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Magnon-photon interaction in antiferromagnets

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The magnon-photon interaction is formulated for a direction of the magnetic vector of the electromagnetic wave perpendicular to the direction of the sublattice magnetization in an antiferromagnet, and from this the one-magnon self-energy is calculated. The linewidth predicted from the one-magnon one-photon process is in good agreement with the experimental value for MnF_2. Higher-order magnon-photon scattering processes are indicated and the corrections arising from such multiple magnon-photon scattering processes are found to depend on temperature and microwave power.

I. INTRODUCTION

The question of electromagnetic-radiation propagation in the medium of precessing magnetic moments has been raised by Kittel.[1] The predicted resonance has been discovered experimentally and has been well reviewed.[2] Efforts have been limited to the solution of Bloch's phenomenological macroscopic equation of motion of the magnetization with Maxwell's equations. Gintzberg[3] has drawn attention to the importance of the dispersion relation in ferromagnets, which has been subsequently studied[4-6] in antiferromagnets. In these works, the radiation is treated classically. It therefore appeared to us that perhaps useful information could be derived from a proper quantum-mechanical treatment. We felt that proper solutions of the Schrödinger equation with field quantization may be desirable. The present work has emerged from such a thought and the outcome is indeed interesting.

In the present paper, a magnon-photon interaction for an electromagnetic-wave magnetic vector perpendicular to the direction of the sublattice magnetization in an antiferromagnet is constructed and from this the self-energy is calculated up to second-order perturbation theory. The imaginary part of the self-energy determines the magnon relaxation time due to decay into photons. There are interesting dependences on the size of the crystal, the frequency, microwave power, and temperature. The predicted lowest-order contribution is in good agreement with the experimental measurement on MnF_2. Some higher-order processes are also predicted.

II. INTERACTION

In the normal configuration, the direction of the magnetic vector of the electromagnetic wave is perpendicular to the direction of the sublattice magnetization. Therefore the interaction may be chosen[7,8] in the form

$$\mathcal{H}' = \sum_{i,i+\delta} g\mu_B (h_{xi} S_{xi} + h_{x,i+\delta} S_{x,i+\delta}) , \qquad (1)$$

where h_x is the magnetic vector of the electromagnetic wave and the two sublattices of the antiferromagnet are located at i and $i+\delta$. The g value is the splitting factor and μ_B is the Bohr magneton. The magnetic vector field of the electromagnetic wave may be expanded in terms of creation ($c_q^\dagger$) and annihilation (c_q) operators of the photon field as

$$h_{xi} = \sum_{\vec{q}} f \left(\frac{2\pi\hbar\omega_{\vec{q}}}{L^3} \right)^{1/2} \times (c_{\vec{q}} e^{i\vec{q}\cdot\vec{r}_i} + c_{\vec{q}}^\dagger e^{-i\vec{q}\cdot\vec{r}_i}) , \qquad (2)$$

where f is a factor representing the standing-wave nature of the field in a rectangular wave guide[8]

$$f = (3\lambda^3 / 2\pi a b \lambda_g)^{1/2} \cos k_g l . \qquad (3)$$

Here a and b give the transverse dimensions of the wave guide, λ_g is the wavelength in the guide and λ that in free space. The distance l of the sample is measured from the end of the guide. The radiation wave vector k_g in the guide is determined from classical electrodynamics.

For the description of the spin waves, we resort to the Holstein-Primakoff representation recently applied by White *et al.*[9] to antiferromagnetic MnF_2. The magnon Hamiltonian is taken in the form

$$\mathcal{H}_m = \mathcal{H}_e + \mathcal{H}_a , \qquad (4a)$$

$$\mathcal{H}_e = -2|J_1| \sum_i \sum_\delta \vec{S}_i \cdot \vec{S}_{i+\delta} + 2J_2 \sum_i \sum_{\delta'} \vec{S}_i \cdot \vec{S}_{i+\delta'} , \qquad (4b)$$

$$\mathcal{H}_a = -\tfrac{1}{2} K \left(\sum_i S_{zi}^2 + \sum_j S_{zj}^2 \right) . \qquad (4c)$$

