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DYSON EQUATION FOR SINGLET-TRIPLET FERROMAGNET

By E. Szczepaniak

Solid State Theory Division, Institute of Physics, A. Mickiewicz University, Poznań*

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The Dyson equation for the singlet-triplet ferromagnet is derived using irreducible double-time temperature Green functions. In the lowest order approximation the results are in agreement with those of the spectral density method, which improves the RPA results.

1. Introduction

In this paper, we concentrate on the singlet-triplet system. Examples of such systems are the rare-earth compounds, which have an A_1 singlet ground state and a first excited T_1 triplet state (e.g. TmN [1], TbSb [2], and Pr_3Tl [3]). The Green function method with random phase approximation (RPA) has been applied to these systems by Hsieh and Blume [4] and by Hsieh [5]. The application of the spectral density method (SDM) [6] to such systems [7, 8] permitted the consideration of two-site correlations.

Here, we propose a derivation of the Dyson equation for the ferromagnetic singlet-triplet system and discuss the resulting equations in the lowest order approximation. We show that SMD is the best method in this order. We use the formalism of irreducible double-time temperature Green functions, previously applied by Plakida to the Heisenberg model [9] and to magnets with spin-phonon coupling [10, 11], and by Micnas and Kowalewski [12] to the Ising model in a transverse field.

2. Dyson equation

The effective spin Hamiltonian for the singlet-triplet system takes the form [4]

$$\mathscr{H} = \Delta \sum_i \vec{S}_i \cdot \vec{T}_i - \sum_{ij} J_{ij}(a\vec{S}_i + b\vec{T}_i) \cdot (a\vec{S}_j + b\vec{T}_j), \tag{1}$$

where S and T are spin-1/2 operators, Δ is the energy gap between the singlet and triplet state, J_{ij} is the effective exchange interaction between ions i and j, and a, b are constants of the model. E.g., we have $a = (1+2\sqrt{14})/2$ and $b = (1-2\sqrt{14})/2$ for Tb^{3+}, or Tm^{3+}, in a octahedral crystalline field [4].

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* Address: Zakład Teorii Ciała Stałego, Instytut Fizyki, Uniwersytet A. Mickiewicza, Matejki 48/49 60-769 Poznań, Poland.

We define the temperature double-time retarded Green functions [13, 14] in terms of the operators

$$\check{B}_q = \begin{pmatrix} S_{-q}^- \\ T_{-q}^- \end{pmatrix}, \quad \check{B}_q^+ = (S_q^+, T_q^+), \tag{2}$$

as follows:

$$\check{G}_q(t, t') = -i\theta(t-t')\langle[\check{B}_q(t), \check{B}_q^+(t')]\rangle = \langle\langle \check{B}_q(t)|\check{B}_q^+(t')\rangle\rangle, \tag{3}$$

where

$$S_q^\alpha = \frac{1}{\sqrt{N}}\sum_i S_i^\alpha e^{iqr_i},$$

$$T_q^\alpha = \frac{1}{\sqrt{N}}\sum_i T_i^\alpha e^{iqr_i}, \quad \alpha = +, -, z., \tag{4}$$

The Fourier time transform is defined by

$$\check{G}_q(t, t') = \int_{-\infty}^{\infty} e^{-i\omega(t-t')}\check{G}_q(\omega)d\omega. \tag{5}$$

The aim of this paper is to derive an exact expression for the matrix Green function $\check{G}_q(\omega)$ analogous to the Dyson equation. We use the formalism of irreducible Green functions. We consider only a ferromagnetic phase where $\langle S^z\rangle \neq 0$ and $\langle T^z\rangle \neq 0$.

The equation of motion for $\check{G}_q(\omega)$ is of the form

$$\omega\check{G}_q(\omega) = -\frac{1}{\pi}\check{\alpha} + \check{H}_q(\omega), \tag{6}$$

where

$$\check{\alpha} = \begin{pmatrix} \langle S^z\rangle & 0 \\ 0 & \langle T^z\rangle \end{pmatrix} \tag{7}$$

and

$$\check{H}_q(\omega) = \langle\langle[\check{B}_q, \mathscr{H}]\,|\check{B}_q^+\rangle\rangle_\omega. \tag{8}$$

The commutator in the higher order Green function of Eq. (8) decomposes into two parts, as follows:

$$[\check{B}_q, \mathscr{H}] = \check{A}(q)\check{B}_q + [\check{B}_q, \mathscr{H}]^{\mathrm{irr}}. \tag{9}$$

