustic Two-Magnon Processes in an Antiferromagnet*

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The attenuation of acoustic phonons due to linear processes involving any pair of magnons (nuclear and electronic) is calculated for some antiferromagnets. Numerical estimates of the intensities from such processes are made for $RbMnF_3$ and for MnF_2. It is predicted that, in certain favorable cases, such effects may be observable with present-day sensitivities at frequencies which are not near any of the single-spin-wave resonant frequencies.

I. FORMALISM

Many magnetoelastic effects have been studied[1,2] lately in the cubic antiferromagnet RbMnFi. In part this is due to the fact that its anisotropy energy is very small because the substance maintains a cubic symmetry below its Néel temperature. Since many effects are inversely proportional to the anisotropy energy, this leads to relatively large signals.

In an earlier paper,[1] a general formalism was developed for treating a cubic antiferromagnet and was applied to some acoustic properties including single-magnon resonances. It is the purpose of this paper to extend that work to investigate the problem of a phonon interacting with the spins via two magnons. A derivation of the contribution to the linear[3] acoustic attenuation from such processes is presented in this section. In the next section the results are evaluated for some of the two-magnon processes and are discussed with respect to $RbMnF_3$. With very slight modifications, the results are also applied to uniaxial antiferromagnets such as MnF_2.

The formalism, notation, and approximations used in Ref. 1 are used here, since the coupling mechanism is essentially the same. That is, the phonons couple to the electronic spins via the single-ion or volume magnetoelastic interaction. Since the nuclear and electronic spins interact via a hyperfine term in the Hamiltonian, this effectively couples the phonons to the nuclear spins also.[4]

To lowest order in the single-ion magnetoelastic coupling, the change in the (complex) phonon dispersion relation due to the spins is

$$\begin{aligned}\omega^2=\omega_0{}^2(\mathbf{q}\lambda)+(NM)^{-1}\{&\textstyle\sum e_i(\mathbf{q}\lambda)\,e_j(\mathbf{q}\lambda)\,G_{il}q_lG_{jm}q_m\\ &\times(-i)(-i\beta)^{-1}\int dt\langle T(F_{il}(\mathbf{l}at)F_{jm}(\mathbf{l}'a't'))\rangle\\ &\times\exp[i\beta\omega_\nu(t-t')-i\mathbf{q}\cdot(\mathbf{l}-\mathbf{l}')]\},\end{aligned}\tag{1}$$

where ω_0 is the phonon frequency in the absence of the spins, $e_i(\mathbf{q}\lambda)$ is the polarization vector of the phonons with wave vector $\mathbf{q}$ and polarization λ, and M is the mass of a magnetic unit cell of the substance. The coupling constants G_{ij} and the combination of spin operators F_{ij} are given in Appendix B of Ref. 1. The correlation functions $\langle T(\;)\rangle$ are also defined in that reference. The corresponding term to lowest order in the volume magnetoelastic coupling[5] is

$$\begin{aligned}\omega^2=\omega_0{}^2(\mathbf{q}\lambda)&\\ &+(NM)^{-1}\textstyle\sum e_i(\mathbf{q}\lambda)\,e_j(\mathbf{q}\lambda)\,Q_i(\boldsymbol{\alpha}-\boldsymbol{\alpha}')\,Q_j(\boldsymbol{\alpha}_1-\boldsymbol{\alpha}_2)\\ &\times(e^{-i\mathbf{q}\cdot\boldsymbol{\alpha}}-e^{-i\mathbf{q}\cdot\boldsymbol{\alpha}'})(e^{i\mathbf{q}\cdot\boldsymbol{\alpha}_1}-e^{i\mathbf{q}\cdot\boldsymbol{\alpha}_2})(-\tfrac{1}{4}i)(-i\beta)\\ &\times\int dt\langle T(\mathbf{S}(\boldsymbol{\alpha}t)\cdot\mathbf{S}(\boldsymbol{\alpha}'t')\,\mathbf{S}(\boldsymbol{\alpha}_1t')\cdot\mathbf{S}(\boldsymbol{\alpha}_2t'))\rangle\\ &\times\exp[i\beta\omega_\nu(t-t')],\end{aligned}\tag{2}$$

where α is a site index,

$$Q_i(\boldsymbol{\alpha}-\boldsymbol{\alpha}')=(\nabla_\alpha)_iJ(\boldsymbol{\alpha}-\boldsymbol{\alpha}'),$$

and J is the nearest-neighbor exchange integral. It is understood that ω_ν approaches the real frequency ω from above the real axis after all time integrals and frequency sums are performed.