The Holstein-Primakoff representation of the spin operators in terms of site variables is given by

$$\begin{aligned} &S_i^+=(2S)^{1/2}f_i a_i\,, \quad S_j^+=(2S)^{1/2}b_j^\dagger f_j\,, \\ &S_i^-=(2S)^{1/2}a_i^\dagger f_i\,, \quad S_j^-=(2S)^{1/2}f_j b_j\,, \\ &S_{zi}=S-a_i^\dagger a_i\,, \quad S_{zj}=-S+b_j^\dagger b_j\,, \\ &f_i=(1-a_i^\dagger a_i/2S)^{1/2}\,. \end{aligned} \tag{5}$$

Introducing the Fourier transforms of the site variables, we have

$$\begin{aligned} a_i &= N^{-1/2}\sum_{\vec{k}} a_{\vec{k}} \exp(i\vec{k}\cdot\vec{r}_i)\,, \\ b_j &= N^{-1/2}\sum_{\vec{k}} b_{\vec{k}} \exp(-i\vec{k}\cdot\vec{r}_j)\,, \end{aligned} \tag{6}$$

and the Bogoliubov transformation

$$\begin{aligned} a_{\vec{k}} &= u_{\vec{k}}\alpha_{\vec{k}}^\dagger - v_{\vec{k}}\beta_{\vec{k}}\,, \\ b_{\vec{k}} &= -v_{\vec{k}}\alpha_{\vec{k}} + u_{\vec{k}}\beta_{\vec{k}}^\dagger\,, \end{aligned} \tag{7}$$

and following the usual procedure of eliminating the two-magnon number-nonconserving terms, one obtains

$$u_{\vec{k}}=[(A+\omega_{\vec{k}})/2\omega_{\vec{k}}]^{1/2}\,, \quad v_{\vec{k}}=[(A-\omega_{\vec{k}})/2\omega_{\vec{k}}]^{1/2}\,, \tag{8}$$

where

$$\hbar A = 2z_2J_2S+KS=\omega_e+\omega_a\,, \tag{9a}$$

$$\omega_k=[(2\omega_e+\omega_a)\omega_a+\omega_e^2(1-\gamma_k^2)]^{1/2}\,, \tag{9b}$$

$$\gamma_{\vec{k}}=z_\delta^{-1}\sum e^{i\vec{k}\cdot\vec{\delta}}. \tag{9c}$$

The lowest-order magnon Hamiltonian then appears in the diagonal form as

$$\mathcal{H}_0=\sum_{\vec{k}}\hbar\omega_{\vec{k}}\alpha_{\vec{k}}^\dagger\alpha_{\vec{k}}+\sum_{\vec{k}}\hbar\omega_{\vec{k}}\beta_{\vec{k}}^\dagger\beta_{\vec{k}}\,,$$

if there is no external magnetic field. However, the Zeeman interaction

$$\mathcal{H}_z=-g\mu_BH_0\Big(\sum_i S_{zi}^a+\sum_{i+\delta}S_{z,i+\delta}^b\Big) \tag{10a}$$

is exactly diagonalizable and for this reason need not be carried through the transformation, although the transformation procedure will be equally valid. Its eigenvalues are simply $\pm\hbar\gamma H_0$, so it simply splits into two branches and thus (10a) can just be absorbed in the unperturbed Hamiltonian

$$\mathcal{H}_0=\sum_{\vec{k}}\hbar\Omega_{\vec{k}}^+\alpha_{\vec{k}}^\dagger\alpha_{\vec{k}}+\sum_{\vec{k}}\hbar\Omega_{\vec{k}}^-\beta_{\vec{k}}^\dagger\beta_{\vec{k}}\,, \tag{10b}$$

with

$$\Omega_{\vec{k}}^\pm=\omega_{\vec{k}}\pm\gamma H_0\,. \tag{10c}$$

There are also scattering terms as given by White *et al.*,[9] but they are of no immediate interest for the present paper. If we make the substitution $\omega_a/\omega_e=\Delta'$, results (8) become

