

Excitation Spectrum in Singlet-Triplet Ferromagnetic Systems*

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The dynamic properties of a single-triplet ferromagnetic system are studied by the method of double-time Green's functions. Utilizing the standard basis operator formalism to describe the singlet-triplet levels, a Dyson equation for the single-particle Green's function is obtained. This equation describes the magnetic excitations in the system for low temperatures. As an application, the self-energy is calculated approximately to yield the damping of the excitations at zero temperature.

I. INTRODUCTION

In recent years there has been considerable interest in magnetic systems in which the single-ion crystal field ground state is a singlet. In such systems, if the exchange interaction is strong enough, magnetic ordering at low temperature occurs through a polarization process. A review of this subject has been given by Fulde and Peschel⁽¹⁾. The magnetic excitations in such systems are believed to be single-ion crystal field transitions propagating through the crystal by virtue of the exchange coupling. Of particular interest is the singlet-triplet system where the first excited state is a triplet. The spherical symmetry of the singlet-triplet system can be explored to derive an exact hydrodynamic theory for the long wavelength excitations. This has been done by Hohenberg and Swift⁽²⁾, by Cheung^(3,4) and by Huber⁽⁵⁾. On the other hand, microscopic calculations for such systems have not been very successful. Because of the calculational difficulties associated with crystal field effects, past studies have been confined to the random phase approximation (RPA). For example, Hsieh and Blume⁽⁶⁾ have calculated the excitation spectrum for the singlet-triplet system and systems with more complicated level schemes have been studied by Peschel et al.⁽⁷⁾ and by Holden and Buyers⁽⁸⁾.

In this paper we develop a theory for the low temperature dynamic properties of the singlet-triplet ferromagnetic system. In order to gain a better understanding of the transitions between the various energy levels, we utilize the standard basis operator formalism introduced by Haley and Erdős⁽⁹⁾. This is different from previous treatments which represent the levels by pseudo-spin operators. The low lying excitations in the system are then studied by the double time Green's function method^(10,11). The equations of motion are written in the form of a Dyson equation for the

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Green's functions. As an application, we calculate the lifetime of the excitations at zero temperature.

In Sec. II we introduce the standard basis operators and express the Hamiltonian in terms of them. In the ferromagnetic state, the presence of the molecular field mixes the singlet-triplet levels. The molecular field levels are obtained by diagonalizing the "molecular field Hamiltonian" and we take them as our new basis states. In Sec. III a Dyson equation for the single-particle Green's function which describes the low lying excitations at low temperatures is derived. In Sec. IV we show that the simplest approximation for the self energy reproduced the RPA result obtained previously by other methods. In Sec. V we calculate approximately the contribution of the two-particle Green's functions to the self energy, which leads to damping of the excitation.

II. HAMILTONIAN

The Hamiltonian for the singlet-triplet system is taken to be of the form

$$\mathcal{H} = \sum_i V^{ic} - \sum_{i,j} \mathcal{J}_{ij} \mathbf{J}^i \cdot \mathbf{J}^j. \quad (2.1)$$

Here V^{ic} is the crystal field Hamiltonian associated with the i th ion and gives rise to a splitting A between the singlet and the triplet. \mathcal{J}_{ij} is the exchange interaction coupling ions i and j , \mathbf{J}^i is the angular momentum of the i th ion. We denote the ground singlet by $|1\rangle$ and the excited triplet by $|2\rangle, |3\rangle, |4\rangle$. The matrix elements of J_z between the levels are⁽⁶⁾

$$\begin{aligned} J_{z21} &= \langle 2 | J_z | 1 \rangle = \alpha, \\ J_{z33} &= \langle 3 | J_z | 3 \rangle = \beta, \\ J_{z44} &= \langle 4 | J_z | 4 \rangle = -\beta, \end{aligned} \quad (2.2)$$

with similar expressions for J_+ and J_- .

The standard basis operator $\hat{L}_{\alpha\beta}^i$ associated with the α and β levels of V^{ic} is interpreted as taking the i th ion from level β to level α . These operators obey the commutation relation

$$[\hat{L}_{\alpha\alpha}^i, \hat{L}_{\beta\beta}^i] = \delta^{ij} (\delta_{\alpha\beta} \hat{L}_{\alpha\beta}^i - \delta_{\beta\alpha} \hat{L}_{\beta\alpha}^i). \quad (2.3)$$

The operator $\hat{L}_{\alpha\alpha}$ measures the probability that the energy level α is occupied. Its ensemble average will be denoted by D_α and satisfies the obvious sum rule

$$\sum_\alpha D_\alpha = 1. \quad (2.4)$$

We note that any operator can be represented as a linear combination of the standard basis operators by the formula

$$O^i = \sum_{\alpha,\beta} \langle \alpha | O | \beta \rangle \hat{L}_{\alpha\beta}^i. \quad (2.5)$$

