Quenched spin tunneling and diabolical points in magnetic molecules. I. Symmetric configurations

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The perfect quenching of spin tunneling that has previously been discussed in terms of interfering instantons, and has recently been observed in the magnetic molecule Fe_8, is treated using a discrete phase integral (or Wentzel-Kramers-Brillouin) method. The simplest model Hamiltonian for the phenomenon leads to a Schrödinger equation that is a five-term recursion relation. This recursion relation is reflection symmetric when the magnetic field applied to the molecule is along the hard magnetic axis. A completely general Herring formula for the tunnel splittings for all reflection-symmetric five-term recursion relations is obtained. Using connection formulas for a nonclassical turning point that may be described as lying ''under the barrier,'' and which underlies the oscillations in the splitting as a function of magnetic field, this Herring formula is transformed into two other formulas that express the splittings in terms of a small number of action and actionlike integrals. These latter formulas appear to be generally valid, even for problems where the recursion contains more than five terms. The results for the model Hamiltonian are compared with experiment, numerics, previous instanton based approaches, and the limiting case of no magnetic field.

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I. INTRODUCTION

A. Motivation

The purpose of this paper is to discuss the tunneling of a spin degree of freedom described by the Hamiltonian

$$\mathcal{H} = -k_2 J_z^2 + (k_1 - k_2) J_x^2 - g\mu_B \mathbf{J}\cdot\mathbf{H}, \qquad (1.1)$$

where $\mathbf{J}$ is dimensionless spin operator with components J_x, J_y, and J_z, $\mathbf{H}$ is an external magnetic field, and $k_1 > k_2 > 0$. Although our immediate goal is to perform a careful mathematical analysis of the tunneling spectrum, it is part of our larger and longer term goal of understanding the low-temperature magnetization dynamics of molecular magnets and small magnetic particles. Thus our motivation for studying the Hamiltonian (1.1) specifically is that it provides a good approximate description of the low-temperature behavior of the spin of the molecule $[(tacn)_6Fe_8O_2(OH)_{12}]^{8+}$ (or just Fe_8 for short).[1–7] This molecule has a ground manifold of Zeeman states with $J=10$, arising from competing intramolecular antiferromagnetic exchange interactions between the eight Fe^{3+} ions in each molecule. The molecules are very well separated in the solid, and there is no evidence for an exchange coupling between different molecules. The dipolar intermolecular couplings are weak and may be ignored in a first approximation. The parameters relevant to Fe_8 are $k_1 \approx 0.33$ K and $k_2 \approx 0.22$ K.[8] The g factor is close to 2.

It has been found by Wernsdorfer and Sessoli[7] that the tunnel splittings between low-lying pairs of energy levels of Fe_8 oscillate as a function of applied static magnetic field. These oscillations are now understood[9] as manifestations of conical intersections[10] or diabolical points[11] in the space of magnetic fields. Such points are rare in physical systems. In addition, the tunneling of a spin is different from that of a massive particle in some ways, and as we shall discuss at the end, naive arguments based on experience with the latter could easily cause one to miss the oscillations. Third, tunneling is clearly an important component of the low-temperature dynamics of the magnetization of other molecular magnets.[1] For example, there are still many open questions about the paramagnetic relaxation of a related molecule, Mn_{12},[12–16] and it is clear that better understanding of Fe_8will help in that case too. For all these reasons, careful theoretical study of the tunneling properties of model Hamiltonians such as Eq. (1.1) seems worthwhile.

The spectrum of the Hamiltonian (1.1) is very rich. To help appreciate this richness, and understand the phenomena that we shall discuss, we show in Fig. 1 the results of a numerical calculation of the energies as a function of H_x, for

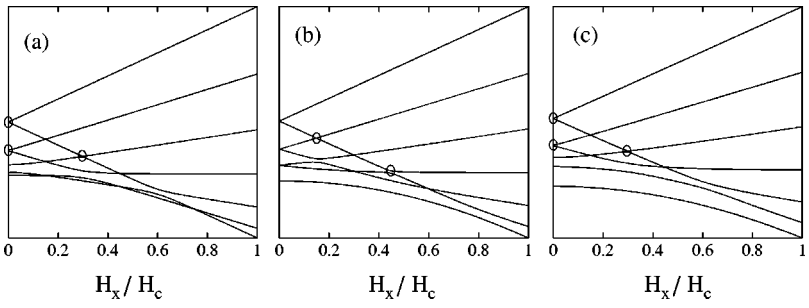

FIG. 1. Spectrum of the Hamiltonian (1.1) for $J=3$, as a function of H_x/H_c. $H_z/H_c = 0$, 0.07454, and 0.1491 in (a), (b), and (c), respectively. The small ovals indicate points that are narrowly avoided anticrossings, but appear to be crossings on low resolution.

TABLE I. Summary of important parameter combinations

Quantity	Formula
H_c	$2k_1J/g\mu_B$
λ	k_2/k_1
$\overline{J}$	$J+\frac{1}{2}$
h_x	$JH_x/\overline{J}H_c$
h_{x0}	H_x/H_c
ω_0	$2J[k_1k_2(1-h_{x0}^2)]^{1/2}$

$J=3$, for three different values of H_z. In all three cases, $H_y=0$. In Fig. 1(a), $H_z=0$, and viewing Eq. (1.1) as a classical energy function for a classical angular momentum vector **J** of fixed length J, we have two degenerate minima for $H_x<H_c=2k_1J/g\mu_B$. (This quantity, along with other important parameter combinations, is listed in Table I.) Correspondingly the lowest two quantum-mechanical energy levels will be split by tunneling. This is evident in the figure. The key features are that (i) the lowest two energy-level curves cross six times (including negative values of H_x), (ii) the crossing points are perfectly periodically spaced, and (iii) a number of higher energy level pairs also cross at some of the field values where the lowest two levels do.

In Fig. 1(b), H_z has a specific nonzero value. The problem is no longer symmetric, and one of the classical minima is lower than the other. Correspondingly, we see that the lowest quantum-mechanical state is always non degenerate. Now, however, if H_z is correctly chosen, the first excited state in the deeper well can be brought into resonance with the lowest state in the shallower well, and these two states can admix by tunneling. And indeed, we see from the figure that the second and third energy levels are quite close. The key features are that (i) they also cross a number of times, (ii) these crossings are shifted by half a period from those in Fig. 1(a), and (iii) crossings between yet higher levels (e.g., the fourth and fifth) occur at some of the same fields where the lower levels cross.

This pattern continues as H_z is increased still further [Fig. 1(c)]. Now the lowest two levels in the deeper well are nondegenerate, and the lowest crossings are between levels 3 and 4. Compared to Fig. 1(b), these crossings are shifted by yet another half period. Again, there is a simultaneous crossing between higher pairs of levels (numbers 5 and 6) at $H_x/H_c\simeq 0.15$.

At the points where the energy levels in Fig. 1 cross, the tunnel splitting goes to zero, and a plot of the splittings as a function of H_x/H_c would show oscillations. It is just these oscillations that Wernsdorfer and Sessoli have seen. The curves marked $n=0$, 1, and 2 in Fig. 2(b) of Ref. 7 correspond precisely to the lowest crossing in Figs. 1(a)–(c), respectively. The higher crossings have also been inferred indirectly by Wernsdorfer *et al.*[17]

Some readers may wonder why an analytic study of the problem is necessary, and why a numerical calculation of the energy levels of the 21×21 Hamiltonian for Fe_8, which could be done just as for the $J=3$ case above, is not the last word on the subject, especially since one can easily include higher-order anisotropy terms[8] in the numerics. The answer is twofold. First, numerics cannot be done easily for larger J, since the number of significant digits required to discern a splitting grows roughly exponentially with J. Thus the calculation has general value. Second, analytic work provides insight beyond numerics in showing how the splitting depends systematically on parameters in $\mathcal{H}$, how it grows as we consider higher pairs of levels and so on. Even if one if succeeeded in fitting the numerical data to explicit functional forms by trial and error, such fits would have only an empirical status. Most importantly, the analytic approach provides a language in which to discuss and understand the ''physics'' of the problem, and gain insight which may be applied to other problems. Thus we shall see that the vanishing of the splitting is linked to an oscillatory exponential behavior of the wave function arising from a nonclassical turning point in a semiclasssical, WKB-like approach. It is extremely unlikely that one could develop such mental pictures based on numerical work alone. These constructs are extremely useful in understanding more complicated problems.[18]

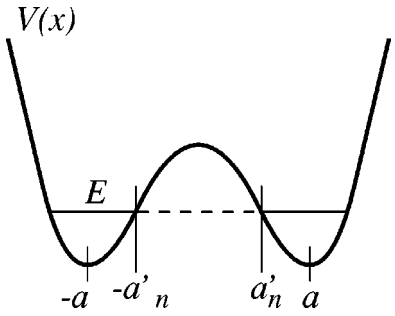

FIG. 2. A symmetric double-well potential $V(x)$.

