Semiclassical mechanics of a nonintegrable spin cluster

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We study detailed classical-quantum correspondence for a cluster system of three spins with single-axis anisotropy exchange coupling. With autoeprogressive spectral estimation, we find oscillating terms in the quantum density of states caused by classical periodic orbits in the slowly varying part of the density of states we see signs of nontrivial topology changes happening to the energy surface as the energy is varied. Also, we can explain the hierarchy of quantum energy levels near the ferromagnetic and antiferromagnetic states with Einstein-Brillouin-Keller quantization to explain large structures and tunneling to explain small structures. [S1063-1829/99/03445-1]

1. INTRODUCTION

When S is large, spin systems can be modeled by classical and semiclassical techniques. Here we reserve “semiclassical” to mean not only that the technique works in the limit of large S (as the term is sometimes used) but that it implements the quantum-classical correspondence (relating classical trajectories to quantum-mechanical behavior).

Spin systems (in particular S = 1/2) are often thought of as the antithesis of the classical limit. Notwithstanding that, classical-quantum correspondence has been studied at large values of S in systems such as an autonomous single spin, kicked single spin, and autonomous two- and three-spin systems.

When the classical motion has a chaotic regime, for example, the dependence of level statistics on the regularity of classical motion has been studied. In regimes where the motion is predominantly regular, the pattern of quantum levels of a spin cluster can be understood with a combination of EBK (Einstein-Brillouin-Keller, also called Bohr-Sommerfeld) quantization and tunneling splitting (Sec. V is such a study for the current system). The latter sort of calculation has potential applications to some problems of current numerical or experimental interest. Numerical diagonalizations for extended spin systems (in ordered phases) on lattices of modest size (10 to 36 spins) may be analyzed by treating the net exchange of each sublattice as a single large spin and thereby reducing the system to an autonomous cluster of a few spins; the clustering of low-lying eigenvalues can probe symmetry breaking that are obscured in a system of such size if only ground-state correlations are examined. Nonlinear self-localized modes in spin lattices, which typically span several sites, have to date been modeled classically, but seem well suited to semiclassical techniques. Another topic of recent experiments is the molecular magnets such as Mn12Ac and Fe8, which are more precisely modeled as clusters of several interacting spins rather than a single large spin; semiclassical analysis may provide an alternative to exact diagonalization techniques for theoretical studies of such models.

In this paper, we will study three aspects of the classical-quantum correspondence of an autonomous cluster of three spins coupled by easy-plane exchange anisotropy, with the Hamiltonian

\[
H = \sum_{i=1}^{3} S_i \cdot S_i - \alpha \sum_{i=1}^{3} S_i^{z}.
\]

This model was introduced in Ref. 9, a study of level repulsion in regions of (E, σ) space, where the classical dynamics is predominantly chaotic. Equation (1) has only two nontrivial degrees of freedom, since it conserves total angular momentum about the z axis. As did Ref. 9 we consider only the case of \(S_i^z = \frac{S_i}{2} = 0\). While studying classical mechanics we set \(S_i^z = 1\); to compare quantum energy levels at different \(S_i^z\), we divide energies by \(S_i^z\) for \(S_i^z > 0\) and \(E - \sum_{i=1}^{3}s_i^z\) for \(S_i^z < 0\).

Below, in Secs. IV and V, we will discuss topology along distorted circles, 120° out of phase. The type of large structures and tunneling to explain small structures. This model was introduced in Ref. 9, a study of level repulsion in regions of (E, σ) space, where the classical dynamics is predominantly chaotic. Equation (1) has only two nontrivial degrees of freedom, since it conserves total angular momentum about the z axis. As did Ref. 9 we consider only the case of \(S_i^z = \frac{S_i}{2} = 0\). While studying classical mechanics we set \(S_i^z = 1\); to compare quantum energy levels at different \(S_i^z\), we divide energies by \(S_i^z\) for \(S_i^z > 0\) and \(E - \sum_{i=1}^{3}s_i^z\) for \(S_i^z < 0\).
example of an orbit spectrum.) Since the classical period \( T = \pi /\lambda \) is large if there exists a periodic orbit with energy \( E \) and period \( T \), the orbit spectrum can be estimated by Fourier transform,\(^{16}\)

\[
O(H, \tau) = \frac{1}{\delta} \sum_{k=-\infty}^{\infty} \delta \int_{E}^{E+\delta} \psi_k^*(E) \psi_k(E+\delta) e^{-i\tau x} dx .
\]

(7)

Variants of Eq. (7) have been used to extract information about classical periodic orbits from quantum spectra.\(^{17,18}\) Unfortunately, the resolution of the Fourier transform is limited by the uncertainty principle, \(-\delta E \delta \tau \approx h/2\).\(^{19}\)

