Light scattering from energy fluctuations in magnetic insulators

G. F. Reiter
City College of the City University of New York, New York 10031
and IBM Zurich Research Laboratory, 8803 Rüschlikon-ZH, Switzerland
(Received 10 April 1975)

It is shown that fluctuations in the total magnetic energy of a magnetic insulator will scatter light, leading to a peak in the scattered intensity around zero energy transfer with a width proportional to the spin-lattice relaxation frequency, and an integrated intensity proportional to the specific heat.

It has been known for some time[1] that fluctuations in the spin density in a magnetic insulator would scatter light. It is the purpose of this paper to point out that the same mechanisms that are responsible for the scattering by the spin fluctuations couple the light to the total magnetic energy of the system. This gives rise to a peak in the differential scattering cross section centered at zero frequency, with a width that can be defined to be the spin-lattice relaxation time, a line shape that depends upon the details of the coupling, and an intensity that is proportional to the specific heat.

The mechanism for this peak is easily understood physically. When the magnetic system is weakly coupled to the lattice, its energy will not be a constant, but will fluctuate about the mean determined by the lattice temperature. If we define a magnetic temperature such that the energy of the magnetic system is the conjugate thermodynamic variable, we can regard this temperature as a fluctuating variable. These temperature fluctuations will have a negligible direct effect on the dielectric constant of the system. The dielectric constant is affected by changes in the local structure of the spin system, through the spin-orbit coupling, so that

$$\delta\epsilon^{\alpha\beta}(\vec{x}) = \frac{\partial\epsilon^{\alpha\beta}}{\partial O^i(\vec{x})}\,\delta O_i(\vec{x})\,, \tag{1}$$

where $O_i(x)$ are the expectation values of some combination of spin operators $\tilde{O}_i(x)$ characterizing the spin configuration around the point x. $\delta O_i(x)$ will depend upon the temperature of the spin system, so that there will be an indirect variation of the dielectric constant given by

$$\delta\epsilon^{\alpha\beta}(\vec{x}) \propto \frac{\partial\epsilon^{\alpha\beta}}{\partial O^i(\vec{x})}\,\frac{\delta O_i(\vec{x})}{\partial T_M}\,\Delta T_M\,, \tag{2}$$

where T_M is the temperature of the magnetic system. As a consequence, the spectrum of the scattered light will have a component proportional to

$$\int_{-\infty}^{\infty} e^{-i\omega t}\langle \Delta T_M(t)\Delta T_M(0)\rangle\, dt\,,$$

which will give rise to a peak at zero frequency with a width that can be used as a definition of the spin-lattice relaxation time, assuming that the coupling coefficients do not vanish. The coupling coefficient may be written more explicity, using

$$\frac{\partial O_i(x)}{\partial T_M} = \beta_M^2\langle \delta\tilde{O}_i(x)\,\delta\mathcal{H}\rangle\,, \tag{3}$$

where $\delta\mathcal{H}$ is the fluctuation in the energy. The operator $\tilde{O}_i(x)$ will generally involve spins located within a few lattice sites of x. In this case, one can expect the correlation function in (3) to be proportional to the specific heat C_H. Since $\Delta T_M \propto \delta\mathcal{H}/C_H$, and $\langle \delta\mathcal{H}(O)\,\delta\mathcal{H}(O)\rangle \propto C_H$, the integrated intensity of the scattering due to the energy fluctuations will be proportional to C_H.

These considerations may be put on a rigorous microscopic basis.

