Quantum Spin Effect and Short-Range Order above the Curie Temperature

R. Y. Gu and V. P. Antropov

Condensed Matter Physics, Ames Laboratory, Ames, IA 50011
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Abstract

Using quantum Heisenberg model calculations with Green’s function technique generalized for arbitrary spins, we found that for a system of small spins the quantum spin effects significantly contribute to the magnetic short-range order and strongly affect physical properties of magnets. The spin dynamics investigation confirms that these quantum spin effects favor the persistence of propagating spin-wave excitations above the Curie temperature. Our investigation suggests a reconsideration of a prevailing point of view on finite temperature magnetism to include quantum effects and the magnetic short-range order.



It is a long-standing debate over the nature of the paramagnetic (PM) state of the ferromagnetic (FM) materials, particularly in the transition metals. Early inelastic neutron experiments\[1, 2, 3\] determined the persistence of spin-wave (SW) like modes above the Curie temperature \(T_C\) in both Ni and Fe, and these modes were interpreted as the evidence of considerable magnetic short-range order (MSRO) in the PM state\[4\]. The presence of MSRO was later supported by the spin and angle-resolved photoemission studies\[2, 4\]. This is rather unusual because majority of magnetism theories are based on the absence of SW excitations above \(T_C\). Moreover, by applying the spherical model (SM) approximation to the Heisenberg model (HM), Shastry et al. concluded that in contrast with the experiment\[2\], this model, with fairly long-ranged interactions, has little MSRO and no SW peaks above \(T_C\) in Fe. A similar conclusion was reached by Monte Carlo (MC) spin dynamics simulation of the classical HM for the same system\[3\]. Very sophisticated technique\[2\] also could not detect any traces of strong MSRO in Fe or Ni. Recent theoretical spin dynamics studies\[10\], however, have demonstrated that a strong MSRO is a ‘must have’ property of the itinerant magnets while such excitations like SW exist above \(T_C\) in both localized and itinerant magnets.

It is puzzling that the applications of HM failed to predict the expected MSRO because BCC Fe has rather good local moments\[11, 12\] and to a large extent HM should be valid\[13\]. What usually is omitted in the classical MC simulation is the quantum nature of spin. In quantum SM \(T_C\), the spin correlations and the susceptibility are proportional to \(S(S + 1)\), while the classical coefficient scales as \(S^2\); so the quantum effect contributes a factor \(Q_S = 1 + S^{-1}\). For small \(S\) this \(Q_S\) can be unreasonably large. For example, for \(S = 1/2\) \(Q_S = 3\) which leads to the unphysical correlation between nearest-neighbor (NN) spins \(\langle S_i \cdot S_j \rangle/S^2 > 1\) in the case of NN coupling of the simple cubic (SC) structure. The same problem appeared in Ref.\[3\] where in the case of strong MSRO and \(S = 1\) \(\langle S_i \cdot S_j \rangle/S^2 = 1.64\). To avoid such difficulties a pragmatical approach is to scale the relevant quantities by \(S(S + 1)\), as was done in Ref.\[3\]. Then, in SM the scaled quantities are independent of \(S\), so the quantitative results for MSRO and region of SW existence for \(S = 1\) and \(S = \infty\) are the same\[3\]. Below we will demonstrate that while the classical HM can describe some degree of MSRO above \(T_C\), QSE properly included strongly increases MSRO and affects its influence on other physical properties.

For the HM Hamiltonian \(H = -\sum_{ij} J_{ij} S_i \cdot S_j\) in the PM state, we use the second-order Green’s function (GF) technique\[14, 15, 16, 17\]. To calculate GF \(G_{ij}^{\omega} = \langle S_i^+ S_j^- \rangle/\omega\) one applies twice the equation of motion and then decouples the high-order GF of forms \(\langle S_i^+ S_j^- S_m^+ S_n^- \rangle/\omega\) and \(\langle S_i^+ S_j^- S_m^+ S_n^- S_j^+ \rangle/\omega\). For \(S = 1/2\) in one-dimensional system Kondo and Yamaji (KY) decoupled them by using a correction parameter \(\alpha\[14\]. Here we extend their method for arbitrary \(S\) by introducing the following decoupling scheme (for \(i \neq \rho\) and \(\rho \neq l\)),

\[
\langle\langle S_i^+ S_j^- - S_i^- S_j^+; S_j^+ \rangle\rangle/\omega \rightarrow (1 - \nu_s \delta_{il}) \bar{C}_{ij} G_{ij}^{\omega} - \bar{C}_{il} G_{ij}^{\omega},
\]

