Nonequilibrium statistical mechanics of the spin-$\frac{1}{2}$ van der Waals model. I. Time evolution of a single spin

Raf Dekeyser
Instituut voor Theoretische Fysica, Katholieke Universiteit Leuven, B-3001 Heverlee, Belgium

M. Howard Lee
Department of Physics and Astronomy, University of Georgia, Athens, Georgia 30602
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The spin van der Waals model has received some attention in recent years as a soluble model, useful for demonstrating collective behavior analytically. By solving the Heisenberg equation of motion, we obtain the time evolution of a single spin in this model when $N\rightarrow\infty$, where N is the number of spins in the model. Our solution is characterized by rotations in spin space having the appearance of a Larmor precession. In the accompanying paper, we use this time-evolution solution to obtain the spin autocorrelation function and determine its long-time behavior.

I. INTRODUCTION

In the late 1960s, molecular-dynamics studies gave evidence of slow decay in the velocity autocorrelation function (VAF) of a particle in a simple fluid.[1] The VAF has a power-law behavior $t^{-\kappa}$ for $t>t_c$, where t_c is some characteristic time of the system and $\kappa=D/2$ where D denotes the dimension of the system. The value of the exponent seems remarkable: It depends on the dimension D but apparently not on the interactions. This kind of slow decay in the VAF is now referred to as long-time tails. The existence of long-time tails, if correct, is physically significant since it can lead to a divergence in the transport coefficients. Conventional transport theories are built on an exponential decay of the VAF.[2]

The evidence of long-time tails observed in computer experiments has stimulated considerable activity both experimentally and theoretically. Measurements in aqueous solutions of polyestyrene spheres by Paul and Pusey[3] and Fedele and Kim[4] appear to indicate a slow-decay component in the VAF. Fox[5] has recently assessed these experimental findings. More recently, Morkel, Gronemeyer, and Glässer[6] have shown strong evidence of long-time tails in liquid sodium at high temperatures.

A qualitative understanding of the origin of long-time tails was initially provided by mode-coupling theory.[2,7] It is a hydrodynamic picture in which slow decay is attributed to shear modes generated by a moving particle, acting on itself at a later time. Presumably, coupling between a particle and its surroundings is an essential source of this anomalous behavior. In an attempt to give a more rigorous derivation based on microscopic models, several workers have applied kinetic theory. Owing to difficulty inherent to fluids, they were ultimately forced to approximate their theories. As a result, they are perhaps not much more rigorous than those based *ab initio* on hydrodynamic pictures.[7] It is probably correct to say that, to date, there are still no rigorous derivations of long-time tails in the VAF of a particle in a fluid.[8]

Spin diffusion is a physical process analogous to particle diffusion. If the spin autocorrelation function (SAF) has a slow decay, the spin transport coefficients may also behave anomalously. In many respects, spin dynamics should be theoretically simpler to study than particle dynamics. Unlike particles in a fluid, spins in a lattice are localized. Also, spin interactions can be limited to those between near neighbors. Spin dynamics is still highly nontrivial. Exact results are limited largely to one-dimensional spin-$\frac{1}{2}$ XY and transverse Ising models, to which we shall confine our remarks.

At $T=\infty$, Perk and Capel[9] have shown that the transverse component of the SAF of a single spin is Gaussian. For the longitudinal component Niemeijer[10] obtained $J_0^2(t)$, where J_0 is the Bessel function of order 0. There is thus a slow decay here, but its origin is obscure. At $T=0$, quantum effects make the determination of the SAF very difficult. The work of Müller and Shrock[11] suggests a nonexponential decay, based on the nonanalytic behavior of the dynamic structure at certain frequencies ω, including $\omega=0$.

In what we might call a canonical approach to dynamics, one would first calculate the time evolution of a dynamical variable (e.g., a spin operator) and then the autocorrelation function and other physical quantities therefrom. This approach can yield information with which to obtain complete dynamical analysis. Not surprisingly, rarely has anyone been able to realize this approach, as our examples in spin dynamics would indicate. We find, however, that there is a certain quantum spin model called the spin van der Waals model for which the canonical approach can actually be realized. For this model we have succeeded in obtaining the exponent κ, which is found to behave discontinuously as a function of interactions.

The spin van der Waals model is a cooperative spin model in which the spin-spin interactions are made independent of the separation distance. In the older literature it was called the molecular-field approximation model.[12] This model may be regarded as the $q \to \infty$ limit of the spin-$\frac{1}{2}$ NN anisotropic Heisenberg model, where q is the coordination number. A spin in the spin van der Waals model is somewhat reminiscent of a Brownian particle in a dense medium. Hence, this model has qualitative features of both quantum cooperativity and Brownian motion.

For this model it is possible to obtain the partition function exactly if $N \to \infty$, where N is the number of spins in the model. Hence, one can know all the static properties of the model, all of which show mean-field character. This model has attracted some attention in recent years as a soluble model, useful for demonstrating collective behavior analytically. Kim and Lee,[13] for example, have used this model to illustrate the correlation inequalities of Falk and Bruch.[14] Pathria[15] has examined the N dependence in the specific heat. Gilmore[16] has shown that this model is equivalent to the Meshkov-Glick-Lipkin model in nuclear physics. Katriel and Kventsel[17] have demonstrated among others a reentrance phenomenon in a generalized version of this model. There are many other works along these lines.[18–22]

Some years ago we were able to realize the canonical approach to the dynamics of the total spin in this model.[23] We have shown that, in the XY regime, the transverse component of the SAF of the *total* spin is Gaussian both above and below the critical temperature T_c. In the Ising regime, it is oscillatory below T_c but Gaussian above T_c. There are no long-time tails at any temperature T. Perhaps most interesting was our finding that there are two time scales, which are the source of the critical anomaly.[24] Kventsel and Katriel[25] have generalized some of these results.