B. Methodology and relation to previous work

The Hamiltonian (1.1) has been the subject of several previous papers by this author.[19] The quenching of ground-state tunnel splittings for $\mathbf{H}\|\hat{\mathbf{x}}$, i.e., those between the lowest pair of levels in Fig. 1(a), were found in the first paper of Ref. 19, on the basis of a path integral or instanton argument involving interference of tunneling Feynman trajectories.[20,21] This work was done before the experiments of Wernsdorfer and Sessoli, and without knowing of the relevance of the Hamiltonian to Fe_8. These quenchings, or those that occur when $\mathbf{H}\|\hat{\mathbf{z}}$, can be understood in light of the von Neumann-Wigner theorem. When $\mathbf{H}\|\hat{\mathbf{x}}$, or $\mathbf{H}\|\hat{\mathbf{z}}$, $\mathcal{H}$ is invariant under a 180° rotation about $\hat{\mathbf{x}}$ or $\hat{\mathbf{z}}$, so energies of levels that are odd and even under this operation can intersect.[19] When **H** has both $\hat{\mathbf{x}}$ and $\hat{\mathbf{z}}$ components, however, $\mathcal{H}$ has no symmetry, and the level crossings [Figs. 1(b) and (c)], corresponding to the $-10\leftrightarrow 9$ and $-10\leftrightarrow 8$ oscillations seen in Ref. 7, were not foreseen. The present study is motivated largely by the need to understand these new oscillations, and also the tunneling between the higher pairs of levels for $\mathbf{H}\|\hat{\mathbf{x}}$.[22]

The approach used here is a discrete phase integral (DPI) or Wentzel-Kramers-Brillouin (WKB) method. This ap-

proach has been used in many other areas of physics and mathematical physics,[23–25] but the first explicit application to spin tunneling of which we are aware is in Ref. 26. The basic idea is that in the J_z basis, Schrödinger's equation for Eq. (1.1) has the form of a recursion relation or difference equation, as opposed to a differential equation for a massive particle in a one dimensional potential $V(x)$. This difference equation can be solved in analogy with the continuum WKB approximation. We will see that compared to previous DPI studies, new types of turning points arise in the study of Eq. (1.1), because the recursion relation has five terms.[27,28] These turning points are central to the oscillations in the splitting, they cannot arise in a three-term problem, and they have no continuum analog. Our present discussion will rely on physical arguments based on viewing the recursion relation as a tight-binding model for an electron in a one-dimensional lattice, and correspondence with the continuum case. A more formal treatment is given in Ref. 27.

Like the instanton method, the DPI method is asymptotically correct in the semiclassical limit, i.e., as $J\to\infty$. While this method does not have the visual and geometrical appeal of interfering instantons, it has the advantage of involving only elementary methods of analysis, and also of yielding wave functions which may be of use in finding matrix elements of perturbations. For the Fe_8 problem, it is especially well suited to studying tunneling when both H_x and H_z are nonzero, and between more than one pair of levels at the same time. In hindsight, these problems can also be addressed using path integrals, but the labor required is far greater, and it is particularly difficult to find the correct $J\to\infty$ asymptotic form of the preexponential factor multiplying the exponentially small Gamow factor in the splitting. Since the DPI method gives this factor naturally, there is no reason not to find it.

C. What is new

In this paper we consider the tunneling problem for Eq. (1.1) for the case $\mathbf{H}\|\hat{\mathbf{x}}$. The problem is then analogous to a massive particle moving in a symmetric double-well potential. The case where H_z is also nonzero corresponds to an asymmetric potential, is more complicated, and will be considered in a second paper.

In the course of our study, we shall develop the DPI formalism for tunneling problems as generally as possible. The payoff is two formulas for the tunnel splitting [Eqs. (4.27) and (4.28)] that can be applied to any problem describable by a five-term recursion relation, and quite possibly, to even higher term recursions.

Brief reports of our work have been given earlier,[19,29] including, in particular, formulas for the locations of the diabolical points.[30] Specifically, we find that the l'th level in the negative J_z well (where $l'=0$ denotes the lowest level) and the l''th level in the positive J_z well are degenerate when

$$\frac{H_z(l',l'')}{H_c}=\frac{\sqrt{\lambda}(l''-l')}{2J}, \tag{1.2}$$

$$\frac{H_x(l',l'')}{H_c}=\frac{\sqrt{1-\lambda}}{J}[J-n-\tfrac{1}{2}(l'+l''+1)], \tag{1.3}$$

with $n=0,1,\ldots,2J-(l'+l''+1)$. Here, $\lambda=k_2/k_1$, and $H_c=2k_1J/g\mu_B$. The result of the first paper of Ref. 19 corresponds to the special case $l'=l''=0$. An approximate version of the DPI method, leading to the same diabolical point locations, has been developed and applied to Fe_8 independently by Villain and Fort.[9] It should be noted that both Villain and Fort's and our calculations entail various approximations, and Eqs. (1.2) and (1.3) are only derived to leading order in $1/J$. However, as has recently been shown by E. Keçecioğlu and the author,[31] these equations are in fact *exact* as written. This suggests that the Hamiltonian (1.1) has extra symmetries which are not yet understood.

In order to describe the part of the paper that pertains to the general DPI formalism, it is useful to recapitulate some results for tunneling of a massive particle in a one-dimensional double well.[32] Given a Hamiltonian

$$\mathcal{H}_{\rm part}=-\frac{\hbar^2}{2m}\nabla^2+V(x), \tag{1.4}$$

where $V(x)=V(-x)$, with minima at $x=\pm a$ (see Fig. 2), the symmetric and antisymmetric combinations of the two states localized in the separate wells with a mean energy E, will be split by an amount given by

$$\Delta E_{\rm part}=\frac{2\hbar^2}{m}\psi_R(0)\psi_R'(0), \tag{1.5}$$

where $\psi_R(x)$ is the wavefunction of the state localized in the right well, normalized to unit total probability.

The formula (1.5) is generally named for Herring[33,34] (but sometimes for Bardeen or Landau). Note that E does not have to be the energy of the lowest level in the separate wells. For the nth level, Eq. (1.5) reduces in the WKB approximation to

$$\Delta E_{{\rm part},n}=g_n\frac{\hbar\omega}{\pi}\exp\left[-\int_{-a_n'}^{a_n'}\frac{|p|}{\hbar}dx\right], \tag{1.6}$$

where $\pm a_n'$ are the inner turning points where $V(x)=E$, $|p|$ is the imaginary momentum in the tunneling region $-a_n'<x<a_n'$, ω is the small oscillation frequency around $x=\pm a$, and

$$g_n=\frac{\sqrt{2\pi}}{n!}(n+\tfrac{1}{2})^{n+1/2}e^{-(n+1/2)} \tag{1.7}$$

is a small correction $[g_0=(\pi/e)^{1/2}\approx 1.075, g_1\approx 1.028, g_2\approx 1.017,\ldots]$ accounting for the curvature of $V(x)$ near the turning point.[35]

For low-lying states, i.e., for n of order 1, the action integral in Eq. (1.6) contains a singular part varying as $\ln[E-V(a)]$, making it difficult to use directly. When this singularity is peeled off, one obtains[36,37]

$$\Delta E_{{\rm part},n}=\frac{1}{n!}F_{\rm part}^{n+1/2}\left(\frac{2}{\pi}\right)^{1/2}\hbar\omega e^{-S_0}, \tag{1.8}$$

with

$$S_0 = \int_{-a}^{a} [2mV(x)]^{1/2} dx, \tag{1.9}$$

and

$$F_{\text{part}} = \frac{2m\omega a^2}{\hbar} \exp\left\{ \int_0^a \left[\frac{m\omega}{\sqrt{2mV(x)}} - \frac{1}{a-x} \right] dx \right\}. \tag{1.10}$$

We will derive discrete analogs of Herring's formula (1.4), and the quasiclassical formulas in terms of action integrals (1.6) and (1.8). These formulas are general, and not limited to the Fe_8Hamiltonian. Of course, we do apply them to the Fe_8 problem too.

D. Plan of paper

In Sec. II, we briefly review the DPI method in order to introduce the basic language of classically allowed and forbidden regions, turning points, and so on, in the discrete case. We will focus particularly on the features associated with the new turning point, and show that there are three *critical energy curves* that collectively play the same role as $V(x)$ in determining the turning points. A longer account of this work, along with connection formulas at the new turning points has been given in Ref. 27.

The discrete analog of Herring's formula for five term recursion relations is given in Sec. III. The algebraic details are given in Appendix A. The result is Eq. (3.2).

In Sec. IV we will use the quasiclassical DPI form of the wave function in Herring's formula to obtain analogs of Eqs. (1.6) and (1.8). The final results are Eqs. (4.27) and (4.28), and the details of the analysis are given in appendixes B and C. By and large, this analysis is an extension of continuum WKB techniques, such as using connection formulas to relate quasiclassical wave functions in different regions, and we are certain that many readers will immediately find it transparent. At the same time, it is quite lengthy, many formulas can not be obtained by simply transcribing continuum case formulas (especially those relating to the new turning point and the oscillatory factor in the splitting), and there is no reason to have readers duplicate all this for themselves. Equation (4.27) is close enough to Eq. (1.6) that one might have guessed it, but the route from Eq. (1.8) to Eq. (4.28) is not at all obvious *a priori*.

The formula (4.28) is applied to Fe_8 in Sec. V, with the actual evaluation of the action integrals deferred to Appendix D. We also discuss the quenching points and other aspects of these results, including comparison with numerics, and work by other authors.[38,39]

We conclude the paper in Sec. VI with some general remarks pertaining to diabolical points in other systems, and some special features of the degeneracies of the Hamiltonian (1.1) that are known exactly.