Eq. (6) now takes the form

$$[\omega\check{I} - \check{A}(q)]\check{G}_q(\omega) = -\frac{1}{\pi}\check{\alpha} + \check{H}_q^{\mathrm{irr}}(\omega), \tag{10}$$

where $\check{I}$ is the unit matrix and

$$\check{H}_q^{\mathrm{irr}}(\omega) = \langle\langle[\check{B}_q, \mathscr{H}]^{\mathrm{irr}}|B_q^+\rangle\rangle_\omega \tag{11}$$

is an irreducible Green function. The unknown matrix $\check{A}(q)$ is determined from the vanishing of non-uniform terms in the equation of motion for $\check{H}_q^{\mathrm{irr}}(\omega)$. This condition is given by

$$\langle[[B_q, \mathscr{H}]^{\mathrm{irr}}, \check{B}_q^+]\rangle = 0, \tag{12}$$

meaning that

$$\check{A}(q)\,\langle[\check{B}_q, \check{B}_q^+]\rangle = \langle[[\check{B}_q, \mathscr{H}], \check{B}_q^+]\rangle. \tag{13}$$

This leads to the equations

$$A_{11}(q) = -\frac{1}{2\langle S^z\rangle}\Big\{\Delta[\langle S^-T^+\rangle+2\langle S^zT^z\rangle] - \frac{2ab}{N}\sum_k J(k)\,\langle S^-_{-k}T_k^+\rangle$$
$$-\frac{2a^2}{N}\sum_k J(k)\,\langle S^-_{-k}S_k^+\rangle + \frac{2a^2}{N}\sum_k J(q-k)\,\langle S^-_{-k}S_k^+\rangle$$
$$-\frac{4a^2}{N}\sum_k J(k)\,\langle S_k^zS^z_{-k}\rangle + \frac{4a^2}{N}\sum_k J(q-k)\,\langle S_k^zS^z_{-k}\rangle - \frac{4ab}{N}\sum_k J(k)\,\langle S_k^zT^z_{-k}\rangle\Big\},$$

$$A_{12}(q) = -\frac{1}{2\langle T^z\rangle}\Big\{-\Delta[\langle S^-T^+\rangle+2\langle S^zT^z\rangle]$$
$$+\frac{2ab}{N}\sum_k J(q-k)\,\langle S^-_{-k}T_k^+\rangle + \frac{4ab}{N}\sum_k J(q-k)\,\langle S_k^zT^z_{-k}\rangle\Big\},$$

$$A_{21}(q) = A_{12}(q)\,\frac{\langle T^z\rangle}{\langle S^z\rangle},$$

$$A_{22}(q) = -\frac{1}{2\langle T^z\rangle}\Big\{\Delta[\langle S^-T^+\rangle+2\langle S^zT^z\rangle] - \frac{2ab}{N}\sum_k J(k)\,\langle S^-_{-k}T_k^+\rangle$$
$$-\frac{2b^2}{N}\sum_k J(k)\,\langle T^-_{-k}T_k^+\rangle + \frac{2b^2}{N}\sum_k J(q-k)\,\langle T^-_{-k}T_k^+\rangle$$
$$-\frac{4b^2}{N}\sum_k J(k)\,\langle T_k^zT^z_{-k}\rangle + \frac{4b^2}{N}\sum_k J(q-k)\,\langle T_k^zT^z_{-k}\rangle - \frac{4ab}{N}\sum_k J(k)\,\langle S_k^zT^z_{-k}\rangle\Big\}, \tag{14}$$

where the one-site averages are assumed to be independent of the localization of the site

The Green functions of zeroth order are defined by neglecting $\check{H}_q^{\mathrm{irr}}(\omega)$ in Eq. (10)

$$[\omega\check{I} - \check{A}(q)]\check{G}_q^0(\omega) = -\frac{1}{\pi}\check{\alpha}. \tag{15}$$

The elementary excitation spectrum, given by

$$\det [\omega \check{I} - \check{A}(q)] = 0 \tag{16}$$

has two branches which are of the forms:

$$\omega_1(q) = \tfrac{1}{2}\{A_{11}(q)+A_{22}(q)+\sqrt{[A_{11}(q)-A_{22}(q)]^2-4A_{12}(q)A_{21}(q)}\}, \tag{17a}$$

$$\omega_2(q) = \tfrac{1}{2}\{A_{11}(q)+A_{22}(q)-\sqrt{[A_{11}(q)-A_{22}(q)]^2-4A_{12}(q)A_{21}(q)}\}. \tag{17b}$$

