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Reaction Field Approximation for the Ordered Phase of Heisenberg Systems[2])

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The approximation method to obtain the static properties of a system of interacting localized magnetic moments, called "reaction field approximation" (RFA), which is previously used for the paramagnetic phase, is extended to cover the ordered phase of a spin system with Heisenberg interaction. The presence of a small anisotropy is also considered and an application to MnF_2 is made.

La méthode d'approximation utilisée pour obtenir les propriétés statiques d'un système de moments magnétiques localisés interactuants, dite „Approximation du Champ de Réaction", déjà appliquée à la phase paramagnetique, est ici éténdue à la phase ordonnée d'un système de spin avec interaction d'Heisenberg. La présence d'une petite anisotropie est aussi considerée et une application au MnF_2 est faite.

1. Introduction

The most well-known method to calculate the susceptibility and other variables of a system of localized magnetic moments in a solid, with given interaction Hamiltonian, is the molecular field approximation, MFA. Being extremely simple, both conceptually and mathematically, it also has the merit of giving very good quantitative results at high temperatures, besides giving rather good qualitative results at all temperatures. A number of effective field theories which are improved versions of MFA can be found in the literature [1 to 6]. The "correlated effective field" (CEF) [3] and the "reaction field approximation" (RFA) [5, 6] are essentially the same method, and we will refer to them in this paper as RFA. Its name comes from Onsager's suggestion [7] that the part of the molecular field on a given moment $\boldsymbol{\mu}_i$ which comes from the reaction of the neighbours, $\boldsymbol{\mu}_j$, to the instantaneous orientation of $\boldsymbol{\mu}_i$, should not be included in the effective orienting field. This "reaction field" simply follows the motion of $\boldsymbol{\mu}_i$ and thus does not favour one orientation over another.

Since the actual value of the reaction field is not known a priori, the procedure followed in practice consists in adding to the MFA effective field a correlation-dependent term, the magnitude of which is determined at the end of the calculation by imposing consistency of the theory with a sum rule for the susceptibility $\chi(\boldsymbol{q})$, obtained from the fluctuation–dissipation theorem. Unfortunately this sum rule is valid only in the paramagnetic phase and in abscence of strong fields. In the present paper we extend the procedure of RFA to the ordered phase of a Heisenberg magnet. The difficulty with the sum rule is avoided by substituting it by another relation (a generalized sum rule), obtained from the internal consistency of the theory, and which reproduces the original sum rule in its region of validity.

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In Section 2 we introduce the model Hamiltonian and describe the method to calculate the spontaneous (or sub-lattice) magnetization and the $\boldsymbol{q}$-dependent susceptibility. In Section 3 an application to MnF_2 is made, and the results are compared with experimental data.

2. RFA for the Heisenberg Magnet

Consider a solid with atomic spins $\boldsymbol{S}_i$ and corresponding magnetic moments $-\gamma\boldsymbol{S}_i$. The interaction Hamiltonian will be assumed to be of the form

$$\mathcal{H} = -\tfrac{1}{2}\sum_{ij} J_{ij}\boldsymbol{S}_i \cdot \boldsymbol{S}_j + \gamma \sum_i \boldsymbol{S}_i \cdot \boldsymbol{H}_i \,, \tag{1}$$

where $\boldsymbol{H}_i$ is a very small dosition-dependent external field. The effective field on $\boldsymbol{S}_i$ is defined in such a way that the Hamiltonian (1) may be approximated by

$$\mathcal{H}^{\mathrm{eff}} = \gamma \sum_i \boldsymbol{S}_i \cdot \boldsymbol{H}_i^{\mathrm{eff}} \,. \tag{2}$$