Readers who are only interested in the spin tunneling problem for its relevance to Fe_8 should skip directly to Sec. V. Readers who wish to get a sense of the DPI methodology and its relation to continuum WKB should also read Secs. II–IV, and skip the appendixes unless they wish to see all of the analysis.

II. DPI METHOD FOR FIVE-TERM RECURSION RELATIONS

The starting point is to write Schrödinger's equation in the J_z basis. Suppose $|\psi\rangle$ is an eigenstate of $\mathcal{H}$ with energy E. Then with $J_z|m\rangle = m|m\rangle$, $\langle m|\psi\rangle = C_m$, $\langle m|\mathcal{H}|m\rangle = w_m$, and $\langle m|\mathcal{H}|m'\rangle = t_{m,m'}$ $(m \neq m')$, we have

$$\sum_{n=m-2}^{m+2}{}' \; t_{m,n} C_n + w_m C_m = E C_m, \tag{2.1}$$

where the prime on the sum indicates that the term $n=m$ is to be omitted. The diagonal terms (w_m) arise from the J_z^2 part of $\mathcal{H}$, the $t_{m,m\pm1}$ terms from the $J_x H_x$ part, and the $t_{m,m\pm2}$ terms from the J_x^2 part.

We can think of Eq. (2.1) as a tight binding model for an electron in a one-dimensional lattice with sites labeled by m, and slowly varying on-site energies (w_m), nearest-neighbor $(t_{m,m\pm1})$, and next-nearest-neighbor $(t_{m,m\pm2})$ hopping terms. Since we can think of dynamics in this model in terms of wave packets, it is clear that there is a generalization of the usual continuum quasiclassical or phase integral method to the lattice case. This is the DPI method.

The general formalism of this method[23–26] and the extension to five terms is discussed at length elsewhere,[27,28] so here we will only give a brief summary. The fundamental requirement for a quasiclassical approach to work is that w_m and $t_{m,m\pm\alpha}(\alpha=1,2)$ vary slowly enough with m that we can find smooth continuum approximants $w(m)$ and $t_\alpha(m)$, such that whenever m is an eigenvalue of J_z, we have

$$w(m) = w_m, \tag{2.2}$$

$$t_\alpha(m) = (t_{m,m+\alpha} + t_{m,m-\alpha})/2, \quad \alpha = 1,2. \tag{2.3}$$

We further demand that

$$\frac{dw}{dm} = O\left(\frac{w(m)}{J}\right), \quad \frac{dt_\alpha}{dm} = O\left(\frac{t_\alpha(m)}{J}\right), \tag{2.4}$$

with m/J being treated as quantity of order 1. We will see that for Eq. (1.1) these conditions will hold in the semiclassical limit $J \gg 1$.

Given these conditions, the basic approximation, which readers will recognize from the continuum case, is to write the wave function as a linear combination of the quasiclassical forms

$$C_m \sim \frac{1}{\sqrt{v(m)}} \exp\left(i \int^m q(m')dm' \right), \tag{2.5}$$

where $q(m)$ and $v(m)$ obey the equations

$$E = w(m) + 2t_1(m)\cos q + 2t_2(m)\cos(2q) \equiv \mathcal{H}_{\text{sc}}(q,m), \tag{2.6}$$

$$v(m)=\partial\mathcal{H}_{\rm sc}/\partial q=-2\sin q(m)[t_1(m)+4t_2(m)\cos q(m)]. \quad (2.7)$$

Equations (2.6) and (2.7) are the lattice analogs of the eikonal (or Hamilton-Jacobi) and transport equations. Equation (2.5) represents the first two terms in an expansion of $\ln C_m$ in powers of $1/J$.

We note in passing that the approximation applies to *any* recursion relation (subject of course to the slow variation conditions), and not just one that originates in a spin problem.[25] This is clear from the tight-binding analogy. In particular, from the viewpoint of understanding the classical limit, there is no need for attaching a specific meaning to the canonically conjugate variables m and q as J_z and the azimuthal angle ϕ in spherical polar coordinates. Indeed, there are some advantages to *avoiding* this description, since the topology of the unit sphere S^2 makes it impossible to construct a classical Hamiltonian formalism in terms of a *globally* nonsingular coordinate and momentum.[40]

As in the continuum case, the approximate DPI wave function is invalid at turning points. These points arise whenever the velocity $v(m)$ vanishes for given energy E, for then the approximation (2.5) diverges. We see from Eq. (2.7) that $v(m)$ can vanish either because $\sin q=0$, i.e., $q=0$ or $q=\pi$, or because $q=q_*\equiv\cos^{-1}(-t_1/4t_2)$. Substituting these values of q in the eikonal equation, we see that a turning point is obtained whenever

$$E=U_0(m),U_\pi(m), \text{ or } U_*(m), \quad (2.8)$$

where

$$U_0(m)=\mathcal{H}_{\rm sc}(0,m)=w(m)+2t_1(m)+2t_2(m), \quad (2.9)$$

$$U_\pi(m)=\mathcal{H}_{\rm sc}(\pi,m)=w(m)-2t_1(m)+2t_2(m), \quad (2.10)$$

$$U_*(m)=\mathcal{H}_{\rm sc}(q_*,m)=w(m)-2t_2(m)-\frac{t_1^2(m)}{4t_2(m)}. \quad (2.11)$$

Note that at a turning point, both m and q are determined. If we denote the values of these quantities generically by m_c and q_c, m_c may be regarded as being fixed by Eq. (2.8), and q_c by the corresponding condition $q_c=0$, $q_c=\pi$, or $q_c=q_*(m_c)$.

To understand the nature of these turning points, let us assume that $t_1<0$, and $t_2>0$. [This is the case for the Hamiltonian (1.1). We can always arrange for t_1 to be negative by means of the gauge transformation $C_m\to(-1)^m C_m$. Thus there is only one other case to be considered, namely, $t_1<0$, $t_2<0$. This is discussed in Ref. 27.] It then follows that $U_\pi>U_0$, and that

$$U_0(m)-U_*(m)=\frac{1}{4t_2(m)}[t_1(m)+4t_2(m)]^2\geq 0. \quad (2.12)$$

Second, let us think of $\mathcal{H}(q,m)$ for fixed m as an energy-band curve. Then U_π is always the upper band edge, while the lower band edge is either U_0 or U_* according as whether $-t_1/4t_2$ is greater than or lesser than 1. To deal with this possibility, it pays to introduce a dual labelling scheme for all three curves U_0, U_π, and U_*. We write $U_\pi(m)\equiv U_+(m)$, and

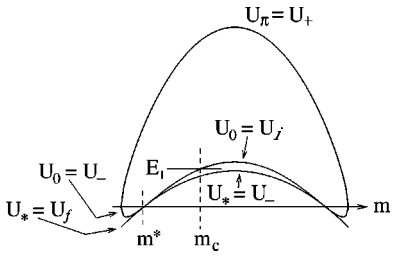

FIG. 3. Critical energy curves for the Hamiltonian (1.1), showing the dual labelling scheme.

$$U_0(m)=U_i(m), \quad U_*(m)=U_-(m), \quad \text{if } q_*\in(0,\pi), \quad (2.13)$$

$$U_0(m)=U_-(m), \quad U_*(m)=U_f(m), \quad \text{if } q_*\notin(0,\pi). \quad (2.14)$$

The subscripts $+$ and $-$ denote upper and lower band edges, while the subscripts i and f denote *internal* and *forbidden*, respectively, since in the first case above, U_0 lies inside the energy band, while in the second case, U_* lies outside. As examples of these curves for a symmetric recursion relation, we show those for Fe_8 in Fig. 3. A magnified view of the lower left-hand portion of this diagram is given in Fig. 4.

Turning points where $E=U_+$, or $E=U_-$ when $U_-=U_0$, are analogous to those encountered in the continuum quasiclassical method, since the energy lies at a limit of the classically allowed range for the value of m in question. Points where $E=U_-$ when $U_-=U_*$ are physically analogous, but mathematically different since the value of q_c is neither 0 nor π. Points where $E=U_i$ (e.g., see the energy E_1 in Fig. 3) are unusual in that the energy is *inside* the classically allowed range for m_c, but the mathematical form of the connection formulas is identical to the case $E=U_-=U_0$ since $q_c=0$. Most interesting are the turning points with $E=U_f$ (the point $m=-m_1$ in Fig. 4, for instance), since now the energy is outside the allowed range for $m=m_c$, and the value of q_c is therefore necessarily complex. These points lie ''under the barrier'' and turn out to be the ones of importance for understanding oscillatory tunnel splittings. As al-

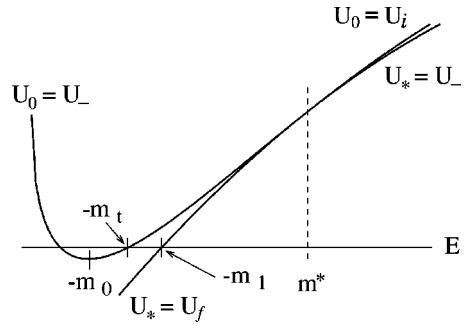

FIG. 4. Magnified view of the lower left-hand region of Fig. 3 showing the point of tangency m^* between U_0 and U_*, and turning points at $m=-m_t$ and $-m_1$ for an energy E.

ready stated, these nonclassical turning points do not arise in problems described by three-term recursion relations, and so the oscillatory effect can not arise in such problems.