In order to determine the irreducible Green function on the right side of Eq. (10), we take the derivative of the Green function $\check{H}_q(\omega)$ (see Eq. (8)) with respect to the time t'. On taking the Fourier time transform, extracting the irreducible part as in Eq. (9), and using the equation of motion for $\check{G}_q(\omega)$ (Eq. (10)) and the properties of the matrix $\check{A}(q)$, we obtain the equation of motion for the irreducible Green function $\check{H}_q^{\mathrm{irr}}(\omega)$ in the form:

$$-\{\omega\check{I}-\check{A}(q)\}\check{H}_q^{\mathrm{irr}}(\omega) = -\frac{1}{2\pi}\langle[[\check{B}_q, \mathscr{H}]^{\mathrm{irr}}, B_q^+]\rangle + \check{H}_q^{\mathrm{irr,irr}}(\omega), \tag{18}$$

where

$$\check{H}_q^{\mathrm{irr,irr}}(\omega) = \langle\langle[\check{B}_q, \mathscr{H}]^{\mathrm{irr}} | [\check{B}_q^+, \mathscr{H}]^{\mathrm{irr}}\rangle\rangle_\omega. \tag{19}$$

With regard to the condition (12), the equation (18) becomes

$$\{\omega\check{I}-\check{A}(q)\}\check{H}_q^{\mathrm{irr}}(\omega) = -\check{H}_q^{\mathrm{irr,irr}}(\omega). \tag{20}$$

On writing the auxiliary equation:

$$\check{G}_q(\omega) = \check{G}_q^0(\omega)+\check{G}_q^0(\omega)\check{P}_q(\omega)\check{G}_q^0(\omega), \tag{21}$$

we obtain from Eqs (10), (15) and (18)

$$\check{P}_q(\omega) = -\pi^2\check{\alpha}^{-1}\check{H}_q^{\mathrm{irr,irr}}(\omega)\check{\alpha}^{-1}. \tag{22}$$

In the derivation of Eq. (22) we used the properties of the matrices $\check{A}$ and $\check{\alpha}$ as well as the fact that Eq. (20) can be written in the form

$$\check{H}_q^{\mathrm{irr}}(\omega)\{\omega\check{I}-\check{A}^+(q)\} = -\check{H}_q^{\mathrm{irr,irr}}(\omega). \tag{23}$$

The mass operator $\check{\Pi}_q(\omega)$, defined as:

$$\check{G}_q(\omega) = \check{G}_q^0(\omega)+\check{G}_q^0(\omega)\check{\Pi}_q(\omega)\check{G}_q(\omega) \tag{24}$$

is related to $\check{P}_q(\omega)$ by the equation

$$\check{P}_q(\omega) = \check{\Pi}_q(\omega)+\check{\Pi}_q(\omega)\check{G}_q(\omega)\check{P}_q(\omega). \tag{25}$$

It is obvious from Eq. (25) that the mass operator $\check{\Pi}_q(\omega)$ is determined by the proper part of $\check{P}_q(\omega)$ i.e. by the part not connected by a single line $\check{G}_q^0(\omega)$. Thus, the mass operator is given by

$$\check{\Pi}_q(\omega) = \check{P}_q^{(p)}(\omega) = -\pi^2\{\check{\alpha}^{-1}\check{H}_q^{\mathrm{irr,irr}}(\omega)\check{\alpha}^{-1}\}^{(p)}. \tag{26}$$

By using the spectral representation for the Green functions, we obtain:

$$\check{\Pi}_q(\omega) = \tfrac{1}{4} \int\limits_{-\infty}^{\infty} \frac{d\omega'}{\omega-\omega'} (e^{\frac{\omega'}{k_B T}}-1) \int\limits_{-\infty}^{\infty} dt e^{-i\omega' t}$$

$$\times \frac{1}{N} \sum_{kp} \begin{bmatrix} \dfrac{\langle \{C_{qk}(t)\}^{\text{irr}} \{C_{qp}^{+}\}^{\text{irr}}\rangle}{\langle S^z\rangle^2} & \dfrac{\langle \{C_{qk}(t)\}^{\text{irr}} \{D_{qp}^{+}\}^{\text{irr}}\rangle}{\langle S^z\rangle \langle T^z\rangle} \\ \dfrac{\langle \{D_{qk}(t)\}^{\text{irr}} \{C_{qp}^{+}\}^{\text{irr}}\rangle}{\langle S^z\rangle \langle T^z\rangle} & \dfrac{\langle \{D_{qk}(t)\}^{\text{irr}} \{D_{qp}^{+}\}^{\text{irr}}\rangle}{\langle T^z\rangle^2} \end{bmatrix}^{(p)}, \tag{27}$$

where

$$C_{qk} = \Delta(S_k^- T_{q+k}^z - S_k^z T_{q+k}^z)$$

$$+2J(k)\,(a^2 S_{q-k}^z S_k^- + ab S_{q-k}^z T_k^- - a^2 S_k^z S_{k+q}^- - ab S_k^z T_{k+q}^z) \tag{28}$$

and

$$D_{qk} = \Delta(S_k^z T_{q+k}^- - S_k^- T_{q+k}^z)$$

$$+2J(k)\,(b^2 T_{q-k}^z T_k^- + ab S_k^- T_{k+q}^z - b^2 T_{q-k}^- T_k^z - ab S_k^z T_{k+q}^-). \tag{29}$$