Using the matrix elements of J between the singlet-triplet levels, we express the angular momentum operator in terms of the standard basis operators

$$\begin{aligned} J_z^i &= \alpha(\hat{L}_{11}^i + \hat{L}_{11}^i) + \beta(\hat{L}_{11}^i - \hat{L}_{11}^i), \\ J_+^i &= \sqrt{2} \alpha(\hat{L}_{11}^i - \hat{L}_{11}^i) + \sqrt{2} \beta(\hat{L}_{11}^i + \hat{L}_{11}^i), \\ J_-^i &= \sqrt{2} \alpha(\hat{L}_{11}^i - \hat{L}_{11}^i) + \sqrt{2} \beta(\hat{L}_{11}^i + \hat{L}_{11}^i). \end{aligned} \quad (2.6)$$

The Hamiltonian now takes the general form

$$\mathcal{H} = \sum_{i,\alpha} \epsilon_\alpha \hat{L}_{\alpha\alpha}^i - \sum_{i,\alpha\alpha'} \sum_{j,\beta\beta'} I_{\alpha\alpha',\beta\beta'}^i \hat{L}_{\alpha\alpha'}^i \hat{L}_{\beta\beta'}^j, \quad (2.7)$$

where $\epsilon_1=0, \epsilon_2=\epsilon_3=\epsilon_4=\Delta$ and $I_{\alpha\alpha',\beta\beta'}^i$ is given by

$$I_{\alpha\alpha',\beta\beta'}^i = \mathcal{J}^{ij} (J_{z\alpha\alpha'} J_{z\beta\beta'} + \frac{1}{2} J_{+\alpha\alpha'} J_{-\beta\beta'} + \frac{1}{2} J_{-\alpha\alpha'} J_{+\beta\beta'}) \quad (2.8)$$

In the ferromagnetic state, each ion is subjected to a field which is generated by the remaining ions. This field which is referred to as the molecular field points in the z direction and mixes the

singlet-triplet levels due to nonzero matrix elements of J_z between them. To investigate this effect we introduce new basis states

$$\begin{aligned} |1'\rangle &= u_1 |1\rangle + v_1 |2\rangle, \\ |2'\rangle &= u_2 |1\rangle + v_2 |2\rangle, \end{aligned} \quad (2.9)$$

in terms of which the angular momentum operators are

$$\begin{aligned} J'_z &= \alpha(u_1 v_1 + u_2 v_2) (\hat{L}'_{1z} + \hat{L}'_{2z}) - 2\alpha u_1 v_1 \hat{L}'_{1z} - 2\alpha u_2 v_2 \hat{L}'_{2z} + \beta(\hat{L}'_{1z} - \hat{L}'_{2z}), \\ J'_+ &= \sqrt{2} [(\beta u_1 - \alpha v_1) \hat{L}'_{1+} + (\beta u_2 + \alpha v_2) \hat{L}'_{2+} - (\beta u_1 - \alpha v_1) \hat{L}'_{1-} - (\beta u_2 + \alpha v_2) \hat{L}'_{2-}], \\ J'_- &= \sqrt{2} [(\beta u_1 - \alpha v_1) \hat{L}'_{1-} + (\beta u_2 + \alpha v_2) \hat{L}'_{2-} - (\beta u_1 - \alpha v_1) \hat{L}'_{1+} - (\beta u_2 + \alpha v_2) \hat{L}'_{2+}], \end{aligned} \quad (2.10)$$

and the Hamiltonian takes the form

$$\mathcal{H} = \sum_{i,\alpha} \epsilon_\alpha \hat{L}'_{\alpha\alpha} - \sum_{i,\alpha\alpha'} h_{\alpha\alpha'} \hat{L}'_{\alpha\alpha'} - \sum_{i,\alpha\alpha'} \sum_{j,\beta\beta'} J'_{\alpha\alpha',\beta\beta'} \hat{L}'_{\alpha\alpha'} \hat{L}'_{\beta\beta'}, \quad (2.11)$$

where from now on we shall drop the primes indicating the new states. The coupling constants are rather complicated and are given in the appendix.

The "molecular field Hamiltonian" is obtained from (2.11) by replacing $\hat{L}'_{\beta\beta'}$ in the third term on the right hand side by its thermal average $\delta_{\beta\beta'} D_\beta$

$$\mathcal{H}_{MF} = \sum_{i,\alpha} \epsilon_\alpha \hat{L}'_{\alpha\alpha} - \sum_{i,\alpha\alpha'} h_{\alpha\alpha'} \hat{L}'_{\alpha\alpha'} - \sum_{i,\alpha\alpha'} C_{\alpha\alpha'} \hat{L}'_{\alpha\alpha'}, \quad (2.12)$$

where

$$\begin{aligned} C_{\alpha\alpha'} &= 2 \sum_{j,r} J'_{\alpha\alpha',rr} D_r, \\ &= 2 \sum_{j,r} I'_{\alpha\alpha',rr} D_r, \end{aligned} \quad (2.13)$$

