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Quasiclassical Equation of Motion for the Heisenberg Spin System*

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Previous results of quasiclassical spin dynamics are extended by presenting a new approximation for quantum corrections to the classical equation of motion for the Heisenberg model. The accuracy of the equation with these corrections is determined by comparing computer calculations based on this formalism with exact quantum and classical results for spin pair correlation functions for a six-spin linear chain at infinite temperatures.

I. INTRODUCTION

The dynamical behavior of the Heisenberg spin system, described by the Hamiltonian

$$H = -\sum_{i \neq i'} J_{ii'} \vec{S}_i \cdot \vec{S}_{i'} , \tag{1}$$

has received a great deal of attention. Dynamical quantities of interest are, for example, spin pair correlation functions of the form

$$\langle S_{i\alpha}(0) S_{I\beta}(t) \rangle = \frac{\mathrm{Tr} e^{-\beta H} S_{i\alpha}(0) S_{I\beta}(t)}{\mathrm{Tr} e^{-\beta H}} . \tag{2}$$

The spin pair correlation functions defined in Eq. (2) are directly related to the neutron scattering cross section.[1] Another quantity of interest which is proportional to the neutron cross section is the Fourier transform of the spin pair correlation function:

$$S^{\alpha\beta}(\vec{K}, \omega) = \frac{1}{2\pi} \int_{-\infty}^{\infty} dt\, e^{-i\omega t} \sum_{i,I} e^{i\vec{K}\cdot(\vec{r}_I - \vec{r}_i)} \langle S_{i\alpha}(0) S_{I\beta}(t) \rangle . \tag{3}$$

In recent years, the technique of computer simulation has been applied to the classical Heisenberg model, and several interesting results have been obtained.[2-6] It is well known that the classical approximation is valid in the large spin limit.[7] However, for finite spin j, the validity of the classical approximation is not understood very well.

In a previous paper[8] (which will be referred to as I), we have employed the Wigner formalism[9,10] to obtain quantum corrections to the classical spin dynamics. These quantum corrections derived in I can be divided into two types. The first results from approximating the thermal average as an integration over a classical distribution of the form $e^{-\beta H_{\mathrm{cl}}}$. The second is a quantum correction to the classical equation of motion.

These two corrections will be discussed separately in Secs. II and III. In Sec. IV, we present computer calculations to test the accuracy of the quasiclassical equation of motion.

II. THERMAL AVERAGE

In this section, for simplicity, we will consider only the spin-$\frac{1}{2}$ nearest-neighbor exchange model. In I, we obtained spin pair correlation functions in terms of a series in classical spin pair correlation functions. The result to second-order classical correlations is

$$\begin{aligned} \tfrac{1}{2}\langle S_{i\alpha}(0) S_{I\beta}(t) + S_{I\beta}(t) S_{i\alpha}(0) \rangle &= \langle \Omega_{i\alpha}\, \Omega_{I\beta}(t) \rangle_{\mathrm{cl}} \\ &- \tfrac{3}{4}(2\beta - \beta^2 J) \sum_{i'} J_{ii'} \langle \Omega_{i'\alpha}\, \Omega_{I\beta}(t) \rangle_{\mathrm{cl}} \\ &- \tfrac{3}{4}\beta^2 \sum_{i',i''} J_{ii'} J_{i'i''} \langle \Omega_{i''\alpha}\, \Omega_{I\beta}(t) \rangle_{\mathrm{cl}} + \cdots . \end{aligned} \tag{4}$$

In Eq. (4), classical pair correlation functions are defined as

$$\langle \Omega_{i\alpha}\, \Omega_{l\beta}(t)\rangle_{\rm cl} = j(j+1)\, \frac{\int d\vec{\Omega}_1 \cdots d\vec{\Omega}_N\, e^{-\beta H_W}\, \Omega_{i\alpha}\, \Omega_{l\beta}(t)}{\int d\vec{\Omega}_1 \cdots d\vec{\Omega}_N\, e^{-\beta H_W}} \ , \tag{5}$$

where

$$H_W = -j^2 \sum_{i\neq i'} J_{ii'}\, \vec{\Omega}_i \cdot \vec{\Omega}_{i'} \tag{6}$$

and $\vec{\Omega}_i$ is a unit vector.