Nonlinear spectral estimation techniques, however, can surpass the resolution of the Fourier transform.\(^{19}\) One such technique, harmonic inversion, has been successfully applied to scaling systems\(^{-16}\)—i.e., systems like billiards or Kepler systems in which the classical and quantum dynamics at one energy are identical to those at any other energy, after a rescaling of time and coordinate scales. In a scaling system, windowing is unnecessary because there are no bifurcations and the scaled periods of orbits are constant. In this section, we will apply nonlinear spectral estimation to our system (1), which is nonscaling.\(^{20}\)

A. Diagonalization

To get the quantum level spectrum, we wrote software to diagonalize arbitrary spin Hamiltonians which are polynomial in \((S^x, S^y, S^z)\), where \(i\) is an index running over arbitrary \(N\) spins of arbitrary (and often large) spin \(S\). The program, written in JAVA, takes advantage of discrete translational and parity symmetries by constructing a basis set in which the Hamiltonian is block diagonal, letting us diagonalize the blocks independently with an optimized version of LAPACK. Illustrating the spins in a ring, the Hamiltonian Eq. (1) is invariant to cyclic permutations of the spins, so the eigenstates are states of definite wave number \(k = 0, \pm 2 \pi /N\) (matrix blocks for \(k = \pm 2 \pi /N\) are identical by symmetry). In the largest system we diagonalized (three-spin cluster with \(S = 65\)), the largest blocks contained \(N = 4620\) states.

B. Autoregressive approach to construct spectrum

The input to an orbit spectrum calculation is the list of discrete eigenenergies with total \(S_z = 0\); no other information on the eigenstates (e.g., the wave number quantum number) is necessary. This level spectrum is smoothed by convolving with a Gaussian (width \(10^{-3}\)) for Fig. 3 and discretely sampling over energy (with sample spacing \(6.5 \times 10^{-3}\)).

We estimate the power spectrum by the autoregressive (AR) method. AR models a discretely sampled input signal, \(y_n\) (in our case the density of states), with a process that attempts to predict \(y_n\) from its previous values,

\[
y_n = \sum_{j=1}^{N} a_j y_{n-j} + \epsilon_n .
\]

(8)

Here \(N\) is a free parameter that determines how many spectral peaks that model can fit. Refs. 19 and 22 discuss guidelines for choosing \(N\). Fast algorithms exist to implement least-squares, i.e., to choose \(N\) coefficients \(a_j\) to minimize (within constraints) \(\sum_j (\epsilon_j)^2\), of these we used the Burg algorithm.\(^{23}\)

To estimate the power spectrum, we discard the original \(x_i\) and model \(x_i\) with uncorrelated white noise. Thinking of Eq. (8) as a filter acting on \(x_i\), the power spectrum of \(y_i\) is computed from the transfer function of Eq. (8) and is

\[
P(\nu) = \frac{\langle \xi^2 \rangle}{1 - \sum a_j e^{i\nu j}} .
\]

(9)

Unlike the discrete Fourier transform, \(P(\nu)\) can be evaluated at any value of \(\nu\). In our application, of course, \(\delta\) has units of energy, so \(\nu\) (more exactly \(\hbar\nu /\delta\)) actually has units of time and is to be identified with \(\tau\) in Eq. (7).

C. Orbit spectrum results and discussion

Figure 3 shows the orbit spectrum of our system with \(S = 65\) and \(S_z = 0\); it is displayed as a 5000x500 array of pixels, colored light where \(O(H, \tau)\) is large. Each horizontal row is the power spectrum in an energy window centered at \(E\); we stack rows of varying \(H\) vertically. With a window width 250 energy samples long (\(50^2 = 11250\)), we fit \(N = 150\) coefficients in Eq. (8). To improve visual resolution, we let windows overlap and space the centers of successive windows 25 samples apart.

Comparing Fig. 3 and Fig. 2 we see that our orbit spectrum detects the fundamental periodic orbits as well as multiple transversals of the orbits. Interestingly, we produced Fig. 3 before we had identified most of the fundamental orbits; Fig. 3 correctly predicted three out of four families of orbits.

We believe that, given the same data, the AR method normally produces a far sharper spectrum than Fourier analysis. This is not surprising, since the Fourier analysis allows the holes that appeared in the energy surface at \(E_s\) close up. A discontinuity in the slope of the area of the energy surface occurs at energy \(E_c\), but is not visible in Fig. 4; in the range \(E_c < E < E_s\), the area of the energy surface (and hence the slowly varying part of the DOS) seems to be constant as a function of energy.

In the special isotropic \((\sigma = 0)\) case, the flat interval is \((-1.5, -1)\) and it can be analytically derived that the DOS is constant there. This is simplest for the smoothed quantum DOS, since for \(\sigma = 1, 2, \ldots\), there are clusters of energy levels with level spacing proportional to \(n\). (A derivation also exists for the classical case, but is less direct.) We have no analytic results for general \(\sigma\).

