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Spin dynamics of an ultra-small nanoscale molecular magnet

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Abstract We present mathematical transformations which allow us to calculate the spin dynamics of an ultra-small nanoscale molecular magnet consisting of a dimer system of classical (high) Heisenberg spins. We derive exact analytic expressions (in integral form) for the time-dependent spin autocorrelation function and several other quantities. The properties of the time-dependent spin autocorrelation function in terms of various coupling parameters and temperature are discussed in detail.

Recent succesful efforts in synthesizing solid lattices of weakly coupled molecular clusters containing few strongly interacting spins has opened up the possibility of experimentally studying magnetism at the nano scale [1]. Due to the presence of organic ligands which wrap the molecular clusters, the inter-cluster magnetic interaction is vanishingly small when compared to intra-cluster interactions, therefore the properties of the bulk sample reflect the properties of independent individual nanoscale molecular clusters. The magnetic ions in each molecular cluster can

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be generally arranged in different ways, giving rise to structures of very high symmetry (for example rings) and/or of lower symmetry presenting other important features. In some cases, the positions of the magnetic ions in the cluster define a nearly planar ring structure within the host lattice, for instance the Fe6 molecule is one of this type [2]. Here, the six Fe^{3+} ions have spin $S = 5/2$ and are coupled by nearest-neighbor antiferromagnetic exchange interactions. Other nanoscale molecular clusters consist of paramagnetic ions whose positions define a three-dimensional structure. Examples of this type are the molecules Fe4 and Cr4, which feature four Fe^{3+} ions [3] ($S = 5/2$) and four Cr^{3+} ions [4] ($S = 3/2$), respectively, which occupy the vertices of a tetrahedron embedded in the host lattice. Smaller clusters are the irregular triangle molecule [5] known as Fe3 which incorporates three Fe^{3+} ions with spin $S = 5/2$ and the dimer [6] system, Fe2 consisting of two Fe^{3+} ions with spin $S = 5/2$.

Low nuclearity complexes, such as Fe2 and Fe3, are likely to represent the "molecular" nanoscale bricks for the formation of high-nuclearity molecular clusters. Therefore, their characterization is an essential step for broader studies targeting larger systems [7]. Because of the high spin value of the Fe^{3+} ions, it turns out that the measured magnetic susceptibility and other related quantities can be reproduced to very high accuracy [8] by using the *classical* Heisenberg model which incorporates interaction between classical unit vectors. Only for very low temperatures need one consider the quantum character of the Fe^{3+} spins.

The spin dynamics of these nanoscale magnetic clusters is of particular interest since it can directly be probed by different experimental methods such as nuclear magnetic resonance (NMR) [9]. In view of the importance of knowing the dynamical behavior of spin–spin correlation

functions it is most desirable to find model systems which can be solved exactly. This way one can test the regimes of validity of various experimental results and theoretical approximation schemes. Among the variety of spin–spin correlation functions, the time-dependent spin autocorrelation function is closely linked with spin dynamics, therefore, it is natural to focus on this quantity. Earlier studies have numerically investigated the time-dependent spin autocorrelation function of many-spin systems such as a classical Heisenberg model with nearest-neighbor exchange interaction between spins [10]. The goal of these simulations was the study of the expected power-law decay of the long-time spin autocorrelation function for many-spin systems at infinite temperature [11].

In this work, we focus on the spin dynamics of ultra-small, nanoscale, molecular magnets of classical (high) Heisenberg spins. In particular, we give exact expressions (in integral form) for the time-dependent spin autocorrelation function at arbitrary temperature for a dimer system of classical (high) spins that interact with both exchange and biquadratic exchange interaction. The mathematical difficulty to solve exactly the equations of motion and to perform the phase–space average for interacting spins makes an exact analytical calculation of the time-dependent spin autocorrelation function very challenging, even for the ultra-small system considered here. To overcome these mathematical difficulties we introduce a method which simplifies the calculation of various quantities through the introduction of suitably chosen auxiliary time-independent variables into an extended phase–space integration [12, 13]. The present analytic results, although derived for the dimer system of spins [14], can provide useful benchmarks for assesing numerical methods that calculate the time-dependent spin dynamics of other magnetic high-spin systems.

