

Time correlation functions and dynamical properties of a Heisenberg fluid

This article has been downloaded from IOPscience. Please scroll down to see the full text article.

2003 J. Phys.: Condens. Matter 15 S83

(<http://iopscience.iop.org/0953-8984/15/1/309>)

View [the table of contents for this issue](#), or go to the [journal homepage](#) for more

Download details:

IP Address: 171.66.16.97

The article was downloaded on 18/05/2010 at 19:23

Please note that [terms and conditions apply](#).

Time correlation functions and dynamical properties of a Heisenberg fluid

I M Mryglod^{1,2,3}, I P Omelyan^{1,2} and R Folk²

¹ Institute for Condensed Matter Physics, National Academy of Sciences of Ukraine,
1 Svientsitskii Street, UA-79011 Lviv, Ukraine

² Institute for Theoretical Physics, Linz University, Altenbergerstr. 69, A-4040 Linz, Austria

E-mail: mryglod@icmp.lviv.ua

Received 14 October 2002

Published 16 December 2002

Online at stacks.iop.org/JPhysCM/15/S83

Abstract

The main aims of this paper are: (i) to present molecular dynamics (MD) results for time correlation functions, calculated with the help of a new symplectic algorithm that was proposed by us recently; (ii) to discuss the results obtained with the main focus on the interplay between the spin and liquid subsystems as well as on the influence of an external magnetic field on the properties of the system considered; (iii) to compare the time correlation functions, calculated in MD simulations, with theoretical predictions. Our results provide evidence of the interesting interplay of the two subsystems, causing new phenomena not found in nonmagnetic fluids and magnetic lattice models.

1. Introduction

One of the simplest models of a continuous system exhibiting ferromagnetic behaviour is a spin fluid with an isotropic Heisenberg-like interaction between spin degrees of freedom [1, 2]. Because of its simplicity as well as its very specific features, the theoretical study of both static and dynamical properties for this model are of general interest and can be considered as a test for any theory developed for an inhomogeneous liquid, by comparison with results of computer simulations. On the other hand, the Heisenberg model ferrofluid is of interest in its own right. Recently, it was demonstrated [3] that a $\text{Co}_{80}\text{Pd}_{20}$ melt can be undercooled below its Curie temperature. Hence, the first evidence of a ferromagnetic behaviour in a liquid metal was obtained under conditions where the Heisenberg exchange interaction absolutely dominates over the contribution of the magnetic dipole–dipole interaction.

The different phase diagrams of the continuum Heisenberg model, depending on the ratio of the strength of the exchange interaction to the spin-independent interaction, have been established within mean-field-like theory [1, 4–6] and using methods of integral equations or

³ Author to whom any correspondence should be addressed.

density functional theory [5–7]. More quantitative results have been obtained from Monte Carlo simulations [7, 8] for the case where the spin-independent interaction is of the hard-sphere type.

Until recently, the theoretical description of magnetic liquid dynamics was based to a great extent on phenomenological approaches (see, e.g., [9–12]). However, some of the results obtained in different approaches were contradictory. For example, one may note that the expressions for the sound velocity found within two main groups of phenomenological theories differ even qualitatively [11]. Hence, study of the hydrodynamic behaviour using a systematic rigorous statistical treatment of the problem became unavoidable.

In the series of our previous papers [13–15] the spectrum of hydrodynamic collective modes for an isotropic Heisenberg-like model of a ferrofluid at constant external magnetic field has been studied. In [13] we used a rigorous microscopic treatment for deriving the generalized transport equations and equations for the time correlation functions (TCFs). These equations were then analysed in the hydrodynamic limit [14] and explicit expressions for the static correlation functions in relation to the well-known thermodynamic quantities as well as expressions for the transport coefficients have been derived. These results were used for the calculation of the hydrodynamic collective mode spectrum [14] and the derivation of analytical expressions for all the hydrodynamic TCFs, constructed on the basis of the conserved dynamical variables [15]. We emphasize that the expressions obtained are asymptotically exact in the small- (k, ω) limit, and all input parameters in such expressions are just thermodynamic quantities and hydrodynamic transport coefficients, so these results have a wider range of application than just the Heisenberg ferrofluid.

The goal of this paper is to discuss in more detail the specific features of the dynamical behaviour of a Heisenberg spin fluid. To achieve this goal, we performed molecular dynamics (MD) simulations and calculated the correlation functions of interest. We finally compare the results obtained with the predictions of theory.