This flat interval is specific to our three-spin cluster, but we expect that the compactness of spin phase space will, generally, cause changes in the energy surface topology of spin systems that do not occur in traditionally studied particle systems.

V. LEVEL CLUSTERING

The quantum levels with total \(S_z = 0\) show rich patterns of clustering, some of which are visible on Fig. 5. The levels that form clusters correspond to different regimes of the classical dynamics in which the motion becomes nearly regular: (1) the FM limit (not visible in Fig. 5); (2) the AFM limit (bottom edge of Fig. 5); and (3) the isotropic limit \(\sigma = 0\) (left edge of Fig. 5). Indeed, the levels form a hierarchy as the clusters break up into subclusters. In this section, we first approximately map the phase space from four coordinates to two coordinates—with the topology of a sphere. (Two of the original six coordinates are trivial, or decoupled, due to symmetry, as noted in Sec. B.) Then, using Einstein- Brillouin-Keller (EKB) quantization and some consideration of quantum tunneling, many features of the level hierarchy will be understood.

A. Generic behavior: The polyad phase sphere

In all three limiting regimes, the classical dynamics become trivial. For small deviations from the limit, the equa-
same, too. Then different tunneling paths connect must differ in phase by a topo-
logical term, with a familiar form proportional to the (real part of) the spherical area between the two paths.28

B. Results

Here we summarize some observations made by exami-
nation of polyads in the three regimes, for a few combina-
tions of $S$ and $\alpha$.

1. Ferromagnetic Limit

This regime is the best-behaved in that regular behavior persists for a wide range of energies. The ferromagnetic
state is the energy maximum, is a fixed number of the dynas-
tics around it are "spin-wave" excitations (viewing our system as the three-site case of a one-dimensional ferromagnet).

The two are oscillators in which the polyad is con-
structed. Thus, the "pole" points in Fig. 6 correspond to
"spin waves" propagating clockwise or counterclockwise
around the ring of three spins, an example of the "three-
phase" type of orbit. The stable and unstable points on
the "equator" are identified, respectively, with the orbits (a) and
(c) of Fig. 1. Classically, in this regime, the three-phase orbit
is the fundamental orbit with lowest frequency $\omega_{3\text{-phase}}$:
thus the corresponding levels in successive polyads have a
somewhat smaller spacing $h\omega_{3\text{-phase}}$ as compared to
the energy end of each polyad consists of twofold subclusters and the low-energy end consists of threefold subclusters.

We see the appearance of fine structure (presumably tunnel splittings) that is just like the pattern in the four-spin problem. Namely, throughout each polyad the degenera-
tions successively levels follow the pattern (2,1,1,2) and im-
prove. (Here—as also for regime 3—every "2" level has $k \equiv 1 \pmod{2}$ and every "1" level has $k = 0$, where wave number $k$ is defined in Sec. III A.) Numerical data show that (inde-
dependent of $S$) the pattern (starting from the lowest energy
begins (2112...) for even $P$, but for odd $P$ it begins (2211...).

In the energy range of twofold subclusters, the levels are grouped as (21)(112), i.e., one tunnel-split subcluster be-
tween two unsplit subclusters (and repat); in the threelfold subcluster regime, the grouping is (21)(12), so that each sub-
cluster gets tunnel-split into a pair and a single level, but the
sense of the splitting alternates from one subcluster to the
next.

An analysis of $\sigma = 0.4, S = 30$ showed that the fraction of the subclusters indeed grows from around 0.3 for small $P$ to nearly 0.5 as $P \to 40$. Furthermore, when $aP^2$ and $bP^3$ were estimated by the method described near the end of Sec. VA, they indeed scaled as $P^2$ and $P^3$, respectively.

2. Antiferromagnetic Limit

This regime occurs as $E = E_{\text{AF}}(\sigma)$, where $E_{\text{AF}}(\sigma)$ is given by Eq. (4). That means the classical energy surface is divided into two disconnected pieces, related by a mirror reflection of all three spins in any plane normal to the easy plane. Analogous to regime one, two degenerate antiferromagnetic

"spin waves" exist around either energy minimum, and the
polyad states are built from the levels of these two oscilla-
tors. Thus the clustering hierarchy outlined in Sec. VA—
polyads clusters, EBK, and tunneling over barriers of $I$ on the polyad phase sphere—is repeated within each disconnected piece, leading to a prediction that all lev-
els should be twofold degenerate.

Consequently, on the level diagram (Fig. 5), there should be half the apparent level density below the line $E = E_{\text{AF}}(\sigma)$ as above. Indeed, a rough inspection of the

apparent level crossing behavior is visible at that line (shown dashed in the figure).