$$u_{\vec{k}}=\left(\frac{(1+\Delta')+[(2+\Delta')\Delta'+(1-\gamma_{\vec{k}}^2)]^{1/2}}{2[(2+\Delta')\Delta'+(1-\gamma_{\vec{k}}^2)]^{1/2}}\right)^{1/2}, \tag{11a}$$

$$v_{\vec{k}}=\left(\frac{(1+\Delta')-[(2+\Delta')\Delta'+(1-\gamma_{\vec{k}}^2)]^{1/2}}{2[(2+\Delta')\Delta'+(1-\gamma_{\vec{k}}^2)]^{1/2}}\right)^{1/2}. \tag{11b}$$

There is an alternative way of expressing the anisotropy. Instead of (4c) we may write

$$\mathcal{H}_a=-g\mu_BH_A\Big(\sum_i S_{zi}+\sum_j S_{zj}\Big)\,. \tag{12}$$

Since $\frac{1}{2}K\langle S_z\rangle=g\mu_BH_A$, the two methods are almost equivalent. It should be noted that in either case the anisotropy appears to depend on temperature outside of spin-wave theory. Ideally the temperature dependence should arise only from the magnon-scattering terms and the coupling constants should be treated as independent of temperature. If (12) is chosen, the form of the functions obtained through the Bogoliubov transformation becomes[10]

$$u_{\vec{k}}=\left(\frac{(1+\Delta)+[(1+\Delta)^2-\gamma_{\vec{k}}^2]^{1/2}}{2[(1+\Delta)^2-\gamma_{\vec{k}}^2]^{1/2}}\right)^{1/2}, \tag{13a}$$

$$v_{\vec{k}}=\left(\frac{(1+\Delta)-[(1+\Delta)^2-\gamma_{\vec{k}}^2]^{1/2}}{2[(1+\Delta)^2-\gamma_{\vec{k}}^2]^{1/2}}\right)^{1/2}, \tag{13b}$$

where $\Delta=g\mu_BH_A/|J|Sz$, if only the nearest-neighbor exchange is considered. Substituting (5) and (6) in succession and (2) in (1) our magnon-photon interaction has the form

$$\begin{aligned} \mathcal{H}' = {} & (2SN)^{1/2}\sum_{kq}A_q g\mu_B[a_kc_q\delta(k+q)+a_kc_q^\dagger\delta(k-q)+b_kc_q\delta(k+q)+b_kc_q^\dagger\delta(k-q)+\text{H.c.}] \\ & +\sum_{k_1k_2k_3q}[2(2SN)^{1/2}]^{-1}[a_{k_1}^\dagger a_{k_2}a_{k_3}c_q\delta(k_1-k_2-k_3-q)+a_{k_1}^\dagger a_{k_2}a_{k_3}c_q^\dagger\delta(q+k_1-k_2-k_3)+a_{k_1}^\dagger a_{k_2}^\dagger a_{k_3}c_q\delta(k_1+k_2-k_3-q) \\ & +a_{k_1}^\dagger a_{k_2}^\dagger a_{k_3}c_q^\dagger\delta(k_3-k_1-k_2-q)+b_{k_1}^\dagger b_{k_2}b_{k_3}c_{-q}\delta(k_1-k_2-k_3+q) \\ & +b_{k_1}^\dagger b_{k_2}b_{k_3}c_{-q}^\dagger\delta(k_1-k_2-k_3-q)+b_{k_1}^\dagger b_{k_2}^\dagger b_{k_3}c_{-q}\delta(k_1+k_2-k_3+q) \\ & +b_{k_1}^\dagger b_{k_2}^\dagger b_{k_3}c_{-q}^\dagger\delta(k_1+k_2-k_3-q)]\,. \end{aligned} \tag{14}$$