The Hamiltonian of a dimer system of spins with exchange and biquadratic interaction is written as

$$H(t) = J\vec{S}_1(t)\vec{S}_2(t) + K\left[\vec{S}_1(t)\vec{S}_2(t)\right]^2, \qquad (1)$$

where J, K represent, respectively, the exchange, biquadratic exchange interaction and $\vec{S}_i(t)$ are time-dependent classical spin vectors of unit length ($i = 1,2$). The orientation of the classical unit vectors $\vec{S}_i(t)$ at a moment of time, t, is specified by polar and azimuthal angles, $\theta_i(t)$ and $\phi_i(t)$, which, respectively, extend from 0 to π and 0 to 2π. The exchange interaction between a pair of spins can be either antiferromagnetic (AF), $J = |J| > 0$, or ferromagnetic (F), $J = -|J| < 0$. The biquadratic exchange interaction, K, can be positive, zero or negative.

At an arbitrary temperature, T, the time-dependent spin autocorrelation function, $C_T(t) = \langle \vec{S}_i(0)\vec{S}_i(t)\rangle$, is evaluated as a phase space average over all possible initial time orientations of the spins:

$$C_T(t) = \frac{\int d\vec{S}_1(0) \int d\vec{S}_2(0) \exp[-\beta H(0)]\vec{S}_i(0)\vec{S}_i(t)}{Z(T)}, \qquad (2)$$

where $i = 1$ or 2 is a selected spin index, $d\vec{S}_j(0) = d\theta_j(0)\sin[\theta_j(0)]d\varphi_j(0)$ is the initial time solid angle element appropriate for the j-th spin, $\beta = 1/(k_B\,T)$, and k_B is Boltzmann's constant. The denominator of Eq. 2 represents the partition function, $Z(T) = \int d\vec{S}_1(0)\int d\vec{S}_2(0)\exp[-\beta H(0)]$, where $H(0)$ is the initial time Hamiltonian of the spin system. In order to evaluate the time-dependent spin autocorrelation function we need first to solve the equations of motions for the spins and then perform the angular average over all possible initial time spin orientations in the phase space.

The dynamics (equations of motion) of classical spins is determined from

$$\frac{d}{dt}\vec{S}_i(t) = -\vec{S}_i(t) \times \frac{\partial H(t)}{\partial \vec{S}_i(t)}, \qquad (3)$$

where the set of solutions, $\{\vec{S}_i(t)\}$ depends on the initial orientation of the spins, $\{\vec{S}_i(0)\}$.

The calculation of $C_T(t)$ follows several steps: (i) solve the equations of motion for the spins to obtain $\vec{S}_i(t)$; (ii) calculate the partition function $Z(T)$; and (iii) compute the integrals appearing in the numerator of Eq. 2.

By applying Eq. 3 to each spin of the dimer, it is not difficult to note that the total spin, $\vec{S}(t) = \vec{S}_1(t) + \vec{S}_2(t)$, is a constant of motion, $\vec{S}(t) = \vec{S}(0) = \vec{S}$, and as a result we can rewrite Eq. 3 as

$$\frac{d}{dt}\vec{S}_i(t) = -[J + K(S^2 - 2)]\vec{S}_i(t) \times \vec{S}, \qquad (4)$$

where $\vec{S}$ represents the constant total spin.

The above differential equations for spins can be exactly solved in a new coordinate system (x' y' z') in which the constant vector $\vec{S}$ lies parallel to the z' axis. Let us denote (α_i, β_i) to be the polar and azimuthal angles of spin $\vec{S}_i(0)$ with respect to the new coordinate system in which the direction of $\vec{S}$ defines the z' (polar) axis. It follows that $S\cos(\alpha_i) = \vec{S}_i(0)\vec{S}$. The solution of Eq. 4 for each spin component of $\vec{S}_i(t)$ depends on the sign of $[J + K\,(S^2–2)]$. Irrespective of the sign of $[J + K\,(S^2–2)]$, we find that the quantity $\vec{S}_i(0)\vec{S}_i(t)$ is given by the expression

$$\vec{S}_i(0)\vec{S}_i(t) = \sin^2(\alpha_i)\cos[\omega(S)t] + \cos^2(\alpha_i), \qquad (5)$$

where $\omega(S) = |J + K\,(S^2–2)|\,S$ denotes a precession frequency, and $0 \leq S = |\vec{S}| \leq 2$. Note that $\vec{S}_i(0)\vec{S}_i(t)$ does not depend on the i-th spin azimuthal angle β_i.