The differential scattering cross section for the scattering of light from a field $\vec{E}_1$, wave vector $\vec{K}_1$, frequency ω_1, to a field $\vec{E}_2$, wave vector $\vec{K}_2$, frequency ω_2 by a magnetic system is given by[2]

$$\frac{d^2h}{d\Omega\, d\omega_2} = \frac{\omega_1\omega_2^3}{2\pi C^4 V}\,\frac{n_2}{n_1}\sum_{\alpha\beta\gamma\delta}\epsilon_1^\alpha\epsilon_2^\beta\epsilon_1^\gamma\epsilon_2^\delta \times\int_{-\infty}^{\infty} dt\, e^{-i\omega t}\langle \tilde{M}^{\alpha\beta}(K)\tilde{M}^{\gamma\delta}(K,t)\rangle\,, \tag{4}$$

where $\omega = \omega_2 - \omega_1$, $\vec{K} = \vec{K}_2 - \vec{K}_1$, $\vec{\epsilon}_1$, $\vec{\epsilon}_2$ are the polarization vectors of the incident and scattered light, n_1, n_2 are the respective indices of refraction, and V is the volume of the crystal. $\langle\ \rangle$ denotes a thermal average. The tensor $M^{\alpha\beta}(K)$ gives the dependence of the polarizability on the spin operators. To terms of second order in the spin operators we have[3]

$$M^{\alpha\beta}(\vec{K}) = \sum e^{i\vec{K}\cdot\vec{r}_i} M_i^{\alpha\beta}\,, \tag{5a}$$

$$M_i^{\alpha\beta} = \alpha_i^{\alpha\beta,u}(\omega_1)S^u + \sum_j \alpha_{ij}^{\alpha\beta,uv}(\omega_1)S_i^u S_j^v\,, \tag{5b}$$

$$\tilde{M}^{\alpha\beta}(\vec{K}) = M^{\alpha\beta}(\vec{K}) - \langle M^{\alpha\beta}(\vec{K})\rangle\,. \tag{5c}$$

The first term in (5b) leads to scattering from single spin fluctuations, i.e., fluctuations of the spin density with a given wave vector, and is well

understood theoretically. We shall not consider it any further. The second term has traditionally been regarded as giving rise to scattering by pairs of spin fluctuations, and it will be our concern to show that it also leads to scattering from the energy fluctuations of the system. We shall not consider the most general tensor $\alpha_{ij}^{\alpha\beta,\mu\nu}$ but restrict ourselves to

$$M_i^{\alpha\beta} = \sum_j P_{ij}^{\alpha\beta}(\omega_1)\vec{S}_i \cdot \vec{S}_j \ . \tag{6}$$

The terms omitted do not contribute to the energy fluctuation scattering for the spin Hamiltonian we shall consider, but could contribute for a Hamiltonian of lower symmetry.

The time evolution in (4) is determined by the Hamiltonian of the spin system, which we take to be

$$\mathcal{H} = -\frac{1}{2}\sum_q V(\vec{q})\vec{S}(\vec{q}) \cdot \vec{S}(-\vec{q}) + \mathcal{H}_{\text{spin lattice}} + \mathcal{H}_{\text{lattice}} \ . \tag{7}$$

The Heisenberg term in (7) is chosen for simplicity. The presence of anisotropy terms would not affect the substance of the argument, although they would change the scattering intensities.

The response function in (4) may be written in a form that will make the argument more transparent. It can readily be shown that

$$I(\omega) = \int_{-\infty}^{\infty} dt\, e^{i\omega t}\langle \tilde{M}^{\alpha\beta}(\vec{K})\tilde{M}^{\gamma\delta}(\vec{K}, t)^*\rangle$$

$$= \frac{2\omega}{e^{\beta\omega}-1}\,\mathrm{Re}\int_0^{\infty} dt\, e^{i(\omega+i\epsilon)t}\langle \tilde{M}^{\gamma\delta}(\vec{K}, t)\,|\,\tilde{M}^{\alpha\beta}(\vec{K})\rangle \ . \tag{8}$$

The notation $\langle\ |\ \rangle$ denotes an inner product on the linear vector space V consisting of all linear operators on the Hilbert space of the spin system [for instance, S_i^α, $S_i^\alpha S_j^\beta$, $(S_i^\alpha)^2$, $S_i^\alpha S_j^\beta S_K^\gamma$, are elements of V], and is defined for any two elements of V, A, and B, by

$$\langle A\,|\,B\rangle = \left\langle \int_0^\beta e^{T\mathcal{H}} A^\dagger e^{-T\mathcal{H}} B\, dT \right\rangle \ . \tag{9}$$