In this work, we undertake the canonical approach to the dynamics of a *single* spin in the van der Waals model when $N \to \infty$. We are interested in determining whether slow decay exists in the SAF of a single spin, absent in the SAF of the total spin. If slow decay exists, we would like to know its origin. Our work is presented in two parts. In the first part, we present the time evolution of a single spin by solving the Heisenberg equation of motion. In the second part (accompanying paper), we use the time-evolution solution of a single spin to calculate the SAF.

II. SPIN-$\frac{1}{2}$ VAN DER WAALS MODEL

Consider a system of $N+1$ lattice spins, denoted by a spin-$\frac{1}{2}$ operator $\mathbf{s}_i = (s_i^x, s_i^y, s_i^z)$, where i labels the lattice sites. The interaction energy of the spin van der Waals model of $N+1$ spins takes the following form:

$$H = -N^{-1} \sum_{i,j=1}^{N+1} [J(s_i^x s_j^x + s_i^y s_j^y) + J_z s_i^z s_j^z] , \tag{1}$$

where the sum is over all pairs of spins, and J and J_z are coupling constants. In the spin van der Waals model, each spin interacts with each other with the same strength J or J_z, independently of the separation distance between any two spins. A division by N is needed to ensure that the energy is extensive.

Among these $N+1$ spins, we shall now single out one spin, which may be any one of the $N+1$ spins, and denote it as $\mathbf{s}$. The remaining N spins are collectively denoted by

$$\mathbf{S} = (S_x, S_y, S_z) = \sum_{i=1}^{N} \mathbf{s}_i . \tag{2}$$

The total spin $\mathbf{S}^{\text{tot}}$ is given by

$$\mathbf{S}^{\text{tot}} = \mathbf{s} + \mathbf{S} . \tag{3}$$

For the system defined by (1), important constants of motion are $(\mathbf{S}^{\text{tot}})^2$, S_z^{tot}, $\mathbf{S}^2$, $\mathbf{s}^2$, and $\mathbf{s}\cdot\mathbf{S}$.

If N is very large, it is useful to formally regard our system to be as if composed of two subsystems: one subsystem containing $\mathbf{s}$ and the other containing $\mathbf{S}$. Let us refer to $\mathbf{s}$ and $\mathbf{S}$ as a small spin and a large spin, respectively. Note that the two spins commute. If N is very large, the subsystem containing the large spin acts somewhat like a reservoir for the subsystem containing the small spin. That is, a small change in the small spin should not affect the state of the large spin, but a small change in the large spin could have a large effect on the state of the small spin. We should emphasize here that by this remark we do not mean to introduce *a priori* any approximation.

We can rewrite our interaction energy (1) in terms of the small and large spins as follows:

$$H = H_0 + V , \tag{4}$$

where

$$H_0 = -(J\mathbf{S}^2 - \lambda S_z^2)/N, \quad \lambda = J - J_z , \tag{5}$$

and

$$V = -2(J\mathbf{s}\cdot\mathbf{S} - \lambda s^z S_z)/N . \tag{6}$$

In obtaining (4) we have dropped an additive constant. Note that $[H_0, V] \neq 0$. The first term H_0 represents the "self-interaction" energy of the large spin; the second term V, the interaction energy between the small and large spins. As already suggested, H_0 has the appearance of the energy of a reservoir; V, the interaction of a spin and a reservoir. Both H_0 and V are characterized by two parameters: $g = J/N$ and $\omega = (J - J_z)/N$. The first parameter may be said to indicate the strength of spherical symmetry; the second, that of axial symmetry. In an earlier work, we have given a complete account of the nonequilibrium behavior of the large spin in the thermodynamic limit;[23] we shall find this work useful for describing the behavior of the small spin.

III. EQUATION OF MOTION

We shall denote the time evolution of the small and large spins as follows (adopting $\hbar = 1$):

$$\mathbf{s}(t) = e^{iHt} \mathbf{s} e^{-iHt} \tag{7}$$

and

$$\mathbf{S}(t)=e^{iHt}\mathbf{S}e^{-iHt} , \tag{8}$$

where H is given by (1) or (4), $\mathbf{s}=\mathbf{s}(t=0)$ and $\mathbf{S}=\mathbf{S}(t=0)$, which may be regarded as initial conditions. The equation of motion for the small spin at $t=0$ is

$$\dot{\mathbf{s}}=i[H,\mathbf{s}]=i[V,\mathbf{s}] . \tag{9}$$

Using V given by (6), we obtain

$$\dot{s}^{x}(t)=2g_z s^{y}(t)S_z(t)-2gs^{z}(t)S_y(t) , \tag{10a}$$

$$\dot{s}^{y}(t)=-2g_z s^{x}(t)S_z(t)+2gs^{z}(t)S_x(t) , \tag{10b}$$

$$\dot{s}^{z}(t)=2gs^{x}(t)S_y(t)-2gs^{y}(t)S_x(t) , \tag{10c}$$

where $g_z=J_z/N=g-\omega$. The above equations show that the time evolution of the small spin depends on its coupling to the large spin, where g and g_z act as the strengths of the coupling. In this work, we propose to solve the equation of motion for the small spin.

