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Quantum correlation effects in the spin dynamics of Gd at high temperatures in the light of complex dilation theory

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Recent experimental results on spin dynamics of Gd at high temperatures above T_c [J. W. Cable and R. M. Nicklow, Phys. Rev. B **39**, 11732 (1989)] are analyzed in the context of general quantum correlation theory of condensed disordered matter. The theory is based on the fermionic second-order density matrix in connection with complex dilation and thermalization. Assuming the validity of this theory, a confrontation with the aforementioned experimental data leads to the conclusion that the spatial extension of the quantum correlations characterizing the considered system is independent of temperature between $T=320$ and 850 K. The analysis further suggests that the dimension of the magnetically ordered regions is independent of T in the considered range.

The existence of propagating spin waves above T_c in ferromagnetic systems and their relation to magnetic short-range order is of current interest (see Refs. 1 and 2 and references cited therein). In particular, neutron inelastic scattering (cf. Ref. 1) and muon spin rotation (cf. Ref. 2) experiments give insight to this important physical process. Recently, it has been shown that many systems [e.g., rare-earth aluminides,[2] EuO, EuS, and Gd (Ref. 1)] clearly exhibit the existence of spin waves above T_c. Of particular importance for our following treatment are the very recent and precise experimental results[1] of neutron scattering on Gd (along the $\langle 110 \rangle$ direction), which clearly demonstrate a crossover from spin diffusion at small scattering wave vector q to damped spin-wave behavior at large q. A surprising result consists of the existence of spin correlations even at $T=850$ K, i.e., far above the Curie point $T_c=293$ K. It has been concluded[1] that, in Gd, it is not the thermal breakup of spin-pair correlations that destroys the spin waves for $T > T_c$.

Motivated by these findings (which appear to be of interest also for the general theory of microdynamical processes in condensed matter) we tried to apply the recent theory[3] of quantum correlations into the present physical context. In that work[3] (and with the aid of its very recent applications[4-6]) it was demonstrated from first principles that a new kind of quantum correlations may be related to the second-order reduced density matrix[7] $\Gamma^{(2)}$ of fermionic systems[8] after (i) proper "thermalization" (i.e., connecting the latter with the canonical ensemble of quantum statistics) and (ii) application of the complex dilation (or scaling) transformation[9-11] to the corresponding reduced second-order Hamiltonian. This treatment is intended to reveal general features of (possibly existing) quantum correlations in amorphous condensed matter, which also means that the explicit form of the actual Hamiltonian appears to be of less importance. The experimental relevance and the predictive power of the theory have been demonstrated through some applications concerning proton transfer and H^+ conductivity in water,[4] conductivity of molten alkali chlorides,[5] and quantum correlation effects in superconductors.[6]

After some short introductory remarks on the theory, we immediately apply the main formula (4) to the physical context and the experimental results[1] under consideration. The following results strongly indicate the existence of the specific quantum correlations[3-6] in the present physical context.

As mentioned above, the present theory has its origin in quantum statistics. In a more detailed setting the theoretical context is as follows. Using the suggestion of Penrose and Onsager[12] for $\Gamma^{(2)}$ corresponding to the so-called extreme case[8] of the matrix representation of $\Gamma^{(2)}$, one obtains the decomposition $\Gamma^{(2)}=\Gamma_L^{(2)}+\Gamma_S^{(2)}$. Here the first term describes the "large component" associated with off-diagonal long-range order (ODLRO) in the terminology of Yang[13] and the second term, the "small part," the background dissipation.[3] The next step is to introduce the theorems of dilation analyticity[9] allowing for a time-irreversible extension of quantum mechanics. The latter has the important property of producing (upon thermalization) an irreducible (Jordan block) pattern of $\Gamma_S^{(2)}$. The minimal size, $s_{\min}$, of this pattern shows the number of (paired) degrees of freedom "needed" to constitute a self-organized, so-called coherent dissipative structure with relaxation time $\tau_{\rm rel}$. A detailed derivation arrived at the relation

$$s_{\min}=\frac{4\pi k_B T}{\hbar}\tau_{\rm rel}\,, \tag{1}$$

giving the aforementioned minimal dimensionality $s_{\min}$, with k_B being the Boltzmann constant, T the temperature, and $\tau_{\rm rel}$ the characteristic decaying (relaxation) time. ($\tau_{\rm rel}$ is connected with the resonances being exposed after complex scaling.[9]) The crucial step, cf. Refs. 4-6, for the