Eqs (24)–(29) and Eq. (15) complete the derivation of the Dyson equation.

3. Analysis of the zeroth-order approximation

The equation of motion for the zeroth order Green function (15) has the solution:

$$G_q^0(\omega)_{11} = \langle\langle S_{-q}^- | S_q^+ \rangle\rangle_\omega = -\frac{\langle S^z\rangle\,[\omega - A_{22}(q)]}{\pi[\omega-\omega_1(q)]\,[\omega-\omega_2(q)]},$$

$$G_q^0(\omega)_{12} = \langle\langle S_{-q}^- | T_q^+ \rangle\rangle_\omega = -\frac{\langle T^2\rangle A_{12}(q)}{\pi[\omega-\omega_1(q)]\,[\omega-\omega_2(q)]},$$

$$G_q^0(\omega)_{21} = G_q^0(\omega)_{12},$$

$$G_q^0(\omega)_{22} = \langle\langle T_{-q}^- | T_q^+ \rangle\rangle_\omega = -\frac{\langle T^2\rangle\,[\omega - A_{11}(q)]}{\pi[\omega-\omega_1(q)]\,[\omega-\omega_2(q)]}, \tag{30}$$

where $A_{11}(q)$, $A_{12}(q)$, $A_{22}(q)$ are given by Eq. (14) and $\omega_1(q)$, $\omega_2(q)$ by Eq. (17). On applying the spectral theorem, we obtain from Eqs (30) the following equations for the correlation functions:

$$\langle S_{-q}^- S_q^+\rangle = \langle S^z\rangle \left\{ \frac{\omega_1(q) - A_{22}(q)}{2\omega_1(q) - A_{11}(q) - A_{22}(q)} \coth\frac{\beta\omega_1(q)}{2} \right.$$

$$\left. + \frac{\omega_2(q) - A_{22}(q)}{2\omega_2(q) - A_{11}(q) - A_{22}(q)} \coth\frac{\beta\omega_2(q)}{2} - 1 \right\}, \tag{31a}$$

$$\langle S_{-q}^{-}T_{q}^{+}\rangle = \langle S^{z}\rangle\left\{\frac{A_{12}(q)}{2\omega_1(q)-A_{11}(q)-A_{22}(q)}\left(\coth\frac{\beta\omega_1(q)}{2}-\coth\frac{\beta\omega_2(q)}{2}\right)\right\}, \tag{31b}$$

$$\langle T_{-q}^{-}T_{q}^{+}\rangle = \langle T^{z}\rangle\left\{\frac{\omega_1(q)-A_{22}(q)}{2\omega_1(q)-A_{11}(q)-A_{22}(q)}\coth\frac{\beta\omega_2(q)}{2}\right.$$
$$\left.+\frac{\omega_2(q)-A_{22}(q)}{2\omega_2(q)-A_{11}(q)-A_{22}(q)}\coth\frac{\beta\omega_1(q)}{2}-1\right\}, \tag{31c}$$

where $\beta = 1/k_B T$ (k_B is the Boltzmann constant). The one-site averages $\langle S^z\rangle$, $\langle T^z\rangle$ and $\langle S^- T^+\rangle$ are cases to evaluate with the aid of the relations

$$\langle S^z\rangle = \tfrac{1}{2}-\frac{1}{N}\sum_q \langle S_{-q}^{-}S_q^{+}\rangle, \tag{32a}$$

$$\langle T^z\rangle = \tfrac{1}{2}-\frac{1}{N}\sum_q \langle T_{-q}^{-}T_q^{+}\rangle, \tag{32b}$$

$$\langle S^- T^+\rangle = \frac{1}{N}\sum_q \langle S_{-q}^{-}T_q^{+}\rangle. \tag{32c}$$

In order to obtain the one-site average $\langle S^z T^z\rangle$ we use the Green function $\check{H}_q(\omega)$ (see Eq. (6)). The equation of motion (6) for $[G_q(\omega)]_{11}$ takes the form

$$\omega\langle\langle S_{-q}^{-}|S_q^{+}\rangle\rangle_\omega = -\frac{1}{\pi}\langle S^z\sigma+\langle\langle[S_{-q}^{-},\mathscr{H}]\,|S_q^{+}\rangle\rangle_\omega. \tag{33}$$

