Large-field versus discontinuous instantons in spin orientation tunneling

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Abstract

Tunnel splitting oscillations in magnetic molecules are reconsidered within the simplest model for the problem, which does not contain fourth order anisotropy. It is shown that at large magnetic field, there is only one instanton, and it is continuous. This is in contrast to the discontinuous instantons that are induced by the fourth order term [Ersin Keçecioğlu and A. Garg, Phys. Rev. Lett. 88, 237205 (2002)].

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I. INTRODUCTION

The purpose of this short note is to clarify the differences between certain types of instantons that arise in the study of spin orientation tunneling in molecular magnets. The problem derives from the specific phenomenon of tunnel splitting or gap oscillations in the molecular ion \([(tacn)_6Fe_8O_2(OH)_{12}]^{8+}\) (abbreviated to Fe$_8$ henceforth) [1], but our considerations should apply to other molecular magnetic system as well.

The simplest model Hamiltonian that leads to gap oscillations is

\[ H_0 = k_1 J_z^2 + k_2 J_y^2 - g \mu_B J \cdot H, \]  

(1.1)

where $J$ is a dimensionless spin operator, $k_1 > k_2 > 0$, and $H$ is an external magnetic field. It is known that for $H \parallel \hat{z}$, the tunnel splitting, or gap, between the two lowest eigen states of $H_0$ oscillates as a function of $H_z$, vanishing whenever

\[ H_z = \frac{1}{J} \left( J - n - \frac{1}{2} \right) H_c. \]  

(1.2)

Here,

\[ \lambda = k_2/k_1, \text{ and } H_c = 2k_1J/g\mu_B. \]  

(1.3)

For Fe$_8$, for which $J = 10$, $k_1 = 0.338$ K, and $k_2 = 0.246$ K, this would imply that the splitting is quenched at 10 values of $H_z > 0$, equally spaced by 0.263 T. This quenching phenomenon is nicely interpreted in terms of interfering semiclassical paths, or instantons [3].

In reality, only four quenches are seen in Fe$_8$, and the spacing between the quenches is $\sim 0.41$ T, 50% larger than predicted by Eq. (1.2). Both these facts are accounted for by a slightly modified Hamiltonian

\[ H_1 = H_0 - C[(J_z + iJ_y)^4 + \text{h.c.}], \]  

(1.4)

with $C = 0.29 \mu K$, as is easily verified by explicit numerical diagonalization. The fact that only 4 quenches are seen is paradoxical, however. The relative phase between the interfering instantons is a topological quantity, so that it must equal $J\pi$ at $H = 0$, and tend to 0 as $H \to H_c$. There is no apparent way to get other than 10 quenches. The resolution is that the $C$ term, though small, has a dramatic effect: any non zero $C$ leads to the existence of instantons that are \textit{discontinuous} at the end points [1]. One of these instantons ends
up being the dominant one by virtue of having the lowest action beyond a certain field ($\sim 0.25H_c$ for Fe$_8$), and since it has no interfering partner, there are no more quenches of the splitting beyond this field. In this way, the discontinuous or jump instantons may be said to shunt the interference effect. The jump instantons have discontinuities, and thus do not have to respect the topology.

In private correspondence to us, E. Chudnovsky has alleged that jump instantons were first introduced by him and Hidalgo in an earlier study of the simpler Hamiltonian (1.1) at high fields [5]. We disagree. The instanton studied by Chudnovsky and Hidalgo (CH) is of exactly the standard continuous type arising in the study of a particle tunneling in a symmetric one-dimensional double well, but its nature is obscured by the subtleties of spin trajectories, and the singularities of the spherical polar coordinate system on the sphere. These subtleties are vexing and the source of innumerable pitfalls, so it seems useful to us to elaborate on the instantons in question, and show explicitly that they are continuous. We also use the opportunity to correct a misconception in a still earlier study of the same problem, i.e., Eq. (1.1) at high fields, by one of us [6].

II. DETAILS

We consider only fields parallel to $\hat{z}$, and define the reduced field

$$h = H/H_c,$$

and the special value

$$h^* = (1 - \lambda)^{1/2}.$$

CH study the problem for $h > h^*$, and Chudnovsky is asserting that the instantons in that study have jumps. We give three arguments to show that this is not so.