2. Theory and computer simulations

We consider a classical system composed of N magnetic particles of mass m , described by the Hamiltonian

$$H = \sum_{i=1}^N \frac{mv_i^2}{2} + \sum_{i<j}^N [V(r_{ij}) - J(r_{ij})\mathbf{s}_i \cdot \mathbf{s}_j] - h \sum_{i=1}^N s_i^z, \quad (1)$$

where r_i and v_i are the translational position and velocity, respectively, of particle i carrying spin \mathbf{s}_i . The liquid potential is denoted by $V(r_{ij})$, and $J(r_{ij}) > 0$ is the exchange integral for a pair of spins with interparticle distance r_{ij} . The classical approach treats \mathbf{s}_i as a three-component continuous vector with a fixed length for each site i . We put for convenience $|\mathbf{s}_i| = 1$, so J is measured in energy units. The last term in (1) describes the interaction with an external homogeneous magnetic field h .

Because the exchange integral $J(r_{ij})$ depends on an interparticle distance r_{ij} , one can expect a strong influence of the spin subsystem on liquid properties of the model (1). For instance, in [16] it has been shown that the position of the ‘gas–liquid’ critical point depends on the value of an external magnetic field. More recently, this problem has been studied in detail in the papers [17–19]. One of our previous findings [15] was the prediction of a strong *dynamic* interplay between the spin and translational degrees of freedom in the spin ordered phase, which can be observed as the appearance of side sound-like peaks in the *magnetic* dynamic structure factor. In particular, this prediction was validated in our recent paper [20], where TCFs of the model (1) were studied in the ferromagnetic state using MD simulations.

The most interesting TCFs are the ‘density–density’ $F_{nn}(k, t)$ and ‘spin density–spin density’ $F_{mm}^{\alpha\beta}(k, t)$ TCFs. The Fourier transform of the first one gives us the dynamic structure factor $S(k, \omega)$, and the second function could be related to the magnetic dynamic structure factor $S_m(k, \omega)$. To be more explicit we have to note that, because of the coupling between spin and liquid subsystems, $S_m(k, \omega)$ is expressed via the Fourier transform of TCF $F_{ss}(k, t)$, where the dynamic variable \hat{s}_k can be written as a linear combination of two density operators, namely, the z -component of the spin density \hat{m}_k^z and the particle number density \hat{n}_k (see, e.g., [14, 15]). In the hydrodynamic limit for $F_{nn}(k, t)$ and $F_{ss}(k, t)$ the following explicit expressions can be used:

$$\begin{aligned} F_{nn}^H(k, t)/F_{nn}(k) &= \sum_{\alpha} G_{\alpha}^{nn}(k) \exp\{-z_{\alpha}(k)t\}, \\ F_{ss}^H(k, t)/F_{ss}(k) &= \sum_{\alpha} G_{\alpha}^{ss}(k) \exp\{-z_{\alpha}(k)t\}, \end{aligned} \quad (2)$$

where the subscript α labels the types of hydrodynamic collective excitation ($\alpha = \{+, -, h, m\}$ corresponds to a pair of complex-conjugated sound excitations ($=, -$) heat (h) and spin (m) diffusion modes); G_{α}^{nn} and G_{α}^{ss} denote the weight coefficients, describing the α th-mode contribution to a relative TCF. Explicit expressions for the weight coefficient as well as for the Landau–Placzek ratios can be found in [15]. The most important point for our following discussion is that, if $h \neq 0$ or the fluid is in its ferromagnetic phase, the contributions from sound modes to the function $F_{ss}^H(k, t)$ are nonzero. In particular, we found that

$$G_{\pm}^{ss} = \frac{1}{2} \frac{(1 - \delta_T)}{\gamma_m} \left\{ 1 \mp ik \frac{b_{ss}}{v_s} \right\}, \quad (3)$$

where v_s , $\delta_T = \kappa_{T,m}/\kappa_{T,h}$, and $\gamma_m = C_{P,m}/C_{V,m}$ are the sound velocity and ratios of the isothermal compressibilities and the specific heats, defined in different ensembles, respectively. The value of b_{ss} depends on thermodynamic quantities and transport coefficients (for details see, e.g., [15]) and describes asymmetric contributions to the hydrodynamic TCF $F_{ss}^H(k, t)$. Note that in the paramagnetic case, when $h = 0$, one has $\delta_T = 1$, so the hydrodynamic sound modes do not contribute to $F_{nn}^H(k, t)$.