$I'_{\alpha\alpha',rr}$ denotes the Fourier transform in space of $J'_{\alpha\alpha',rr}$. After some algebra, we find that the "molecular field Hamiltonian" can be diagonalized by the following choices of u_i and v_i

$$u_i = \frac{-C}{\sqrt{E_i + C^2}}, \quad v_i = \frac{E_i}{\sqrt{E_i + C^2}} \quad i=1,2 \quad (2.14)$$

with

$$E_1 = \frac{1}{2} (\Delta - \sqrt{\Delta^2 + 4C^2}), \quad (2.15)$$

$$E_2 = \frac{1}{2} (\Delta + \sqrt{\Delta^2 + 4C^2}), \quad (2.16)$$

and

$$C = 2\alpha \mathcal{G}(O)M. \quad (2.17)$$

$\mathcal{G}(\mathbf{k})$ is the Fourier transform in space of \mathcal{G}_{ij} . The magnetization \mathbf{M} is the nonzero thermal average of the z component of the angular momentum operator and is given implicitly by the equation

$$M = \frac{2\alpha C}{\sqrt{\Delta^2 + 4C^2}} D_{1z} + \beta D_{3z}. \quad (2.18)$$

In the above formula $D_{\alpha\beta}$ denotes $D_\alpha - D_\beta$. The molecular field levels are $E_1, E_2, E_3 = \mathbf{A} \cdot \mathbf{d}$ and $E_4 = \Delta + d$ with $d = 2\beta \mathcal{G}(O)M$ respectively.

Finally, the Hamiltonian in the new representation takes the form

$$\mathcal{H} = \sum_{i,\alpha} E_\alpha \hat{L}'_{\alpha\alpha} - \sum_{i,\alpha\alpha'} \sum_{j,\beta\beta'} J'_{\alpha\alpha',\beta\beta'} \hat{L}'_{\alpha\alpha'} \hat{L}'_{\beta\beta'}, \quad (2.19)$$

with the understanding that the "molecular field" term has been subtracted from the second term on the right hand side.

The molecular field ground state depends on which energy level is lowest at zero temperature.

If E_3 is lowest we have the case of fully magnetized ground state with magnetization $M=\beta$. On the other hand if E_1 is lowest the magnetization is not saturated but given by

$$M = \frac{[16\alpha^4 g^2(O) - \Delta^2]^{1/2}}{4\alpha g(O)}. \quad (2.20)$$

which situation actually occurs depends on the choice of parameters $\alpha, \beta, g(O)$ and A . We refer to the paper of Hsieh and Blume⁽⁶⁾ for a detailed discussion. In the following we will be mainly interested in the case of unsaturated magnetic ground state. The other case will be commented on briefly.

III. EQUATIONS OF MOTION OF GREEN' S FUNCTIONS

In this section we apply the method of double time Green' s functions to the system. For arbitrary operators A and B , the retarded Green' s function is defined by

$$G^{AB}(t-t') \equiv \langle\langle A(t) | B(t') \rangle\rangle = -i\theta(t-t') \langle[A(t), B(t')]\rangle. \quad (3.1)$$

Its temporal Fourier transform

$$G^{AB}(w) = \langle\langle A | B \rangle\rangle_w = \int_{-\infty}^{\infty} dt e^{i\omega(t-t')} \langle\langle A(t) | B(t') \rangle\rangle, \quad (3.2)$$

satisfies the equation of motion

$$wG^{AB}(w) = \langle[A, B]\rangle + \langle\langle[A, \mathcal{H}] | B \rangle\rangle_w. \quad (3.3)$$

For the case we are considering E_1 is the lowest level and is predominately occupied at very low temperatures. An examination of (2. 6) shows the transverse magnetic excitations at low temperatures are chiefly due to transitions between E_1 and E_4 and between E_1 and E_3 . Accordingly we introduce a column vector

$$A^k = \begin{pmatrix} \hat{L}_{13}^k \\ \hat{L}_{41}^k \end{pmatrix}, \quad (3.4)$$

and its conjugate row vector

$$A^{k+} = (\hat{L}_{31}^{-k} \hat{L}_{41}^{-k}), \quad (3.5)$$

where k is the momentum vector in reciprocal space and $\hat{L}_{13}^k = 1/\sqrt{N} \sum_i \hat{L}_{13}^i e^{-i\mathbf{k}\cdot\mathbf{r}_i}$, etc. The Green' s function to be studied form a 2×2 matrix which is represented symbolically as

$$Q(\mathbf{k}, w) = \langle\langle A^k | A^{k+} \rangle\rangle_w = \begin{pmatrix} \langle\langle \hat{L}_{13}^k | \hat{L}_{31}^{-k} \rangle\rangle_w & \langle\langle \hat{L}_{13}^k | \hat{L}_{41}^{-k} \rangle\rangle_w \\ \langle\langle \hat{L}_{41}^k | \hat{L}_{31}^{-k} \rangle\rangle_w & \langle\langle \hat{L}_{41}^k | \hat{L}_{41}^{-k} \rangle\rangle_w \end{pmatrix} \quad (3.6)$$

The equations of motion of the Green' s functions are determined by the commutator of the operators \hat{L}_{13}^k and \hat{L}_{41}^k with the Hamiltonian. Using the commutation relation (2. 3) we obtain

$$[\hat{L}_{13}^k, \mathcal{H}] = (E_3 - E_1) \hat{L}_{13}^k + F_1^k, \quad (3.7)$$

$$[\hat{L}_{41}^k, \mathcal{H}] = (E_1 - E_4) \hat{L}_{41}^k + F_4^k, \quad (3.8)$$

where

$$F_1^k = -2 \sum_{rr', q} I_{1v, rr'}^q \hat{L}_{1v}^q \hat{L}_{rr'}^{k-q} + 2 \sum_{rr', q} I_{11, rr}^q \hat{L}_{13}^q \hat{L}_{rr'}^{k-q}, \quad (3.9)$$

$$F_4^k = -2 \sum_{rr', q} I_{4v, rr'}^q \hat{L}_{4v}^q \hat{L}_{rr'}^{k-q} + 2 \sum_{rr', q} I_{44, rr}^q \hat{L}_{41}^q \hat{L}_{rr'}^{k-q}, \quad (3.10)$$