In this interaction the magnons are not in the proper form. Therefore we substitute (7) in (14) to obtain the correct magnon description. The results of this calculation can be grouped as follows:

$$\mathcal{H}' = \mathcal{H}_1' + \mathcal{H}_2' + \mathcal{H}_3' + \mathcal{H}_4' , \tag{15}$$

$$\mathcal{H}_1' = (2SN)^{1/2} g\mu_B \sum_{kq} A_q(u_k - v_k)[\alpha_k c_q \delta(k+q) + \alpha_k c_q^\dagger \delta(k-q) + \beta_k c_q \delta(k+q) + \beta_k c_q^\dagger \delta(k-q) + \text{H.c.}] , \tag{16}$$

$$\begin{aligned}\mathcal{H}_2' = \sum_{qk_1k_2k_3} g\mu_B A_q [& (u_{k_1}u_{k_2}u_{k_3} - v_{k_1}v_{k_2}v_{k_3})(\alpha_{k_1}^\dagger \alpha_{k_2}\alpha_{k_3} c_q^\dagger + \beta_{k_1}\beta_{k_2}^\dagger \beta_{k_3}^\dagger c_q)\,\delta(q+k_1-k_2-k_3) \\ & + (u_{k_1}u_{k_2}u_{k_3} - v_{k_1}v_{k_2}v_{k_3})(\alpha_{k_1}^\dagger \alpha_{k_2}^\dagger \alpha_{k_3} c_q + \beta_{k_1}\beta_{k_2}\beta_{k_3}^\dagger c_q^\dagger)\,\delta(k_1+k_2-k_3-q) \\ & + (v_{k_1}u_{k_2}v_{k_3} - u_{k_1}v_{k_2}u_{k_3})(\alpha_{k_1}^\dagger \beta_{k_2}^\dagger \alpha_{k_3} c_q + \beta_{k_1}\alpha_{k_2}\beta_{k_3}^\dagger c_q^\dagger)\,\delta(k_1+k_2-k_3-q) \\ & + (v_{k_1}v_{k_2}u_{k_3} - u_{k_1}u_{k_2}v_{k_3})(\beta_{k_1}\beta_{k_2}^\dagger \alpha_{k_3} c_q^\dagger + \alpha_{k_1}^\dagger \alpha_{k_2}\beta_{k_3}^\dagger c_q)\,\delta(k_1+k_3-k_2-q) \\ & + (u_{k_1}v_{k_2}v_{k_3} - v_{k_1}u_{k_2}u_{k_3})(\beta_{k_1}\alpha_{k_2}^\dagger \alpha_{k_3} c_q^\dagger + \alpha_{k_1}^\dagger \beta_{k_2}\beta_{k_3}^\dagger c_q)\,\delta(k_1+k_3-k_2-q) \\ & + (v_{k_1}u_{k_2}v_{k_3} - u_{k_1}v_{k_2}u_{k_3})(\alpha_{k_1}^\dagger \beta_{k_2}\alpha_{k_3} c_q^\dagger + \beta_{k_1}\alpha_{k_2}^\dagger \beta_{k_3}^\dagger c_q)\,\delta(k_1+q-k_2-k_3)] . \end{aligned} \tag{17}$$