As noted, the above equations express a linear coupling of the two spins. It is thus natural to introduce bilinear spin operators P, Q, and R defined at $t=0$ as follows:

$$P=s^xS_x+s^yS_y , \tag{11a}$$

$$iQ=s^xS_y-s^yS_x , \tag{11b}$$

$$R=s^zS_z . \tag{11c}$$

Observe that the bilinear spin operators may be combined with the large spin to give the small spin:

$$s^xU=PS_x+iQS_y+is^yS_z , \tag{12a}$$

$$s^yU=PS_y-iQS_x-is^xS_z , \tag{12b}$$

$$s^zS_z=R , \tag{12c}$$

where $U=\mathbf{S}^2-S_z^2$. Hence, if one knows the time evolution of the bilinear spin and large-spin operators, one can obtain the time evolution of the small-spin operators. Thus, we turn to obtain the time evolution of the bilinear spin operators, rather than to directly solve the equation of motion for the small spin.

Using (10a)–(10c) we find that the bilinear spin operators satisfy the following equations of motion:

$$\dot{P}(t)=-2igS_z(t)Q(t)-igP(t) , \tag{13a}$$

$$\dot{Q}(t)=-2igS_z(t)P(t)+2igs_z(t)U(t) -igQ(t)-igS_z(t) , \tag{13b}$$

$$\dot{R}(t)=2igS_z(t)Q(t)+igP(t) . \tag{13c}$$

Observe that now only the parameter g enters into these equations. We immediately note that

$$\dot{P}(t)+\dot{R}(t)=0 . \tag{14}$$

Hence,

$$P(t)+R(t)=\mathbf{s}\cdot\mathbf{S} , \tag{15}$$

recalling that the right-hand side of (15) is a constant of motion mentioned earlier. To solve (13a)–(13c) we need to know the time evolution of the large spin, in particular $S_z(t)$, and also $S_x(t)$ and $S_y(t)$ in (12). Hence, we first look for the time evolution of the large spin. Since we have in mind the case $N\rightarrow\infty$, we need only to look for asymptotically exact solutions. Let us write the equation of motion for the large spin as follows:

$$\dot{\mathbf{S}}(t)=i[H,\mathbf{S}(t)]=i[H_0+V,\mathbf{S}(t)]\equiv\dot{\mathbf{S}}^{(0)}(t)+\dot{\mathbf{S}}^{(1)}(t) . \tag{16}$$

The first term refers to the time evolution induced by the "self-interaction;" the second term, that induced by the interaction with the small spin. In the limit $N\rightarrow\infty$, the second term may be neglected, as we shall see. By evaluating the commutators at $t=0$ (for simplicity), we obtain

$$\dot{S}_x^{(0)}=-2\omega S_zS_y-i\omega S_x , \tag{17a}$$

$$\dot{S}_y^{(0)}=2\omega S_zS_x-i\omega S_y , \tag{17b}$$

$$\dot{S}_z^{(0)}=0 , \tag{17c}$$

and

$$\dot{S}_x^{(1)}=-2gs^yS_z+2g_zs^zS_y , \tag{18a}$$

$$\dot{S}_y^{(1)}=2gs^xS_z-2g_zs^zS_x , \tag{18b}$$

$$\dot{S}_z^{(1)}=-2g(s^xS_y-s^yS_x) . \tag{18c}$$

The linear term on the right-hand side of (17a) or (17b) is an effect of noncommutativity in the large spin; if the spins were classical, it would not be present. Also, the terms of the right-hand sides of (18a)–(18c) are all linear in the large-spin operators, comparable to the "quantum fluctuation" terms of (17a) and (17b).

IV. ASYMPTOTIC SOLUTIONS FOR THE LARGE SPIN

If $N\rightarrow\infty$, we find it sufficient to obtain asymptotic solutions for the large-spin operators and also for the bilinear spin operators. These solutions are much simpler to obtain and are physically more intuitive than the general solutions. Hence, we proceed to obtain the asymptotic solutions here and in the following section. For purposes of comparison we have given the general solutions in Appendix A. To obtain the asymptotic solutions, the large-spin operators are regarded as terms of order N. In our differential equations we retain terms to leading order in N only. On the right-hand sides of Eqs. (17)–(18), we drop all the linear terms in the large spin. Valid to this order, we obtain

$$S_x(t)=[\cos(\Omega_zt)]S_x-[\sin(\Omega_zt)]S_y , \tag{19a}$$

$$S_y(t)=[\cos(\Omega_zt)]S_y+[\sin(\Omega_zt)]S_x , \tag{19b}$$

$$S_z(t)=S_z , \tag{19c}$$

where $\Omega_z=2\omega S_z$. To this order, S_z is now a constant of motion. Our zeroth-order or asymptotic solutions may be written simply in a matrix form:

$$\mathbf{S}(t)=\mathsf{M}(t)\mathbf{S} \tag{20}$$

where

$$\mathsf{M}(t)=\begin{bmatrix}\cos(\Omega_z t) & -\sin(\Omega_z t) & 0\\ \sin(\Omega_z t) & \cos(\Omega_z t) & 0\\ 0 & 0 & 1\end{bmatrix}. \tag{21}$$

Observe that $\det\mathsf{M}(t)=\det\mathsf{M}(0)=1$. The time evolution of the large spin is a simple plane rotation in spin space and it is represented by an orthogonal matrix. The rotation is proper.

V. ASYMPTOTIC SOLUTIONS FOR THE BILINEAR SPIN OPERATORS

Now turning to the equations for the bilinear spins (13a)–(13c), we may similarly reduce them to

$$\dot{P}(t)=-2igS_zQ(t)\ , \tag{22a}$$

$$S_z\dot{Q}(t)=-2igS_z^2P(t)+2igUR(t)\ , \tag{22b}$$

$$\dot{R}(t)=2igS_zQ(t)\ . \tag{22c}$$

Observe that (14) remains valid, i.e., $\mathbf{s}\cdot\mathbf{S}$ is still a constant of motion. Also observe that only g still appears in these equations. Asymptotically we may regard the large-spin and bilinear spin operators as classical operators, i.e., commuting operators.