The thermal average of the commutator $[A^k, A^{k+}]$ is simply

$$Q = \begin{pmatrix} D_{13} & 0 \\ 0 & D_{41} \end{pmatrix}. \quad (3.11)$$

The equation of motion of the Green' s function can now be written as

$$Q^{-1} \begin{pmatrix} w+d - \frac{A}{2} - \frac{1}{2} \sqrt{\Delta^2 + 4c^2} & 0 \\ 0 & w+d + \frac{A}{2} + \frac{1}{2} \sqrt{\Delta^2 + 4c^2} \end{pmatrix} Q(\mathbf{k}, w) = I + \mathcal{F}(\mathbf{k}, w), \quad (3.12)$$

where Z is the unit matrix and Q^{-1} is the matrix inverse to Q . $\mathcal{F}(\mathbf{k}, w)$ is a higher-order 2x2 Green' s function

$$\mathcal{F}(\mathbf{k}, w) = Q^{-1} \begin{pmatrix} \langle\langle F_1^{\mathbf{k}} | \hat{L}_{11}^{-\mathbf{k}} \rangle\rangle_w & \langle\langle F_1^{\mathbf{k}} | \hat{L}_{14}^{-\mathbf{k}} \rangle\rangle_w \\ \langle\langle F_1^{\mathbf{k}} | \hat{L}_{11}^{-\mathbf{k}} \rangle\rangle_w & \langle\langle F_1^{\mathbf{k}} | \hat{L}_{14}^{-\mathbf{k}} \rangle\rangle_w \end{pmatrix}. \quad (3.13)$$

If the term $\mathcal{F}(\mathbf{k}, w)$ is neglected completely, (3.12) can be inverted to obtain

$$Q_0(\mathbf{k}, w) = \begin{pmatrix} E+d - \frac{\Delta}{2} - \frac{1}{2} \sqrt{\Delta^2 + 4c^2} & 0 \\ 0 & E+d + \frac{\Delta}{2} + \frac{1}{2} \sqrt{\Delta^2 + 4c^2} \end{pmatrix} \quad (3.14)$$

which is just the molecular field result.

In order to go beyond the molecular field approximation an equation of motion for the higher order Green' s function $\mathcal{F}(\mathbf{k}, w)$ must be obtained. To do this we take the commutator of the second argument of $\mathcal{F}(\mathbf{k}, w)$ with the Hamiltonian. We obtain

$$\mathcal{F}(\mathbf{k}, w) Q_0^{-1}(\mathbf{k}, w) = P(\mathbf{k}, w), \quad (3.15)$$

where the polarization operator on the right hand side is the sum of two terms

$$P(\mathbf{k}, w) = P_0(\mathbf{k}) + P_1(\mathbf{k}, w), \quad (3.16)$$

the first term is independent of w and is given by

$$P_0(\mathbf{k}) = Q^{-1} \begin{pmatrix} \langle [F_1^{\mathbf{k}}, \hat{L}_{11}^{-\mathbf{k}}] \rangle & \langle [F_1^{\mathbf{k}}, \hat{L}_{14}^{-\mathbf{k}}] \rangle \\ \langle [F_1^{\mathbf{k}}, \hat{L}_{11}^{-\mathbf{k}}] \rangle & \langle [F_1^{\mathbf{k}}, \hat{L}_{14}^{-\mathbf{k}}] \rangle \end{pmatrix} Q^{-1}. \quad (3.17)$$

The second term is a still higher-order Green' s function,

$$P_1(\mathbf{k}, w) = Q^{-1} \begin{pmatrix} \langle\langle F_1^{\mathbf{k}} | F_1^{+\mathbf{k}} \rangle\rangle_w & \langle\langle F_1^{\mathbf{k}} | F_1^{+\mathbf{k}} \rangle\rangle_w \\ \langle\langle F_1^{\mathbf{k}} | F_1^{+\mathbf{k}} \rangle\rangle_w & \langle\langle F_1^{\mathbf{k}} | F_1^{+\mathbf{k}} \rangle\rangle_w \end{pmatrix} Q^{-1}. \quad (3.18)$$

Combining (3.15) with the equation of motion for the one-particle Green' s function

$$Q_0^{-1}(\mathbf{k}, w) Q(\mathbf{k}, w) = I + \mathcal{F}(\mathbf{k}, w), \quad (3.12')$$

gives

$$Q(\mathbf{k}, w) = Q_0(\mathbf{k}, w) + Q_0(\mathbf{k}, w) P(\mathbf{k}, w) Q_0(\mathbf{k}, w). \quad (3.19)$$

If we introduce a self-energy operator defined by

$$\Sigma(\mathbf{k}, w) = P(\mathbf{k}, w) [I + Q_0(\mathbf{k}, w) P(\mathbf{k}, w)]^{-1}, \quad (3.20)$$

then a Dyson equation for the single-particle Green' s function

$$[Q_0^{-1}(\mathbf{k}, w) - \Sigma(\mathbf{k}, w)] Q(\mathbf{k}, w) = I, \quad (3.21)$$

is obtained. Our problem is now to calculate the self-energy operator $\Sigma(\mathbf{k}, w)$.