In the classical approximation, the right-hand side of Eq. (4) is simply replaced by $\langle \Omega_{i\alpha}\, \Omega_{l\beta}(t)\rangle_{\rm cl}$. The quantum correction terms in Eq. (4) have a rather complicated appearance. However, the physical implications of these terms can be more clearly seen if we examine the contribution of these terms to $\mathcal{S}^{\alpha\beta}(\vec{\kappa}, \omega)$ in Eq. (3).

We note that $\mathcal{S}^{\alpha\beta}(\vec{\kappa}, \omega)$ is related to the Fourier transform of spin pair correlation functions by the relationship

$$\mathcal{S}^{\alpha\beta}(\vec{\kappa}, \omega) = \frac{2\coth(\frac{1}{2}\beta\hbar\omega)}{1-e^{-\beta\hbar\omega}}\, \frac{1}{2\pi} \int_{-\infty}^{\infty} dt\, e^{-i\omega t} \times \sum_{i,l} e^{i\vec{\kappa}\cdot(\vec{r}_l - \vec{r}_i)}\, \tfrac{1}{2}\langle S_{i\alpha}(0) S_{l\beta}(t) + S_{l\beta}(t) S_{i\alpha}(0)\rangle \ . \tag{7}$$

Now substituting the Fourier transform of Eq. (4) into Eq. (7), we obtain

$$\mathcal{S}^{\alpha\beta}(\vec{\kappa}, \omega) = \frac{2\coth(\frac{1}{2}\beta\hbar\omega)}{1-e^{-\beta\hbar\omega}}\, \mathcal{S}^{\alpha\beta}_{\rm cl}(\vec{\kappa}, \omega) \times [1 - \tfrac{3}{2}\beta J \xi_1(\vec{\kappa}) - \tfrac{3}{4}\beta^2 J^2 \xi_2(\vec{\kappa}) + \cdots] \ , \tag{8}$$

where

$$\mathcal{S}^{\alpha\beta}_{\rm cl}(\vec{\kappa}, \omega) = \frac{1}{2\pi} \int_{-\infty}^{\infty} dt\, e^{-i\omega t} \sum_{i,l} e^{i\vec{\kappa}\cdot(\vec{r}_l - \vec{r}_i)} \langle \Omega_{i\alpha}\, \Omega_{l\beta}(t)\rangle_{\rm cl} \ , \tag{9}$$

$$\xi_1(\vec{\kappa}) = \sum_{(i')} e^{i\vec{\kappa}\cdot(\vec{r}_{i'} - \vec{r}_i)} \ , \tag{10}$$

$$\xi_2(\vec{\kappa}) = \sum_{(i',i'')} e^{i\vec{\kappa}\cdot(\vec{r}_{i''} - \vec{r}_i)} - \xi_1(\vec{\kappa}) \ . \tag{11}$$

In Eqs. (10) and (11), the summation on i' is restricted to the nearest neighbors of i and the summation on i'' is restricted to the nearest neighbors of i' such that $i'' \neq i$.

It is clear from Eq. (8) that the essential dynamical features of $\mathcal{S}^{\alpha\beta}(\vec{\kappa}, \omega)$ which can be seen in its ω dependence are determined by $\mathcal{S}^{\alpha\beta}_{\rm cl}(\vec{\kappa}, \omega)$. The corrections appear as a multiplicative factor which depends only upon $\vec{\kappa}$. One is tempted then to conclude that, apart from the detailed balance factor $\coth(\frac{1}{2}\beta\hbar\omega)/(1-e^{-\beta\hbar\omega})$, the ω dependence of $\mathcal{S}^{\alpha\beta}(\vec{\kappa}, \omega)$ is determined by the classical equations of motion.