The time evolution in (8) can be expressed in terms of the Liouville operator for the system, defined by its action on any element A in V as

$$L[A] = (1/\hbar)[\mathcal{H}, A] \ , \tag{10}$$

i.e., $A(t) = e^{iLt}[A]$. In terms of L, we see that the differential scattering intensity is proportional to

$$I(\omega) = 2\beta^{-1}\,\mathrm{Re}\, i\langle \tilde{M}^{\alpha\beta}(\vec{K})\,|\,(\omega + i\epsilon - L)^{-1}\,|\,\tilde{M}^{\gamma\delta}(\vec{K})\rangle \ . \tag{11}$$

We have assumed $\beta\omega \ll 1$ in (11). L is Hermitian in the inner product defined by (9). Equation (11) expresses the scattering intensity in terms of the spectral density of L.

The correlation functions appearing in (8) are of the form

$$\sum_{qq'} P_q^{\alpha\beta}(\omega_1)P_{q'}^{\gamma\mu}(\omega_1)\langle \vec{S}(\tfrac{1}{2}\vec{K}+\vec{q}, t)\cdot\vec{S}$$
$$\times(\tfrac{1}{2}\vec{K}-\vec{q}, t)\,|\,\vec{S}(\tfrac{1}{2}\vec{K}+\vec{q}')\cdot\vec{S}(\tfrac{1}{2}\vec{K}-\vec{q}')\rangle \ , \tag{12}$$

where

$$P_q^{\alpha\beta}(\omega_1) = \sum e^{i\vec{q}\cdot\vec{r}_{ij}} P_{ij}^{\alpha\beta}(\omega_1) \ .$$

Standard treatments of light scattering proceed by approximating the four-spin correlation function in (12) by a product of pair correlation functions describing the spin-density fluctuation, i.e.,

$$\langle \vec{S}(\tfrac{1}{2}\vec{K}+\vec{q}, t)\cdot\vec{S}(\tfrac{1}{2}\vec{K}-\vec{q}, t)\,|\,\vec{S}(\tfrac{1}{2}\vec{K}+\vec{q})\cdot\vec{S}(\tfrac{1}{2}\vec{K}-\vec{q})\rangle$$
$$= \beta^{-1}\langle S^i(\tfrac{1}{2}\vec{K}+\vec{q}, t)\,|\,S^i(\tfrac{1}{2}\vec{K}+\vec{q})\rangle$$
$$\times\langle S^j(\tfrac{1}{2}\vec{K}-\vec{q}, t)\,|\,S^j(\tfrac{1}{2}\vec{K}-\vec{q})\rangle\delta(\vec{q}-\vec{q}') \ . \tag{13}$$

At low temperature ($T \ll T_c$), such an approximation leads naturally to the interpretation of the scattering as due to pairs of magnons. It has been possible to extend the calculations in the low-temperature regime[4] to include magnon interactions omitted by the approximation (13). In principle, such calculations should also show the effects that we shall describe, but existing calculations have not been detailed enough to do so. Richards and Brya[5] have taken another approach, which is to assume a form for the correlation function and use a calculation of the first two moments of the spectral density to determine the parameters in the assumed form. Either the moment method or the factorization approach misses an essential fact about the correlation function in (12) which is that it does not vanish, when $K=0$, as $t\to\infty$, if spin-lattice interactions are ignored. To see this most readily, we shall insert a complete set of states in V in the matrix element appearing in (11). Included in such a set would be states corresponding to all the operators that were constants of the motion of the spin system. If, as we shall assume temporarily, the spin-lattice interaction is neglected, then these would be the magnitude of the total spin on each site, $\vec{S}_i\cdot\vec{S}_i$, which is just $S(S+1)I$, with I the identity operator on V,

$$\sum_i S_i^z = N^{1/2}S^z(0)$$

the total spin, and $\mathcal{H}_{\text{Heisenberg}}$ the Hamiltonian. Below T_c, one would also have to include the staggered magnetization in an antiferromagnet.