The above asymptotic equations may be solved in a number of ways. Perhaps the simplest is to note that

$$\ddot{Q}(t)+4g^2\mathbf{S}^2Q(t)=0\ . \tag{23}$$

Hence, at once,

$$Q(t)=a\sin(\Omega t)+b\cos(\Omega t),\quad \Omega^2=4g^2\mathbf{S}^2\ , \tag{24}$$

where a and b are integration constants, i.e., constants of motion, to be determined from boundary conditions. Substituting (24) into (22c), we obtain

$$R(t)=(2igS_z/\Omega)[-a\cos(\Omega t)+b\sin(\Omega t)]+c\ . \tag{25}$$

Also by substituting (24) into (22a), we obtain

$$P(t)=-(2igS_z/\Omega)[-a\cos(\Omega t)+b\sin(\Omega t)]+d\ . \tag{26}$$

The two constants c and d are, of course, simply related through (15). That is,

$$P+R=\mathbf{s}\cdot\mathbf{S}=c+d\ . \tag{27}$$

Also by evaluting (22b) by (24) and also by (25) and (26), one gets

$$S_z^2d=Uc\ . \tag{28}$$

Hence, it follows that

$$\mathbf{S}^2c=\mathbf{s}\cdot\mathbf{S}S_z^2 \tag{29}$$

and

$$\mathbf{S}^2d=\mathbf{s}\cdot\mathbf{S}U\ . \tag{30}$$

One can evaluate the remaining two constants a and b similarly:

$$Q(0)=b \tag{31}$$

and

$$R(0)=-(2igS_z/\Omega)a+c\ . \tag{32}$$

Hence,

$$b=-i(s^xS_y-s^yS_x) \tag{33}$$

and

$$\mathbf{S}^2a=(\Omega/2ig)(\mathbf{s}\cdot\mathbf{S}S_z-s^z\mathbf{S}^2)\ . \tag{34}$$

We have thus completely determined the time evolution of the bilinear spin operators, valid to leading order in N. Observe that there is only one frequency Ω, which depends on g, but not on g_z. [See Eqs. (22a)–(22c).]

VI. TIME EVOLUTION OF THE SMALL SPIN: ASYMPTOTIC SOLUTIONS

We now use our asymptotic solutions obtained for the large-spin and bilinear spin operators given in Secs. IV and V to obtain the time evolution of the small-spin operators via Eqs. (12a)–(12c). For the asymptotic solutions, one may neglect the linear terms in (12a) and (12b) [the third term on the right-hand sides of (12a) and (12b)]. After some lengthy but straightforward algebraic manipulations, we obtain our solutions as given below (denoting $X=S_x$, $Y=S_y$, and $Z=S_z$):

$$\begin{aligned}\mathbf{S}^2s^x(t)=&s^x\{[X^2+(\mathbf{S}^2-X^2)\cos\Omega t]\cos\Omega_z t-[XY(1-\cos\Omega t)-SZ\sin\Omega t]\sin\Omega_z t\}\\&+s^y\{[XY(1-\cos\Omega t)+SZ\sin\Omega t]\cos\Omega_z t-[Y^2+(\mathbf{S}^2-Y^2)\cos\Omega t]\sin\Omega_z t\}\\&+s^z\{[XZ(1-\cos\Omega t)-SY\sin\Omega t]\cos\Omega_z t-[YZ(1-\cos\Omega t)+SX\sin\Omega t]\sin\Omega_z t\}\ ,\end{aligned} \tag{35a}$$

$$\begin{aligned}\mathbf{S}^2s^y(t)=&s^x\{[XY(1-\cos\Omega t)-SZ\sin\Omega t]\cos\Omega_z t+[X^2+(\mathbf{S}^2-X^2)\cos\Omega t]\sin\Omega_z t\}\\&+s^y\{[Y^2+(\mathbf{S}^2-Y^2)\cos\Omega t]\cos\Omega_z t+[XY(1-\cos\Omega t)+SZ\sin\Omega t]\sin\Omega_z t\}\\&+s^z\{[YZ(1-\cos\Omega t)+SX\sin\Omega t]\cos\Omega_z t+[XZ(1-\cos\Omega t)-SY\sin\Omega t]\sin\Omega_z t\}\ ,\end{aligned} \tag{35b}$$

$$\mathbf{S}^2s^z(t)=s^x\{XZ(1-\cos\Omega t)+SY\sin\Omega t\}+s^y\{YZ(1-\cos\Omega t)-SX\sin\Omega t\}+s^z\{Z^2+(\mathbf{S}^2-Z^2)\cos\Omega t\}\ , \tag{35c}$$

where $\mathbf{S}^2=S(S+1)\cong S^2$, $\Omega=2gS$, and $\Omega_z=2\omega S_z$. Observe that $s^x(t)$ and $s^y(t)$ both depend on the parameters g and g_z, but $s^z(t)$ depends on g only, as their equations of motion (10a)–(10c) would suggest.