Actually, tunneling is possible between the disconnected pieces of the energy surface and may split these degenerate pairs. In fact this hyperbolic splitting happens to 1/3 of the pairs, again following the (2112) pattern within a given polyad. This (2112) pattern starts to break up as the energy moves away from the AFM limit; even for large $S$ (30 or 65), this breakup happens already around the polyad with $P = 10$, so it is much harder than in the FM case to ascertain the asymptotic pattern of subclustering. We conjecture that the breakup may happen near the energies where, classically, the stable periodic orbits bifurcate and a small bit of phase space goes chaotic.

The barrier for tunneling between the disconnected energy surfaces has the energy scale of the bare Hamiltonian, which is much larger (at least, for small $P$) than the scale of effective Hamiltonian I, which provides the barrier for tun-
neling among the states in a subcluster. Hence, the hyperbolic
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by Fig. 6 and Eq. (11). For example, an umbrellalike configuration in which the three-spin directions are equally tilted out of their plane corresponds to a three-phase type orbit, with two cases depending on the handedness of the arrangement. A configuration where one spin is parallel/antiparallel to the net moment, and the other two spins offset symmetrically around it, follows one of the threefold degenerate orbits. Numerically, the level behavior in the near-isotropic limit is similar to the near-6FM limit. The fine-structure degeneracies are a repeat of the (2121) pattern as in the other regimes; the lowest levels of any polyad always begin with (1221). The fraction of threedfold subclusters is large here and, as expected, grows with $S$ (from 0.5 to 0.7 in the case $S = 15$). However, the energy scales of $\alpha^2P_1$ and $\beta^2P_1$ behave numerically as $\sigma^{1/2}$ and $\beta^{1/2}$. What is different about the isotropic limit is that the precession frequency—hence the oscillator frequency $\omega$—is not a constant, but is proportional to $S_{2z}$. Since perturbation techniques give formulas for $I$ with inverse powers of $\omega$, it is plausible that $\alpha$ and $\beta$ in Eq. (11) include factors of $P^{-2}$ here, which were absent in the other two regimes.

VI. CONCLUSION AND SUMMARY

To summarize, by using detailed knowledge of the classical mechanics of a three spin cluster, we have studied the semiclassical limit of spins in three ways. First, using autoregressive spectral analysis, we identified the oscillating contributions that the fundamental orbits of the cluster make to the density of states; in fact, we detected the quantum signature of the orbits before discovering them. Second, we verified that the quantum DOS is proportional to the area of the energy surface; we also observed kinks in the smoothed quantum DOS, which are the quantum manifestation of topology changes of the energy surface; such topology changes, we expect, are more common in spin systems than particle phase space, since even a single spin has a nontrivial topology. Finally, we have identified three regimes of near-regular behavior in which the levels are clustered according to a four-level hierarchy, and we explained many features qualitatively in terms of a reduced, one degree-of-freedom system. This system appears promising for two extensions analogous to Ref. 5: normal amplitudes (and their topological phases) could be computed more explicitly; also, the low-energy levels from exact diagonalization of a finite system with $\theta$ that is the anisotropic exchange antiferromagnet on the triangular lattice could probably be mapped to three large spins and analyzed in the fashion sketched above in Sec. V.

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References

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[12] This is an example of the general phenomenon of symmetry-breaking bifurcations of orbits. For a systematic discussion, see M. A. de Aguiar and C. P. Malta, Ann. Phys. (N.Y.) 180, 167 (1987); our case corresponds to (b) in their Table I.
[13] As Ref. 13 notes, the third-order expansion of the Hamiltonian near a ground state is the same for our model (in the right coordinates) as for the famous Hénón-Heiles model [M. Hénón and C. Hélie, Astron. J. 69, 73 (1964)]. Hence, we see similar behaviors near the limit [e.g., the dependence on excitation energy of the frequency splitting between orbits (b) and (c)].
[17] $\alpha(\varepsilon, E)$ contains a phase factor, $\exp(\varepsilon)$, where $\alpha$ is the Maslov index and depends on the topology of the linearized dynamics near the orbit; see J. M. Robbins, Nonlinearity 4, 343 (1991). As we do not consider amplitude or phase, the Maslov index is irrelevant for this paper.
[18] Equation (7) is for illustration. The square window aggravates artifacts of the Fourier transform that could be reduced by using a different window function (see Ref. 19).
[24] In fact, the classical dynamics of our system do scale if one rescales the spin length $S$ simultaneously. However, in contrast to the mentioned scaling systems, $S$ in a spin system is not just a numerical parameter but a discrete quantum number.
[25] As Ref. 13 notes, the third-order expansion of the Hamiltonian near a ground state is the same for our model (in the right coordinates) as for the famous Hénón-Heiles model [M. Hénón and C. Heiles, Astron. J. 69, 73 (1964)]. Hence, we see similar behaviors near the limit [e.g., the dependence on excitation energy of the frequency splitting between orbits (b) and (c)].