However, the equation satisfied by $\vec{\Omega}_l(t)$ is not the usual classical equation of motion. One must add correction terms to the classical equation for $\vec{\Omega}_l(t)$, as we shall see in Sec. III.

III. QUASICLASSICAL EQUATION OF MOTION

The corrections to the classical equation of motion for arbitrary spin j were obtained in I as a power series in t;

$$\frac{d}{dt}\, \vec{\Omega}_l(t) = \frac{2j}{\hbar} \sum_{l'} J_{ll'}\, \vec{\Omega}_l(t) \times \vec{\Omega}_{l'}(t) + \vec{\gamma}_l(t) \ , \tag{12}$$

where

$$\vec{\gamma}_l(t) = -\frac{2jJt}{\hbar^2} \sum_{l'} J_{ll'} [\vec{\Omega}_l(t) - \vec{\Omega}_{l'}(t)] \times [1 - \vec{\Omega}_l(t) \cdot \vec{\Omega}_{l'}(t)] + O(t^2) \ . \tag{13}$$

Retaining only the first term in the right-hand side of Eq. (13) should be sufficient for short times, say $jJt/\hbar < 1$. However, for large t, the short-time approximation for $\vec{\gamma}_l(t)$ does not apply. In fact the linear t dependence of our approximation to $\vec{\gamma}_l(t)$ causes our approximation to diverge at large t. To avoid this spurious effect of the linear time dependence of our approximation to the quantum correction terms for the long-time behavior, we now make a different approximation for $\vec{\gamma}_l(t)$.

To estimate $\vec{\gamma}_l(t)$, we can evaluate $(d/dt)\, \vec{\gamma}_l(t)$ at $t=0$ and then replace the $t=0$ argument by t. Then, neglecting higher order terms, we obtain

$$\frac{d}{dt}\, \vec{\Omega}_l(\tau) = \sum_{l'} \vec{\Omega}_l(\tau) \times \vec{\Omega}_{l'}(\tau) - \frac{1}{2j} \int_0^{\tau} d\tau' \sum_{(l')} [\vec{\Omega}_l(\tau') - \vec{\Omega}_{l'}(\tau')] \times [1 - \vec{\Omega}_l(\tau') \cdot \vec{\Omega}_{l'}(\tau')] \ , \tag{14}$$

where the scaled time τ is defined as

$$\tau = 2jJt/\hbar \ . \tag{15}$$

The quantum correction term in Eq. (14) is of the order of $1/j$, which vanishes in the large j limit and hence correctly reproduces the classical equation of motion. Of course, higher-order corrections can be written in terms of multiple integrals of the form

$$\int_0^{\tau} d\tau_1 \int_0^{\tau_1} d\tau_2 \cdots \int_0^{\tau_{n-1}} d\tau_n f(\vec{\Omega}_1(\tau_n) \cdots \vec{\Omega}_N(\tau_n)) \ . \tag{16}$$

We should hasten to note that the principal reason for introducing the more complicated corrections in Eq. (14) is that it does not suffer from spurious divergence problems at large t. Clearly we have no reason to expect that this correction will accurately predict $\vec{\Omega}_l(\tau)$ for values of τ greater than unity. We still only have an approximation