The approximation used in Ref. 1 for the average of four spin operators was the sum of all possible factorizations in terms of lower-order correlation function. However, in that paper, only resonant terms, those involving a single correlation function, were studied. In this paper those terms proportional to a product of two correlation functions are now considered [those terms in the first set of brackets in Eq. (B4) of Ref. 1], and cross terms between the single-ion and volume magnetostriction are neglected. After some lengthy but straightforward algebra (very similar to that of Ref. 1) the contribution to the phonon dispersion from nonresonant terms is obtained:

$$\begin{aligned}\omega^2=\omega_0{}^2(\mathbf{q}\lambda)-16(NM)^{-1}\sum_k\{&[C_{xx}{}^2+(Qa\mathbf{q}\cdot\mathbf{e})^2]\mathfrak{D}_{xxxx}\\ &+[C_{yy}{}^2+(Qa\mathbf{q}\cdot\mathbf{e})^2]\mathfrak{D}_{yyyy}\\ &+2[C_{xx}C_{yy}-(Qa\mathbf{q}\cdot\mathbf{e})^2]\mathfrak{D}_{xyxy}+2C_{xy}{}^2\mathfrak{D}_{xxyy}\},\end{aligned}\tag{3}$$

where $\mathfrak{D}(\omega)$ is the limit of $\mathfrak{D}(\omega_\nu)$ as ω_ν approaches ω from above the real axis and

$$\mathfrak{D}_{ijlm}(\omega_\nu)=\beta^{-1}\sum_{\bar\nu}D_{\bar i\bar j}(aa,\,q+k,\,\omega_\nu+\omega_{\bar\nu})\,D_{\bar l\bar m}(aa,\,k,\,\omega_{\bar\nu}).\tag{4}$$

In these equations the D's are the spin response functions defined and evaluated in Ref. 1, Q is defined in

Ref. 5, a is the spacing between magnetic ions, and the C's are complicated functions of q, $e(\mathbf{q}, \lambda)$, and the magnetic sublattice orientations. The C's are given for cases A and B of Ref. 1 in Eqs. (5) and (6), respectively:

$$\begin{aligned}C_{xx}&=G_{11}(\tfrac{3}{2}q_ze_z-\mathbf{q}\cdot\mathbf{e})(S_x^2-\tfrac{1}{2}S_z^2)/S_0^2\\&\quad+G_{44}[(\mathbf{q},\mathbf{e})_zS_z^2/2S_0^2\\&\quad-[(\mathbf{q},\mathbf{e})_x+(\mathbf{q},\mathbf{e})_y]S_xS_z/\sqrt{2}S_0^2],\\C_{yy}&=-\tfrac{1}{2}G_{11}(\tfrac{3}{2}q_ze_z-\mathbf{q}\cdot\mathbf{e})-\tfrac{1}{4}G_{44}(\mathbf{q},\mathbf{e})_z,\\C_{xy}&=\tfrac{3}{4}G_{11}(q_ye_y-q_xe_x)S_z/S_0\\&\quad+G_{44}[(\mathbf{q},\mathbf{e})_y-(\mathbf{q},\mathbf{e})_x]S_x/2^{3/2}S_0,\end{aligned}\tag{5}$$

where $(\mathbf{q}, \mathbf{e})_x=q_ye_z+q_ze_y$ with cyclic variations for y and z; and for case B

$$\begin{aligned}C_{xx}&=\tfrac{1}{2}G_{11}(\mathbf{q}\cdot\mathbf{e}-\tfrac{3}{2}q_ze_z)+\tfrac{1}{2}G_{44}(\mathbf{q},\mathbf{e})_z,\\C_{yy}&=\tfrac{1}{2}G_{11}(\tfrac{3}{2}q_ze_z-\mathbf{q}\cdot\mathbf{e})\\&\quad+G_{44}[-\tfrac{1}{6}(\mathbf{q},\mathbf{e})_z+\tfrac{1}{3}[(\mathbf{q},\mathbf{e})_y-(\mathbf{q},\mathbf{e})_x]\epsilon].\end{aligned}\tag{6}$$

The frequency sum in Eq. (4) can easily be performed. However, because of the complexity of the results, we shall now specialize to the "spin-flop" case, where the net sublattice magnetization is almost parallel to the applied field. In this case D_{xy} is negligible and the remaining D's can be expressed simply as

$$\begin{aligned}&D_{\xi\xi}(aa,\mathbf{q},\omega)\\&\quad=\sum_i A_{\xi i}(\mathbf{q})\{[\omega-\omega_i(\mathbf{q})]^{-1}-[\omega+\omega_i(\mathbf{q})]^{-1}\},\end{aligned}\tag{7}$$

where ξ is $\bar{x}$ or $\bar{y}$ (in the notation of Ref. 1) and i runs over the two electronic modes (denoted $e+$ and $e-$) and the two nuclear modes (denoted $n+$ and $n-$). For small wave vectors the only nonzero A's [as seen from Eqs. (3.1) of Ref. 1] are

$$\begin{aligned}A_{\bar{x}e+}&=\omega_ES_0/2\omega_{e+}(q),\\A_{\bar{y}e-}&=\omega_ES_0/2\omega_{e-}(q),\\A_{\bar{x}n+}&=\omega_E^2\omega_N^2\omega_{NE}S_0/\omega_{e+}^4(q)\,\omega_{n+}(q),\\A_{\bar{y}n-}&=\omega_E^2\omega_N^2\omega_{NE}S_0/\omega_e^4(q)\,\omega_{n-}(q).\end{aligned}\tag{8}$$