IV. RANDOM PHASE APPROXIMATION

The simplest approximation is to consider the term $P_0(\mathbf{k})$ only as it is independent of frequency. We further approximate the self-energy by

$$\Sigma(\mathbf{k}, \omega) \cong P(\mathbf{k}, \omega) \cong P_0(\mathbf{k}). \quad (4.1)$$

To evaluate the thermal averages of the commutation in (3.17), it is best to calculate it first in real space. We obtain in a straight-forward manner

$$\begin{aligned} \langle [F'_i, \hat{L}'_{ij}] \rangle = & -2 \sum I''_{i', r'} \{ \delta^{ij} \delta_{r'} \langle \hat{L}'_{i'} \hat{L}'_{r'} \rangle - \delta^{ij} \delta_{r'} \langle \hat{L}'_{i'} \hat{L}'_{r'} \rangle \\ & + \delta^{ij} \delta_{i'} \langle \hat{L}'_{i'} \hat{L}'_{r'} \rangle - \delta^{ij} \delta_{i'} \langle \hat{L}'_{i'} \hat{L}'_{r'} \rangle \}, \end{aligned} \quad (4.2)$$

with similar expressions for the other commutators. We next factorize the thermal averages of the products of two operators into products of thermal averages of single operators. Keeping in mind that the molecular field term has already been subtracted out, we obtain

$$\langle [F'_i, \hat{L}'_{ij}] \rangle = -2D_{i3} I''_{i', i}, \quad (4.3)$$

$$\langle [F'_i, \hat{L}'_{ij}] \rangle = -2D_{i3} D_{i1} I''_{i', i}, \quad (4.4)$$

$$\langle [F'_i, \hat{L}'_{ij}] \rangle = -2D_{i3} D_{i1} I''_{i', i}, \quad (4.5)$$

$$\langle [F'_i, \hat{L}'_{ij}] \rangle = -2D_{i3} I''_{i', i}. \quad (4.6)$$

The various I 's are given in the appendix.

Substituting these values in (3.21), we obtain the following expression for the single-particle Green's function

$$G^{-1}(\mathbf{k}, \omega) = \begin{pmatrix} D_{i3}^{-1} \left(\omega + d - \frac{\Delta}{2} - \frac{1}{2} \sqrt{\Delta^2 + 4c^2} \right) + 2I''_{i', i} & 2I''_{i', i} \\ 2I''_{i', i} & D_{i1}^{-1} \left(\omega + d + \frac{\Delta}{2} + \frac{1}{2} \sqrt{\Delta^2 + 4c^2} \right) + 2I''_{i', i} \end{pmatrix} \quad (4.7)$$

This is essentially the same as that obtained by Hsieh and Blume⁽⁶⁾ using pseudospin operators. The poles of the above Green's function can only be found explicitly at zero temperature. At absolute zero $D_{i3}=1$, $D_{i1}=-1$ and using the value of M in (2.20) we find after some algebra that the two poles of the Green's function are located at

$$\begin{aligned} R_{1,2}(k) = & -\beta \left(4\alpha^2 g^2(O) - \frac{\Delta^2}{4\alpha^2} \right)^{1/2} \left(1 - \frac{g(k)}{g(O)} \right) \pm \frac{1}{2} \left\{ [\Delta + 4g(O)\alpha^2]^2 \right. \\ & \left. - \frac{g(k)}{g(O)} \left[16\alpha^2 \beta^2 g^2(O) - \left(\frac{\beta}{\alpha} \right)^2 \Delta^2 \right] \right\}^{1/2} \left(1 - \frac{g(k)}{g(O)} \right)^{1/2}. \end{aligned} \quad (4.8)$$

The Green's functions then take the following forms at absolute zero

$$\langle \langle \hat{L}'_{ij} | \hat{L}'_{ij} \rangle \rangle_{\omega} = \frac{B_1(k)}{\omega - R_1(k)} + \frac{B_2(k)}{\omega - R_2(k)}, \quad (4.9)$$

where

$$B_1(k) = \frac{1}{R_1(k) - R_2(k)} \left[R_1(k) + d + \frac{\Delta}{2} + \frac{1}{2} \sqrt{\Delta^2 + 4c^2} - 2I''_{i', i} \right], \quad (4.10)$$

$$B_2(k) = \frac{-1}{R_1(k) - R_2(k)} \left[R_2(k) + d + \frac{\Delta}{2} + \frac{1}{2} \sqrt{\Delta^2 + 4c^2} - 2I''_{i', i} \right], \quad (4.11)$$