comparison of the theory with experimental results is given by

$$d_{\min} = F(H_{\rm rel}) W_i^{\rm dB} s_{\min} , \quad (2)$$

showing the spatial dimension of a coherent-dissipative structure depending linearly on the thermal de Broglie wavelength

$$W_i^{\rm dB} = \hbar (2\pi / k_B T m_i)^{1/2} \quad (3)$$

and on the functional F, which relies on the relevant Hamiltonian $H_{\rm rel}$ (as well as some thermodynamical variables and/or external parameters). The quantity m_i is the effective mass of "particle i," in this case corresponding to the inertial mass associated with the motion of a particular magnetic cluster in the presence of other surrounding clusters. This mass depends on factors describing domain-wall motion; magnetic exchange and anisotropy energy. For more details on the derivations and underlying analysis that leads to Eqs. (1)–(3), we refer to earlier work.[4,5]

Before proceeding we emphasize the following fundamental points:

(i) We are speaking of quantum correlations at high temperatures. They are to be distinguished from those at low (or zero) temperatures.

(ii) Our formulation is based on the small component $\Gamma_S^{(2)}$ of $\Gamma^{(2)}$, see above, and hence it *does not refer to a wave function.*

(iii) Equation (1) is obtained from the thermalization of $\Gamma_S^{(2)}$, which in the present high-temperature regime is the relevant part of $\Gamma^{(2)}$.

(iv) One can prove the general decomposition $\Gamma^{(2)} = \Gamma_L^{(2)} + \Gamma_S^{(2)}$ under quite general conditions. The above (extreme case) decomposition is standard in condensed-matter theories (usually perceived in connection with superconductivity), but the present application leading to Eq. (1) in the region of high temperatures is, to our knowledge, new.

(v) The first part of the decomposition of $\Gamma^{(2)}$ above is the so-called *large part* and the second the *small component.* The large part obtains from a (macroscopic) wave function, and, e.g., quantum correlations associated with $\Gamma_L^{(2)}$ do not vanish for $T = 0$ K.

With Eqs. (1)–(3) one obtains

$$d_{\min} = \text{const} \times F(H_{\rm rel}) \sqrt{T} \tau_{\rm rel} . \quad (4)$$

In the following, we make the connection of the main formula (4) with the experimental results presented in Ref. 1. In this reference, the size of spin-ordered regions above $T_c = 293$ K making possible the appearance of short-wavelength spin waves as seen by neutron inelastic scattering is qualitatively represented by the inverse of the reduced crossover scattering wave vector ζ_c. The experimental findings, as given in the inset of Fig. 6 of Ref. 1, exhibit only a slight, nearly linear increase of ζ_c with temperature above T_c. This result is clearly in contrast with predictions of standard theories (cf. Refs. 1 and 2 for details).

Our present treatment is based on the assumption that coherent-dissipative structures[3–6] may properly describe the (short-ranged) magnetically ordered regimes above T_c, i.e., in the temperature region where the ferromagnetic long-range order is broken. The derived main formula (4) allows for the following treatment of the experimental data:

(a) From Fig. 6 of Ref. 1 we can extract the numerical values of the crossover wave vector ζ_c at the considered five different temperatures ($T = 293$, 320, 440, 586, and 850 K).

(b) The scattering results being parametrized in Tables I and II of Ref. 1 [on the basis of the so-called damped harmonic oscillator (DHO) model] allow us to determine the values of the damping parameter β_q corresponding to the crossover wave vector $\zeta_c(T)$ at the temperatures given; see Table I. Note that Tables I and II of Ref. 1 represent parametrizations of two different experimental series, which are based on slightly different final neutron energies (and different mean intensities I_0).

(c) To apply our theoretical result (4) we need explicitly the quantity $\tau_{\rm rel}$, i.e., the characteristic relaxation time of the microscopic quantum "units" (or systems) which contribute to the creation of a coherent-dissipative structure. In the present physical context, it seems reasonable to associate the inverse of the damping parameter β_q corresponding to ζ_c with this relaxation time, as β_q represents the "lifetime" of the magnetically ordered clusters (in the framework of the DHO model[1]). With this choice, i.e., $\tau_{\rm rel} = \beta_q^{-1}$, we calculate the quantity $\sqrt{T}/\beta_q$ as desired by Eq. (4); see our Table I.

(d) The aforementioned different parametrizations of Tables I and II of Ref. 1 contain data at $T_c = 293$ K. This allows us to determine two numerical values for the desired quantity $\sqrt{T}/\beta_q$ at T_c. The data at $T = 320$ K (cf. our Table I) are extracted from Table II of Ref. 1.