\[
\langle\langle S_i^+ S_j^- S_m^+ S_n^-; S_j^+ \rangle\rangle/\omega \rightarrow (1 - \nu_s \delta_{il}) \bar{C}_{ij}^{zz} G_{mj}^{\omega} G_{nj}^{\omega},
\]

where \(\bar{C}_{ij}^{zz} = \alpha_m C_{ij}^{zz}\) and \(\bar{C}_{ij} = \alpha_d C_{ij}\), with \(\alpha_m = \alpha(1 - \delta_{mp}) + \delta_{mp}\) and \(C_{ij}^{zz} = \langle S_i^+ S_j^- S_m^+ S_n^- \rangle, C_{ij} = \langle S_i^+ S_j^- \rangle = 2\langle S_i^+ S_i^- \rangle\) are the spin correlations, \(\{A, B\} = (AB + BA)/2\) is the symmetric product of operators, and \(\nu_s\) is a \(S\)-dependent constant which will be determined later. For \(S = 1/2\) spin operator identities require \(\nu_s = 1\), and Eq.\[14\] is reduced to the KY decoupling.

Decoupling the high order GF in the equation of motion with Eq.\[14\] one can obtain the following expression for the dynamic susceptibility

\[
\chi^{\pm\pm}(q, \omega) = -\frac{2 \sum_n z_n J_n C_n (1 - \gamma_n^q)}{\omega^2 - \omega_q^2},
\]

where \(n\) is the shell index, \(J_n\), and \(C_n\) are \(J_{ij}\) and \(C_{ij}\), correspondingly. \(\gamma_n^q = z_n^{-1} \sum_{\delta_{ij}} (1 - e^{i q \delta_{ij}})\) with \(z_n\) being the total number of sites on \(n\)-th shell and \(\delta_{ij}\) being sites on that shell.

The SW excitation spectrum is

\[
\omega_q = \left\{ \sum_n z_n (1 - \gamma_n^q) \left[ D_n - \nu_s J_n C_n - J_n^q C_n \right] \right\}^{1/2}
\]
where $D_n = N^{-1} \sum_{k} J^k \gamma_n^{k} \tilde{c}^k$ with $J^k$ and $\tilde{C}^k$ being the Fourier transforms of $J_i$ and $\tilde{C}_{ij}$, correspondingly. At this stage $\nu_s = (2 - S)/3S$ is obtained by comparing Eq. (4) with the well-known result $\omega_q = \left(J^0 - J^s\right)S$ in the FM spin correlation limit $\nu_s = 2S^2/3$.

From Eq. (4) and the spectral theorem, the spin correlation can be written as

$$C^q = \sum_n z_n n J_n (1 - \gamma_n^q \frac{C_n}{\omega_q} \coth \frac{\omega_q}{2T}).$$

With the requirements $C_n = 1/N \sum_q C^q \gamma_n^q$ and $C_0 = 1/N \sum_q C^q = 2S(S+1)/3$, Eqs. (3) and (4) can be solved self-consistently. $T_C$ is determined by $\chi^{-1} = 0$ ($\chi = \chi^{++}(0,0)/2$). To check the validity of our method, in Fig. 1 we compare our calculated $T_C/\gamma^0$ and $E_c/E_0$ for the BCC structure with the accurate results obtained by the high-temperature-expansion (HTE) methods \cite{19} and the SM results in the NN coupling case. Here $T_C^{\text{MF}}$ is the Curie temperature in the mean field (MF) approximation, $E_c$ and $E_0$ are the total energies at $T_C$ and zero temperature, correspondingly. The parameter $E_c/E_0$ is a proper measure of MSRO at $T_C$, and in the NN coupling case $E_c/E_0$ is identical to the average cosine of angles between NN spins. In the MF approximation there is no MSRO ($E_c/E_0 = 0$) at and above $T_C^{\text{MF}}$. The existence of MSRO suppresses $T_C$ with respect to $T_C^{\text{MF}}$. Such suppression exists also in the SM and is identical for all $S$ in that case. In more accurate calculations, however, $T_C$ is more suppressed at smaller $S$.

The MSRO parameter $E_c/E_0$ demonstrates the increase in MSRO for smaller $S$. Although in the SM increases even faster ($E_c/E_0 \propto Q_S$), this quantity already is not well defined owing to the appearance of $E_c/E_0 > 1$ e.g. for the SC structure for $S = 1/2$ and in Ref. \cite{21} for $S = 1$. At this stage the scaling should be introduced which leads to the elimination of real QSE.