and

$$\langle \langle \hat{L}'_{ij} | \hat{L}'_{ij} \rangle \rangle_{\omega} = \frac{C_1(k)}{\omega - R_1(k)} + \frac{C_2(k)}{\omega - R_2(k)}, \quad (4.12)$$

where

$$C_1(k) = \frac{-1}{R_1(k) - R_2(k)} \left[R_1(k) + d - \frac{\Delta}{2} - \frac{1}{2} \sqrt{\Delta^2 + 4c^2} + 2I''_{i', i} \right], \quad (4.13)$$

$$C_2(k) = \frac{1}{R_1(k) - R_2(k)} \left[R_2(k) + d - \frac{\Delta}{2} - \frac{1}{2} \sqrt{\Delta^2 + 4c^2} + 2I''_{i', i} \right]. \quad (4.14)$$

c and d in the above formulas are evaluated at absolute zero.

It is interesting to examine the behaviour of the poles at small k . For a crystal of cubic symmetry, the following relation holds near $k=0$

$$\mathcal{G}(k) = \mathcal{G}(O) (1 - \eta k^2 \delta^2 \dots), \quad (4.15)$$

η depends on the geometry of the crystal and δ is the lattice constant. Using the above expression we found a linear spectrum for small k

$$R_{1,2}(k) = \pm \sqrt{\eta} \delta \left[\left(1 + \frac{\beta^2}{\alpha^2}\right) \Delta^2 + \frac{1}{2} \Delta \mathcal{G}(O) \alpha^2 - 16\alpha^2(\beta^2 - \alpha^2) \mathcal{G}^2(O) \right]^{1/2} k + O(k^2). \quad (4.16)$$

We note that one of the poles is positive and the other is negative indicating that the higher levels are populated even at absolute zero. This shows that RPA is inconsistent and has to do with the molecular field ground state being not the true ground state of the system^(2,6).

Another branch of the magnetic excitations which involves transitions from the lowest level is the longitudinal excitations which are the poles of the Green's function $\langle\langle \hat{L}_{11}^k | \hat{L}_{11}^{-k} \rangle\rangle_w$. Similar considerations give the following for the Green's function at absolute zero

$$\langle\langle \hat{L}_{11}^k | \hat{L}_{11}^{-k} \rangle\rangle_w = \frac{w^4 - 4\alpha^2 \mathcal{G}(O) + \frac{1}{8} \Delta^2 \mathcal{G}(k)}{w^2 - 16\alpha^4 \mathcal{G}^2(O) + \Delta^2 \mathcal{G}(O)} \frac{\mathcal{G}(k)}{\mathcal{G}(O)} \quad (4.17)$$

The longitudinal excitations have a gap at zero momentum.

To calculate the spectrum at finite temperatures, we have to know the occupation of the levels. This can be obtained from the Green's functions using the formula⁽⁹⁾

$$D_\beta = \langle \hat{L}_{\beta\alpha}^t \hat{L}_{\alpha\beta}^t \rangle = \langle \hat{L}_{\beta\beta}^t \rangle, \quad (4.18)$$

and the spectrum can be determined numerically by iteration.

V. DAMPING OF THE EXCITATIONS

In this section, we consider the effect of the frequency dependent contribution to the polarization operator on the excitation spectrum. $P_1(\mathbf{k}, w)$ has a branch cut along the real axis. As such, it contributes an imaginary as well as a real part to the single-particle Green's function. Both parts are second order in the interaction. We shall be interested only in the imaginary part as it is the leading order contribution to the damping.

It is a rather formidable task to calculate the two-particle Green's functions involved in $P_1(\mathbf{k}, w)$. We shall make the simplest approximation by decoupling the two-particle Green's functions into products of single-particle Green's functions. For example, we decouple in the following manner

$$\begin{aligned} \langle \hat{L}_{\alpha\beta}^t(t) \hat{L}_{\gamma\delta}^t(t) \hat{L}_{\alpha'\beta'}^{t'}(t') \hat{L}_{\gamma'\delta'}^{t'}(t') \rangle &= \langle \hat{L}_{\alpha\beta}^t(t) \hat{L}_{\alpha'\beta'}^{t'}(t') \rangle \langle \hat{L}_{\gamma'\delta'}^{t'}(t') \rangle \\ &+ \langle \hat{L}_{\alpha\beta}^t(t) \hat{L}_{\gamma'\delta'}^{t'}(t') \rangle \langle \hat{L}_{\gamma\delta}^t(t) \hat{L}_{\alpha'\beta'}^{t'}(t') \rangle \end{aligned} \quad (5.1)$$