Approximating $G_q(\omega)_{11}$ by $G_q^0(\omega)_{11}$, we obtain:

$$\langle\langle[S_{-q}^{-},\mathscr{H}]\,|S_q^{+}\rangle\rangle_\omega = \frac{\langle S^z\rangle}{\pi[\omega_2(q)-\omega_1(q)]}\left\{\frac{\omega[\omega-A_{22}(q)]}{\omega-\omega_2(q)}-\frac{\omega[\omega-A_{22}(q)]}{\omega-\omega_1(q)}\right\}+\frac{1}{\pi}. \tag{34}$$

Using the spectral theorem and evaluation the commutator in the average $\langle[S_{-q}^{-},\mathscr{H}]S_q^{+}\rangle$, we obtain the following expression

$$\langle S^z T^z\rangle = \tfrac{1}{2}\langle T^z\rangle-\tfrac{1}{2}\langle S^- T^+\rangle+\frac{1}{\Delta}\left\{\frac{2a^2}{N}\sum_q J(q)\langle S_q^z S_{-q}^z\rangle\right.$$
$$+\frac{2ab}{N}\sum_q J(q)\langle S_q^z T_{-q}^z\rangle-a^2\langle S^z\rangle J(0)-ab\langle T^z\rangle J(0)$$
$$+\frac{\langle S^z\rangle}{N}\sum_q\frac{\omega_1(q)\,[\omega_1(q)-A_{22}(q)]}{\omega_2(q)-\omega_1(q)}\coth\frac{\beta\omega_1(q)}{2}$$

$$+\frac{a^2}{N}\sum_q J(q)\langle S^-_{-q}S^+_q\rangle+\frac{ab}{N}\sum_q J(q)\langle S^-_{-q}T^+_q\rangle$$

$$-\frac{\langle S^z\rangle}{N}\sum_q \frac{\omega_2(q)\,[\omega_2(q)-A_{22}(q)]}{\omega_2(q)-\omega_1(q)}\coth\frac{\beta\omega_2(q)}{2}+\frac{\langle S^z\rangle}{N}\sum_q A_{11}(q)\Big\}. \quad (35)$$

Eqs (14), (17), (31), (32), (35) are as yet not self-consistent because of $\langle S^z_{-q}S^z_q\rangle$, $\langle S^z_{-q}T^z_q\rangle$ and $\langle T^z_{-q}T^z_q\rangle$. We achieve self-consistency by performing the very simple approximation

$$\langle S^z_{-q}S^z_q\rangle \to \langle S^z_{-q}\rangle\langle S^z_q\rangle,$$
$$\langle S^z_{-q}T^z_q\rangle \to \langle S^z_{-q}\rangle\langle T^z_q\rangle,$$
$$\langle T^z_{-q}T^z_q\rangle \to \langle T^z_{-q}\rangle\langle T^z_q\rangle. \quad (36)$$

Using the above approximation we obtain the same results as when applying SDM to our problem [8].

4. *Discussion*

We have derived the Dyson equation of the double-time temperature Green functions for the singlet-triplet system in the ferromagnetic region. The poles of the Green functions in the lowest order approximation give the elementary excitation spectrum of our system. We have shown that the best zeroth-order approximation is equivalent to the spectral density method [8]. To make the procedure self-consistent, however, it is necessary to apply the higher-order Green function $\langle\langle[S^-_{-q},\mathscr{H}]|S^+_q\rangle\rangle$. The simplest way is to express this higher-order Green function in terms of the zeroth-order Green function $G^0_q(\omega)$. The above procedure reproduces exactly the results of SDM. This shows that the evaluation of $\langle S^zT^z\rangle$ has to be performed in the next order of the approximation. Similarly, the evaluation of $\langle S^z_{-q}S^z_q\rangle$ for the Heisenberg model applying SDM in the lowest order of the approximation [6] has, in fact, to be carried out in the next-higher order. It is not possible to solve such problems self-consistently in the lowest order. Of course, the singlet-triplet system is much more complicated than the Heisenberg model and our procedure allows to calculate only all the one-site averages and transversal two-site correlations. We have decoupled the two-site correlations $\langle S^z_{-q}S^z_q\rangle$, $\langle S^z_{-q}T^z_q\rangle$ and $\langle T^z_{-q}T^z_q\rangle$. After this decoupling, our set of equations is self-consistent.

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