The above discussion shows that the curves U_0, U_π, and U_* collectively play the same role as the potential energy in the continuum quasiclassical method. We refer to them as *critical curves*. We have already noted that $U_\pi > U_0 \geqslant U_*$. Let us suppose that the case of equality in Eq. (2.12) occurs at $m=m^*$. Clearly $t_1(m^*)/4t_2(m^*)=-1$, which is precisely the condition found above for the lower band edge to change from $q=0$ to $q=q_*$. Second, expanding t_1 and t_2 about m^*, we see that U_0 and U_* have a common tangent when they meet.

III. HERRING'S FORMULA FOR FIVE-TERM RECURSION RELATIONS

As discussed in Sec. I, Herring's formula greatly simplifies the calculation of tunnel splittings in a symmetric double well.[33,34] An analogous formula can be derived in the discrete case[41,26] following the simplified treatment of Landau and Lifshitz.[32]

We have already noted the importance of the critical curves. For low-lying energy levels, in particular, the curve U_- is very much like the potential energy in the continuum case, and it is clear that we will have an entire series of approximate energy eigenstates with wave functions localized in any one of the two wells, in the vicinity of $\pm m_0$, the minima of $U_-(m)$ (see Fig. 3). Let C_m be the nth such wave function localized in the right-hand well, normalized to unit total probability, and let it satisfy the Schrödinger equation (2.1) with an energy E_0 for all values of m well to the right of the left well, including in particular the region around $m=0$. More precisely, we take C_m to decay away from the right well in *both* directions. Such a function could be obtained, e.g., as the energy eigenfunction of a modified problem in which the on-site energy is increased by a large positive amount for all $m<m_a$, where $-m_0 \ll m_a \ll 0$, it being understood that m_a is far away from all turning points for the energy concerned. However, this problem need not be solved explicitly, as the exact behavior of C_m near $m=-m_0$ is never needed, and therefore need not be examined too closely.

Given such a function, Herring shows that the true symmetric and antisymmetric eigenfunctions, s_j and a_j, with energies E_1 and E_2, respectively, are given very accurately by

$$a_m = \frac{1}{\sqrt{2}}(C_m - C_{-m}),$$

$$s_m = \frac{1}{\sqrt{2}}(C_m + C_{-m}). \tag{3.1}$$

The product $C_m C_{-m}$ is exponentially small everywhere, so these functions are normalized to unit total probability to exponentially high accuracy.

The remaining steps are very similar to manipulations involving the use of Green's identity in the theory of self-adjoint operators. One writes the Schrödinger equations for C_m and a_m, constructs inner products over half the region $-J \leqslant m \leqslant J$, and subtracts corresponding terms. The final result for the splitting $\Delta = E_2 - E_1$ is derived in Appendix A. Up to an irrelevant over all sign, we get

$$\Delta = \begin{cases} 2[t_{0,1}C_0(C_1 - C_{-1}) + t_{0,2}C_0(C_2 - C_{-2}) + t_{-1,1}(C_1^2 - C_{-1}^2)], & \text{integer } J, \\ 2\,t_{-1/2,1/2}(C_{1/2}^2 - C_{-1/2}^2) + 4\,t_{-3/2,1/2}(C_{1/2}C_{3/2} - C_{-1/2}C_{-3/2}), & \text{half integer } J. \end{cases} \tag{3.2}$$

We remind readers that this result is not limited to the ground-state splitting. Further, we can take C_m to be the wave function of either the left- or the right-localized state, as that only changes the sign of Δ.

IV. QUASICLASSICAL FORMULAS FOR TUNNEL SPLITTING

A. Singularity-unextracted quasiclassical formula

We now apply our Herring formula within the DPI approximation. For this, we need the left-localized wave function C_m in the central region near $m=0$.

From the discussion in Sec. II we know that for an energy E as depicted in Fig. 4, the classically allowed region is in the vicinity of $-m_0$, the minimum of $U_-(m)$. Turning points exist at $m=-m_t$ and $-m_1$, the latter being under the barrier. (There is also a turning point to the left of $-m_0$, but that is not of interest at the moment.) The local wave vector $q(m)$ is purely imaginary in the region $-m_t < m < -m_1$, and complex in the region $-m_1 < m < m_1$. To see this, we solve the eikonal equation (2.6) for $\cos q$:

$$\cos q(m) = \frac{-t_1(m) \pm [t_1^2(m) - 4t_2(m)f(m)]^{1/2}}{4t_2(m)}, \tag{4.1}$$

where $f(m) = w(m) - 2t_2(m) - E$. Since $\cos q = -t_1/4t_2$ at $m=-m_1$, the discriminant in Eq. (4.1) must vanish, and we conclude that as we cross $-m_1$, $\cos q$ changes from real to complex, and q changes from imaginary to complex. Incidentally, it may be verified that the condition for vanishing discriminant, i.e.,

$$t_1^2(m) = 4t_2(m)[w(m) - 2t_2(m) - E], \tag{4.2}$$

is identical to $E = U_*(m)$.

We now note that if we choose the phase of C_m so that C_m is real in the forbidden region to the left of $-m_0$, then

because the recursion relation is real, C_m must be real for all m. Because $q(m)$ is complex in the central region, we can meet this demand only by taking a linear combination of two DPI forms with complex conjugate q's. We write these as

$$q_{1,2}(m) = i\kappa(m) \pm \chi(m), \tag{4.3}$$

with κ and χ both real. Since C_m is left-localized by hypothesis, it must decay with increasing m, and so we must choose $\kappa > 0$. Further, we choose q_1 to be the solution with a positive real part, i.e., we also take $\chi > 0$. Then, the DPI form which proves convenient for $m > -m_1$ is given by

$$C_m = \mathrm{Re} \frac{A}{\sqrt{s_1(m)}} \exp\left(i \int_{-m_1}^{m} q_1(m')dm' \right), \tag{4.4}$$

where $s_1(m) = -iv[q_1(m)]$.

The next step is to substitute Eq. (4.4) into Eq. (3.2). Before doing this, we note that

$$\cosh\kappa \cos\chi = -t_1/4t_2, \tag{4.5}$$

$$\sinh\kappa \sin\chi = (4t_2 f - t_1^2)^{1/2}/4t_2, \tag{4.6}$$

so that

$$s_1 = 8t_2(m)\sinh\kappa(m)\sin\chi(m)\sin q_1(m). \tag{4.7}$$

We now substitute Eqs. (4.4)–(4.7) into Eq. (3.2). In doing this, we may neglect the variation of quantities $t_\alpha(m)$, $q(m)$, and $v(m)$ among the sites near the center of the lattice, since the number of sites involved is of order 1, and so the variation leads to higher-order corrections in powers of $1/J$. To save writing, we denote quantities evaluated at $m=0$ by a bar: $q_1(0) \equiv \bar{q_1}$, $\kappa(0) \equiv \bar{\kappa}$, etc. We thus get

$$C_m = \mathrm{Re}\, A_2 \frac{e^{i(\Omega + m\bar{q_1})}}{\sqrt{\sin\bar{q_1}}}, \tag{4.8}$$

where

$$\Omega = \int_{-m_1}^{0} q_1(m')dm', \tag{4.9}$$

$$A_2 = (8\bar{t_2}\sinh\bar{\kappa}\sin\bar{\chi})^{-1/2} A. \tag{4.10}$$

The cases of integer and half integer J are best tackled separately. We do the integer case first. The requisite algebra is lengthy, and we provide the following intermediate formulas for reference:

$$C_1 - C_{-1} = iA_2[e^{i\Omega}\sqrt{\sin\bar{q_1}} - \mathrm{c.c.}], \tag{4.11}$$

$$C_1 + C_{-1} = A_2\left[e^{i\Omega}\sqrt{\frac{\cos^2\bar{q_1}}{\sin\bar{q_1}}} + \mathrm{c.c.} \right], \tag{4.12}$$

$$C_0(C_1 - C_{-1}) = i\frac{A_2^2}{2}\left[\left(e^{2i\Omega} - e^{-2\,\mathrm{Im}\,\Omega}\sqrt{\frac{\sin\bar{q_1}^*}{\sin\bar{q_1}}} \right) - \mathrm{c.c.} \right], \tag{4.13}$$

$$C_2 - C_{-2} = 2iA_2[e^{i\Omega}\cos\bar{q_1}\sqrt{\sin\bar{q_1}} - \mathrm{c.c.}], \tag{4.14}$$

$$C_0(C_2 - C_{-2}) = iA_2^2\left[\left(e^{2i\Omega}\cos\bar{q_1} - e^{-2\,\mathrm{Im}\,\Omega}\cos\bar{q_1}^*\sqrt{\frac{\sin\bar{q_1}^*}{\sin\bar{q_1}}} \right) - \mathrm{c.c.} \right], \tag{4.15}$$

$$C_1^2 - C_{-1}^2 = iA_2^2\left[\cos\bar{q_1}\left(e^{2i\Omega}\cos\bar{q_1} - e^{-2\,\mathrm{Im}\,\Omega}\sqrt{\frac{\sin\bar{q_1}^*}{\sin\bar{q_1}}} \right) - \mathrm{c.c.} \right]. \tag{4.16}$$