We shall further neglect correlation functions like $\langle \hat{L}_{11}^t(t) \hat{L}_{11}^{t'}(t') \rangle$, $\langle \hat{L}_{11}^t(t) \hat{L}_{11}^{t'}(t') \rangle$ and $\langle \hat{L}_{11}^t(t) \hat{L}_{11}^{t'}(t') \rangle$ as well as nondiagonal ones as they are small compared with those we keep. An examination of the coupling constants enables us to obtain

$$\begin{aligned} \langle\langle F_1^k(t) | F_1^{k+}(t') \rangle\rangle &= -i\theta(t-t') \{ 4 \sum_{\mathbf{q}} (I_{11,11}^{\mathbf{q}} - I_{11,11}^{\mathbf{q}}) (I_{11,11}^{\mathbf{q}} - I_{11,11}^{\mathbf{q}} + I_{11,11}^{\mathbf{q}}) \langle \hat{L}_{11}^{k-\mathbf{q}}(t) \hat{L}_{11}^{-k+\mathbf{q}}(t') \rangle \langle \hat{L}_{11}^{\mathbf{q}}(t) \hat{L}_{11}^{-\mathbf{q}}(t') \rangle \\ &+ 4 \sum_{\mathbf{q}} I_{11,11}^{\mathbf{q}} (I_{11,11}^{\mathbf{q}} + I_{11,11}^{\mathbf{q}} - I_{11,11}^{\mathbf{q}} - I_{11,11}^{\mathbf{q}}) \langle \hat{L}_{11}^{k-\mathbf{q}}(t) \hat{L}_{11}^{-k+\mathbf{q}}(t') \rangle \langle \hat{L}_{11}^{\mathbf{q}}(t) \hat{L}_{11}^{-\mathbf{q}}(t') \rangle \}, \end{aligned} \quad (5.2)$$

and

$$\begin{aligned} \langle\langle F_1^k(t) | F_1^{k+}(t') \rangle\rangle &= i\theta(t-t') \{ 4 \sum_{\mathbf{q}} (I_{11,11}^{\mathbf{q}} - I_{11,11}^{\mathbf{q}}) (I_{11,11}^{\mathbf{q}} - I_{11,11}^{\mathbf{q}} - I_{11,11}^{\mathbf{q}}) \langle \hat{L}_{11}^{-k+\mathbf{q}}(t) \hat{L}_{11}^{k-\mathbf{q}}(t') \rangle \langle \hat{L}_{11}^{\mathbf{q}}(t) \hat{L}_{11}^{-\mathbf{q}}(t') \rangle \\ &- 4 \sum_{\mathbf{q}} I_{11,11}^{\mathbf{q}} (I_{11,11}^{\mathbf{q}} - I_{11,11}^{\mathbf{q}} - I_{11,11}^{\mathbf{q}} - I_{11,11}^{\mathbf{q}}) \langle \hat{L}_{11}^{-k+\mathbf{q}}(t) \hat{L}_{11}^{k-\mathbf{q}}(t') \rangle \langle \hat{L}_{11}^{\mathbf{q}}(t) \hat{L}_{11}^{-\mathbf{q}}(t') \rangle \}. \end{aligned} \quad (5.3)$$

The off-diagonal elements of $P_1(\mathbf{k}, \omega)$ can be neglected as their contribution to the damping is of higher order.

To evaluate the above Green's functions, we introduce the following spectral functions

$$J_{1\alpha}(\mathbf{q}, \omega) = \frac{-2 I_m \langle \hat{L}_{1\alpha}^{\uparrow} | \hat{L}_{1\alpha}^{-\uparrow} \rangle_{\omega+i\epsilon}}{e^{\omega/\theta} - 1}, \quad (5.4)$$

and

$$J_{\alpha 1}(\mathbf{q}, \omega) = \frac{-2 I_m \langle \hat{L}_{1\alpha}^{\uparrow} | \hat{L}_{1\alpha}^{-\uparrow} \rangle_{\omega+i\epsilon}}{e^{\omega/\theta} - 1}, \quad (5.5)$$

$\alpha=2, 3, 4$ and $\theta=k_B T$ with k_B the Boltzmann constant. In terms of the spectral functions, the correlation functions are

$$\langle \hat{L}_{1\alpha}^{\uparrow}(t) \hat{L}_{1\alpha}^{-\uparrow}(t') \rangle = \int_{-\infty}^{\infty} \frac{d\omega}{2\pi} J_{1\alpha}(\mathbf{q}, \omega) e^{\omega t/\theta} e^{-i\omega(t-t')}, \quad (5.6)$$

and

$$\langle \hat{L}_{1\alpha}^{-\uparrow}(t) \hat{L}_{1\alpha}^{\uparrow}(t') \rangle = \int_{-\infty}^{\infty} \frac{d\omega}{2\pi} J_{\alpha 1}(\mathbf{q}, \omega) e^{-i\omega(t-t')}. \quad (5.7)$$

Employing (5.6) and (5.7), we obtain

$$\begin{aligned} I_m \langle F_1^{\uparrow} | F_1^{\uparrow+} \rangle_{\omega+i\epsilon} = & -4\pi \left\{ \sum_{\mathbf{q}} (I_{11,11}^{\uparrow} - I_{11,11}^{\downarrow}) (I_{11,11}^{\uparrow} - I_{11,11}^{\downarrow} + I_{11,11}^{\uparrow\downarrow}) \right. \\ & \cdot \int \frac{d\omega'}{2\pi} \int \frac{d\omega''}{2\pi} J_{11}(\mathbf{k}-\mathbf{q}, \omega') J_{11}(\mathbf{q}, \omega'') e^{\omega'+\omega''/\theta} \delta(\omega-\omega'-\omega'') \\ & + \sum_{\mathbf{q}} I_{11,11}^{\uparrow\downarrow} (I_{11,11}^{\uparrow} + I_{11,11}^{\downarrow} - I_{11,11}^{\uparrow\downarrow}) \\ & \left. \cdot \int \frac{d\omega'}{2\pi} \int \frac{d\omega''}{2\pi} J_{11}(\mathbf{k}-\mathbf{q}, \omega') J_{11}(\mathbf{q}, \omega'') e^{\omega'+\omega''/\theta} \delta(\omega-\omega'-\omega'') \right\}, \quad (5.8) \end{aligned}$$