These states may be orthonormalized, and denoted by $|\mathcal{H}i\rangle$ so that the identity operator on V may be resolved as

$$\sum_{i=1}^{3} |\mathcal{H}_i\rangle\langle\mathcal{H}_i| + \sum_j |R_j\rangle\langle R_j| \ . \tag{14}$$

The states $|R_j\rangle$ denote the remaining states needed to form a basis in V. A particular choice for the

operators corresponding to the states $|\mathcal{H}_i\rangle$, above T_c, would be

$$\mathcal{H}_1 = \beta^{-1/2} I \,, \tag{15}$$

where I is the identity operator on the Hilbert space of the spins,

$$\mathcal{H}_2 = S^z(O)/\chi^z(O)^{1/2} \,, \tag{16}$$

where $\chi^z(O)$ is the static susceptibility, $\langle S^z(O)\,|\,S^z(O)\rangle$,

$$\mathcal{H}_3 = (\mathcal{H} - \langle\langle\mathcal{H}\rangle\rangle)/(KT^2 C_H)^{1/2} \,, \tag{17}$$

where C_H is the specific heat at constant field

$$KT^2 C_H = \langle\mathcal{H} - \langle\langle\mathcal{H}\rangle\rangle\,|\,\mathcal{H} - \langle\langle\mathcal{H}\rangle\rangle\rangle \,.$$

We then have from Eq. (8)

$$\mathrm{Re}i\langle\tilde{M}^{\alpha\beta}(K)\,|\,(\omega + i\epsilon - L)^{-1}\,|\,\tilde{M}^{\mu\nu}(K)\rangle = \mathrm{Re}i\sum_i \langle\tilde{M}^{\alpha\beta}(K)\,|\,\mathcal{H}_i\rangle\langle\mathcal{H}_i\,|\,(\omega + i\epsilon - L)^{-1}\,|\,\mathcal{H}_i\rangle\langle\mathcal{H}_i\,|\,\tilde{M}^{\mu\nu}(K)\rangle$$

$$+ \mathrm{Re}i\sum_{i,j} \langle\tilde{M}^{\alpha\beta}(K)\,|\,R_i\rangle\langle R_i\,|\,(\omega + i\epsilon - L)^{-1}\,|\,R_j\rangle\langle R_j\,|\,\tilde{M}^{\mu\nu}(K)\rangle \,. \tag{18}$$

Cross terms of the form $\langle\mathcal{H}_i\,|\,(\omega + i\epsilon - L)^{-1}\,|\,R_j\rangle$ vanish since $L\,|\,\mathcal{H}_i\rangle = 0$ and $\langle\mathcal{H}_i\,|\,R_j\rangle = 0$, as do terms of the form

$$\langle\mathcal{H}_i\,|\,(\omega + i\epsilon - L^{-1}\,|\,\mathcal{H}_j\rangle \,, \quad i \neq j \,.$$

With $M^{\alpha\beta}(K)$ given by Eq. (7), the only nonvanishing coupling coefficient is, for $K = 0$,

$$\langle\tilde{M}^{\alpha\beta}(0)\,|\,\mathcal{H}_3\rangle = \sum_{\vec{q}'} P^{\alpha\beta}_{\vec{q}'}(\omega_1)\langle\vec{S}(\vec{q}')\cdot\vec{S}(-\vec{q}')\,|\,\mathcal{H}_3\rangle \,. \tag{19}$$