The first and mathematically simplest way is to consider fields just less than $H_c$. We write

$$h = (1 - \eta)$$

where $0 < \eta \ll 1$. The expectation value of $J_z$ in the low energy states is then extremely close to $J$, so we can approximate

$$J_x \approx Jq, \quad J_y \approx p,$$
where $p$ and $q$ are momentum and position operators with the commutator $[q, p] = i$. It is self-consistently verifiable that $J_y \sim \eta$, $J_x \sim \eta^{1/2}$, so that we can write $J_z \simeq (J^2 - J_x^2 - J_y^2)^{1/2}$. Expanding the term $-g\mu_b H J_z$ to order $J_y^2$ and $J_x^4$, rewriting in terms of $q$ and $p$, and omitting an additive constant, we arrive at the equivalent Hamiltonian

$$\mathcal{H}_{\text{equiv}} = \frac{1}{2m}p^2 + \frac{1}{4}k_1 h J^2 q^4 - \eta k_1 J^2 q^2.$$  \hspace{1cm} (2.5)

Here,

$$m = [2(k_2 - \eta k_1)]^{-1} \approx (2k_2)^{-1}. \hspace{1cm} (2.6)$$

Equation (2.3) is the Hamiltonian for the particle in a quartic double well. The minima of the well are at $\pm q_0 = \pm (2\eta)^{1/2}$. The instantons for this problem are known, and the one that runs from $q_0$ to $-q_0$ is given by

$$q(t) = -q_0 \tanh \gamma t, \hspace{1cm} (2.7)$$

$$p(t) = -im\gamma q_0 \tanh \gamma t \operatorname{sech}^2 \gamma t. \hspace{1cm} (2.8)$$

Here, $t$ is an imaginary time, and $\gamma = (2k_1 k_2 \eta)^{1/2}J$. If we transcribe these results back into $J_x$, $J_y$, and $J_z$, it is manifest that the spin trajectory goes continuously from one minimum of the classical energy to the other without any jumps. There is nothing in the physical character of the problem to indicate a qualitative change as $H$ is reduced, so as long as one is in the field range where the gap does not oscillate ($h > h^*$), the instantons should be continuous.

In our second argument, we study the instantons for all $H$. Since these lie on the complex unit sphere, it is highly advantageous to use stereographic coordinates, $z = \tan \frac{1}{2}\theta e^{i\phi}$, $\bar{z} = \tan \frac{1}{2}\theta e^{-i\phi}$, \hspace{1cm} (2.9)

where $\theta$ and $\phi$ are the customary spherical polar coordinates. If a direction $\hat{n}$ has coordinates $(\theta, \phi)$, and if $|\hat{n}\rangle$ is the spin coherent state with maximal spin projection along $\hat{n}$, then up to normalization and phase, $|\hat{n}\rangle$ is identical to $|z\rangle \equiv e^{z J^-} |J, J\rangle$ where $J^2 |J, J\rangle = J(J + 1) |J, J\rangle$ and $J_z |J, J\rangle = J |J, J\rangle$. A point on the complex unit sphere is parametrized by complex $\theta$ and $\phi$, which means that $z$ and $\bar{z}$ need not be complex conjugates; rather, they are independent complex variables. Either coordinate system shows that the complex unit sphere is a four-dimensional manifold, but the $(z, \bar{z})$ system handles coordinate singularities better. In terms
of $z$ and $\bar{z}$, the cartesian components of $J$ are given by

$$(J_x, J_y, J_z) = \frac{J}{1 + \bar{z}z}(z + \bar{z}, -i(z - \bar{z}), 1 - \bar{z}z).$$  (2.10)

The south pole is the point $z = \bar{z} = \infty$; if $z \to \infty$ with $\bar{z}$ finite, $J \to J(\bar{z}^{-1}, -i\bar{z}^{-1}, -1)$; if $\bar{z} \to \infty$ with $z$ finite, $J \to J(z^{-1}, iz^{-1}, -1)$.

We have found the instantons for this problem in Ref. 7 among other places, but we restate the results along with some attendant formulas for ready reference, and to enable readers to verify the key results for themselves. Let us define

$$E(\bar{z}, z) = \langle z | \mathcal{H} | z \rangle / \langle z | z \rangle.$$  (2.11)

Then the Euler-Lagrange equations of motion for the instantons are given by

$$\dot{\bar{z}} = \frac{(1 + \bar{z}z)^2}{2J} \frac{\partial E}{\partial \bar{z}}, \quad \dot{z} = -\frac{(1 + \bar{z}z)^2}{2J} \frac{\partial E}{\partial z},$$  (2.12)