and

$$\begin{aligned} I_m \langle F_1^{\downarrow} | F_1^{\downarrow+} \rangle_{\omega+i\epsilon} = & 4\pi \left\{ \sum_{\mathbf{q}} (I_{11,11}^{\downarrow} - I_{11,11}^{\uparrow}) (I_{11,11}^{\downarrow} - I_{11,11}^{\uparrow} - I_{11,11}^{\uparrow\downarrow}) \right. \\ & \cdot \int \frac{d\omega'}{2\pi} \int \frac{d\omega''}{2\pi} J_{11}(\mathbf{k}-\mathbf{q}, \omega') J_{11}(\mathbf{q}, \omega'') \delta(\omega-\omega'-\omega'') \\ & + \sum_{\mathbf{q}} I_{11,11}^{\uparrow\downarrow} (I_{11,11}^{\downarrow} - I_{11,11}^{\uparrow} + I_{11,11}^{\uparrow\downarrow}) \\ & \left. \cdot \int \frac{d\omega'}{2\pi} \int \frac{d\omega''}{2\pi} J_{11}(\mathbf{q}, \omega') J_{11}(\mathbf{k}-\mathbf{q}, \omega'') \delta(\omega-\omega'-\omega'') \right\}. \quad (5.9) \end{aligned}$$

At absolute zero the first expression is nonzero only for $\omega > 0$ while the second is nonzero only for $\omega < 0$. Since the spectrum of $\langle \hat{L}_{11}^{\uparrow} | \hat{L}_{11}^{-\uparrow} \rangle_{\omega}$ and $\langle \hat{L}_{11}^{\downarrow} | \hat{L}_{11}^{-\downarrow} \rangle_{\omega}$ contains a gap, the delta functions in the above formulas are satisfied only for large values of k . Using these expressions in (3.21), the damping of the positive pole is proportional to (5.8) and is finite only for $R_1(k) > [16g^2(O)\alpha^4 - \Delta^2]^{1/2}$ while the damping of the negative pole is proportional to (5.9) and is finite only for $|R_1(k)| > [16g^2(O)\alpha^4 - \Delta^2]^{1/2}$.

VI. CONCLUSION

We have studied the singlet-triplet ferromagnetic system using double-time Green's functions through a Dyson equation approach. The imaginary part of the self energy to second order in the interaction is calculated approximately to obtain the damping of the excitations. The damping turns out to be zero for small values of k . This is somewhat in agreement with the hydrodynamic theory which predicts a damping proportional to k^2 .⁽⁴⁾ Of course, at zero temperature, hydrodynamic region ceases to exist.

For the case where the magnetic ground state is saturated, similar calculations can be carried out. However, one of the low lying excitations shows a gap and the other has a quadratic spectrum.

APPENDIX

$I_{\alpha\beta, \gamma\delta}^{ij}$ have the following symmetry properties

$$I_{\alpha\beta, \gamma\delta}^{ij} = I_{\gamma\delta, \alpha\beta}^{ij} = I_{\beta\alpha, \gamma\delta}^{ij} = I_{\beta\alpha, \delta\gamma}^{ij} \quad (\text{A. 1})$$

We list here a few typical I^{ij} 's

$$I_{11, 11}^{ij} = -2\alpha^2 u_2 v_2 (u_1 v_2 + u_2 v_2) \mathcal{I}_{ij} \quad (\text{A. 2})$$

$$I_{11, 12}^{ij} = \alpha\beta (u_1 v_2 + u_2 v_1) \mathcal{I}_{ij} \quad (\text{A. 3})$$

$$I_{12, 11}^{ij} = -\alpha\beta (u_1 v_2 + u_2 v_1) \mathcal{I}_{ij} \quad (\text{A. 4})$$

$$I_{11, 13}^{ij} = -(\beta u_1 + \alpha v_1) (\beta u_2 + \alpha v_2) \mathcal{I}_{ij} \quad (\text{A. 5})$$

$$I_{11, 14}^{ij} = (\beta u_2 + \alpha v_2)^2 \mathcal{I}_{ij} \quad (\text{A. 6})$$

$$I_{11, 41}^{ij} = (\beta u_2 + \alpha v_2) (\beta u_2 - \alpha v_2) \mathcal{I}_{ij} \quad (\text{A. 7})$$

$$I_{11, 44}^{ij} = (\beta u_2 - \alpha v_2)^2 \mathcal{I}_{ij} \quad (\text{A. 8})$$

$$I_{12, 41}^{ij} = -(\beta u_1 - \alpha v_1) (\beta u_2 - \alpha v_2) \mathcal{I}_{ij} \quad (\text{A. 9})$$

u_i and v_i , $i=1, 2$ are given by (2.14).