$$\begin{aligned}\mathcal{H}_3' = \sum_{qk_1k_2k_3} g\mu_B A_q \{ & (u_{k_1}u_{k_2}u_{k_3} - v_{k_1}v_{k_2}v_{k_3})[(\alpha_{k_1}^\dagger \alpha_{k_2}\alpha_{k_3} c_q + \beta_{k_1}\beta_{k_2}^\dagger \beta_{k_3}^\dagger c_q^\dagger)\,\delta(k_1-k_2-k_3-q) \\ & \qquad + (\alpha_{k_1}^\dagger \alpha_{k_2}^\dagger \alpha_{k_3} c_q^\dagger + \beta_{k_1}\beta_{k_2}\beta_{k_3}^\dagger c_q)\delta(k_3-k_1-k_2-q)] \\ & + (v_{k_1}u_{k_2}v_{k_3} - u_{k_1}v_{k_2}u_{k_3})[(\alpha_{k_1}^\dagger \beta_{k_2}\alpha_{k_3} c_q + \beta_{k_1}\alpha_{k_2}^\dagger \beta_{k_3}^\dagger c_q^\dagger)\delta(k_1-k_2-k_3-q) \\ & \qquad + (\alpha_{k_1}^\dagger \beta_{k_2}^\dagger \alpha_{k_3} c_q^\dagger + \beta_{k_1}\alpha_{k_2}\beta_{k_3}^\dagger c_q)\delta(k_3-k_1-k_2-q)] \\ & + (u_{k_1}v_{k_2}v_{k_3} - v_{k_1}u_{k_2}u_{k_3})[(\beta_{k_1}\alpha_{k_2}\alpha_{k_3} c_q^\dagger + \alpha_{k_1}^\dagger \beta_{k_2}^\dagger \beta_{k_3}^\dagger c_q)\delta(q-k_1-k_2-k_3) \\ & \qquad + (\beta_{k_1}\alpha_{k_2}^\dagger \alpha_{k_3} c_q + \alpha_{k_1}^\dagger \beta_{k_2}\beta_{k_3}^\dagger c_q^\dagger)\,\delta(k_2-k_1-k_3-q)] \\ & + (v_{k_1}v_{k_2}u_{k_3} - u_{k_1}u_{k_2}v_{k_3})[(\beta_{k_1}\beta_{k_2}^\dagger \alpha_{k_3} c_q + \alpha_{k_1}^\dagger \alpha_{k_2}\beta_{k_3}^\dagger c_q^\dagger)\,\delta(k_2-k_1-k_3-q) \\ & \qquad + (\beta_{k_1}\beta_{k_2}\alpha_{k_3} c_q^\dagger + \alpha_{k_1}^\dagger \alpha_{k_2}^\dagger \beta_{k_3}^\dagger c_q)\delta(q-k_1-k_2-k_3)]\} , \end{aligned} \tag{18}$$

$$\begin{aligned}\mathcal{H}_4' = \sum_{qk_1k_2k_3} A_q g\mu_B [& (u_{k_1}v_{k_2}v_{k_3} - v_{k_1}u_{k_2}u_{k_3})(\beta_{k_1}\alpha_{k_2}\alpha_{k_3} c_q + \alpha_{k_1}^\dagger \beta_{k_2}^\dagger \beta_{k_3}^\dagger c_q^\dagger) \\ & + (v_{k_1}v_{k_2}u_{k_3} - u_{k_1}u_{k_2}v_{k_3})(\beta_{k_1}\beta_{k_2}\alpha_{k_3} c_q + \alpha_{k_1}^\dagger \alpha_{k_2}^\dagger \beta_{k_3}^\dagger c_q^\dagger)]\delta(q+k_1+k_2+k_3) , \end{aligned} \tag{19}$$

$$\mathcal{H}_0' = \sum_k \hbar\Omega_k^+ \alpha_k^\dagger \alpha_k + \sum_k \hbar\Omega_k^- \beta_k^\dagger \beta_k + \sum_q \hbar\omega_q c_q^\dagger c_q . \tag{20}$$

This last expression defines the unperturbed Hamiltonian of the system, and $A_q = (2\pi\hbar\omega_q/L^3)^{1/2} f$.

III. LIFETIME

From the quasiparticle number-conserving terms of (16) we obtain the second-order contribution to the system energy as

$$\Sigma_s^{(1)} = \sum_{kq} \frac{2SNg^2\mu_B^2 A_q^2(u_k - v_k)^2(n_k - N_q)\delta(k-q)}{\hbar\omega_q - \Omega_k} . \tag{21}$$