We can simplify the above expressions somewhat by adopting a matrix-like notation which will be useful when comparing with the general solutions later.

$$\mathbf{S}^2 s^x(t)=s^x[m^{xx}\cos(\Omega_z t)-m^{yx}\sin(\Omega_z t) + s^y[m^{xy}\cos(\Omega_z t)-m^{yy}\sin(\Omega_z t)] + s^z[m^{xz}\cos(\Omega_z t)-m^{yz}\sin(\Omega_z t)] , \tag{36a}$$

$$\mathbf{S}^2 s^y(t)=s^x[m^{yx}\cos(\Omega_z t)+m^{xx}\sin(\Omega_z t)] + s^y[m^{yy}\cos(\Omega_z t)+m^{xy}\sin(\Omega_z t)] + s^z[m^{yz}\cos(\Omega_z t)+m^{xz}\sin(\Omega_z t)] , \tag{36b}$$

$$\mathbf{S}^2 s^z(t)=s^x m^{zx}+s^y m^{zy}+s^z m^{zz} , \tag{36c}$$

where

$$m^{xx}=S_x^2+(\mathbf{S}^2-S_x^2)\cos(\Omega t) , \tag{37a}$$

$$m^{xy}=S_xS_y[1-\cos(\Omega t)]+SS_z\sin(\Omega t) , \tag{37b}$$

$$m^{xz}=S_xS_z[1-\cos(\Omega t)]-SS_y\sin(\Omega t) , \tag{37c}$$

$$m^{yx}=S_xS_y[1-\cos(\Omega t)]-SS_z\sin(\Omega t) , \tag{37d}$$

$$m^{yy}=S_y^2+(\mathbf{S}^2-S_y^2)\cos(\Omega t) , \tag{37e}$$

$$m^{yz}=S_yS_z[1-\cos(\Omega t)]+SS_x\sin(\Omega t) , \tag{37f}$$

$$m^{zx}=S_xS_z[1-\cos(\Omega t)]+SS_y\sin(\Omega t) , \tag{37g}$$

$$m^{zy}=S_yS_z[1-\cos(\Omega t)]-SS_x\sin(\Omega t) , \tag{37h}$$

$$m^{zz}=S_z^2+(\mathbf{S}^2-S_z^2)\cos(\Omega t) . \tag{37i}$$

Through these coefficients m^{ij}, one can readily recognize some of the symmetries present in our solutions (36a)–(36c).

Our solutions may be written more compactly in a vector form:

$$\mathbf{S}^2\mathbf{s}(t)=\mathbf{\Sigma}(t)+\{\hat{\mathbf{z}}\times[\hat{\mathbf{z}}\times\mathbf{\Sigma}(t)]\}[1-\cos(\Omega_z t)] + [\hat{\mathbf{z}}\times\mathbf{\Sigma}(t)]\sin(\Omega_z t) , \tag{38}$$

where $\hat{\mathbf{z}}$ is the unit vector in the direction of the z axis in spin space and

$$\mathbf{\Sigma}(t)=\mathbf{S}^2\mathbf{s}\cos(\Omega t)+\mathbf{S}(\mathbf{s}\cdot\mathbf{S})[1-\cos(\Omega t)] + S(\mathbf{s}\times\mathbf{S})\sin(\Omega t) . \tag{39}$$

In discussing the equation of motion for the small spin, it was pointed out that the time evolution of the small spin may be viewed as being induced by its coupling to the large spin, in which g and g_z play the role of coupling strengths. Thus, the motion of the small spin is bound to the plane rotation of the large spin in spin space. If $g=0$ but $g_z\neq 0$, the time evolution of the small spin (35a)–(35c) reduces to

$$s^x(t)=s^x\cos(\Omega_z t)-s^y\sin(\Omega_z t) , \tag{40a}$$

$$s^y(t)=s^x\sin(\Omega_z t)+s^y\cos(\Omega_z t) , \tag{40b}$$

$$s^z(t)=s^z , \tag{40c}$$

where now $\Omega_z=-2g_zS_z$. The small spin undergoes the same rotation as the large spin, i.e., the large spin "drags" the small spin along. As g increases from $g=0$, the motion of the small spin gains an additional independent rotation. The time evolution of the z component of the small spin illustrates the extra rotation quite clearly. It is worth noting here that if $g=0$, then there can be no long-time tails in the SAF of the small spin. As noted, there are no long-time tails in the SAF of the large spin.[23] Coupling between the small and large spins does not necessarily imply the existence of a long-time tail in the SAF of the small spin. To develop a long-time tail, as will be seen in our second paper, an independent rotation must superpose the simple plane rotation in a certain way.

The validity of our solutions may be tested by satisfying self-consistency requirements. For example, if under T, $s_i^x\leftrightarrow s_i^y$ and $s_i^z\to -s_i^z$ for every i, then $TH=H$. Hence, $Ts^y(t)=s^x(t)$, etc. Our solutions satisfy this requirement. To leading order in N, our solutions also satisfy the following requirements:

(1) $\mathbf{s}(t=0)=\mathbf{s}$.

(2) $\mathbf{s}(t)\times\mathbf{s}(t)=i\mathbf{s}(t)$, i.e., the commutation relations of $\mathbf{s}(t)$ are recovered.

(3) $\dot{\mathbf{s}}(t)=i[H,\mathbf{s}(t)]$, i.e., the original equation of motion is recovered.

(4) If $\omega=0$, the components of $\mathbf{s}(t)$ are cyclic.

(5) $\mathbf{s}^2(t)=3/4$, i.e., a constant of motion.

(6) $\mathbf{s}(t)\cdot\mathbf{S}(t)=\mathbf{s}\cdot\mathbf{S}$, i.e., also a constant of motion.

(7) $[\mathbf{s}(-t)]^*=\mathbf{s}^*(t)$, i.e., invariant under the time-reversal operation.

Finally, our asymptotic solutions are recovered from our general solutions given in the Appendix if $N\to\infty$ therein. We are thus satisfied that our solutions are exactly valid, to leading order in N.

VII. DISCUSSION

Our solutions for the time evolution of the small spin are purely formal and without specific physical content (i.e., all possibilities are given, unweighted). The physical significance will emerge when we evaluate the SAF. We may still lend them some qualitative interpretation.