With the Hamiltonian (1.1),

$$E(\bar{z}, z) = k_1 J^2 \left[ \frac{(1 - \bar{z}z)^2 - \lambda(z - \bar{z})^2 - 2h(1 - \bar{z}^2 z^2)}{(1 + \bar{z}z)^2} \right],$$  (2.13)

so that

$$\dot{z} = -\frac{k_1 J}{(1 + \bar{z}z)} \left[ -2z(1 - \bar{z}z) + \lambda(z - \bar{z})(1 + z^2) + 2hz(1 + \bar{z}z) \right],$$  (2.14)

$$\dot{\bar{z}} = \frac{k_1 J}{(1 + \bar{z}z)} \left[ -2\bar{z}(1 - \bar{z}z) + \lambda(\bar{z} - z)(1 + \bar{z}^2) + 2h\bar{z}(1 + \bar{z}z) \right].$$  (2.15)

The minima of $E$ are at $\bar{z} = z = \pm z_0$ where $z_0 = [(1 - h)/(1 + h)]^{1/2}$. There are two solutions to the equations of motion that run from $z_0$ to $-z_0$. The first is given by

$$z^{(1)}(t) = -z_0 \tanh \tau,$$  (2.16)

$$\bar{z}^{(1)}(t) = -z_0 \frac{\sqrt{\lambda} \tanh \tau + \sqrt{1 - h^2}}{\sqrt{\lambda} + \sqrt{1 - h^2} \tanh \tau},$$  (2.17)

while the second is given by

$$z^{(2)}(t) = -z_0 \frac{\sqrt{\lambda} \tanh \tau - \sqrt{1 - h^2}}{\sqrt{\lambda} - \sqrt{1 - h^2} \tanh \tau},$$  (2.18)

$$\bar{z}^{(2)}(t) = -z_0 \tanh \tau.$$  (2.19)
We have defined $\tau = \omega t/2$, with $\omega = 2k_1J[\lambda(1 - h^2)]^{1/2}$. Readers can verify by direct substitution into the equations of motion that these are indeed solutions. Also, we have

$$z^{(2)}(t) = -\bar{z}^{(1)}(-t), \quad \bar{z}^{(2)}(t) = -z^{(1)}(-t). \quad (2.20)$$

For $h < h^*$, the two solutions can be seen to be distinct by noting that in the first $\bar{z}$ diverges at some $t$ with $z$ remaining finite, while in the other the converse happens. The action for these two instantons differs by a field dependent phase, which gives rise to the gap oscillation.

For $h > h^*$, on the other hand, the two solutions are physically identical. To see this, let us note that we can write

$$z^{(1)} = -z_0 \tanh \tau, \quad \bar{z}^{(1)} = -z_0 \tanh(\tau + \tau_0), \quad (2.21)$$

$$z^{(2)} = -z_0 \tanh(\tau - \tau_0), \quad \bar{z}^{(2)} = -z_0 \tanh \tau, \quad (2.22)$$

where,

$$\tau_0 = \tanh^{-1} \sqrt{\frac{1 - h^2}{\lambda}}. \quad (2.23)$$

If $h > h^*$, $\tau_0$ is real, so the two solutions are related by a simple translation in time. Such translations are automatically included in the sum over multiinstanton paths, so there is only one type of instanton.

More importantly, the instanton is entirely continuous. We plot the components of $\mathbf{J}$ versus $\tau$ in Fig. 1. As can be seen, there is no discontinuity in any component. This figure should be compared with Fig. 2 of Ref. 4, which pertains to the problem with the $C$ term in Eq. (1.4), and shows instantons with patent jumps.

Thirdly, let us consider the problem in spherical polar coordinates, as is also done by CH, and in Ref. 6. CH proceed by integrating out $\cos \theta$, which is feasible as the integral is Gaussian. The resulting action for $\phi$ is like that of a massive particle. If one now seeks instantons for this problem analogously to the particle in a quartic double well say, and insists on a solution $\phi(\tau)$ in which $\phi$ is real, one discovers that the variable $\tau$ cannot be real, but rather must follow a Z-shaped contour in the complex plane, consisting of two segments parallel to the real axis, joined by a segment along the imaginary axis [8]. This is very different, however, from a discontinuity in $\phi$ itself. Indeed, CH seem aware of this point, for they say “It is clear from the shape of the potential that all three parts of the trajectory
FIG. 1: Components of the complex vector $\mathbf{J}(\tau)$ for the instanton solution, versus $\tau$. The $x$ and $z$ components are real, the $y$ component is pure imaginary.