In as much as the spins are equivalent, without loss of generality we fix $i = 1$ and concentrate on the calculation

of $C_T(t) = \langle \vec{S}_1(0)\vec{S}_1(t) \rangle$. From the definition of the total spin variable, $\vec{S} = \vec{S}_1(0) + \vec{S}_2(0)$, recalling that $S\cos(\alpha_1) = \vec{S}_1(0)\vec{S}$, we easily find that $1 + \vec{S}_1(0)\vec{S}_2(0) = S\cos(\alpha_1)$. Since the product $\vec{S}_1(0)\vec{S}_2(0)$ is expressable in terms of the total spin as $\vec{S}_1(0)\vec{S}_2(0) = S^2/2 - 1$, it follows that $\cos(\alpha_1) = S/2$, and it depends only on the total spin magnitude. Through these simple mathematical transformations we reach the first goal to express $\vec{S}_1(0)\vec{S}_1(t)$ as

$$\vec{S}_1(0)\vec{S}_1(t) = F(t,S) = \left(1 - \frac{S^2}{4}\right)\cos[\omega(S)t] + \frac{S^2}{4}. \tag{6}$$

In the same way, the Hamiltonian can be written in terms of the total spin variable as

$$H(t) = H(0) = \frac{J}{2}\left(S^2 - 2\right) + \frac{K}{4}\left(S^2 - 2\right)^2 \tag{7}$$

and is a constant of motion.

By expressing all relevant quantities in terms of the total spin variable which is a constant of motion, we now apply our calculation method whose success is based on the observation that the values of all multi-dimensional integrals, for example $Z(T)$, are left unchanged if multiplied by unity written as

$$\int d^3S \int \frac{d^3q}{(2\pi)^3} \exp\left[i\vec{q}\left(\vec{S} - \vec{S}_1(0) - \vec{S}_2(0)\right)\right] = 1. \tag{8}$$

Note that the above identity originates from the well known formula, $\int d^3S \delta^{(3)}\left[\vec{S} - \vec{S}_1(0) - \vec{S}_2(0)\right] = 1$, that applies to three-dimensional Dirac delta functions. Subsequent calculations are straightforward given that both $H(0)$ and $\vec{S}_1(0)\vec{S}_1(t)$ appearing in Eq. 2 can be expressed solely in terms of S. As a result, the integrations over individual spin variables pose no problems. For the partition function, we obtain

$$Z(T) = (4\pi)^2 \int_0^2 dS D(S) \exp\left[-\frac{\beta J}{2}(S^2 - 2) - \frac{\beta K}{4}(S^2 - 2)^2\right], \tag{9}$$

where $D(S) = 4\pi S^2 \int \frac{d^3q}{(2\pi)^3}\exp(i\vec{q}\vec{S})(\sin q/q)^2$ can be calculated analytically and is

$$D(S) = \begin{cases} S/2 & 0 < S < 2 \\ S/4 & S = 2 \\ 0 & S > 2 \end{cases} \tag{10}$$

The vanishing of $D(S)$ for $S > 2$ reflects the constraint that the total spin cannot exceed 2. Note that for $K \equiv 0$, the partition function becomes $Z(T) = (4\pi)^2 \frac{\sinh(\beta J)}{\beta J}$. In the most general case, $J \neq 0$ and $K \neq 0$, the integral in

Eq. 9 can be expressed analytically in terms of error functions. From the perspective of numerical calculations, the above one-dimensional integral form is better suited.

The integral appearing in the numerator of Eq. 2 is generally very difficult to calculate. However, using the method illustrated above, integration is simplified, and one obtains

$$C_T(t) = \frac{(4\pi)^2}{Z(T)} \int_0^2 dS\, D(S) \exp\left[-\frac{\beta J}{2}(S^2 - 2) - \frac{\beta K}{4}\cdot(S^2 - 2)^2\right]F(t,S). \tag{11}$$

The integrals appearing in Eq. 11 can be carried out analytically. The final result can be written in a closed form in terms of error functions. The expressions are quite lengthy and cumbersome. Because of such undesired complexity, the one-dimensional integral representation in Eq. 11 not only suffices, but is preferable for all practical needs. The preceding formula for $C_T(t)$ represents the exact expression (in integral form) for the time-dependent spin autocorrelation function of a dimer system of classical spins with exchange and biquadratic exchange interaction at an arbitrary temperature.