In these equations ω_E is the exchange frequency, ω_N is the unpulled nuclear frequency, and ω_{NE} is the hyperfine frequency.

Thus, in the spin-flop case,

$$\begin{aligned}\mathfrak{D}_{\xi\xi\eta\eta}&=\tfrac{1}{2}\sum_{i,j}A_{\xi i}(q+k)A_{\eta j}(k)\{[\coth\tfrac{1}{2}\beta\omega_i(q+k)\\&\quad-\coth\tfrac{1}{2}\beta\omega_j(k)]\{[\omega_j(k)-\omega_i(q+k)+\omega+i\delta]^{-1}\\&\quad-[\omega_i(q+k)-\omega_j(k)+\omega+i\delta]^{-1}\}\\&\quad+[\coth\tfrac{1}{2}\beta\omega_j(k)+\coth\tfrac{1}{2}\beta\omega_i(q+k)]\\&\quad\times\{[\omega_j(k)+\omega_i(q+k)+\omega+i\delta]^{-1}\\&\quad-[-\omega_j(k)-\omega_j(q+k)+\omega+i\delta]^{-1}\}\},\end{aligned}\tag{9}$$

where δ is an arbitrarily small positive quantity. With the frequencies which we will consider (less than 10^{12} sec^{-1}) q may be set equal to zero in Eq. (9).

II. DISCUSSION AND CONCLUSIONS

It is now easy to obtain the contributions to the absorption from the various two-magnon processes. As a function of the dimensionless variable $x=ka$, the nuclear and electronic eigenfrequencies may be written as [Eq. (3.2) of Ref. 1]

$$\begin{aligned}\omega_{n\pm}(x)&=\omega_N[1-2\omega_E\omega_{NE}/\omega_{e}{}^2{}_{\pm}(x)]^{1/2},\\\omega_{e\pm}(x)&=[\omega_{e\pm}{}^2(0)+\xi\omega_E^2x^2]^{1/2}.\end{aligned}\tag{10}$$

These are valid if $x\ll1$ and ξ is a dimensionless parameter of order one. These formulas are probably only valid for even smaller x because of spin-wave interactions. Since the imaginary part of D contains δ functions and, for small wave vectors, the k dependence of the modes depends only on $|k|$, the remaining integrals are trivial.

There are many possible two-magnon processes. For example, the contribution to the absorption coefficient[6] α from the formation of two $e\pm$ magnons is

$$\alpha(2e\pm)=Al_\pm[1-(\omega_{e\pm}(0)/\omega)^2][\omega_{e\pm}(0)/\omega],\tag{11}$$

where $\omega_e(0)$ is the electronic frequency at zero wave vector and

$$\begin{aligned}A&=2S_0^2G_{11}^2/\pi\beta\hbar\omega_E\xi^{3/2}Mv_0^3\hbar,\\l_\pm&=(C_{xxyy}^2/q^2G_{11}^2)+(Qa/G_{11})^2\hat{q}\cdot\mathbf{e},\end{aligned}\tag{12}$$

where S_0 is the average spin, v_0 is the velocity of the sound wave, and β is 1 over the Boltzmann constant times the temperature.

The absorption increases as $\omega/2\omega_{e\pm}(0)$ increases because the density of available states becomes larger.

The contribution to α from two $n\pm$ magnons is

$$\begin{aligned}&\alpha(2n_\pm)=Al_\pm[\omega_N^2/2\omega_E\omega_{NE}]^{1/2}f(y;y_0),\\&f(y,y_0)=(1-y^2)^{3/2}[1-(y_0/y)]^{1/2}(1-y_0^2)^{-1/2},\\&y=\omega/2\omega_N,\qquad y_0=\omega_{n\pm}(0)/\omega_N.\end{aligned}\tag{13}$$

This formula has been expressed in terms of the frequency in units of twice the unpulled nuclear frequency and the real nuclear frequency at zero wave vector divided by the unpulled nuclear frequency. Thus y always lies between y_0 and one because $\omega_n(q)$ lies between $\omega_n(0)$ and ω_N.