In our MD study of the Heisenberg fluid, we have used [20] the Yukawa potential,

$$J(r) = (\epsilon\sigma/r) \exp[(\sigma - r)/\sigma],$$

and a soft-core potential,

$$V(r) = \begin{cases} 4\epsilon[(\sigma/r)^{12} - (\sigma/r)^6] + \epsilon, & \text{if } r < 2^{1/6}\sigma \\ 0, & \text{if } r > 2^{1/6}\sigma, \end{cases}$$

for the description of spin and liquid interactions with the intensities ϵ and ϵ , respectively. The function $J(r)$ was truncated at $R = 2.5\sigma$ and shifted to be zero at the truncation point to avoid force singularities. The simulations were carried out for $N = 864$ particles (employing periodic boundary conditions) at a reduced density $n^* = N\sigma^3/V = 0.6$, a reduced core intensity $\epsilon/\epsilon = 1$, and a *dynamical coupling parameter* $d = \sigma(m\epsilon)^{1/2}/\hbar = 2$. This last parameter is, in fact, the ratio τ_{tr}/τ_s , where $\tau_{tr} = \sigma(m/\epsilon)^{1/2}$ and $\tau_s = \hbar/\epsilon$ are the characteristic time intervals of varying translational and spin variables, respectively. At the fixed density n^* , we have considered three thermodynamic points in our calculations, namely: the ferromagnetic phase (A) with a reduced temperature $T^* = k_B T/\epsilon = 1.5 < T_c^*$ and at zero magnetic field $h = 0$; the paramagnetic phase (B) with a reduced temperature $T^* = k_B T/\epsilon = 2.475 > T_c^*$ and at zero magnetic field $h = 0$; and the state (C) with spins ordered by a nonzero external field $h^* = h/\epsilon = 2.00$ at $T^* = k_B T/\epsilon = 2.479 > T_c^*$. (Here, $T_c^* \simeq 2.06$ denotes the temperature of the ferromagnetic phase transition [21].) The results, obtained for the point (A) were partly reported in [20].

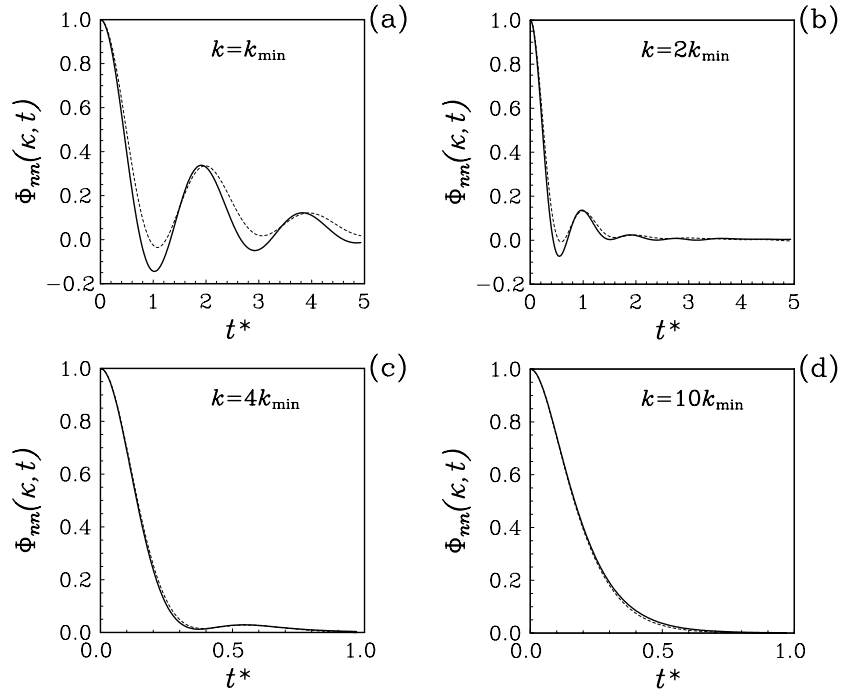


Figure 1. The normalized ‘density–density’ TCF $\Phi_{nm}(k, t) = F_{nm}(k, t)/F_{nm}(k)$ obtained in the MD simulations for four values of the wavenumber: (a) $k = k_{min}$; (b) $k = 2k_{min}$; (c) $k = 4k_{min}$; (d) $k = 10k_{min}$. $k_{min} = 2\pi/V^{1/3}$ denotes the value of a minimal wavenumber achieved in our MD study. Results obtained for the case $h \neq 0$ are shown by the dashed curves.

MD simulations have been performed with the help of a new symplectic time-reversible algorithm [20], developed recently for numerical integration of the equations of motion in magnetic liquids. This algorithm exactly conserves spin lengths, is stable, and can be used with much larger time steps than those inherent in standard predictor–corrector schemes.