The one-magnon self-energy is just $\Sigma_k = \partial\Sigma_s/\partial n_k$ so that this corresponds to the relaxation time of the uniform magnon,

$$1/\tau_1 = (2\pi/\hbar)2SNg^2\mu_B^2 f^2(u_{10} - v_{10})^2\omega_0^3(\pi c^3)^{-1} , \tag{22}$$

which is just the imaginary part of the self-energy as $1/\tau = (-2/\hbar)\,\text{Im}\Sigma_k$. Here u_{10} and v_{10} are the values of u_k and v_k, respectively, evaluated at resonance for $k=0$. The resonance value of the radiation wave vector is taken to be $q_0 = \omega_0/c$. We replace SN by the appropriate magnetization in a given volume through the boundary conditions and change the units from inverse time to Gauss; using (3) the width from (22) is then

$$\Delta H = (2\pi^2)(4\pi M_s)V\left(\frac{\omega}{c}\right)^3\left(\frac{3\lambda^3}{2\pi ab\lambda_g}\right)\cos^2 k_g l\left(\frac{\Delta}{2}\right)^{1/2} . \tag{23}$$

If (13) is used the required factor is $(u_{10} - v_{10})^2 = (\frac{1}{2}\Delta)^{1/2}$. If instead (11) is employed the factor is $(\frac{1}{2}\Delta')^{1/2}$. In the former case[10,11] if we use $J = -2.45\ \text{cm}^{-1}$, $S = \frac{5}{2}$, $z = 8$, $g\mu_B H_A = 0.8\ \text{cm}^{-1}$, so that $|J|Sz \simeq 50\ \text{cm}^{-1}$, the anisotropy parameter is $(\frac{1}{2}\Delta)^{1/2} = 0.09$ and there is a happy coincidence with the factor $H_A/H_C \simeq 0.09$ used by Sanders *et al.*.[12]

However, there is no way of obtaining H_C in our calculation since this is not a parameter of the Hamiltonian. The critical field H_C at which the spin flop occurs and the ordered antiferromagnet changes into the flopped phase with canted spins, is defined in a complicated way by Keffer.[2] If we use (11), then the required factor is $(\frac{1}{2}\Delta')^{1/2} \simeq 0.10$ for the parameters of White *et al.*[9] So the two results differ only by about 10%. If we take the approximate expression

$$H_C = (2H_a H_e + H_a^2)^{1/2}$$

then

$$H_a/H_C = [\Delta'/(2+\Delta')]^{1/2} \simeq (\tfrac{1}{2}\Delta')^{1/2} ,$$

and once again agreement results. The small difference in the estimates probably occurs owing to the choice of numerical values for the anisotropy. White *et al.* give $J_1 = 2.29$ cm^{-1}, $J_2 = 1.26$ cm^{-1}, and the anisotropy energy $K = 0.3$ cm^{-1} at $T = 4$ K. However, the later depends on temperature[13] as remarked before, $K(T)/K(0) \simeq [M(T)/M(0)]^3$. Therefore a small difference in the numerical estimates is very likely. If we compare our result with that of Sanders *et al.* a factor of $\frac{4}{3}$, which they have, does not appear to arise in our calculation. So for the choice of parameters appropriate to MnF_2, the radiative linewidth for $\cos^2 k_z l = 1$ is 13.5 G whereas the experimental linewidth is 12.7 G. In view of the uncertainty in the data and the measured volume of the MnF_2 crystal, 1.77 mm^3, the difference of 0.8 G between our calculated value and that measured by Sanders *et al.* is understandable. Sanders *et al.* actually calculate a width of 19 G, which is probably masked either by some error or by the choice of parameters. For the values which they take, the width from their formula is calculated to be 18.06 G. So it appears that errors of the order of 5% can easily arise.

Our method of calculation treats the radiation problem at par with the spin waves. Instead, the method of Sanders *et al.*[12] can not achieve this correspondence. Besides, the advantage of our method of calculation is that higher-order radiation processes are predicted. Although While *et al.* claim to understand the linewidth of the antiferromagnetic resonance in MnF_2 above $T = 5$ K, it appears to us that the region near and below 4 K is not well understood. At such temperatures the thermal scattering of magnons is considerably suppressed and, as noted from the experiment of Sanders *et al.*, the radiation process becomes important. A detailed study of the temperature dependence of the antiferromagnetic resonance in MnF_2 below 4 K does not appear to have been published. However, our interactions predict a temperature-dependent radiative linewidth. At this stage it may be thought that these terms are small and the calculation is more or less of academic interest. We therefore limit ourselves only to the largest of the terms in (17) which conserve the number of quasiparticles in the system and exclude the mode-mixing contribution which is small. The largest of the remaining terms then give the following contribution to the one-magnon self-energy,