Depending on the magnitude and sign of the coupling constants, J and K, the quantity $C_T(t)$ approaches a unique non-zero value at infinite time ($t \to \infty$) given by

$$C_T(t \to \infty) = \frac{1}{2}\left[1 + \frac{\int_{-1}^1 dx\, x \exp(-\beta Jx - \beta Kx^2)}{\int_{-1}^1 dx \exp(-\beta Jx - \beta Kx^2)}\right], \tag{12}$$

where the auxiliary variable, $x = (S^2-2)/2$ was introduced to facilitate calculations. The final expression is rather lengthy and can be expressed in terms of error functions.

For vanishing biquadratic exchange interaction ($K \equiv 0$), the infinite-time limit of the spin autocorrelation function is

$$C_T(t \to \infty) = \frac{1}{2}[1 - L(\beta J)] \text{ for } K \equiv 0, \tag{13}$$

where $L(z) = \coth(z) - 1/z$ is Langeven's function.

Let us now study in detail the time dependence of $C_T(t)$ for two extreme cases: very low temperature (we choose a typical value, $k_B T/|J| = 0.1$) and very high temperature ($T \to \infty$).

Figures 1–4 display the time-dependent spin autocorrelation function for the classical dimer of spins with exchange and biquadratic exchange interaction as a function of $|J|\, t$ at $k_B T/|J| = 0.1$. In Figs. 1 and 2 we consider an AF exchange interaction, $J = |J| > 0$, and, respectively, non-negative $K = |K| \geq 0$ and non-positive
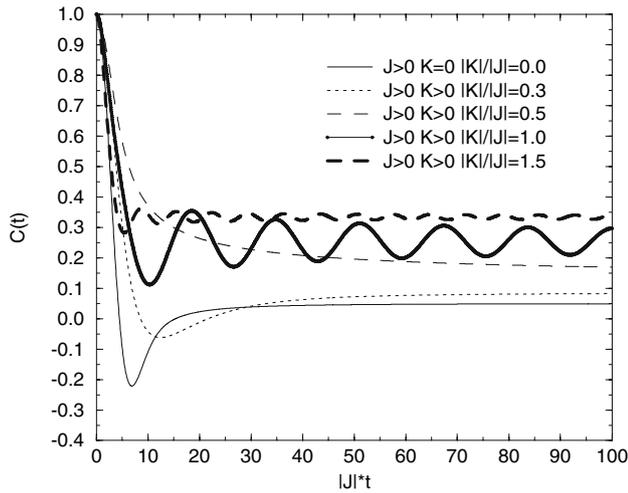
Fig. 1 Time-dependent spin autocorrelation function, $C_T(t)$ for the classical dimer of spins with exchange and biquadratic exchange interaction at a very low temperature, $k_B T/|J| = 0.1$. Given an AF exchange interaction between spins, $J = |J| > 0$, we consider several non-negative values of the biquadratic exchange interation, $K = |K| \geq 0$. Note how $C_T(t)$ changes from a very smooth function of $|J| t$ for small values of $|K|/|J|$, to a strongly oscillatory function of $|J| t$ as $|K|/|J|$ becomes comparable or greater than unity

Fig. 3 Time-dependent spin autocorrelation function, $C_T(t)$, for the classical dimer of spins with exchange and biquadratic exchange interaction at a very low temperature, $k_B T/|J| = 0.1$. Several non-negative values of the biquadratic exchange interaction, $K = |K| \geq 0$, are considered for a given F exchange interaction, $J = -|J| < 0$. When $|K|/|J|$ increases from 0.0 to 0.1, the oscillations of $C_T(t)$ amplify, but for larger values of $|K|/|J|$ the function gradually transforms into a smooth function of $|J| t$ with fast decaying oscillations

$K = -|K| \leq 0$. The AF case of $J = |J| > 0$ and $K = |K| \geq 0$ shown in Fig. 1 is rather interesting. One notes that $C_T(t)$ dramatically changes its time dependence from a smooth function to a strongly oscillatory function of $|J| t$ when

$|K|/|J|$ increases and becomes larger or of the order of unity.