3. Results and discussion

From the MD run we can obtain the positions $r_i(t)$, the velocities $v_i(t)$, and the values of spins $s_i(t)$ at times t fixed by the condition $t = n\tau$ with $n = 1, 2, \dots, n_{max}$. This allows one to calculate all the quantities needed. For instance, if one defines dynamic variables

$$\hat{n}_k(t) = \frac{1}{\sqrt{N}} \sum_{i=1}^N \exp(ik \cdot r_i(t)), \quad \hat{m}_k^\alpha(t) = \frac{1}{\sqrt{N}} \sum_{i=1}^N s_i^\alpha(t) \exp(ik \cdot r_i(t)), \quad (4)$$

being the particle number density and spin densities, respectively, we can construct on the basis of these several correlation functions that are commonly of interest both for theory and computer simulations.

Note that in the case of a nonzero value of an external field one has to distinguish the component \hat{m}_k^z , describing the fluctuations along the direction of a magnetic field h , and the transverse components $\hat{m}_k^{(t)} \equiv \hat{m}_k^x \equiv \hat{m}_k^y$. In the case $h = 0$, all the components are equivalent because of the magnetic isotropy of the system.

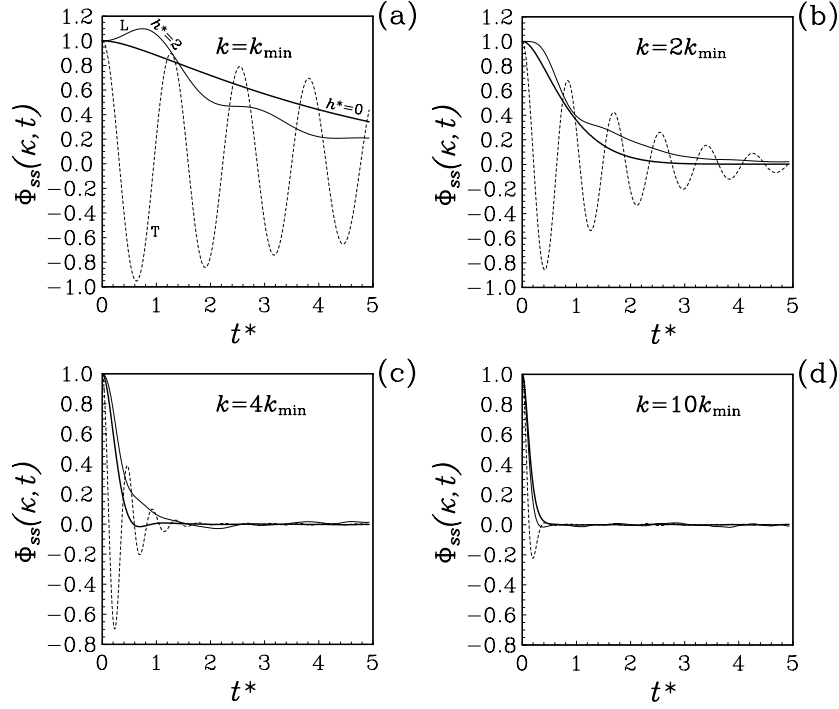


Figure 2. The normalized ‘spin density–spin density’ TCF $\Phi_{ss}(k, t) = F_{ss}(k, t)/F_{ss}(k)$ obtained in the MD simulations for four values of the wavenumber k . Results found for a paramagnetic case (the point (B)) are shown by bolder solid curves.

In [14, 15] we have derived a few useful relations that can be used as definitions of generalized k -dependent thermodynamic quantities. For example, the generalized magnetic susceptibility $\chi_{T,n}(k)$ per particle can be introduced as follows:

$$f_{ss}(k) = \langle \hat{s}_k \hat{s}_{-k} \rangle = \frac{1}{\beta} \chi_{T,n}(k) |_{k \rightarrow 0} \rightarrow \frac{1}{\beta} \chi_{T,n} = k_B T \chi_{T,n}, \quad (5)$$

where $\hat{s}_k = \hat{m}_k^z - f_{mn}^l(k)/f_{nn}(k) \hat{n}_k$ is the spin density, orthogonalized with respect to \hat{n}_k . In fact, the expression (5) may be considered as an example of a generalized thermodynamic relation, well known in the theory of simple liquids [22]. Equilibrium TCFs $F_{ab}(k, t)$, constructed on the basis of the dynamic variables (4), can be calculated from the standard definition, namely, $F_{ab}(k, t) = \langle \hat{a}_k(t) \hat{b}_{-k}(0) \rangle$.