At high temperatures this is readily evaluated as

$$\beta^{1/2}\sum_{\vec{q}'} P^{\alpha\beta}_{\vec{q}'}(\omega_1) V(\vec{q}') \Big/ \sum_{q'} V(\vec{q}')^2 \,,$$

where in evaluating the coupling constant, we have neglected the spin-lattice interaction. There is, therefore, a term in $I(\omega)$ of the form

$$\mathrm{Re}i\langle\tilde{M}^{\alpha\beta}(0)\,|\,\mathcal{H}_3\rangle\langle\mathcal{H}_3\,|\,M^{\mu\nu}(0)\rangle\langle\mathcal{H}_3\,|\,(\omega + i\epsilon - L)^{-1}\,|\,\mathcal{H}_3\rangle \,. \tag{20}$$

Since $L\,|\,\mathcal{H}_3\rangle = 0$, this term has a pole at zero frequency, indicating that the correlation function in (12) does not vanish as $t \to \infty$. The physical origin of this singularity is that the perturbation produced by the light changes the total energy of the system [assuming that $\langle M^{\alpha\beta}(0)\,|\,\mathcal{H}_3\rangle \neq 0$], and since the perturbation cannot decay if we neglect the coupling to the lattice, the system relaxes to an equilibrium determined by the perturbed energy, not the original energy of the system.

This term in (18) actually vanishes, since in the absence of any fluctuations in the energy, there can be no absorption. It is the coupling to the lattice that produces these fluctuations, of course, that eventually return the system energy to its original value, and causes the correlation functions (12) to decay to zero as $t \to \infty$. The time scale for this to occur is by definition the spin-lattice relaxation time, and may depend upon the particular correlation function being considered. (It need not be the same for the magnetization and the energy, for instance.[6]) We shall account for these relaxation processes by phenomenologically including a relaxation frequency ν_L in the denominator in (20). We obtain, therefore, that the contribution to $I(\omega)$ due to the energy fluctuations is

$$I^{\alpha\beta,\mu\nu}(\omega)_E = 2\beta^{-1}\langle\tilde{M}^{\alpha\beta}(0)\,|\,\mathcal{H}_3\rangle\langle\mathcal{H}_3\,|\,\tilde{M}^{\mu\nu}(0)\rangle\nu_L/(\omega^2 + \nu_L^2). \tag{21}$$

There is, in addition the contribution coming from the second term on the right-hand side of (18), which does not contain any conserved operators, and which includes the scattering due to pairs of spin fluctuations. Whereas the characteristic frequency in (21) is ν_L, the characteristic frequency for this term will be

$$\omega_{ex} \simeq \left(\tfrac{2}{3}S(S+1)\sum_j V_{ij}^2\right)^{1/2} \,,$$

and for systems of greatest interest at this time $\omega_{ex} \gg \nu_L$. Thus the cross section for scattered light should appear schematically as in Fig. 1, assuming that the coupling coefficients do not vanish. We shall assume that if the coupling is allowed by symmetry, then it will actually exist. Fleury and Loudon[1] have investigated the symmetry of $P^{\alpha\beta}_{ij}(\omega_1)$ for tetragonal crystals of the perovskite structure and find that $\epsilon_1^\alpha P^{\alpha\beta}_{ij}(\omega_1)\epsilon_2^\beta$ can contain terms of the form

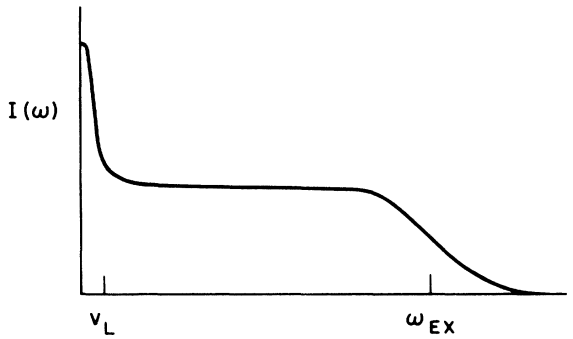

FIG. 1. Schematic light-scattering intensity in a polarization that allows coupling to energy-density fluctuations.