In Figs. 3 and 4 we consider an F exchange interaction, $J = -|J| < 0$, and, respectively, non-negative $K = |K| \geq 0$

Fig. 2 Time-dependent spin autocorrelation function, $C_T(t)$ for the classical dimer of spins with exchange and biquadratic exchange interaction at a very low temperature, $k_B T/|J| = 0.1$. Given an AF exchange interaction between spins, $J = |J| > 0$, we consider several non-positive values of the biquadratic exchange interaction $K = -|K| \leq 0$. Note that there are no relevant qualitative changes on the dependence of $C_T(t)$ as a function of $|J| t$ as $|K|/|J|$ varies. Qualitatively speaking, $C_T(t)$ remains a smooth function of $|J| t$ with a minimum that deepens and occurs sooner as $|K|/|J|$ increases

Fig. 4 Time-dependent spin autocorrelation function, $C_T(t)$, for the classical dimer of spins with exchange and biquadratic exchange interaction at a very low temperature, $k_B T/|J| = 0.1$. Several non-positive values of the biquadratic exchange interaction, $K = -|K| \leq 0$, are considered for a given F exchange interaction, $J = -|J| < 0$. Note that $C_T(t)$ approaches its long-time asymptotic limit value (that is larger for larger values of $|K|/|J|$) with less pronounced oscillations as $|K|/|J|$ increases

and non-positive $K = -|K| \leq 0$. Contrary to what is seen in Fig. 1, the case described in Fig. 3 for $J = -|J| < 0$ and $K = |K| \geq 0$ shows a very different behavior, in the sense that the strong oscillatory dependence of $C_T(t)$ as function of $|J|\ t$ is supressed when $|K|/|J|$ increases.

At infinite temperature $(T \rightarrow \infty)$ and arbitrary time, the time-dependent spin autocorrelation may be expressed as

$$C_{T\rightarrow\infty}(t) = \int_0^2 dS D(S) F(t, S). \tag{14}$$

Using Eq. 6 and Eq. 10 one can rewrite $C_{T \rightarrow \infty}(t)$ in a suitable form as

$$C_{T\rightarrow\infty}(t) = \frac{1}{2} + \int_0^1 dx\, x \cos\left(2|J + 2K(1 - 2x)|\sqrt{1 - xt}\right), \tag{15}$$