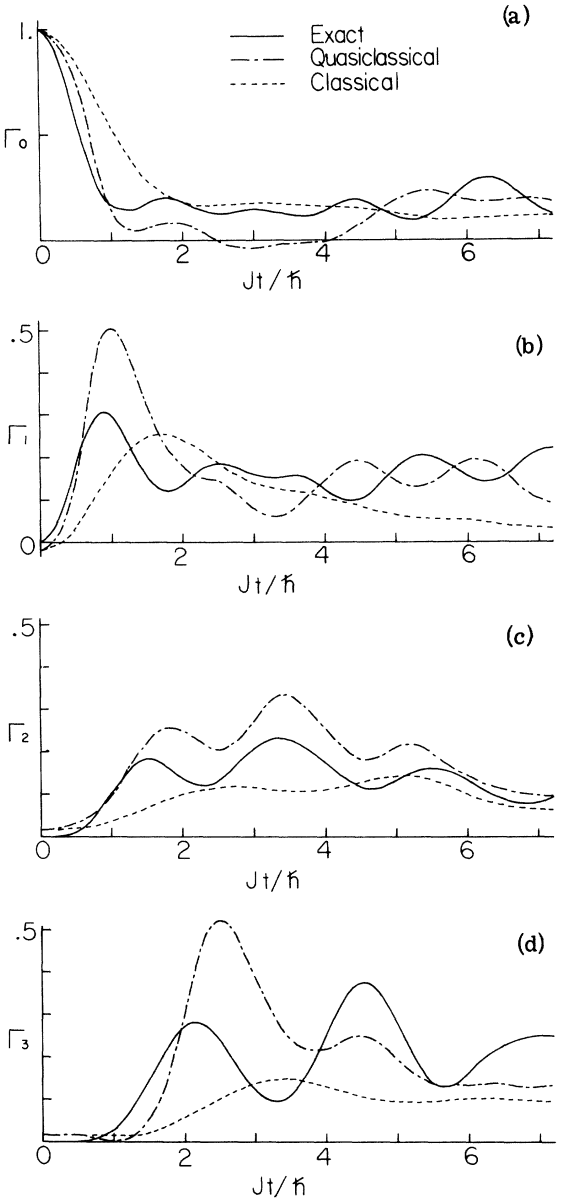

FIG. 1. Comparison of exact quantum correlation functions with classical and quasiclassical approximations for a spin-$\frac{1}{2}$ Heisenberg linear chain with six spins at infinite temperatures. $\Gamma_n = \langle \vec{S}_0 \cdot \vec{S}_n(t) \rangle / \langle \vec{S}_0 \cdot \vec{S}_0 \rangle$.

that is accurate for small τ. It is simply not divergent at large τ.

IV. COMPUTER CALCULATION

Eqs. (8) and (14) establish an explicit relationship between quantum and classical spin dynamics. This relationship provides us with a technique to perform quantum calculations in terms of "classical" variables. This classical procedure has the advantage that using it one can easily perform numerical calculations with the aid of a computer.

Let us now consider a spin-$\frac{1}{2}$ linear chain containing six spins. We employ Eq. (14) to compute spin pair correlation functions for this system at infinite temperatures. The reason for the choice of a small system is that exact quantum calculations are easily accessible for the comparison with our approximation. Hence the results obtained in this section are mainly aimed at determining the accuracy of the quasiclassical equation of motion in Eq. (14).

In Fig. 1, we compare quantum correlation functions with quasiclassical and classical results. The procedure of the computation for quasiclassical and classical results is essentially the same as that employed by Windsor.[2] Here the results were averaged over 100 randomly chosen sets of initial $\vec{\Omega}$'s. Of course we used the same initial sets for both classical and quasiclassical cases.

From Fig. 1 we can see that the short-time behavior is much better described by the quasiclassical approximation than the classical approximation, and so is the over-all structure of correlation functions. For a particular set of initial $\vec{\Omega}$'s the classical equation of motion gives a very smooth time dependence for $\vec{\Omega}_i(t)$, which can be characterized by a precessional period T of a spin in the effective field of neighboring spins,

$$T = 2\pi\hbar/zJ \ , \tag{17}$$

where $z = 2$ is the number of nearest neighbors.