$$A_{ij}(\epsilon_1^x\epsilon_2^x+\epsilon_1^y\epsilon_2^y)+A_{ij}^1\epsilon_1^z\epsilon_2^z+B_{ij}(\epsilon_1^x\epsilon_2^y+\epsilon_2^x\epsilon_1^y)\sigma_{ij}^x\sigma_{ij}^y$$
$$+[x,z]+[y,z]+C_{ij}(\epsilon_1^x\epsilon_2^y-\epsilon_2^x\epsilon_1^y)\sigma_{ij}^x\sigma_{ij}^y$$
$$+[x,z]+[y,z]\,, \tag{22}$$

where $\sigma_{ij}^{\alpha}=\pm 1$ depending upon whether $r_{ij}/|r_{ij}|$ is positive or negative, and A_{ij}, B_{ij}, C_{ij}, etc., are coefficients that depend only on $|\vec{r}_{ij}|$, and were assumed to be nonvanishing only for nearest neighbors. From (19), using (22) we have

$$\langle \tilde{M}^{xx}(0)|\mathcal{H}_3\rangle=\beta^{1/2}\sum_j A_{ij}V_{ij}\Big/\sum_j V_{ij}^2\,,$$
$$\langle \tilde{M}^{xy}(0)|\mathcal{H}_3\rangle=\beta^{1/2}\sum_j B_{ij}\sigma_{ij}^x\sigma_{ij}^y V_{ij}\Big/\sum_j V_{ij}^2\,. \tag{23}$$

Since $\sum_j \sigma_{ij}^y\sigma_{ij}^x V_{ij}=0$, the energy-fluctuation scattering cannot be seen in the configuration in which the incident and scattered beams have crossed polarizations. It should, however, be visible when the polarizations are the same and can appear in the crossed polarized configuration if the Hamiltonian has lower symmetry. The relative intensity of the spin-fluctuation and energy-fluctuation scattering, in this configuration, depends upon the range of A_{ij}. If it extends only to nearest neighbors, the scattering is entirely energy fluctuations, and the larger the range, the larger the proportion of spin-fluctuation scattering. At infinite temperatures, we can calculate the ratio. The independent mode approximation holds [Eq. (10)] at $t=0$, so that the total intensity of the scattering is $[S(S+1)]^2\sum_2(A_{ij})^2$. The energy-fluctuation scattering is proportional to

$$[S(S+1)]^2\Big(\sum_j A_{ij}V_{ij}\Big)^2\Big/\sum_j (V_{ij})^2\,.$$

At finite temperatures, the intensity of the pole near $\omega=0$ is proportional to $|\langle M^{\alpha\alpha}(0)|\mathcal{H}\rangle|^2/KT^2C_H$, while the total intensity is proportional to $\langle M^{\alpha\alpha}(0)|M^{\alpha\alpha}(0)\rangle$. Assuming that $P_{ij}^{\alpha\alpha}$ is short-ranged, both terms should be proportional to the specific heat.

The relaxation times ν_L for $RbMnF_3$ and MnF_2 have been measured by Moran and Luthi, [7] in acoustic-attenuation experiments. They find $2\pi/\nu_L\simeq 3\times10^{-9}$ sec for MnF_2, $2\pi/\nu_L\simeq 3\times10^{-10}$ sec for $RbMnF_3$, near T_c. If the coefficients A_{ij} are comparable to B_{ij}, these widths ought to be resolvable with existing interferometers. (For the spectra in the cross-polarized configuration, see Ref. 1.)

It might be objected that the light scattering is not exactly at $K=0$. The above argument is only slightly modified in this case, since we can replace $|\mathcal{H}_3\rangle$ by $|\mathcal{H}_K\rangle$, where the operator corresponding to the state $|\mathcal{H}_K\rangle$,

$$\mathcal{H}_K=\frac{1}{2}\sum V(\vec{q})\vec{S}(\tfrac{1}{2}\vec{K}+\vec{q})\cdot\vec{S}(\tfrac{1}{2}\vec{K}-\vec{q})\,.$$

The light will couple to $\mathcal{H}_K$, which is not conserved, but decays with a lifetime given by D_TK^2, where D_T is the thermal diffusion coefficient. The remaining analysis is essentially unchanged; we simply replace ν_L by $\nu_L+D_TK^2$.