Substituting these and the formula $\bar{t_1} = -4\bar{t_2}\cosh\bar{\kappa}\cos\bar{\chi}$ into Eq. (3.2), we get

$$\Delta = -8A_2^2\bar{t_2}\,\mathrm{Im}\left(e^{2i\Omega}\Theta - e^{-2\,\mathrm{Im}\,\Omega}\sqrt{\frac{\sin\bar{q_1}^*}{\sin\bar{q_1}}}\,\mathrm{Re}\,\Theta \right); \tag{4.17}$$

$$\Theta = \cos\bar{q_1} - \cosh\bar{\kappa}\cos\bar{\chi}. \tag{4.18}$$

But, it follows from Eq. (4.3) that

$$\cos\bar{q_1} = \cosh\bar{\kappa}\cos\bar{\chi} - i\sinh\bar{\kappa}\sin\bar{\chi}, \tag{4.19}$$

so $\mathrm{Re}\,\Theta = 0$, the second term in Eq. (4.17) vanishes altogether, and

$$\Delta = 4A_2^2\bar{t_2}\sinh\bar{\kappa}\sin\bar{\chi}(e^{2i\Omega} + e^{-2i\Omega^*}). \tag{4.20}$$

For half integer J, we get

$$C_{\pm 1/2}^2 = \frac{A_2^2}{4}\left[\left(\frac{e^{2i\Omega}}{\sin\bar{q_1}}e^{\pm i\bar{q_1}} + \frac{e^{-2\,\mathrm{Im}\,\Omega}}{|\sin\bar{q_1}|} \right) + \mathrm{c.c.} \right], \tag{4.21}$$

$$C_{\pm 1/2}C_{\pm 3/2} = \frac{A_2^2}{4}\left[\left(\frac{e^{2i\Omega}}{\sin\bar{q_1}}e^{\pm 2i\bar{q_1}} + \frac{e^{-2\,\mathrm{Im}\,\Omega}}{|\sin\bar{q_1}|}e^{\mp i\bar{q_1}} \right) + \mathrm{c.c.} \right]. \tag{4.22}$$

Thus

$$C_{1/2}^2 - C_{-1/2}^2 = \frac{i}{2}A_2^2(e^{2i\Omega} - \mathrm{c.c.}), \tag{4.23}$$

$$C_{1/2}C_{3/2} - C_{-1/2}C_{-3/2} = iA_2^2(\cos\bar{q_1}e^{2i\Omega} - \mathrm{c.c.}), \tag{4.24}$$

and

$$\Delta = iA_2^2[(\bar{t_1} + 4\bar{t_2})e^{2i\Omega} - \mathrm{c.c.}] = 4A_2^2\bar{t_2}\sinh\bar{\kappa}\sin\bar{\chi}(e^{2i\Omega} + e^{-2i\Omega^*}), \tag{4.25}$$

which is identical to Eq. (4.20).

Using Eq. (4.10), we thus obtain, for both integer and half integer J,

$$\Delta = \tfrac{1}{2}A^2(e^{2i\Omega}+e^{-2i\Omega^*}). \tag{4.26}$$

The quantity $i\Omega$ is an action integral from $-m_1$ to 0 [see Eq. (4.9)]. From experience with continuum WKB, we expect the amplitude A to be related to the amplitude inside the well by a barrier penetration factor, $\exp[-\int_{-m_t}^{-m_1}|q(m)|dm]$. When we use this result in Eq. (4.26), we will obtain a total action integral that runs between the classical turning points, $-m_t$ and m_t. The precise prefactor is determined by systematic use of connection formulas, and with the final answer (B12) for A, we get

$$\Delta = \frac{\omega_0 g_n}{\pi}\left[\exp\left(i\int_{-m_t}^{m_t} q(m')dm'\right)+\text{c.c.}\right]. \tag{4.27}$$

Here $q(m')$ is chosen to have a positive real part χ in the first term. We reiterate that this result applies to higher pairs of excited states, and not just the ground pair. The essential dependence on n, the excitation number, enters through the n dependence of m_t, the turning point.

The similarity of Eq. (4.27) to Eq. (1.6) is striking, and one can ask whether one should not have anticipated it right away. For the ground-state pair, the instanton approach[19] makes it very easy to understand the presence of two complex conjugate tunneling actions, and the fact that they should be superposed, but does not give the prefactor. The action integrals in the instanton approach, however, run not from turning point to turning point but from one minimum of the energy to the other. Further, properly justifiying the prefactor using instantons has proven very difficult.[42,43] Purely as a recipe for calculations, however, a hybrid approach, in which one adds the tunneling actions from all equivalent instantons, and uses the DPI approach to determine the form of the prefactor, would appear to be valid for all problems. Thus we strongly suspect that Eq. (4.27) is correct even when the recursion relation has seven or more terms.

B. Singularity-extracted quasiclassical formula

While the formula (4.27) is very general, it has the disadvantage that the action integral runs between turning points. The integrand is therefore close to a singularity, and for low-lying states, this gives rise to terms in the action that depend on $\ln J$. Hence the formula does not reveal the asymptotic behavior as a function of J in a transparent way.

We can obtain a formula without this drawback, analogous to Eq. (1.8), by systematically subtracting away the singular terms in the action integral. This is done in Appendix C, and the final result is

$$\Delta_n = \frac{1}{n!}\sqrt{\frac{8}{\pi}}\omega_0 F^{n+1/2}e^{-\Gamma_0}\cos\Lambda_n, \tag{4.28}$$

where

$$\Gamma_0 = 2\int_{-m_0}^{0}\kappa_0(m)dm, \tag{4.29}$$

$$\Lambda_n = 2\int_{-m_1}^{0}[\chi_0+(n+\tfrac{1}{2})\omega_0\chi_0']dm, \tag{4.30}$$

$$F = 2M\omega_0(m-m_1)^2\exp\left[-2\left(Q_1+\omega_0\int_{-m_1}^{0}\kappa_0' dm\right)\right], \tag{4.31}$$

$$Q_1 = \int_{-m_0}^{-m_1}\left(\frac{\omega_0 B_0'}{\sqrt{B_0^2-1}}+\frac{1}{m+m_0}\right)dm. \tag{4.32}$$

In Eqs. (4.29)–(4.32), the irregular turning points $\pm m_1$ may be evaluated by setting $E=U_-(\pm m_0)$, and it should be recalled that $\pm m_0$ are the minima of $U_-(m)$. Further,

$$\kappa_0=\kappa(m,\epsilon=0); \quad \kappa_0'=\left.\frac{\partial\kappa(m,\epsilon)}{\partial\epsilon}\right|_{\epsilon=0}, \tag{4.33}$$

$$\chi_0=\chi(m,\epsilon=0); \quad \chi_0'=\left.\frac{\partial\chi(m,\epsilon)}{\partial\epsilon}\right|_{\epsilon=0}, \tag{4.34}$$

$$B_0=\cos q(m,\epsilon=0); \quad B_0'=\left.\frac{\partial\cos q(m,\epsilon)}{\partial\epsilon}\right|_{\epsilon=0}, \tag{4.35}$$

with

$$\epsilon\equiv E-U_-(-m_0). \tag{4.36}$$

The problem of finding the low level splittings is thus reduced to the evaluation of a handful of integrals. The proliferation of notation masks the actual simplicty of these formulas.

V. TUNNEL SPLITTINGS FOR Fe_8

We now apply our general result, Eq. (4.28) to the specific problem of Fe_8, with the model Hamiltonian (1.1). The action integrals are evaluated in Appendix D, and we find that the splitting of the nth pair of levels is given by

$$\Delta_n = \frac{1}{n!}\sqrt{\frac{8}{\pi}}\omega_0 F^{n+1/2}e^{-\Gamma_0}\cos\Lambda_n, \tag{5.1}$$

where

$$\omega_0 = 2J[k_1k_2(1-h_{x0}^2)]^{1/2}, \tag{5.2}$$

$$F = 8J\frac{\lambda^{1/2}(1-h_x^2)^{3/2}}{1-\lambda-h_x^2}, \tag{5.3}$$

$$\Gamma_0 = J\left[\ln\left(\frac{\sqrt{1-h_x^2}+\sqrt{\lambda}}{\sqrt{1-h_x^2}-\sqrt{\lambda}}\right)-\frac{h_x}{\sqrt{1-\lambda}}\times\ln\left(\frac{\sqrt{(1-h_x^2)(1-\lambda)}+h_x\sqrt{\lambda}}{\sqrt{(1-h_x^2)(1-\lambda)}-h_x\sqrt{\lambda}}\right)\right], \tag{5.4}$$

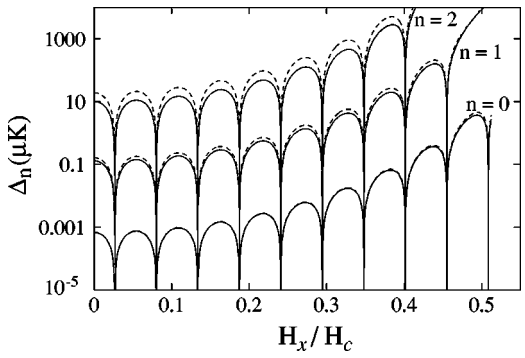

FIG. 5. Comparison between numerical (solid lines) and analytic [Eqs. (5.1)–(5.5), dashed lines] results for the splitting between the first three pairs of levels for $H_z=0$. The parameters are $k_1=0.321$ K, $k_2=0.229$ K, close to those for Fe_8.

$$\Lambda_n=\max\left\{0,\pi J\left(1-\frac{H_x}{\sqrt{1-\lambda}H_c}\right)-n\pi\right\}. \tag{5.5}$$

In these equations, $\overline{J}=J+\frac{1}{2}$, $h_x=JH_x/\overline{J}H_c$, and $h_{x0}=H_x/H_c$. Recall that $H_c=2k_1J/g\mu_B$ and $\lambda=k_2/k_1$ (see Table I), and note that unlike Eq. (5.4), what appears in Eq. (5.5) is the ratio H_x/H_c, i.e., h_{x0}, not h_x. This fact is important for the location of the diabolical points.