In RFA $\boldsymbol{H}_i^{\mathrm{eff}}$ is written in the form

$$\boldsymbol{H}_i^{\mathrm{eff}} = -\frac{1}{\gamma}\sum_j J_{ij}\langle \boldsymbol{S}_j\rangle + \frac{\Lambda_i}{\gamma}\langle \boldsymbol{S}_i\rangle + \boldsymbol{H}_i \,, \tag{3}$$

where $\langle \ldots \rangle$ means ensemble average. The second term above is the negative of the reaction field. It is natural to assume it to be parallel to $\langle \boldsymbol{S}_i\rangle$ because this is the only preferred direction of the system of spins with isotropic interaction. The form in which equation (3) is written does not imply that the reaction field is linear in $\langle \boldsymbol{S}_i\rangle$ because we will allow Λ_i to be a function of $|\langle \boldsymbol{S}_i\rangle|$.

Equation (3) may be separated in its unperturbed and perturbation part:

$$\boldsymbol{H}_{0i}^{\mathrm{eff}} = -\frac{1}{\gamma}\sum_j J_{ij}\langle \boldsymbol{S}_j\rangle_0 + \frac{\lambda_0}{\gamma}\langle \boldsymbol{S}_i\rangle_0 \,, \tag{4}$$

$$\delta\boldsymbol{H}_i^{\mathrm{eff}} = -\frac{1}{\gamma}\sum_j J_{ij}\,\delta\langle \boldsymbol{S}_j\rangle + \frac{\boldsymbol{\lambda}}{\gamma}\cdot\delta\langle \boldsymbol{S}_i\rangle + \boldsymbol{H}_i \,. \tag{5}$$

Here $\langle \ldots \rangle_0$ means ensemble average in the absence of the perturbation field $\boldsymbol{H}_i$. Since the unperturbed system is translationally invariant, λ_0 is independent of i. The tensor $\boldsymbol{\lambda}$ could be dependent on the position i because $\boldsymbol{H}_i$ destroys the homogeneity of the system. However, if we write

$$\boldsymbol{\lambda}_i \cdot \delta\langle \boldsymbol{S}_i\rangle = (\boldsymbol{\lambda} + \delta\boldsymbol{\lambda}_i)\cdot\delta\langle \boldsymbol{S}_i\rangle \,, \tag{6}$$

where $\delta\boldsymbol{\lambda}_i$ is the deviation of $\boldsymbol{\lambda}_i$ from its unperturbed value $\boldsymbol{\lambda}$, then the second term of (6) has to be neglected because it is quadratic in $\boldsymbol{H}_i$ and we are considering the limit of linear response. On the other hand, if the only preferred direction of the unperturbed system, that of the spontaneous magnetization $\langle \boldsymbol{S}_i\rangle_0$, is chosen as one of the axis (say z) of the reference frame, then all physical tensors, including $\boldsymbol{\lambda}$, are diagonal. We will denote by $\lambda_\parallel$ and $\lambda_\perp$ the elements of $\boldsymbol{\lambda}$. Since the sum of (4) and (5) has to reproduce (3), choosing $\boldsymbol{H}_i$ in the x-direction we obtain

$$\Lambda_i\langle \boldsymbol{S}_i\rangle = \lambda_0\langle S_i^z\rangle_0\,\hat{z} + \lambda_\perp\,\delta\langle \boldsymbol{S}_i^x\rangle\,\hat{x} \,. \tag{7}$$

Equation (7) is a vector equation which implies that both λ_0 and $\lambda_\perp$ are equal to Λ_i, which yields

$$\lambda_0 = \lambda_\perp \,, \tag{8}$$

leaving us with only two undetermined parameters, say $\lambda_\perp$ and $\lambda_\parallel$, to be determined later.

Fourier-transforming (5) in space we obtain

$$\delta H_\mu^{\text{eff}}(\boldsymbol{q}) = -\frac{1}{n\gamma}[J(\boldsymbol{q}) - \lambda_\mu]\,\delta\langle S_\mu(\boldsymbol{q})\rangle + H_\mu(\boldsymbol{q})\,, \tag{9}$$

where μ stands for $\parallel$ or $\perp$ and $n = N/V$ is the number of spins per unit volume.