where $x = 1 - S^2/4$ is a dummy variable introduced to simplify the final expression. One notes that, at infinite temperature $(T \rightarrow \infty)$ and arbitrary time, the expression for $C_T(t)$ remains unchanged when the two coupling constants, J and K simultaneosly reverse sign to $-J$ and $-K$. A simultaneous sign change of the two couplings J and K leaves the same expression for $C_{T \rightarrow \infty}(t)$ since as seen in Eq. 15 both J and K occur under the absolute value sign. Figs. 5 and 6 show $C_T(t)$ as a function of $|J|\ t$ for infinite temperature $(T \rightarrow \infty)$.
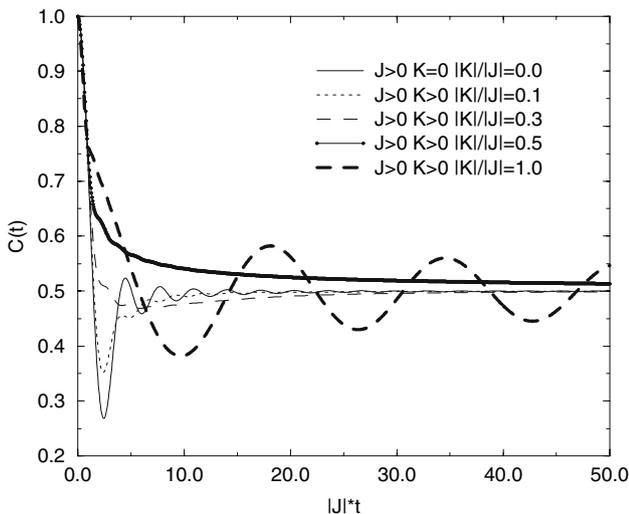
Fig. 5 Time-dependent spin autocorrelation function, $C_T(t)$, for the classical dimer of spins with exchange and biquadratic exchange interaction at infinite temperature, $T \rightarrow \infty$. For an AF exchange interaction, $J = |J| > 0$, several non-negative values of the biquadratic exchange interation, $K = |K| \geq 0$, are considered. Depending on the value of $|K|/|J|$, different behaviors of $C_{T \rightarrow \infty}(t)$ as a function of $|J|\ t$ arise. Note that when $|K|/|J|$ becomes comparable to unity, "large" oscillations occur on $C_{T \rightarrow \infty}(t)$ that otherwise are not present for smaller values of $|K|/|J|$
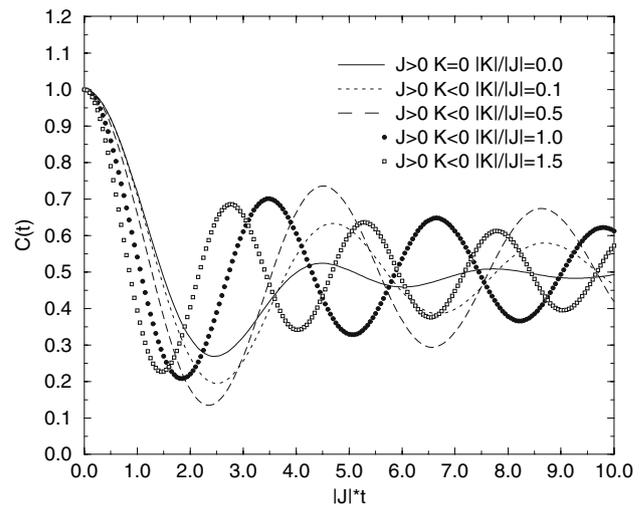
Fig. 6 Time-dependent spin autocorrelation function, $C_T(t)$, for the classical dimer of spins with exchange and biquadratic exchange interaction at infinite temperature, $T \rightarrow \infty$. For an AF exchange interaction, $J = |J| > 0$, several non-positive values of the biquadratic exchange interation $K = -|K| \leq 0$ are considered. Note that $C_{T \rightarrow \infty}(t)$ has a stronger oscillatory dependence on $|J|\ t$ as $|K|/|J|$ increases

Let us now consider the case of an AF exchange interaction, $J = |J| > 0$, and non-negative, $K = |K| \geq 0$, and non-positive, $K = -|K| \leq 0$, biquadratic exchange. The situation shown in Fig. 5 for $J = |J| > 0$ and $K = |K| \geq 0$ is of particular interest since one observes the appearance of large and very slowly decaying oscillations on the spin autocorrelation function as $|K|/|J|$ becomes of the order of unity. For a vanishing biquadratic exchange interaction, $K \equiv 0$, one has the special case of a dimer with only exchange interaction, and in this case $\omega(S) = |J|\ S$.

Figure 7 shows $C_T(t)$ when $K \equiv 0$ for several temperatures and for both AF and F exchange interactions. One clearly notes that for low temperatures the spin autocorrelation function is dominated by the lowest frequency $(S \approx 0)$ when we have AF coupling and by the highest frequency $(S \approx 2)$ for the F case. This very different behavior of the time-dependent spin autocorrelation function at low temperatures is better illustrated in Figs. 8 and 9 where one notes that, for the same temperature, there is a strong oscillatory dependence on $|J|\ t$ for an F exchange interaction, while such dependence is very smooth for an AF exchange coupling.

For zero biquadratic exchange interaction $(K \equiv 0)$ and at infinite temperature, $T \rightarrow \infty$, one calculates the spin autocorrelation function directly from Eq. 15 and obtains