$$\Sigma_k^{(2)} = \sum_{qk_1k_2k_3} 4g^2\mu_B^2 A_q^2 \Bigg[(u_{k_1}u_{k_2}u_{k_3} - v_{k_1}v_{k_2}v_{k_3})^2 \Bigg(\frac{n_{k_2}N_q + n_{k_1}n_{k_2} - n_{k_1}N_q - n_{k_2}}{\Omega_{k_1} - \Omega_{k_2} - \Omega_{k_3} + \hbar\omega_q} \, \delta(k_1 + q - k_2 - k_3) + \frac{(n_{k_2} + n_{k_1} + 1)N_q - n_{k_1}n_{k_2}}{\Omega_{k_1} + \Omega_{k_2} - \Omega_{k_3} - \hbar\omega_q} \, \delta(k_1 + k_2 - k_3 - q) \Bigg) \Bigg] , \tag{24}$$

the imaginary part of which gives a contribution to the lifetime of the magnon of,

$$\frac{1}{T_1'} = \frac{2\pi}{\hbar} \sum_{qk_1k_2k_3} 4g^2\mu_B^2 A_q^2 ((u_{k_1}u_{k_2}u_{k_3} - v_{k_1}v_{k_2}v_{k_3})^2 \{ (n_{k_2}N_q + n_{k_1}n_{k_2} - n_{k_1}N_q - n_{k_2}) \delta(k_1 + q - k_2 - k_3) \times \delta(\Omega_{k_1} - \Omega_{k_2} - \Omega_{k_3} + \hbar\omega_q) + [(n_{k_2} + n_{k_1} + 1)N_q - n_{k_1}n_{k_2}] \times \delta(k_1 + k_2 - k_3 - q) \delta(\Omega_{k_1} + \Omega_{k_2} - \Omega_{k_3} - \hbar\omega_q) \}) \tag{25}$$

which has a factor like $M_s T^{5/2}[\exp(\hbar\omega_0/k_B T) - 1]$. At 4 K, this gives a 10% correction to (22). The $\cos^2 k_z l$ factor is again present, which has been verified experimentally.[12] The result (22) is independent of the temperature and of the photon density N_q, and hence of the rf power. However (25) depends on the temperature and on the number of photons present in the system.

IV. CONCLUSIONS

From a second-quantized approach we have calculated a radiative antiferromagnetic-resonance linewidth in which one of the terms is in accord with the experimental measurements on MnF_2. Some additional magnon-photon scattering processes are predicted. This kind of radiative effect has not been calculated previously in the literature. Our Hamiltonian is new, and interesting effects may be associated with it. Its pertinence in connection with the super-radiance problem in a magnetically ordered system is rather obvious.

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APPENDIX: CLASSICAL CALCULATION

It is of interest to trace the origin of the factor of $\frac{4}{3}$ in the classical calculation of the radiative linewidth. The power radiated classically by a precessing dipole moment $\mu_\perp$, as written by Sanders *et al.*,[12] is

$$P_r = \tfrac{2}{3}ck^4\mu_\perp^2, \tag{A1}$$

whereas the power absorbed is

$$P_a = \tfrac{1}{2}\omega\chi'' Vh_1^2, \tag{A2}$$

where k is the radiation wave vector, c is the velocity of light, ω is the frequency, V is the volume, h_1 is the amplitude of the magnetic vector field of the electromagnetic wave, and χ'' is the susceptibility function

$$\chi'' = \gamma^2 M\Delta H/[(\omega-\omega_0)^2+(\Delta H)^2], \tag{A3}$$

where at resonance, $\omega=\omega_0$,

$$\chi'' = M/\Delta H; \tag{A4}$$

ΔH is the phenomenological linewidth at resonance. We define the dynamic susceptibility as