These results possess several points of interest. The first concerns the fields where the nth tunnel splitting vanishes. Taking account of the fact that Λ_n is necessarily positive as indicated by Eq. (5.5), we see that this happens whenever[29,19]

$$\frac{H_x}{H_c}=\frac{\sqrt{1-\lambda}}{J}\left[J-l-\frac{1}{2}\right], \tag{5.6}$$

with $l=n,\ n+1,\ \ldots,\ 2J-n-1$, yielding $2(J-n)$ quenching points in all for Δ_n. When $n=0$, these are the results quoted in Sec. I.

In Fig. 5 we compare Eq. (5.1) with the numerically evaluated splittings for the first three pairs of levels. Within our numerical precision, we always find the zeros of Δ_n to agree with Eq. (5.6). Note, however, that for other values of H_x, the discrepancy between the numerics and Eq. (5.1) is well outside our numerical error, so that Eq. (5.1) is not exact, even though as an asymtotic estimate of the splitting it is rather good. This means that in general the leading semiclassical approximation is not exact for the Hamiltonian (1.1), and only the quenching points appear to be so reproduced. The second point to note is that for $n=1$ (the pair of first excited states in each well), the highest field quenching point is lost, for $n=2$, the highest two points are lost, and so on, exactly as indicated by Eq. (5.6).

Next, let us compare our answers with previous work. Let us consider the tunneling action Γ_0 in the Gamow factor $\exp(-\Gamma_0)$ first. Except for the replacement of J by $\overline{J}$ and h_{x0} by h_x, this is precisely Eq. (3.10) of the third paper of Ref. 19. This agreement is unsurprising, because if we write Δ_n in the form of a prefactor c_1 times a Gamow factor $\exp(-Jc_2)$ where $c_2=O(1)$, then the $J\to\overline{J}$, and $h_{x0}\to h_x$ corrections in Eq. (5.4) represent terms that should be included in the prefactor c_1, which we did not seek in the third paper of Ref. 19. The detailed form of the prefactor is perhaps more interesting. Up to multiplicative terms of order J^0, our answer for Δ_n agrees precisely with that in Refs. 38 and 39. We do not understand, however, how these papers have succeeded in sidestepping the difficulties in the path integral treatment that were noted by Enz and Schilling,[42] and by Belinicher, Providencia, and da Providencia.[43] In Ref. 38, for instance, the problem is treated by writing the spin coherent-state expectation value of the Hamiltonian (1.1) in spherical polar coordinates, and integrating out $\cos\theta$ (the J_z projection) exactly, and then addressing the resulting effective Lagrangian for the ϕ coordinate exactly as for a massive particle in one dimension. In performing the integration over θ, however, it is not clear to us why S^2 is replaced by $S(S+1)$ in the scalar potential $V(\phi)$ [see Eq. (12) there], but not in the vector potential $\Theta(\phi)$.

A related point, which is relatively minor, but has scope for creating confusion, is that if the Gamow factor is written as $\exp(-Jc_2)$ with $c_2=O(1)$, then it is safest to write the J dependence of the prefactor as $\omega_0 J^{n+1/2}$, since ω_0 depends on parameters such as k_1 and k_2, whose scaling with J is a matter of choice, at least as far as model Hamiltonians are concerned. We may note that Villain and Fort's treatment[9] does not readily yield the complete prefactor.

One further check is obtained by considering the limiting case $H_x=0$, answers for which are known [see, e.g., Eq. (16) of Ref. 42, Eq. (48) of Ref. 43, or Ref. 44.] Transcribing Eqs. (4.30) and (4.31) from Ref. 44 in terms of the present parameters, we get

$$\Delta_n=\frac{1}{n!}F_0^n\Delta_0; \tag{5.7}$$

$$F_0=8J\frac{\sqrt{\lambda}}{1-\lambda}, \tag{5.8}$$

$$\Delta_0=8\omega_{00}\left(\frac{J}{\pi}\right)^{1/2}\frac{\lambda^{1/4}}{1+\sqrt{\lambda}}\left(\frac{1-\sqrt{\lambda}}{1+\sqrt{\lambda}}\right)^J, \tag{5.9}$$

with $\omega_{00}=2J(k_1k_2)^{1/2}$. It follows from Eqs. (5.2)–(5.5) that as $H_x\to 0$, $\omega_0\to\omega_{00}$, $F\to F_0$ [see Eq. (5.3)], $\cos\Lambda_n\to\pm 1$, and

$$\Gamma_0\to(J+\tfrac{1}{2})\ln\frac{1+\sqrt{\lambda}}{1-\sqrt{\lambda}}. \tag{5.10}$$

It is then easy to see that our present answers for Δ_n go over precisely into Eqs. (5.7)–(5.9).

VI. DISCUSSION

As stated in Sec. I, diabolical points require special conditions, and are rare for that reason. We contrast here the way in they come about in our problem and those involving a massive particle with $p^2+V(\mathbf{x})$ type Hamiltonian, where the dimensionality of $\mathbf{x}$ is arbitrary. Take the path integral view first. Wilkinson[45] notes the possibility of interfering tunneling trajectories when the tunneling action is complex,

and also notes that this can only happen in two or more dimensions, i.e., for phase space of dimension 4 or more. The spin system shows interference[20,21] even though its phase space is two dimensional (the unit sphere S^2). The same conclusion is reached from the operator point of view. General results from Sturm-Liouville theory forbid the occurrence of *any* degeneracy in the discrete part of the spectrum for a $p^2+V(\mathbf{x})$ Hamiltonian if the boundary conditions at the end points are independent, whereas our difference equation is one dimensional yet has degeneracy. An entertaining example of a continuum one-dimensional problem with diabolical points has been invented by Berry and Mondragon,[46] but the Hamiltonian is now p^4, quartic in the momentum, and the boundary conditions compatible with hermiticity form a four parameter system, which is large enough to admit diabolicity.

We conclude by mentioning some remarkable exact properties of the special Hamiltonian (1.1).[31] First [see Eqs. (1.2) and (1.3)], the diabolical points form part of a perfect centered rectangular lattice in the H_x–H_z plane. Second, in general, we have a *simultaneous* degeneracy of more than one pair of levels—if we arrange the points on a set of concentric rhombi, one pair is degenerate on the outermost rhombus, two on the next, three on the one next to that, and so on. Both these facts are captured by the leading order DPI analysis. The exact results suggest the existence of a higher dynamical symmetry, but that is not yet established. Further, when higher-order anisotropy terms are included in the Hamiltonian to obtain quantitative agreement with experimentally observed period,[7] the simultaneous degeneracy of several pairs of levels and the perfect lattice of diabolical points are no longer exact properties, but they continue to hold to fair approximation.[17]

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APPENDIX A: DERIVATION OF HERRING'S FORMULA

The Schrödinger equations obeyed by C_m and a_m defined in Sec. III are

$$(w_m-E_0)C_m+\sum_{n=m-2}^{m+2}{}' t_{m,n}C_n=0, \tag{A1}$$

$$(w_m-E_1)a_m+\sum_{n=m-2}^{m+2}{}' t_{m,n}a_n=0. \tag{A2}$$

We define m_r to be 1 if J is integral, and 1/2 when J is half integral. Multiplying Eq. (A1) by a_m, Eq. (A2) by C_m, summing over m from m_r to J, and subtracting corresponding terms, we get

$$(E_1-E_0)\sum_{m=m_r}^{J} C_m a_m+\Sigma_1-\Sigma_2=0, \tag{A3}$$

where

$$\Sigma_1=\sum_{m=m_r}^{J}\sum_{n=m-2}^{m+2}{}' a_m t_{m,n}C_n, \tag{A4}$$

$$\Sigma_2=\sum_{m=m_r}^{J}\sum_{n=m-2}^{m+2} C_m t_{m,n}a_n. \tag{A5}$$

To simplify Eq. (A3), we first note that by Eq. (3.1)

$$\sum_{m=m_r}^{J} C_m a_m\approx\frac{1}{\sqrt{2}}\sum_{m=m_r}^{J} C_m^2\approx\frac{1}{\sqrt{2}}, \tag{A6}$$

since the product C_mC_{-m} is everywhere exponentially small, and since C_m^2 is concentrated almost completely in the right well. Second, most of the terms in the sums Σ_1 and Σ_2 can be seen to be identical by shifting the summation indices in various terms suitably, and making use of the symmetry $t_{m,n}=t_{n,m}$. For example, the difference between the terms in Σ_1 with $n=m+2$, and those in Σ_2 with $n=m-2$ equals

$$\begin{aligned}&\sum_{m=m_r}^{J}(a_m t_{m,m+2}C_{m+2}-C_m t_{m,m-2}a_{m-2})\\&=\sum_{m=m_r}^{J} a_m t_{m,m+2}C_{m+2}-\sum_{m=m_r-2}^{J-2} C_{m+2}t_{m+2,m}a_m\\&=-a_{m_r-1}t_{m_r-1,m_r+1}C_{m_r+1}-a_{m_r-2}t_{m_r-2,m_r}C_{m_r},\end{aligned} \tag{A7}$$

where we have made use of the obvious facts that $t_{J,J+2}$ and $t_{J-1,J+1}$ are identically zero. The differences between the other terms in Σ_1 and Σ_2 can be similarly evaluated, and reduce to a small number of terms involving the product of an a with a C, which can then be written entirely in terms of C's using Eq. (3.1). Finally, we can see that $E_1-E_0=E_0-E_2=\pm\Delta/2$, where Δ is as quoted in the main text [Eq. (3.2)].