$$C_{T\rightarrow\infty}(t) = \frac{1}{2} + \frac{3}{2}\frac{\sin(2|J|t)}{(|J|t)^3} + \frac{3}{4}\frac{\cos(2|J|t) - 1}{(|J|t)^4} - \frac{1}{2}\frac{2\cos(2|J|t) + 1}{(|J|t)^2} \text{ for } K \equiv 0, \tag{16}$$
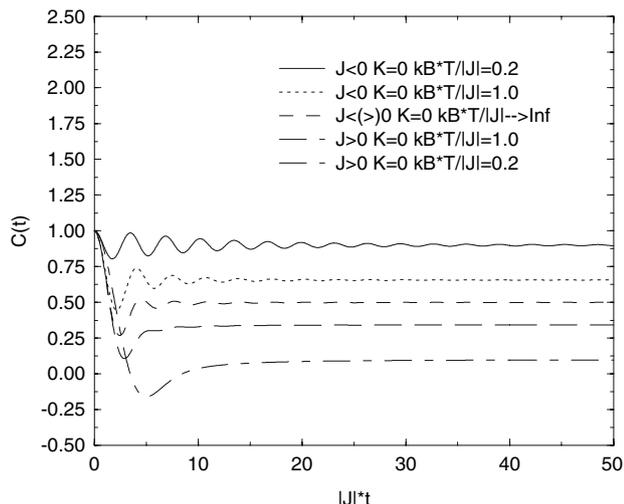
Fig. 7 Time-dependent spin autocorrelation function, $C_T(t)$, for the classical dimer of spins with AF/F exchange interaction and no biquadratic exchange ($K \equiv 0$) as a function of $|J| t$ at some arbitrary temperatures. In the $T \rightarrow \infty$ limit, $C_{T \rightarrow \infty}(t)$ is the same irrespective of the sign of J

Fig. 9 Time-dependent spin autocorrelation function, $C_T(t)$, for the classical dimer of spins with only exchange interaction and no biquadratic exchange interaction ($K \equiv 0$) for very low temperatures and for an AF exchange interaction, $J = |J| > 0$. Contrary to the F case, $C_T(t)$ is a very smooth function of $|J| t$ and approaches its long-time asymptotic temperature-dependent value much faster

a result that coincides with the formula derived by Muller [15]. We observe that $C_{T \rightarrow \infty}(t)$ first goes through a deep minimum and then approaches its long-time asymptotic value, $C_{T \rightarrow \infty}(t \rightarrow \infty) = 1/2$. Such value remains the same whether we have $K \equiv 0$ or $K \neq 0$.

In conclusion, we studied the spin dynamics and time-dependent spin autocorrelation function for a nanoscale molecular magnet consisting of a dimer system of

Fig. 8 Time-dependent spin autocorrelation function, $C_T(t)$, for the classical dimer of spins with only exchange interaction and no biquadratic exchange interaction ($K \equiv 0$) for very low temperatures and for an F exchange interaction, $J = -|J| < 0$. $C_T(t)$ approaches its long-time asymptotic temperature-dependent value very slowly with many slowly decaying oscillations around that value

Heisenberg spins interacting with exchange and biquadratic exchange interaction. By using a method which introduces the total spin variable into the defining expression of the time-dependent spin autocorrelation function, we obtain the exact analytic expression (in integral form) for this quantity at an arbitrary temperature. The results elucidate the spin dynamics of nanoscale molecular magnets consisting of dimer systems of magnetic ions with high (classical) spin values (for instance, Fe^{3+} ions). Such is the iron(III) $S = 5/2$ dimer (in short Fe2) described by the spin Hamiltonian $H = J\vec{S}_1\vec{S}_2$ where $J \sim 22$ K is an AF exchange coupling constant. Experimental studies of Fe2 dimer at room temperature show that the measured proton nuclear spin-lattice relaxation rate, T_1^{-1} is frequency independent [6]. This result is consistent with the behavior of the spin autocorrelation function, $C_T(t)$, for an AF coupling $J > 0$ and $K \equiv 0$ as shown in Fig. 7 (three lower curves). An initial fast decay of $C_T(t)$ followed by a much slower decay at long time generates a narrow Lorentzian-type peak in the spectral density (which is basically defined as a Fourier transform of spin autocorrelation function) a feature that is in agreement with the above experimental work. The mathematical method we employed can be extended to certain other larger high-spin nanoscale magnetic clusters with more complicated geometries such as rings and/or polyhedra that are described by a spin Hamiltonian of the form $H(t) = J \sum_{i<j}^{N} \vec{S}_i(t)\vec{S}_j(t)$, where N is the total number of spins in the magnetic nano-cluster. One can always express such a spin Hamiltonian in terms of the total spin $\vec{S}(t) = \sum_{i=1}^{N} \vec{S}_i(t) = \vec{S}$, which is a constant of motion and then proceed to calculate spin–spin correlation and

autocorrelation functions by following the method outlined in this work.

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