*join smoothly, because $\phi(\tau)$ and $\dot{\phi}(\tau)$ coincide at the joints.* We agree with this. And, it is clear that if one finds the dominant path for $\cos \theta(\tau)$, i.e., the value of $\cos \theta$ for which the original Gaussian factor is maximal, one will discover that $\cos \theta(\tau)$ is also continuous. To eliminate any doubt, we show this by examining the explicit solution for the instanton from Ref. 6. In our present notation, this reads

$$
cos \phi = -i \frac{(h^2 + \lambda - 1)^{1/2} \tanh 2\tau}{(1 - h^2 - \lambda \tanh^2 2\tau)^{1/2}}, \tag{2.24}
$$

$$
cos \theta = \frac{h(1 - h^2 - \lambda \tanh^2 2\tau) - (1 - h^2)\lambda^{1/2}(h^2 + \lambda - 1)^{1/2}\sech 2\tau}{1 - h^2 - \lambda + h^2\lambda \sech^2 2\tau}. \tag{2.25}
$$

From these one can construct the combinations $\tan \frac{1}{2} \theta e^{\pm i\phi}$ to obtain $z$ and $\bar{z}$. After a certain amount of algebra, we obtain

$$
\begin{pmatrix}
  z(t) \\
  \bar{z}(t)
\end{pmatrix} = -z_0 \frac{\tanh \tau \pm \alpha}{1 \pm \alpha \tanh \tau}, \tag{2.26}
$$

where

$$
\alpha = \frac{(h^2 + \lambda - 1)^{1/2} - \lambda^{1/2}}{(1 - h^2)^{1/2}}. \tag{2.27}
$$

If we now note that

$$
\tanh^{-1} \alpha = -\tau_0 / 2, \tag{2.28}
$$
we see that Eq. (2.26) is the same as Eqs. (2.16) and (2.17) up to a shift in $\tau$. Again, we see that there is no discontinuity in the instanton.

It is a separate issue as to what contour what chooses for $\tau$. If, instead of the real line, one lets $\tau$ run on the Z-shaped contour in Ref. 6, $z$ will run from $z_0$ to $-z_0$ along some contour other than the real line, while $\bar{z}$ will run along some other contour, such that

$$\bar{z}(t) = \frac{\sqrt{\lambda} z(t) - (1 - h)}{\sqrt{\lambda} - (1 + h) z(t)}$$

(2.29)

[This can be verified directly from Eq. (2.26), and it is a consequence of energy conservation along the instanton.] Since the kinetic term in the action can be written as a contour integral over $z$,

$$S = J \int \frac{1}{1 + \bar{z}z} \left[\bar{z}(z) - z \frac{d\bar{z}}{dz}\right] dz$$

(2.30)

it follows that any deformation of the $z$-contour that does not cross any singularities of $\bar{z}(z)$ is allowed. This is indeed the case for us, as for $h > h^*$, the pole at $z = \sqrt{\lambda}/(1 + h)$ lies outside the segment $[-z_0, z_0]$ on the real line.

We conclude by clarifying the misconception in Ref. 6. The discussion above shows that there is only one instanton when $h > h^*$, whereas, in the earlier work it is implicit that there are two, with equal, real Euclidean actions [9]. However, the central thesis of that work, and the calculation of the action itself, are completely unaffected.

One further point is that at exactly at $h = h^*$, standard instanton calculations of the Hamiltonian (1.1) cease to be valid, because two distinct instantons merge into one. This is rather like the phenomenon of merging critical points in the steepest descent approximation for ordinary integrals [10]. We expect existing formulas for the tunnel splitting to be inaccurate in a narrow window of fields near $h = h^*$. The gap can undoubtedly be bridged via a uniform asymptotic approximation, which could be developed using the discrete phase integral method as in Ref. 11. In the methodology of that paper, what happens as we cross the point $h = h^*$ from below is that the width of the unusual forbidden region, in which the wavefunctions are exponentially damped with oscillations, shrinks to zero. In this case, one should join the wavefunctions on the two sides of this region using one quadratic turning point connection formula as opposed to two linear turning point formulas. We leave this as an exercise for the reader.

In summary, we have shown that that the high-field instantons in the Fe$_8$ problem are continuous, and thus qualititatively distinct from the discontinuous instantons induced by
the fourth order anisotropy in Eq. (1.4).

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