Our formalism allows to obtain the following important result for $T_C$ for $S = \infty$:

$$T_C = \alpha T_C^{\text{SM}} = 3T_C^{\text{MF}}/(2F + 1),$$

where $\alpha = 3F/(2F + 1)$ with $F = N^{-1} \sum_q (1 - \gamma_1^q)^{-1}$. This new and transparent expression provides another immediate and accurate check of applicability of our generalized GF formalism. For instance, it gives $T_C/(J_1S^2) = 1.49, 2.11, \text{and } 3.25$ for SC, BCC and FCC structures which are very close to the corresponding HTE results 1.45, 2.06, and 3.18 \cite{16}. Eq. (6) clearly indicates the importance of the correction parameter $\alpha$ introduced above in the GF decoupling. For $S = \infty$ the parameter $E_c/E_0 = 1 - F^{-1}$ is the same as the one obtained in the SM.

The good agreement between our and HTE results indicates the applicability of this formalism for the case of arbitrary $S$ and NN interaction. We also studied a Heisenberg Hamiltonian corresponding to a realistic material: we used extended (four NN) interactions in BCC Fe: $J_2/J_1 = 0.5221$, $J_3/J_1 = 0.0056$, and $J_4/J_1 = -0.0879$ \cite{21}, where $J_1S^2 = 2.44$ mRy. For $S = \infty$ MC simulation gives $T_C^{\text{MC}}/T_C^{\text{MF}} = 0.68 \pm 0.70$ and $E_c/E_0 = 0.38 \pm 0.41$. In SM $T_C^{\text{SM}}/T_C^{\text{MF}} = 0.59$ and $E_c/E_0 = 0.40Q_S$ for all $S$. In our formalism, for $S = 1/2, 1$ and $\infty$, $T_C/T_C^{\text{MF}} = 0.51, 0.57$ and 0.67, and their $E_c/E_0 = 0.79, 0.68$ and 0.41, correspondingly. At $S = 1$, $T_C^{\text{SM}} = 2334K$ is more than twice higher than the experimental 1040K of Fe, our calculated $T_C$ is suppressed to the much lower value 1330K. Comparing with Fig.1, one can see the additional suppression of $T_C$ with $E_c/E_0$ being considerably larger, thus indicating stronger MSRO than in the corresponding NN coupling case. However, the parameters in Ref. \cite{21} have been obtained in the long-wavelength approximation and can only describe a small MSRO in classical case. The inset of Fig.2 shows directly $\cos \theta_n = (S_i \cdot S_{i+k})/S^2$, giving the details of the QSE enhancement of MSRO between several neighboring spins.

The spin correlation length $\xi$ is often used to describe the strength of MSRO. Despite the magnitude of $\cos \theta_n$, $\xi$ always tends to infinity when temperature approaches $T_C$ from above, so near $T_C$ $\xi$ may not be a parameter that properly reflects MSRO. Above $T_C$ the evaluation of $\xi$ in our formalism is straightforward from the long-wavelength behavior of the spin-correlation function $C^q \sim 1/(q^2 + \xi^2)$. We found that at fixed $T/T_C$, $\xi$ always increases as $S$ becomes smaller, in contrast to the SM where $\xi$ is independent of $S$. At $T = 1.1 T_C$, $\xi = 4.1, 3.7$ and 2.8 for $S = 1/2, 1$ and $\infty$, again demonstrating the QSE enhancement of MSRO from another perspective.

Now let us analyze the SW excitations. In the standard magnetism theories such as the random phase approximation \cite{22} and its various modified versions \cite{23}, SW exist due to the magnetic long-range order, so its spectrum is renormalized to zero at $T_C$. In our formalism SW comes from the short-range spin correlations and the long-range order is no longer a prerequisite for its existence, so SW spectrum can be finite at $T_C$. In Fig.2 we plot the calculated SW spectrum obtained from Eq. (6) at $T_C$. To demonstrate the $S$-dependence of the SW renormalisation, the SW spectrum at $T = 0$ (the FM case) is also plotted with all $\omega_q$ scaled by $S$.

Let us estimate the renormalisation factor in the BCC Fe \cite{2} where SW modes have been observed above the middle of the Brillouin zone along the (110) direction, $Q = (\pi/2, 0)$ (lattice constant $a = 1$). The SW renormalisation factors $\omega Q(T_C)/\omega_0^Q$ ($\omega_0^Q$ is $\omega_Q$ at $T = 0$) are 0.86, 0.76 and 0.60 for $S = 1/2, 1$ and $\infty$, correspondingly. Experimentally in the BCC Fe $\omega Q(T_C)/\omega_0^Q(0.3T_C) \approx 0.84$ \cite{2} and the difference between $\omega_Q$ and $\omega_0^Q(0.3T_C)$ is about 15\% \cite{2}, so the overall SW renormalisation factor becomes 0.71, and our result for $S = 1$ is close to that. Fig. 2 also indicates that $\omega_q$ for smaller spins is less affected at elevated temperatures, implying that QSE favors the persistence of SW modes.