By the same symmetry argument as above we see that the susceptibility is also a diagonal tensor and that the induced part of the magnetization may be written as

$$\delta M_\mu(\boldsymbol{q}) = \chi_\mu(\boldsymbol{q})\, H_\mu(\boldsymbol{q}) = \chi_\mu^{\text{eff}}\,\delta H_\mu^{\text{eff}}(\boldsymbol{q})\,. \tag{10}$$

The quantity χ_μ^{eff} is the susceptibility of a system of non-interacting localized spins in presence of an external field equal to $\boldsymbol{H}_{0i}^{\text{eff}}$.

From (9) and (10) and the identification

$$\delta\langle S_\mu(\boldsymbol{q})\rangle = -\frac{1}{\gamma}\,\delta M_\mu(\boldsymbol{q}) \tag{11}$$

we obtain

$$\chi_\mu(\boldsymbol{q}) = \frac{\chi_\mu^{\text{eff}}}{1 - (\chi_\mu^{\text{eff}}/n\gamma^2)\,[J(\boldsymbol{q}) - \lambda_\mu]}\,. \tag{12}$$

This expression is similar to that for the paramagnetic phase [5], when one has χ_0 instead of χ_μ^{eff} and λ instead of λ_μ. A sum rule relating λ with χ_0 was derived from the fluctuation–dissipation theorem for the paramagnetic phase. A similar relation may be obtained invoking again the concept of reaction field, which reproduces in the paramagnetic phase the original sum rule, but is also valid in the ordered phase. The susceptibility in $\boldsymbol{R}$-space,

$$\chi_{ij}^{\mu\nu} = \frac{1}{N}\sum_{\boldsymbol{q}} \chi_{\mu\nu}(\boldsymbol{q})\, e^{i\boldsymbol{q}\cdot(\boldsymbol{R}_i - \boldsymbol{R}_j)}\,, \tag{13}$$

relates the "response" at site i to a local field applied at site j. In particular $\chi_{ii}^{\mu\mu}$, which relates $\delta\langle S_i^\mu\rangle$ with the field H_i^μ applied at the same site i, may be identified with χ_μ^{eff} because the deviation of the internal field on $\boldsymbol{S}_i$ from $\boldsymbol{H}_{0i}^{\text{eff}}$ due to the influence of $\delta\langle S_i^\mu\rangle$ on the neighbour spins is precisely the reaction field, which does not contribute to the effective orienting field. From this identification and (13) we obtain

$$\frac{1}{N}\sum_{\boldsymbol{q}} \chi_\mu(\boldsymbol{q}) = \chi_\mu^{\text{eff}}\,. \tag{14}$$

The susceptibility χ_μ^{eff} is a well-defined function of H_{0i}^{eff}, which is related to $\langle S_i^z\rangle_0$ by (4) and (8). Since the spontaneous magnetization is assumed to be along the z-axis, we have

$$H_{0i}^{\text{eff}} = -\frac{1}{\gamma}\sum_j J_{ij}\langle S_j^z\rangle_0 + \frac{\lambda_\perp}{\gamma}\langle S_i^z\rangle_0\,. \tag{15}$$

For a homogeneous ferromagnet all $\langle S_i^z\rangle_0$ are equal, and for an antiferromagnet there is a sign change when we go from one sub-lattice to the other. In both cases we may suppress the subscript i in (15) and write

$$H_0^{\text{eff}} = -\frac{1}{\gamma}[J(\boldsymbol{Q}) - \lambda_\perp]\langle S^z\rangle_0\,, \tag{16}$$

where $\boldsymbol{Q}$ is the wave vector that characterizes the spontaneous magnetization ($\boldsymbol{Q} = \boldsymbol{0}$ for the ferromagnet). $\langle S^z \rangle_0$ is a known function of H_0^{eff}, namely

$$\langle S^z \rangle_0 = \operatorname{tr} \varrho_0 S^z = \frac{\sum_{m=-S}^{S} m \exp(-\beta\gamma H_0^{\text{eff}} m)}{\sum_{m=-S}^{S} \exp(-\beta\gamma H_0^{\text{eff}} m)}. \tag{17}$$