This smooth motion for $\vec{\Omega}_i(t)$ clearly cannot yield the detailed structure occuring in the quantum correlation functions as shown in Fig. 1. This structure in the correlation functions has much shorter "periods" than the precessional period given in Eq. (17). Of course we expect this detailed structure to be less evident for a large system. However, this feature should result from the averaging process due to the interactions among many particles in the system, and not from the smooth motion of each of the $\vec{\Omega}$'s. Quite simply the classical equation for $\vec{\Omega}_i(t)$ gives much too smooth a time dependence.

To estimate the accuracy of the quasiclassical equation of motion, we have chosen a case where the quantum corrections would be dominant, namely, the spin-$\frac{1}{2}$ case and a small number of spins. Examining the figures we can see that the quasiclassical results are not too bad for short times and the first few moments. We should note here the failure of the classical and quasiclassical Γ_n's for $n > 0$ to go to zero at $t = 0$. This is due to the fact that our 100 initial sets of $\vec{\Omega}$'s are not large enough to fulfill this condition.

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de Haas-van Alphen Effect in Antiferromagnetic Cr-Rich Alloys*

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de Haas–van Alphen measurements have been made on Cr-rich alloys with up to 2-at.% V and 1-at.% Mn. In the crystals in which the antiferromagnetic wave vector $\vec{q}$ is incommensurate with a reciprocal-lattice vector, the changes in three of the frequencies associated with hole ellipsoids at N are consistent with changes in $\vec{q}$, rather than with a geometrical change in the size of the hole ellipsoids upon alloying. In the crystal in which $\vec{q}$ is commensurate with a reciprocal-lattice vector, several frequency branches were observed. The angular dependence of these branches is consistent with simple ellipsoid models of the Fermi surface.

I. INTRODUCTION

In recent years the antiferromagnetism of Cr has been extensively studied and the experimental results are in agreement with an itinerant electron model of antiferromagnetism. In this model the electron gas acts collectively to produce a spatially varying spin density with wave vector $\vec{q}$ which in general is not commensurate with a reciprocal-lattice vector.

The effects on the physical properties of Cr due to alloying V, Mn, and other metals with Cr have also been studied. The effect on the Néel temperature T_N, for example, due to alloying V, Mn, and Mo with Cr is shown in Fig. 1. It is seen that V decreases the Néel temperature while Mn increases T_N.[1-11] These two metals have a similar effect on the magnitude of $\vec{q}$.[8] The changes in T_N and $\vec{q}$ have been interpreted as arising from a modification of the Fermi surface (FS) of Cr due to alloying.[1] Since, as shown in Fig. 1, as little as 4-at.% V in Cr lowers T_N below 4 K, one may be able to observe the modified FS of Cr in the paramagnetic state.

Lomer[12,13] first proposed a model of the FS of Cr and suggested that the anitferromagnetic state was stablized by flat portions of the FS. Loucks[14] calculated the FS of Cr using an augmented-plane-wave method, and his result for the (100) plane is shown in Fig. 2. Loucks did not find hole ellipsoids at N (shown by dashes in Fig. 2), although experimental evidence[15] indicates they are present. The flat portions of the hole octahedron and body of the electron jack are separated by $q = (2\pi/a)(1-\delta)$, $\delta \sim 0.05$.

One method used to determine the FS of a metal is the de Haas–van Alphen (dHvA) effect. dHvA measurements have been made on pure Cr in a single-$\vec{q}$ state.[15] Experiments have shown that usually there are three spin-density wave vectors present in a Cr crystal. However, if the crystal is cooled through T_N in the presence of a large magnetic field (field cooling) the crystal will contain only one $\vec{q}$.[16] The resulting dHvA spectrum obtained from a crystal with single $\vec{q}$ is quite complex, and not all of the observed branches in the spectrum have been interpreted. dHvA measurements in Cr-alloy crystals[17] will provide more insight into the FS and antiferromagnetic state of Cr. In the present paper, we report the first detailed FS measurements on a series of antiferromagnetic Cr-V and Cr-Mn alloys.

II. EXPERIMENT

The dHvA samples were prepared from single- or