Below T_c, the conserved operators no longer correspond to orthogonal states, and in an antiferromagnet one must also include the staggered magnetization in the set of operators that correspond to states that are eigenfunctions of L with zero eigenvalue in the inner product appropriate to $T<T_c$. We can therefore choose as a basis, for an antiferromagnet,

$$|\mathcal{H}_1\rangle\sim\beta^{-1/2}I\,,$$
$$|\mathcal{H}_2\rangle\sim S^z(0)/[\chi^z(0)]^{1/2}\,,$$
$$|\mathcal{H}_3\rangle\sim[S^z(\vec{K}_0)-\langle S^z(\vec{K}_0)\rangle]/\chi^z(\vec{K}_0)\,,$$
$$|\mathcal{H}_4\rangle\sim(\mathcal{H}-\langle\mathcal{H}\rangle-\langle\mathcal{H}|\mathcal{H}_2\rangle\mathcal{H}_2-\langle\mathcal{H}|\mathcal{H}_3\rangle\mathcal{H}_3)/(KT^2C_1)^{1/2}\,.$$

Where

$$KT^2C^1=KT^2C_H+|\langle\mathcal{H}|\mathcal{H}_2\rangle|^2+|\langle\mathcal{H}|\mathcal{H}_3\rangle|^2\,.$$

In general, one can expect that $\langle\tilde{M}^{\alpha\beta}(0)|\mathcal{H}_i\rangle\neq 0$ for $i=2,3,4$ and hence the line observed near $\omega=0$ can have components with three different widths if the spin-lattice coupling is such that the decay times for the energy, staggered magnetization, and magnetization are different. The relative intensities will be determined by the appropriate coupling coefficients, which can be written as thermodynamic derivatives: $\langle M^{\alpha\beta}(0)|\mathcal{H}_3\rangle$, for instance, is $\propto\partial\langle\mathcal{H}\rangle/\partial H(K_0)$, where $H(\vec{K}_0)$ is a staggered field. We note that the coupling between the staggered magnetization and the energy has been observed in neutron scattering experiments[8] and computer simulations.[9] If the finite $\vec{K}$ value of the fluctuations is considered, additional relaxation times, due to magnetization and staggered magnetization diffusion as well as cross relaxation, enter. We will not consider these effects here. They can be described by a straightforward generalization of the previous results. We would also point out that below T_c there is also a contribution from the energy fluctuations in what has been called the one magnon scattering spectrum, i.e., that arises from the first term in (5b). This may also be calculated by analogous procedures, and will have a very different temperature dependence and selection rules since the coupling coefficient will be proportional to $\langle\vec{S}(0)\delta H\rangle$.

ACKNOWLEDGMENTS

I would like to thank H. Cummins for valuable conservations regarding this work, G. Potter for her assistance in preparing the manuscript, and M. Goldman and the University of Colorado for their hospitality.

[1]P. A. Fleury and R. Loudon, Phys. Rev. 166, 514 (1968).
[2]R. Loudon, J. Phys. C 3, 872 (1970).
[3]T. Morita, J. Appl. Phys. 39, 1042 (1968).
[4]R. J. Elliott, M. F. Thorpe, G. F. Imbusch, R. Loudon, and J. B. Parkinson, Phys. Rev. Lett. 21, 147 (1968).
[5]P. Richards and W. J. Brya, Phys. Rev. B 9, 3044 (1974).
[6]This was first pointed out to me by D. Huber.
[7]T. J. Moran and B. Lüthi, Phys. Rev. B 4, 122 (1974).
[8]M. P. Schulhof, R. Nathans, P. Heller, and A. Linz, Phys. Rev. B 4, 2254 (1971).
[9]D. L. Huber, AIP Conf. Proc. 5, 1261 (1971).