Equations (16) and (17) are not sufficient to determine H_0^{eff} and $\langle S^z \rangle_0$ because $\lambda_\perp$ is also unknown. We have to use the "sum rule" (14) to complete the set. The susceptibility $\chi_\mu(\boldsymbol{q})$, equation (12), may be written in a more convenient form by introducing new variables s_μ, defined by

$$s_\mu = \frac{1}{J(\boldsymbol{Q})}\left[\frac{n\gamma^2}{\chi_\mu^{\text{eff}}} + \lambda_\mu\right]. \tag{18}$$

Equations (12) and (18) give

$$\chi_\mu(\boldsymbol{q}) = \frac{n\gamma^2/J(\boldsymbol{Q})}{s_\mu - J(\boldsymbol{q})/J(\boldsymbol{Q})}, \tag{19}$$

and equation (14) may be written as

$$G(s_\mu) = J(\boldsymbol{Q})\,\frac{\chi_\mu^{\text{eff}}}{n\gamma^2}, \tag{20}$$

where

$$G(s) = \frac{1}{N}\sum_{\boldsymbol{q}} \frac{1}{s - J(\boldsymbol{q})/J(\boldsymbol{Q})} \tag{21}$$

is known as "lattice Green's function" and is tabulated for the cubic lattices [8]. Eliminating $\chi_\mu^{\text{eff}}/n\gamma^2$ between (18) and (20), we obtain a simple relation between λ_μ and s_μ, namely

$$\lambda_\mu = J(\boldsymbol{Q})\left[s_\mu - \frac{1}{G(s_\mu)}\right]. \tag{22}$$

For a system of magnetic moments in isotropic environment, except for the presence of a magnetic field H_0 having magnetization M_0, the susceptibility to a test field perpendicular to H_0 is

$$\chi_\perp^0 = \frac{M_0}{H_0}. \tag{23}$$

Recalling the definition ot $\chi_\perp^{\text{eff}}$, we see that an equation analogous to (23) applies to it, namely

$$\chi_\perp^{\text{eff}} = -\frac{n\gamma\langle S^z \rangle_0}{H_0^{\text{eff}}}. \tag{24}$$

From (16), (18), and (24) we obtain

$$s_\perp = 1 \tag{25}$$

for any temperature in the ordered phase. This result agrees with the intuitive notion that the perpendicular uniform susceptibility of the Heisenberg ferromagnet should diverge (see (19)) at any temperature.

From (16), (22), and (25) it follows that

$$H_0^{\text{eff}} = -\frac{J(\boldsymbol{Q})}{G(1)}\langle S^z\rangle_0 . \tag{26}$$

Equations (17) and (26) may be solved numerically for $\langle S^z\rangle_0$, giving the spontaneous or sub-lattice magnetization. The only difference between this set of equations and the usual MFA result is that the constant $G(1)$ appearing in (26) is not present in MFA. The values of $G(1)$ for s.c., b.c.c., and f.c.c. lattices with nearest-neighbour interaction are, respectively, 1.516, 1.393, and 1.345. Once H_0^{eff} is determined, $\chi_\mu^{\text{eff}} \equiv \chi_\mu^0(H_0^{\text{eff}})$ is also known. From (20) and (19) we then determine s_μ and $\chi_\mu(\boldsymbol{q})$, respectively.