In figure 1 the numerical results for the normalized ‘density–density’ TCF $\Phi_{nn}(k, t) = F_{nn}(k, t)/F_{nn}(k)$, calculated at the thermodynamic points (B) (solid curves) and (C) (dashed curves), are presented. It is clearly seen that for small wavenumbers sound excitations are well defined and produce the oscillating behaviour of this function like for other simple fluids. Under the influence of an external field, the frequency of the sound modes is slightly shifted, and the corresponding functions go above the curves obtained for $h = 0$. This means that the integrated intensity of the central Rayleigh peak in the dynamic structural factor increases if the external field is nonzero, which is in agreement with our prediction [15].

Another kind of result can be observed for the normalized ‘spin density–spin density’ TCF $\Phi_{ss}(k, t) = F_{ss}(k, t)/F_{ss}(k)$ (see figure 2). As far as one can judge from figure 2, in this case an external field changes the time dependence of $F_{ss}(k, t)$ crucially and as a result we see:

- (i) the appearance due to anisotropy of two components of spin fluctuations with different time behaviour (the transverse $F_{ss}^T(k, t)$ and longitudinal $F_{ss}^L(k, t)$ functions are shown in figure 2 by dashed and thin solid curves);
- (ii) oscillating behaviour of $F_{ss}^L(k, t)$ at small wavenumbers k with time periods close to those observed for $F_{nn}(k, t)$ (compare with figure 1).

These results, together with the calculations performed for the ferromagnetic case (A) (see [20]), have already demonstrated the interesting dynamic interplay between liquid and spin subsystems in a Heisenberg ferrofluid. A more complete presentation of our results (including the investigation of the collective mode spectrum and the calculations of the weight coefficients) will be given in a future paper [23].

Acknowledgments

Part of this work was supported by the Fonds zur Förderung der wissenschaftlichen Forschung under Project No P15247. IM and IO thank the Fundamental Researches Fund of the Ministry of Education and Sciences of Ukraine for support under Project No 02.07/00303.

References

- [1] Hemmer P C and Imbro D 1977 *Phys. Rev. A* **16** 380
- [2] Hoye J S and Stell G 1976 *Phys. Rev. Lett.* **36** 1569
- [3] Albrecht T, Bühner C, Fähnle M, Maier K, Platzek D and Reske J 1997 *Appl. Phys. A* **65** 215
- [4] Feijoo L, Woo C W and Rajan V T 1980 *Phys. Rev. B* **22** 2404
- [5] Tavares J M, Telo da Gama M M, Teixeira P I C, Weis J J and Nijmeijer M J P 1995 *Phys. Rev. E* **52** 1915
- [6] Weis J J, Nijmeijer M J P, Tavares J M and Telo da Gama M M 1997 *Phys. Rev. E* **55** 436
- [7] Lomba E, Weis J J, Almarza N, Bresme F and Stell G 1994 *Phys. Rev. E* **49** 5169
- [8] Nijmeijer M J P and Weis J J 1995 *Phys. Rev. Lett.* **75** 2887
- Nijmeijer M J P and Weis J J 1996 *Phys. Rev. E* **53** 591
- [9] Sliomis M I 1972 *Sov. Phys.-JETP* **34** 1291
- [10] Rosensweig R E 1985 *Ferrohydrodynamics* (Cambridge: Cambridge University Press)
- [11] Hubbard J B and Stiles P J 1986 *J. Chem. Phys.* **84** 6955
- [12] Henjes K and Liu M 1993 *Ann. Phys., NY* **223** 243
- [13] Mryglod I M, Tokarchyk M V and Folk R 1995 *Physica A* **220** 325
- [14] Mryglod I M and Folk R 1996 *Physica A* **234** 129
- [15] Mryglod I, Folk R, Dubyk S and Rudavskii Yu 2000 *Physica A* **277** 389
- [16] Vakarchuk I V, Rudavskii Yu K and Ponedilok G V 1984 *Theor. Math. Phys.* **58** 291
- Vakarchuk I V, Rudavskii Yu K and Ponedilok G V 1985 *Phys. Status Solidi b* **128** 231
- [17] Schinagl F, Iro H and Folk R 1999 *Eur. Phys. J. B* **82** 113
- [18] Lado F, Lomba E and Weis J J 1998 *Phys. Rev. E* **58** 3478
- [19] Sokolovska T G and Sokolovskii R 1999 *Phys. Rev. E* **59** R3819
- [20] Omelyan I P, Mryglod I M and Folk R 2001 *Phys. Rev. Lett.* **86** 898
- [21] Mryglod I M, Omelyan I P and Folk R 2001 *Phys. Rev. Lett.* **86** 3156
- [22] Hansen J-P and McDonald I R 1986 *Theory of Simple Liquids* (London: Academic)
- [23] Mryglod I M, Omelyan I P and Folk R 2003 *Phys. Rev. E* in preparation