This treatment of the experimental data reveals a remarkable result that may be shortly stated by the following:

(e) The experimental facts show that the quantity $\sqrt{T}\tau_{\rm rel}$ (which is due to quantum correlations of general character and in our theory contributes to the size $d_{\min}$ of the magnetically ordered regions above T_c) is nearly independent of the temperature. (This is graphically presented in Fig. 1.)

Let us now consider this finding in more detail.

(f) The value of $\sqrt{T}\tau_{\rm rel}$ at $T_c = 293$ K is clearly larger than that at all other temperatures, and this is of course to

TABLE I. Magnetic data for Gd [Ref. 1, Table I, data for $Q = (\zeta,\zeta,2)$]: T = temperature; ζ_c = reduced crossover wave vector (in units of $4\pi/a$); β_q = damping parameter corresponding to ζ_c (in THz). [Data in parentheses are taken from Table II of Ref. 1, $Q = (\zeta,\zeta,2)$.]

T (K)	ζ_c	β_q	$\sqrt{T}/\beta_q$
293	0.114	0.67	25.5
(293)		(0.745)	(23.0)
(320)	0.177	(1.16)	(15.4)
440	0.184	1.325	15.8
586	0.228	1.59	15.2
850	0.290	1.79	16.3

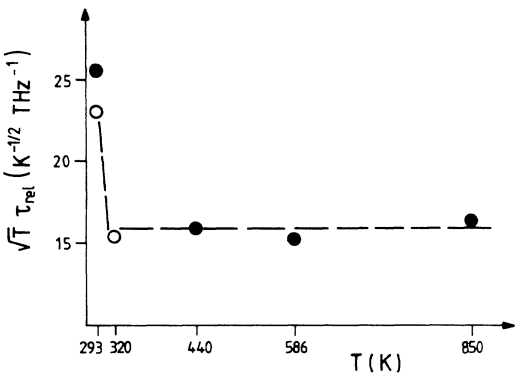

FIG. 1. Temperature dependence of the quantity $\sqrt{T}\tau_{rel}$ which is associated with the spatial size of the quantum correlations in the Gd spin system. The constancy of this quantity above T_c is clearly shown. Solid (open) circles correspond to the data of Table I (II) of Ref. 1. The dashed line is a guide to the eye. (For details, see the text.)

be expected, since this region is dominated by the well established critical phenomenon of the spin system.

(g) In the light of our theory, the above result (e) also reveals the interesting point that the relevant Hamiltonian H_{rel} (representing the magnetically ordered regions during their lifetime) may depend at most very slightly on the temperature. To see this, one may compare the aforementioned temperature dependence of the crossover wave vector with the expression of d_{min} and the above finding (e). The linear dimension of a magnetically ordered region (say, D_{magn}) is of the order ζ_c^{-1} and, at the same time, is represented by the theoretical quantity d_{min}. This comparison immediately shows that the value of the functional $F(H_{rel})$ may decrease very little with increasing temperature. In this context it should be observed that H_{rel} is a specific quantity of a realistic physical system, i.e., it, of course, cannot be determined by our general quantum correlations theory.

(h) The scattering of the experimental data (in the temperature range between 320 and 850 K) with respect to the constant function in Fig. 1 is about $\pm 4\%$ (i.e., not larger than the experimental errors).

The remarks (e)–(h) suggest the following additional possibility for an interpretation of the experimental data:[1]

(i) The crossover wave vector ζ_c represents, of course, a rough measure for the actual size of the magnetically ordered regions D_{magn} under consideration. This thus implies that the slight temperature dependence of this quantity (as shown in Fig. 6 of Ref. 1) does not strictly prove that this size, D_{magn}, does really decrease with increasing temperature. This remark, in connection with the revealed constancy (e), suggests that the functional $F(H_{rel})$ may be a T-independent quantity, which just leads to the conclusion that the size d_{min} may then be independent of the temperature, too. In other words: The present theoretical analysis of the experimental results of Ref. 1, which is based on our theory of coherent-dissipative structures[3–6] and therefore assumes the validity of the equality

$$D_{magn} = d_{min} ,$$

suggests that the actual size D_{magn} of the considered magnetically ordered regions remains constant between 320 and 850 K—which clearly is a surprising finding.

It should be observed that the above treatment does not use any specific feature of the underlying system Hamiltonian H_{rel}. Therefore, the indicated T independence of the quantum correlation factor $\sqrt{T}\tau_{rel}$ and the suggested constancy should be considered to be due to the general character of quantum correlations in condensed matter as being revealed with the aid of our general theoretical framework, i.e., without introducing additional *ad hoc* conditions.

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