3. The Anisotropic Case — Application to MnF_2

The completely isotropic Hamiltonian studied above is an idealization which rarely describes well a real physical situation. For many systems, the presence of some anisotropy may be simulated by adding to the Hamiltonian a single-particle term of the form $D(S_i^z)^2$. In this case the use of (8), which was obtained as a consequence of the rotational invariance of the interaction Hamiltonian (see (3) and the comments following it), is no longer justified. However, when the anisotropy energy is small compared to the exchange energy, considering that the reaction field is a correction to the exchange field, one may argue that the anisotropy has only a secondary effect on the values of the λ's and equation (8) may be maintained as a reasonable approximation. So, the term $D(S_i^z)^2$ will only be introduced where it has a direct effect, namely, on the explicit form of the single-particle susceptibilities χ_μ^{eff} and on (17), which will be substituted by

$$\langle S^z\rangle_0 = \frac{\sum_m m \exp\left[-\beta(\gamma H_0^{\text{eff}} m + Dm^2)\right]}{\sum_m \exp\left[-\beta(\gamma H_0^{\text{eff}} m + Dm^2)\right]} . \tag{27}$$

The antiferromagnetic MnF_2 has, indeed, a very small anisotropy. Thus, the method described above may be used to determine its properties. The magnetic lattice of MnF_2 is body-centered tetragonal such that the nearest neighbours to a Mn atom are the two Mn just above and below it along the c-axis. The most important interaction of an atom, say, at the centre of the unit cell is an antiferromagnetic exchange with the eight n.n.n. at the corners. There is also a weak ferromagnetic exchange with the two n.n. The exchange constants are represented by J and J', respectively. The values usually found in the literature for J (−3.52 K), J' (0.64 K), and D (−0.212 K) were determined in a slightly inconsistent way: First, Trapp and Stout [9] determined J from the experimental value of the Curie-Weiss temperature, $\theta = -82$ K, neglecting J' and D; then Okazaki and Turberfield [10] determined J' and D from inelastic neutron scattering experiment at low temperatures using a spin wave theory and the J-value

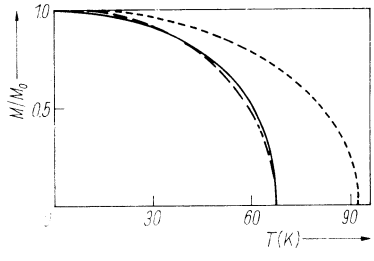

Fig. 1. Sub-lattice magnetization of MnF_2 versus temperature. The solid line represents the experimental result [13], the dashed line is our RFA-calculated value, and the dotted line is the MFA result with the same values of J, J', and D

obtained by Trapp and Stout. We prefer to derive the values of these three parameters from the experimental results for the Curie-Weiss temperature $\theta = -82$ K, the Néel temperature $T_N = 67.3$ K [11], and the spin flop field $H_{SF} = 9.3 \times 10^{-4}$ Oe [12], obtaining $J = -3.73$ K, $J' = 0.86$ K, and $D = -0.20$ K. Using these values and the procedure described above, we calculated the sub-lattice magnetization of MnF_2, which we show in Fig. 1 together with the experimental result [13] and the MFA result, for comparison.

4. Conclusions

From the results of this work and of previous publications [3 to 6] we conclude that RFA is a good method to treat systems of localized spins with exchange and other types of interactions. The agreement with the experimental results is considerably better than the usual mean field approximation for temperatures close to the phase transition temperature T_c, and tends to the same result as MFA for temperatures far from T_c. In particular, we have shown in this paper how to apply RFA for the ordered phase when the spin-Hamiltonian is isotropic or has a very small anisotropy. The applicability of RFA at $T < T_c$ on systems with a strong anisotropy is still an unsolved problem. Other extensions of the theory, e.g., to compounds with non-equivalent spins, alloys or glasses, still remain to be done. Finally we want to remark that the same results as obtained by RFA may be obtained by other techniques, like Green's function equations with RPA decoupling, but the advantage of the former is its conceptual and mathematical simplicity.

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