It was pointed out that the time evolution of the large spin represents a plane rotation in spin space and the rotation matrix is orthogonal. In analogy, we may also write the time evolution of the small spin in a matrix form:

$$\mathbf{s}(t)=\mathsf{L}(t)\mathbf{s} \tag{41}$$

where $\mathsf{L}(t)$ is a 3×3 matrix whose elements may be identified from (35) or (36). Although somewhat tedious, one can show that

$$\det\mathsf{L}(t)=1 , \tag{42}$$

as it must since $\mathbf{s}(t)$ is a "vector" with constant "length."

The time evolution of the small spin also represents a rotation in spin space, and its rotation matrix is orthogonal. The relationship between the time evolutions of the small and large spins may thus be viewed from the relationship between their rotations in spin space.

The large spin, being treated as a classical spin, may be regarded as an ordinary real vector in a three-dimensional space. The small spin is not a similar vector in the same space. It is a spinor. Allowing for this difference, we see that the small-spin vector maintains a constant angle relative to the large-spin vector since $\mathbf{s}(t)\cdot\mathbf{S}(t)=\mathbf{s}\cdot\mathbf{S}$. That is, the motion of the small-spin vector forms a cone with a constant angle about the large-spin vector. But this motion is not orthogonal to the other vector, i.e., $\dot{\mathbf{s}}(t)\cdot\mathbf{S}(t)\neq 0$. One can find another ordinary vector, say $\tau(t)$, which is orthogonal to the motion of the small-spin vector. Such a vector is

$$\tau=(2gS_x,2gS_y,2g_zS_z) \ . \tag{43}$$

Then the equation of motion for the small spin may be written in the form

$$\dot{\mathbf{s}}(t)=\mathbf{s}(t)\times\tau(t) \ . \tag{44}$$

The above is exactly in the form of the equation of motion for, e.g., a classical angular momentum of constant magnitude, rotating about the direction of a magnetic field.[26] This is the Larmor precession. In this semiclassical analogy, $\tau(t)$ acts as the external magnetic field, about which the small-spin vector may be said to precess.[27] The relative angle between $\mathbf{s}(t)$ and $\tau(t)$, however, is not a constant of motion (except at $g=0$ and $g=g_z$), i.e., $\mathbf{s}(t)\cdot\tau(t)\neq\mathbf{s}\cdot\tau$, which apparently is an indication that the motion of the small spin is a Larmor-like precession.

Finally we shall turn to the meaning of our asymptotic solutions. Recall that we have divided our system into two subsystems. When the larger subsystem is much larger than the smaller subsystem (i.e., $N\gg 1$), we have assumed that the time evolution of the larger subsystem is unaffected by the interaction between the two subsystems. That is,

$$\begin{aligned}\mathbf{S}(t)&=\exp(iHt)\mathbf{S}\exp(-iHt)\\ &=\exp[i(H_0+V)t]\mathbf{S}\exp[-i(H_0+V)t]\\ &=\exp(iH_0t)\mathbf{S}\exp(-iH_0t)\{1+O(N^{-1})\} \ .\end{aligned} \tag{45}$$

The large spin evolves in time according to the conditions of its own system only.

The significance of the above approximation may be best seen through the following: For the total system of H, $S_z^{\rm tot}$ is a constant of motion but s^z and S_z individually are not. That is,

$$s^z(t)+S_z(t)=s^z+S_z \ . \tag{46}$$

Their time evolutions must be such as to exactly cancel out one another. This implies that $[V,s^z]=-[V,S_z]$, as may be verified. According to (46),

$$S_z(t)=S_z+[s^z-s^z(t)] \ . \tag{47}$$

With respect to S_z, the terms inside the square brackets contribute to lower orders in N, i.e., fluctuations may be neglected if $N\to\infty$. Also, according to (46),

$$s^z(t)=s^z+[S_z-S_z(t)] \ . \tag{48}$$

Now the terms inside the square bracketes are of the same order in N as s^z. They no longer represent fluctuations and thus they may not be neglected.

The small spin evolves in time not only according to the conditions of its own system, but also governed by its interaction with the large spin. To obtain the proper asymptotic solutions of the time evolution of the small spin, one must thus treat its equation of motion exactly.

It may be worth pointing out that this is in contrast to obtaining time evolutions according to stochastic theories of nonequilibrium statistical mechanics. In those theories, one assumes *a priori* that the random force $\mathbf{F}(t)$ exerted on the small spin $\mathbf{s}(t)$ is stationary and Gaussian and also that it has an infinitely short correlation time, $\langle\mathbf{F}(t)\cdot\mathbf{F}(t')\rangle\sim\delta(t-t')$, i.e., a white-noise power spectrum of $\mathbf{F}(t)$.[28] The underlying equation of motion is the *classical* Langevin equation, whose solutions would yield a simple exponential decay form for the autocorrelation function $\langle\mathbf{s}(t)\cdot\mathbf{s}(t')\rangle$. There is a built-in irreversibility in the time-evolution solutions. In our work, which may be classified as an example of Hamiltonian dynamics, we have made no such assumptions. In our time-evolution solutions, there is always time reversibility, i.e., the solutions are even in t.[29] See Eqs. (35a)–(35c). They are admissible solutions of the *generalized* Langevin equation.[30] We shall show in our second paper that our time-reversible solutions can give rise to long-time tails in the autocorrelation function.