In contrast to the case for two $e\pm$ magnons, the absorption is largest for ω near $2\omega_{n\pm}(0)$ for two $n\pm$ magnons. The reason for this is that the wave-vector-dependent coupling to the nuclear modes is inversely proportional to $[\omega_{e\pm}(q)]^4$. (This can be thought of as arising from energy denominators in a perturbation scheme.) Since $\omega_{e\pm}(q)$ increases with increasing q, the interaction is considerably stronger for nuclear magnons of small wave vector. This stronger coupling more than compensates for the reduced density of available states at small q. In addition, since the absorption depends only on magnons of very small q ($x \ll 1$) except for ω very near to $2\omega_N$, spin-wave interaction should be unimportant.

The contributions from one n magnon plus one e magnon can also be easily calculated. However, the process is probably not separated enough in frequency from the much larger single e-magnon resonance to be distinguished experimentally.

Besides the approximations mentioned earlier (including those of Ref. 1), it has been assumed that $\beta\hbar\omega$ is much less than 1 in Eqs. (11) and (13). Using published values [2] for the parameters involved, A is approximately 0.042 cm^{-1} at 4°K for RbMnF$_3$. The quantity ξ was estimated to be about 0.12 by assuming only nearest- and next-nearest-neighbor exchange integrals and fitting them via a Weiss theory to the two parameters $T_N = 83.6$°K and $T_\theta = -160$°K. The quantity $\omega_N(2\omega_E\omega_{NE})^{-1/2}$ is about 0.13 in this substance.

In RbMnF$_3$ the double nuclear magnon decay of longitudinal phonon could contribute more than 0.005 cm^{-1} to α because presumably[7] the volume magnetostriction is considerably larger than the single-ion magnetostriction. The contribution is largest when the nuclear frequency is pulled the most. This is reflected in $f(y;\, y_0)$, which has a maximum of 1 if $y_0 = 0$. However, its maximum approaches zero as

$$(3^{3/2}/16)(1-y_0^2)$$

as y_0 approaches 1. Thus this function depends strongly on the strength of the coupling of the nuclear and electronic modes.

Equations (11)–(13) are almost exactly the same for an almost cubic uniaxial antiferromagnet at low temperatures with a magnetic field along the special axis. The angular factors from the single-ion magnetostriction are, of course, different. For example, the contribution to α due to the volume magnetostriction formation of an $n+$, $n-$ pair are the same as given by Eq. (13) except that

$$y_0 = [\omega_{n+}(0) + \omega_{n-}(0)]/2\omega_N,$$
$$\omega_{n\pm}(0) = \omega_n \pm \omega_0\gamma_n/\gamma_e \qquad (14)$$

and

$$\omega_n = \omega_N(1 - 2\omega_E\omega_{NE}/\omega_{e+}\omega_{e-})^{1/2},$$
$$\omega_{e+}\omega_{e-} = 2[\omega_E(\omega_A + \omega_{NE})] - \omega_0^2 \qquad (15)$$

where ω_0 is the usual Larmour frequency of the electrons. Except for the anisotropy energy, the parameters for MnF$_2$ are close to those for RbMnF$_3$. However, one would have to have a large magnetic field (10^5 Oe) in order to pull y_0 small enough to make the effect comparable in size to that for RbMnF$_3$.

Thus we have estimated the contributions from two magnon processed to the ultrasonic attenuation in antiferromagnets. The basic approximations were the random-phase approximation for the single-particle response functions and the decomposition made for the product of four spin operators. Effects due to the lifetimes of the modes should be unimportant except near the edges of the absorption spectra. However, inhomogeneities that cause a spread of anisotropy energies or soften the directions of sublattice magnetizations will smear the effect out.

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[1] P. A. Fedders, Phys. Rev. B **1**, 3756 (1970).

[2] See, for instance, R. L. Melcher and D. I. Bolef, Phys. Rev. **186**, 491 (1969); W. J. Ince, *ibid.* **184**, 574 (1969); L. W. Hinderks and P. N. Richards, *ibid.* **183**, 575 (1969).

[3] This is *not* phonon pumping as discussed by A. Platzker and F. R. Morganthaler, Phys. Rev. Letters **22**, 1051 (1969).

[4] In fact the coupling between the electronic and nuclear spins in RbMnF$_3$ is so strong that it is technically incorrect to speak of either electronic or nuclear modes. The eigenmodes of the system are strongly coupled combinations. Thus when we speak of the electronic (or nuclear) modes, we really mean those lying higher (or lower) in frequency.

[5] Herbert S. Bennet and Erling Pytte, Phys. Rev. **155**, 553 (1967).

[6] In this paper the attenuation coefficient α is defined such that the amplitude of a wave decreases as $e^{-\alpha x}$, where x is a length.

[7] R. L. Melcher and D. I. Bolef, Phys. Rev. **178**, 864 (1969).