Let us now estimate the influence of dynamic effects and obtain the relaxation function $F(q, \omega)$. Among vari-
ous analytical approximations for $F(q, \omega)$ the three-pole approximation \[23\] seems to be one of the best and it has been successfully applied to the typical Heisenberg system with large spin $S = 7/2$ \[24\]. In this approximation $F(q, \omega)$ is expressed in terms of $\delta_q^0 = (\omega^2)_q$ and \(\delta_q^2 = (\omega^4)_q/\delta_q^2 - \delta_q^0\), where $(\omega)^s_q$ are frequency moments of $F$ depending on the static correlation. The evaluation of $\langle \omega^2 \rangle_q$ is straightforward \[22\]. $\langle \omega^4 \rangle_q \propto \langle (S^z_q)^2 S^z_{-q} \rangle$ \[20\] contains four-spin correlation terms which have to be properly decoupled as a product of two-spin correlations. In the literature the conventional decoupling $(S^+_i S^+_j S^-_m S^-_n)$ $\rightarrow$ $C_{ij}C_{im}C^*_{jm}$ and $(S^+_i S'^-_j S'^-_m S'_n)$ $\rightarrow$ $C_{im}C_{ij} + C_{ij}C_{lm}$, appropriate for large $S$, have been applied to obtain $\langle \omega^4 \rangle_q$ \[23\]. For small $S$, the spin kernel effect, which is neglected in this decoupling, becomes important. This QSE can be clearly seen in $S = 1/2$ case, where for $i = l$ or $m = j$ the left side of the decoupled equation vanishes while the right side is finite. To take into account this QSE we introduce the following decoupling procedure

\[
\langle (S^z_i S^z_j) \{ S^z_m S^z_n \} \rangle \rightarrow f^r_{il} f^s_{ij} C_{ij} C^*_{im} \quad \text{for } R_{il} \leq R_{im}, R_{ij}, \] \[
\langle (S^+_i S^+_j S^-_m S^-_n) \rangle \rightarrow f^r_{ij} f^s_{im} f^s_{jm} C_{ij} + f^r_{ij} f^s_{im} C_{ij} + C_{ij} C_{lm} + (\delta_{ij} \delta_{lm} + \delta_{im} \delta_{lj}) C_{il} C^*_{im}, \] \(6\)

where $f^r_{il} = 1 - \delta_{il}/2S$. If $i, l, m$ and $j$ are different sites then Eq. \(6\) is the same as in the conventional decoupling. QSE occurs when two or more out of these four sites are the same. In this case Eq. \(6\) at $S = 1/2$ is exact and is reduced to the conventional decoupling for $S \rightarrow \infty$. With these results for two opposite limits of $S$ and the introduced earlier quantum correction in $f^r_{il} \sim 1/S$, one can expect that Eq. \(6\) will be a reasonable interpolation for arbitrary $S$. By applying this decoupling procedure one can obtain $\langle \omega^4 \rangle_q = \langle \omega^{4(0)}_q \rangle + \langle \omega^{4(1)}_q \rangle$, where $\langle \omega^{4(0)}_q \rangle$ corresponds to the conventional decoupling \[23\] while $\langle \omega^{4(1)}_q \rangle$ is the quantum correction given by

\[
\langle \omega^{4(1)}_q \rangle = \frac{1}{4S^3} \sum_n \left\{ \frac{1}{N} \sum_k [Jk(4g^2_k - 6g_k q_k + k) + 2g^2_{k+q}] \right\} \] \[-C^k (h_k - h_{k+q}) (13J^k - 7J^{q+k}) \] \[-(11g_0 - 9g_q)(h_0 - h_q) \] \[+ S^{-1} \sum_n z_n J^0_n C_n (1 - \gamma^0_q) (5C_n + 7C_0 - 6S)] \] \(7\)