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APPENDIX: TIME EVOLUTION OF A SINGLE SPIN, WHEN $N<\infty$

We shall now obtain solutions for the time evolution of a single spin when N, the number of spins in our system, is finite. When N is finite, there is no advantage to be gained by dividing the system into two subsystems. Hence, we shall let $H=H_0$, $V=0$. Also, $\mathbf{S}^{\rm tot}=\mathbf{S}$. Now the small spin $\mathbf{s}$ is any one of the spins that constitute the large spin $\mathbf{S}$. It is now more appropriate to refer to $\mathbf{s}$ and $\mathbf{S}$ as a single spin and a total spin. Observe that the single and total spins no longer commute, e.g., $[\mathbf{S}^2,s^z]\neq 0$. Important constants of motion for this system are $\mathbf{S}^2$, S_z, $\mathbf{s}^2$, $\mathbf{s}\cdot\mathbf{S}$, and U.

One immediate advantage of making $H=H_0$ is that the time evolution of the total spin is already known. We have shown that[23]

$$S_x(t)=e^{i\omega t}[S_x\cos(\Omega_z t)-S_y\sin(\Omega_z t)] , \quad (A1a)$$

$$S_y(t)=e^{i\omega t}[S_y\cos(\Omega_z t)+S_x\sin(\Omega_z t)] , \quad (A1b)$$

$$S_z(t)=S_z , \quad (A1c)$$

where $\omega=(J-J_z)/N$ and $\Omega_z=2\omega S_z$. The above form is convenient for our purposes here, obtained by taking Hermitian conjugation of our original results.

Our method of solution is the same as before when $N\to\infty$. But now the total-spin operators cannot be treated as classical spin operators, i.e., their noncommutativity must be respected. As a result, it is much more complicated to obtain solutions for the time evolution of a single spin. In what follows we shall indicate necessary steps and present our solutions without showing many intermediate steps.

The equation of motion for a single spin is given below:

$$\dot{s}^x(t)=-2gs^z(t)S_y(t)+2g_zs^y(t)S_z-i(g+g_z)s^x(t) , \quad (A2a)$$

$$\dot{s}^y(t)=2gs^z(t)S_x(t)-2g_zs^x(t)S_z-i(g+g_z)s^y(t) , \quad (A2b)$$

$$\dot{s}^z(t)=2g[s^x(t)S_y(t)-s^y(t)S_x(t)]-2igs^z(t) . \quad (A2c)$$

Observe that the above equations reduce to the original equations (10a)–(10c) if we let $\mathbf{S}\to\mathbf{s}+\mathbf{S}$. To solve these equations, we introduce, as before, the same bilinear spin operators P, Q, and R [see (11)], where now the large spin means the total spin. Note that P, Q, and R commute with S_z, but not with U. We then obtain $P(t)$, $Q(t)$, and $R(t)$ from their equations of motion, whence $\mathbf{s}(t)$ via (12).

The equations of motion for the bilinear spin operators are

$$\dot{P}(t)=-2igS_zQ(t)+2igR(t) , \quad (A3a)$$

$$S_z\dot{Q}(t)=-2igS_z^2P(t)+2igR(t)U , \quad (A3b)$$

$$\dot{R}(t)=2igS_zQ(t)-2igR(t) . \quad (A3c)$$

The above equations appear slightly simpler than their analogs, Eqs. (13a)–(13c). Observe that one still has

$$\dot{P}(t)+\dot{R}(t)=0 . \quad (A4)$$

Hence,

$$P(t)+R(t)=\mathbf{s}\cdot\mathbf{S} . \quad (A5)$$

To solve the equations of motion for the bilinear spin operators, it is perhaps simplest to begin by taking one more derivative of (A3c). It results in an equation entirely of R, hence, soluble at once. Using this solution one may obtain $P(t)$ via (A5), thereupon $Q(t)$ via (A3b). Constants appearing in these solutions may be identified through the boundary conditions at $t=0$ or by connection to the constants of motion mentioned earlier. We will not exhibit them here, but the solutions for $P(t)$, $Q(t)$, and $R(t)$ are analogous to those given in Sec. V.

To obtain the time evolution of a single spin, we use the above results in (12a)–(12c). It is straightforward to obtain $s^z(t)$. But it is very complicated to obtain $s^x(t)$ and $s^y(t)$ because of the presence of the third term on the right-hand side of (12a) or (12b), which could be neglected when $N\to\infty$. As a result, each term contributing to the final solution contains the factor $(U^2-S_z^2)$, which finally cancels out. Let us first define the following quantities:

$$A=e^{-igt}\cos(\Omega t) , \quad (A6)$$

$$B=e^{-igt}g\sin(\Omega t)/\Omega , \quad (A7)$$

$$\Omega=g(4\mathbf{S}^2+1)^{1/2} . \quad (A8)$$

In terms of the above quantities, also with $X=S_x$, $Y=S_y$, and $Z=S_z$, our final solutions are

$$\begin{aligned} s^x(t)\mathbf{S}^2e^{-i\omega t}=&s^x\{[X^2+(\mathbf{S}^2-X^2)A-i(\mathbf{S}^2+X^2)B]\cos(\Omega_z t)-[XY(1-A)-(2Z\mathbf{S}^2+iXY)B]\sin(\Omega_z t)\}\\ &+s^y\{[YX(1-A)+(2Z\mathbf{S}^2-iYX)B]\cos(\Omega_z t)-[Y^2+(\mathbf{S}^2-Y^2)A-i(\mathbf{S}^2+Y^2)B]\sin(\Omega_z t)\}\\ &+s^z\{[ZX(1-A)-(2Y\mathbf{S}^2+iZX)B]\cos(\Omega_z t)-[ZY(1-A)+(2X\mathbf{S}^2-iZY)B]\sin(\Omega_z t)\} , \end{aligned} \quad (A9a)$$