Herring gives a more careful justification of his formula by employing the Temple-Kato error bound on energy eigenvalues.[47,48] His argument can be adapted word for word to the present problem, and shows that the error in the splitting as calculated via Eq. (3.2) is exponentially smaller than the splitting itself, by a factor such as e^{-cJ} where $c>0$. As $J\to\infty$, therefore, Eq. (3.2) is asymptotically correct.

Mathematically oriented writings sometimes frame the argument in terms of an abstract eigenoperator I^{op} for all left and right localized states with eigenvalues -1 and $+1$ respectively,[49,41] or a projection operator Θ^{op} for the right localized states.[50] The splitting then appears as the matrix element of the commutator $[I^{\mathrm{op}},\mathcal{H}]$ or $[\Theta^{\mathrm{op}},\mathcal{H}]$ between the left and right localized states. Evaluation of this matrix element is completely equivalent to finding the difference $\Sigma_1-\Sigma_2$, however, so the physical idea is exactly the same.

APPENDIX B: CALCULATION OF THE DPI WAVE FUNCTION AMPLITUDE

Our objective in this appendix is to calculate the amplitude A in the central region DPI wave function (4.4), given that the state C_m is normalized. This is accomplished by relating the wave function across the turning points $-m_t$ and $-m_1$ by connection formulas. In fact, as we shall see, the connection formula near $-m_t$ can be sidestepped, and only that near $-m_1$ is actually applied.

1. DPI form near potential-well minimum

Let us first find C_m in the classically allowed region near $-m_0$, the minimum of $U_-(m)$ (see Fig. 4). We assume, as will be seen to be true for Eq. (1.1), that in this region $U_- = U_0$. For energies close to $U_-(-m_0)$, and m close to $-m_0$, the eikonal equation can only be satisfied if q is close to zero. We can therefore expand $\mathcal{H}_{\rm sc}$ in powers of $m+m_0$ and q:

$$\mathcal{H}_{\rm sc}(q,m) \approx U_-(-m_0) + \frac{1}{2M}q^2 + \frac{1}{2}M\omega_0^2(m+m_0)^2 + \ldots, \tag{B1}$$

where

$$M = -[2t_1(-m_0) + 8t_2(-m_0)]^{-1} > 0, \tag{B2}$$

$$\omega_0^2 = -2(t_1+4t_2)\left.\frac{\partial^2 U_-}{\partial m^2}\right|_{m=-m_0}. \tag{B3}$$

Note that by virtue of Eq. (2.4), and its natural extension to second derivatives, ω_0 is of order $1/J$ relative to t_1 and t_2.

Since the eikonal equation is also the Hamilton-Jacobi equation with $q = \partial\Phi/\partial m$, where Φ is the action, the problem is identical to that of a harmonic oscillator in the approximation (B1). (Alternatively, we could arrive at the same result by approximating the original recurrence relation by a differential equation in the vicinity of $-m_0$.) For the nth state therefore

$$E_0 = U_-(-m_0) + (n+\tfrac{1}{2})\omega_0, \tag{B4}$$

and

$$C_m = [2^{2n}(n!)^2\pi\xi^2]^{-1/4} e^{-x^2/2\xi^2} H_n(x/\xi), \tag{B5}$$

where $x = m+m_0$, H_n is the nth Hermite polynomial, and $\xi = (M\omega_0)^{-1/2}$. The wave function is already normalized, and the additional tails from the forbidden region only modify the normalization by an exponentially small amount.

It is apparent that the expansion (B1) is invalid unless the point $-m_0$ is sufficiently far from the edge $m=-J$. Since the width of the wave function (B5) is $\sqrt{n}\xi$, a necessary condition for the validity of our procedure is

$$J - m_0 \gg \sqrt{n}\xi. \tag{B6}$$

If this condition does not hold, then the recursion relation must be solved near the edge by a different method, which is tantamount to using the Holstein-Primakoff or Bogoliubov transformation. An example of the latter approach is given in Sec. IV of Ref. 44.

From the viewpoint of the DPI method, we have two turning points very close to $-m_0$, one to the left, and one to the right, since the condition $E = U_-(m)$ is then satisfied. The one to the left has been discussed above. Let us now consider the one to the right, at $m=-m_t$. We have

$$-m_t + m_0 = \left[\frac{2n+1}{M\omega_0}\right]^{1/2} \sim (nJ)^{1/2}. \tag{B7}$$

The neglected terms in Eq. (B1), on the other hand, are of relative orders q^4, $(m+m_0)^3/J^3$, and $(m+m_0)q^2/J$, and thus smaller than $n\omega_0$ for $x \ll (nJ^2)^{1/3}$. Thus provided $n \ll J$, the solution (B5) holds well past $-m_t$, and can be matched directly onto the DPI solution under the barrier, without any need of connection formulas at $m=-m_t$.[51] This argument is given at greater length in Sec. V of Ref. 44.

2. DPI form in ordinary forbidden region

In the region $-m_t < m < -m_1$, the DPI solution that decays with increasing m can be taken as

$$C_m = \frac{B}{\sqrt{|v(m)|}} \exp\left(-\int_{-m_t}^{m} \kappa(m')dm'\right), \tag{B8}$$

where $\kappa(m) = \mathrm{Im}\, q(m) > 0$. This solution must be matched on to Eq. (B5) to determine B. For $J^{1/3} \ll (m+m_t) \ll (nJ^2)^{1/3}$, we can continue to use the harmonic approximation (B1), and a simple calculation,[36] traceable to Furry,[35] leads to

$$B = \left(\frac{\omega_0 g_n}{2\pi}\right)^{1/2}, \tag{B9}$$

where g_n is defined in Eq. (1.7).

3. DPI form in central region

Step 3 is to find the wave function in the central region near $m=0$. This is already done if there are no turning points between $-m_t$ and $m=0$. For the Hamiltonian (1.1), it turns out that we encounter another turning point where $E = U_f(m)$ (the only possibility) at an intermediate point $m = -m_1$ (see Fig. 4), The wavefunction in the central region then has the form (4.4), and the coefficient A in this form must be related to B in Eq. (B8) via a connection formula at $-m_1$. This formula was found in Ref. 27. If we rewrite Eq. (B8) as

$$C_m = \frac{\tilde{A}}{2\sqrt{|v(m)|}} \exp\left(-\int_{-m_1}^{m} \kappa(m')dm'\right), \tag{B10}$$

$$\tilde{A} = 2B \exp\left(-\int_{-m_t}^{-m_1} \kappa(m')dm'\right), \tag{B11}$$

we have

$$A=\tilde{A}=\left(\frac{2\omega_0 g_n}{\pi}\right)^{1/2}\exp\left(-\int_{-m_t}^{-m_1}\kappa(m')dm'\right), \tag{B12}$$

where we have used Eqs. (B9) and (B11) in the last step[52].

APPENDIX C: EXTRACTION OF SINGULAR PARTS OF TUNNELING ACTION INTEGRALS

The purpose of this appendix is to derive the singularity extracted formulas for the tunnel splitting in Sec. IV B. The procedure is similar to that used for the continuum case in Ref. 36. We begin by defining

$$\Phi(\epsilon)=-i\int_{-m_t(\epsilon)}^{0}q(m,\epsilon)dm, \tag{C1}$$

where the energy dependence is made explicit. The splitting for the nth pair of states is then given by

$$\Delta_n=\frac{\omega_0 g_n}{\pi}(e^{-2\Phi(\epsilon_n)}+\text{c.c.}), \tag{C2}$$

with $\epsilon_n=(n+\frac{1}{2})\omega_0$. Writing $x=m+m_0$ as in Eq. (B5), the integrand in Φ behaves as $(x^2-x_t^2)^{1/2}$ near the lower limit, with $x_t=-m_t+m_0\sim\epsilon^{1/2}$. Thus there is a singular part in Φ of the form $\epsilon\ln\epsilon$, which it is our goal to extract. To this end, we differentiate Eq. (C1) to get

$$\Phi'(\epsilon)=\frac{d\Phi}{d\epsilon}=-i\int_{-m_t(\epsilon)}^{0}\frac{\partial q}{\partial\epsilon}dm. \tag{C3}$$

Note that the term arising from differentiating the lower limit vanishes, nor is there any explicit contribution from the singular behavior $q\sim(m+m_c)^{1/2}$ for m near $-m_c$.