where $\chi_q$ is the $q$-dependent susceptibility, $g_k = \sum_n z_n J^0_n C_n z_n$ and $h_k = \sum_n z_n J^2_n C_n z_n$. As a function of $\omega$ the relaxation function $F(q, \omega)$ has either one maximum at $\omega = 0$, if $\delta_q^2 > 2\delta_q^0$, or three maxima at $\omega = 0$ and $\omega = \pm \omega_{q, \text{max}}$, if $\delta_q^2 < 2\delta_q^0$. The latter case is often referred as the SW peak at $\omega_{q, \text{max}}$ \[24\] 25. With such a definition the criteria of the SW existence for given $q$ is $\delta_q^2/\delta_q^0 < 2$. Usually $\omega_{q, \text{max}}$ is slightly larger than $\omega_q$. In the literature the SW peak was also defined as $\langle \omega^2 \rangle_q$ \[27\] which is slightly smaller than $\omega_q$. Near the critical value $\delta_q^2/\delta_q^0 < 2$, the maximum of $F$ at $\omega_{q, \text{max}}$ is broad. When $\delta_q^2/\delta_q^0$ is decreased, the SW peak is more pronounced. In Fig. 3 we plot the magnitude of $\delta_q^2/\delta_q^0$ for different $S$ as a function of $q$ at $T_C$. At fixed $q$, $\delta_q^2/\delta_q^0$ is always decreased if $S$ becomes smaller. Along the $(q, q)$ direction, the critical values of $q$, when $\delta_q^2/\delta_q^0 = 2$, are $q_c \approx 0.30\pi, 0.51\pi$ and $0.61\pi$ for $S = 1/2, 1$ and $\infty$, correspondingly. Our value of $q_c$ for $S = 1$ agrees with the experiment result in BCC Fe, where SW modes above $T_C$ exist only above $q \sim \pi/2$ in (110) direction (Fig. 2 of Ref. \[2\]). The SW peaks were also obtained in the SM\[7\] (spin independent $\delta_q^2/\delta_q^0$), but the value of $q_c$ is considerably higher. Our calculations indicate that this theory, which correspond to $S = \infty$, will be applicable if QSE is properly taken into account. At $q = (\pi/2, 0)$ for $S = 1/2, 1$ and $\infty$ the ratio $\delta_q^2/\delta_q^0$ is approximately $0.93, 2.2$ and $3.6$, which are respectively well below, close to, and well above the critical value $\delta_q^2/\delta_q^0 = 2$. The corresponding dynamic structure factor $S(q, \omega) = \omega (1 - e^{-\omega/T_c})^{-1} \chi_q F(q, \omega)$ as a function of $\omega$ is shown in the inset of Fig. 3. It is clear that at this $q$ the well-defined SW exist in the case of $S = 1/2$, the tendency of SW appears for $S = 1$, and there is no SW signal at all for $S = \infty$. Fig. 3 shows that QSE favors the persistence of SW with increasing impact for smaller spins. In many real magnets $S$ is not large ($S \approx 1$ in BCC Fe and $S \approx 0.3$ in FCC Ni) and we believe that QSE plays an important role in the MSRO and the magnetic excitations above $T_C$, especially in the itinerant magnets.

In conclusion, we analytically demonstrated the presence of MSRO in the Heisenberg model and identified the importance of quantum spin effect on MSRO for ferromagnets above $T_C$. By extending the second-order Green’s function technique to arbitrary $S$ we found that for a system of small spins the quantum effects greatly contribute to the MSRO and enhance its influence. The spin dynamics investigation developed from the conventional method of moments further confirms that QSE favors the persistence of spin wave excitations. We demonstrated that this previously neglected QSE removes the long-standing controversy between theory and experiment regarding the presence of MSRO and SW in Fe and Ni above $T_C$ and clearly indicates that the current prevailing point of view of finite temperature magnetism should be reconsidered to properly include MSRO and quantum effects.

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Fig.1. $T_C/T_{C}^{MF}$ and $E_C/E_0$ as a function of $S$ from SM (lines), the HTE methods (close symbols) and our formalism (open symbols) in BCC structure in the NN coupling case.

Fig.2. The calculated SW spectrum $\omega_\mathbf{q}$ for the different $S$ at $T_C$. The dashed line is $\omega_\mathbf{q}$ at $T = 0$ (FM case). The inset shows $cos \theta_n$ from nearest to fifth-nearest neighbors.

Fig.3. The calculated $\delta_\mathbf{q}^2/\delta_\mathbf{q}^1$ for the different $S$ at $T_C$ as a function of $\mathbf{q}$. The criteria of SW $\delta_\mathbf{q}^2/\delta_\mathbf{q}^1 = 2$ is marked by the dashed line. The inset shows $S(\mathbf{q}, \omega)$ at $\mathbf{q} = (\frac{\pi}{2}, \frac{\pi}{2})$. 

4
(a) 

(b) 

\[ \frac{T_C}{T_{C_{MF}}} \]

\[ \frac{E}{E_0} \]

SM