$$\begin{aligned} s^y(t)\mathbf{S}^2e^{-i\omega t}=&s^x\{[XY(1-A)-(2Z\mathbf{S}^2+iXY)B]\cos(\Omega_z t)+[X^2+(\mathbf{S}^2-X^2)A-i(\mathbf{S}^2+X^2)B]\sin(\Omega_z t)\}\\ &+s^y\{[Y^2+(\mathbf{S}^2-Y^2)A-i(\mathbf{S}^2+Y^2)B]\cos(\Omega_z t)+[YX(1-A)+(2Z\mathbf{S}^2-iYX)B]\sin(\Omega_z t)\}\\ &+s^z\{[ZY(1-A)+(2X\mathbf{S}^2-iZY)B]\cos(\Omega_z t)+[ZX(1-A)-(2Y\mathbf{S}^2+iZX)B]\sin(\Omega_z t)\} , \end{aligned} \quad (A9b)$$

$$\begin{aligned} s^z(t)\mathbf{S}^2=&s^x\{(XZ(1-A)+(2Y\mathbf{S}^2-iXZ)B\}+s^y\{(YZ(1-A)-(2X\mathbf{S}^2+iYZ)B\}\\ &+s^z\{(Z^2+(\mathbf{S}^2-Z^2)A-i(\mathbf{S}^2+Z^2)B\} . \end{aligned} \quad (A9c)$$

One can express the above results somewhat more simply in a matrix-like notation:

$$s^x(t)\mathbf{S}^2e^{-i\omega t}=s^x[m^{xx}\cos(\Omega_z t)-m^{yx}\sin(\Omega_z t)]+s^y[m^{xy}\cos(\Omega_z t)-m^{yy}\sin(\Omega_z t)]+s^z[m^{xz}\cos(\Omega_z t)-m^{yz}\sin(\Omega_z t)] , \quad (A10a)$$

$$s^y(t)\mathbf{S}^2 e^{-i\omega t}=s^x[m^{yx}\cos(\Omega_z t)+m^{xx}\sin(\Omega_z t)]+s^y[m^{yy}\cos(\Omega_z t)+m^{xy}\sin(\Omega_z t)]+s^z[m^{yz}\cos(\Omega_z t)+m^{xz}\sin(\Omega_z t)]\ , \tag{A10b}$$

$$s^z(t)\mathbf{S}^2=s^x m^{zx}+s^y m^{zy}+s^z m^{zz}\ , \tag{A10c}$$

where

$$m^{xx}=S_x^2+(\mathbf{S}^2-S_x^2)A-i(\mathbf{S}^2+S_x^2)B\ , \tag{A11}$$

$$m^{xy}=S_yS_x(1-A)+(2S_z\mathbf{S}^2-iS_yS_x)B\ , \tag{A12}$$

$$m^{xz}=S_zS_x(1-A)-(2S_y\mathbf{S}^2+iS_zS_x)B\ , \tag{A13}$$

$$m^{yx}=S_xS_y(1-A)-(2S_z\mathbf{S}^2+iS_xS_y)B\ , \tag{A14}$$

$$m^{yy}=S_y^2+(\mathbf{S}^2-S_y^2)-i(\mathbf{S}^2+S_y^2)B\ , \tag{A15}$$

$$m^{yz}=S_zS_y(1-A)+(2S_x\mathbf{S}^2-iS_zS_y)B\ , \tag{A16}$$

$$m^{zx}=S_xS_z(1-A)+(2S_y\mathbf{S}^2-iS_xS_z)B\ , \tag{A17}$$

$$m^{zy}=S_yS_z(1-A)-(2S_x\mathbf{S}^2+iS_yS_z)B\ , \tag{A18}$$

$$m^{zz}=S_z^2+(\mathbf{S}^2-S_z^2)-i(\mathbf{S}^2+S_z^2)B\ . \tag{A19}$$

The correctness of our solutions may be tested through self-consistency requirements mentioned in connection with our asymptotic solutions. There is one additional possibility: $\sum \mathbf{s}=\mathbf{S}$. If all the single spins, each given by (A9a)–(A9c) or (A10a)–(A10c,) are summed, indeed our results for the total spin (A1a)–(A1c) are recovered.

Also, our asymptotic solutions are recovered if $S\rightarrow\infty$, where $\mathbf{S}^2=S(S+1)$, i.e., $N\rightarrow\infty$. If $S\rightarrow\infty$, $A\rightarrow\cos(2gSt)$ and $B\rightarrow\sin(2gSt)/2S$. Also, $g\rightarrow 0$ and $\omega\rightarrow 0$ but $gS\rightarrow$const. Hence B is asymptotically one order lower in N than A. If we substitute these asymptotic forms into (A11)–(A19), we find that all the imaginary terms may be dropped. If the noncommutativity in the total-spin operators is ignored, we recover the asymptotic solutions of Sec. V.

Finally, our solutions may be compactly expressed in a vector form as follows:

$$\mathbf{s}(t)\mathbf{S}^2=\mathbf{\Sigma}(t)+[\hat{\mathbf{z}}\times\mathbf{\Sigma}(t)]\sin(\Omega_z t)e^{i\omega t} -\{\hat{\mathbf{z}}\times[\mathbf{\Sigma}(t)\times\hat{\mathbf{z}}]\}[1-\cos(\Omega_z t)e^{i\omega t}]\ , \tag{A20}$$

where $\hat{\mathbf{z}}$ is the unit vector in the direction of the z axis and

$$\mathbf{\Sigma}(t)=(\mathbf{s}\cdot\mathbf{S})\mathbf{S}(1-A)+s\mathbf{S}^2A +[2(\mathbf{s}\times\mathbf{S})\mathbf{S}^2-i(\mathbf{s}\mathbf{S}^2+(\mathbf{s}\cdot\mathbf{S})\mathbf{S}]B\ . \tag{A21}$$

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