$$\chi'' = \mu_\perp/Vh_1 \tag{A5}$$

so that from (A4) and (A5),

$$\mu_\perp = Vh_1M/\Delta H. \tag{A6}$$

If all the power is radiated at resonance, then

$$P_r = P_a \tag{A7}$$

leading to $\frac{1}{2}\omega(M/\Delta H)Vh_1^2 = \frac{2}{3}ck^4(Vh_1M/\Delta H)^2$ and we obtain, for the width,

$$\Delta H = \tfrac{4}{3}\,VM\omega^3/c^3, \tag{A8}$$

so that the factor of $\frac{4}{3}$ in the width arises from the factor $\frac{2}{3}$ in the classical radiation formula (A1) and the factor $\frac{1}{2}$ in the classical absorbed power as given by (A2). Actually the expressions (A3) and (A7) are not good since all the absorbed power need not be radiated at resonance nor all the linewidth be radiative. There are several factors causing the error in the calculation. (i) Part of the power absorbed may be lost in terms of nonradiative relaxation mechanisms such as those considered by White *et al.*, invalidating (A7). (ii) Some of the part of the power absorbed is radiated electromagnetically but at frequencies away from the antiferromagnetic resonance. Thus, by considering that all the power is radiated at the antiferromagnetic resonance, one over estimates the radiative linewidth. (iii) In the expression (A3) ΔH represents the total width, not just the radiative contribution. (iv) In the quantum-mechanical treatment presented in the present paper, there is the momentum-conservation requirement which means that in the lowest order the emitted radiation is anisotropic being practically in the same direction as the original incident photon. On the other hand, in the classical result, the emitted radiation goes in all directions and thus one has a factor of the form $\int\int d(\cos\theta)\,d\phi$. The classical radiation formula is

$$\frac{dP}{d\Omega} = \frac{1}{8\pi}ck^4|p|^2\sin^2\psi.$$

If we put $p\sin\psi=\mu_\perp$, where ψ is the precession angle, and assume that the $\cos\psi$ component does not radiate, then

$$P = \frac{1}{8\pi}ck^4\mu_\perp^2\int\int d(\cos\theta)\,d\phi.$$

Since $\int d(\cos\theta)=2$, $\int d\phi = 2\pi$,

$$P=\tfrac{1}{2}ck^4\mu_\perp^2. \tag{A9}$$

Comparing with (A1) as written by Sanders *et al.*,[12] we see that one should have a factor of $\frac{1}{2}$ rather than of $\frac{2}{3}$. If (A9) is used rather than (A1) the factor of $\frac{4}{3}$ in (A8) disappears. The error is thus traced to the interpretation of the precession of the dipole moment. Sanders *et al.* have perhaps not realized that the angle ψ is fixed and is not the same as θ. If we allow ψ to be variable, identical with θ, then we obtain the integral $\int d\phi\int\sin^2\theta \times d(\cos\theta)=\frac{8}{3}\pi$ leading to $\frac{1}{3}$ not $\frac{2}{3}$ in (A1). The classical result (A8) will then have $\frac{8}{3}$ instead of $\frac{4}{3}$. However ψ is uniquely determined at resonance, $\cos\psi = H_0/\mu$, and we need not consider the nonresonant precession of the moment around the field. Classically, a continuum is emitted and a continuum is absorbed. This follows if we obtain the classical Bohr limit by setting $\hbar\to 0$. However, quantum mechanically the photons must be resonant for any radiation to be absorbed. The quantum-mechanical treatment presented by us is free of the difficulties encountered in the classical treatment and predicts multiple magnon-photon processes. It is expected that only the one-photon one-magnon process will give a contribution smaller than some exact results. Alternatively, the self-energies are normalized, and since in the quantum treatment there are several terms, only the lowest-order term will give a smaller contribution than will result upon summing all the possible diagrams. In any case, the classical treatment[12] is incorrect. Since the Compton wavelength of the photon is infinite, the radiation field should be treated quantum mechanically.

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