Next, let us divide $\Phi'(\epsilon)$ into two integrals, Φ_1', in which the limits of integration are $-m_t$ and $-m_1$, and Φ_2', which runs from $-m_1$ to 0. Defining

$$B_\epsilon(m)=\cos(q(m,\epsilon)), \tag{C4}$$

we have

$$\Phi_1'(\epsilon)=\int_{-m_t(\epsilon)}^{0}\frac{B_\epsilon'}{\sqrt{B_\epsilon^2(m)-1}}dm, \tag{C5}$$

where $B_\epsilon'=\partial B_\epsilon/\partial\epsilon$. It follows from Eq. (B1) that near $m=-m_t$,

$$B_\epsilon\approx 1+(\tfrac{1}{2}M\omega^2x^2-\epsilon)M+\ldots, \tag{C6}$$

so the integrand in Eq. (C5) behaves as $-1/\omega_0(x^2-x_t^2)^{1/2}$. If we add and subtract the integral of this expression, we obtain

$$\Phi_1'(\epsilon)=-\frac{1}{\omega_0}\int_{x_t}^{x_1}\frac{dx}{\sqrt{x^2-x_t^2}}+\int_{x_t}^{x_1}\left[\frac{B_\epsilon'}{\sqrt{B_\epsilon^2(m)-1}}+\frac{1}{\omega_0\sqrt{x^2-x_t^2}}\right]dx, \tag{C7}$$

where $x_1=m_0-m_1$. The first integral can be evaluated exactly. In the second integral we can put $\epsilon=0$ both in the limits and in the integrand, since we are not interested in terms of $O(\epsilon)$. Ignoring terms of this order throughout, and making use of the relation

$$x_t^2=2\epsilon/M\omega_0^2, \tag{C8}$$

we obtain

$$\Phi_1'(\epsilon)=\frac{1}{2\omega_0}\left[\ln\frac{\epsilon}{2M\omega_0(m_0-m_1)^2}+2Q_1\right], \tag{C9}$$

where Q_1 is given by Eq. (4.32). Also, we can evaluate m_1 at $\epsilon=0$.

The remaining contribution to $\Phi'(\epsilon)$, $\Phi_2'(\epsilon)$, can be evaluated simply by putting $\epsilon=0$, since the neglected part is $O(\epsilon)$. Recalling the definitions (4.33) and (4.34), we have

$$\Phi_2'(\epsilon)\approx\int_{-m_1}^{0}(\kappa_0'-i\chi_0')dm. \tag{C10}$$

We now integrate the expression for $\Phi'(\epsilon)$ and obtain Φ. It is useful to separate the real and imaginary parts of the answer at this stage. For the real part, we get

$$\begin{aligned}\Gamma&=2\,\mathrm{Re}\Phi\\&=\Gamma_0+\frac{\epsilon}{\omega_0}\left[2Q_1-1+\ln\frac{\epsilon}{2M\omega_0(m_0-m_1)^2}+2\omega_0\int_{-m_1}^{0}\kappa_0'\,dm\right],\end{aligned} \tag{C11}$$

with Γ_0 given by Eq. (4.29), while for the imaginary part, $\Lambda_n\equiv-2\,\mathrm{Im}\,\Phi$, we get Eq. (4.30).

Substituting Eqs. (C11), (4.30), and the definition (1.7) of g_n in the formula (C2) for Δ_n, and recalling Eq. (4.31) and that $\epsilon_n=(n+\frac{1}{2})\omega_0$, we finally obtain the answer quoted in Eq. (4.28).

APPENDIX D: EVALUATION OF ACTION INTEGRALS FOR Fe_8

In this appendix, we evaluate the action integrals for the specific problem described by the Hamiltonian (1.1). The first step is to find the various matrix elements of this Hamiltonian. We have

$$w_m=\frac{1}{2}(k_1+k_2)[J(J+1)-m^2], \tag{D1}$$

$$t_{m,m+1}=-\frac{1}{2}g\mu_B H_x[J(J+1)-m(m+1)]^{1/2}, \tag{D2}$$

$$t_{m,m+2}=\frac{1}{4}(k_1-k_2)\{[J(J+1)-m(m+1)]\times[J(J+1)-(m+1)(m+2)]\}^{1/2}. \tag{D3}$$

We must now replace these by continuous functions $w(m)$, $t_1(m)$, and $t_2(m)$. Since our formalism requires knowing the first two terms in the action in an expansion in powers of $1/J$,

it follows that we need only determine the functions $w(m)$, etc., to the same order. Furthermore, this determination need not be made in the form of a power series, and any functional representation that gives the first two terms correctly will be adequate. The most convenient way to do this is to replace the combination $J(J+1)$ in the above expressions by $\overline{J^2}$, with $\overline{J}=J+\frac{1}{2}$.

It is convenient to measure energies (including ω_0) in units of $k_1\overline{J^2}$, and introduce the scaled variable $\mu=m/\overline{J}$. In terms of these variables,

$$w(m)=(1+\lambda)(1-\mu^2)/2, \tag{D4}$$

$$t_1(m)=-h_x(1-\mu^2)^{1/2}, \tag{D5}$$

$$t_2(m)=(1-\lambda)(1-\mu^2)/4. \tag{D6}$$

The turning points $\mu_0=m_0/\overline{J}$, and $\mu_1=m_1/\overline{J}$ (for $\epsilon=0$) are given by

$$\mu_0=(1-h_x^2)^{1/2}, \tag{D7}$$

$$\mu_1=[(1-\lambda-h_x^2)/(1-\lambda)]^{1/2}. \tag{D8}$$

It is simplest to express everything in terms of μ_0 and μ_1, so we give inverse formulas as well:

$$h_x=(1-\mu_0^2)^{1/2}, \tag{D9}$$

$$\lambda=(\mu_0^2-\mu_1^2)/(1-\mu_1^2). \tag{D10}$$

The mass and the small oscillation frequency are given by

$$M=\frac{1}{2\lambda h_x^2}=\frac{1}{2}\,\frac{1-\mu_1^2}{(1-\mu_0^2)(\mu_0^2-\mu_1^2)}, \tag{D11}$$

$$\omega_0=\frac{2}{\overline{J}}[\lambda(1-h_x^2)]^{1/2}=\frac{2\mu_0}{\overline{J}}\left(\frac{\mu_0^2-\mu_1^2}{1-\mu_1^2}\right)^{1/2}. \tag{D12}$$

To evaluate the integrals, we need expressions for κ_0, χ_0, κ_0', etc., in the ranges $\mu_1<\mu<\mu_0$, and $0<\mu<\mu_1$. The requisite calculations are straightforward so we give the main results only. First, in the range $\mu_1<\mu<\mu_0$, we get

$$B_0=\cosh\kappa_0=\frac{1-\mu_1^2-[(\mu_0^2-\mu_1^2)(\mu^2-\mu_1^2)]^{1/2}}{[(1-\mu_0^2)(1-\mu^2)]^{1/2}}, \tag{D13}$$

$$\sqrt{B_0^2-1}=\sinh\kappa_0=\frac{(\sqrt{\mu_0^2-\mu_1^2}-\sqrt{\mu^2-\mu_1^2})\sqrt{1-\mu_1^2}}{[(1-\mu_0^2)(1-\mu^2)]^{1/2}}, \tag{D14}$$

$$B_0'=-\frac{1}{2}\,\frac{1-\mu_1^2}{[(1-\mu_0^2)(1-\mu^2)(\mu_0^2-\mu_1^2)(\mu^2-\mu_1^2)]^{1/2}}, \tag{D15}$$

$$\frac{\omega_0 B_0'}{\sqrt{B_0^2-1}}=-\frac{\mu_0}{\overline{J}}\,\frac{1}{\sqrt{\mu^2-\mu_1^2}(\sqrt{\mu_0^2-\mu_1^2}-\sqrt{\mu^2-\mu_1^2})}. \tag{D16}$$

Next, in the range $0<\mu<\mu_1$, we first solve Eqs. (4.5) and (4.6) with $\epsilon=0$, to obtain

$$\cosh\kappa_0=[(1-\mu_1^2)/(1-\mu_0^2)]^{1/2}, \tag{D17}$$

$$\cos\chi_0=[(1-\mu_1^2)/(1-\mu^2)]^{1/2}, \tag{D18}$$

$$\sin\chi_0=[(\mu_1^2-\mu^2)/(1-\mu^2)]^{1/2}. \tag{D19}$$

To find κ_0' and χ_0', we differentiate Eqs. (4.5) and (4.6), and set $\epsilon=0$. Solving the resulting equations along with Eqs. (D17)–(D19), we obtain

$$\begin{pmatrix}\kappa_0'\\ \chi_0'\end{pmatrix}=-\frac{(1-\mu_1^2)^{1/2}}{2(\mu_0^2-\mu^2)}\begin{pmatrix}(\mu_0^2-\mu_1^2)^{-1/2}\\ (\mu_1^2-\mu^2)^{-1/2}\end{pmatrix}. \tag{D20}$$

The first integral that we wish to evaluate is Γ_0. Let us break the integration range into two at m_1. From the right-hand part, an integration by parts gives

$$\begin{aligned}\Gamma_{01}&=2\overline{J}\int_{\mu_1}^{\mu_0}\kappa_0 d\mu\\ &=2\overline{J}\left[\kappa_0(\mu)\mu\Big|_{\mu_1}^{\mu_0}-\int_{\mu_1}^{\mu_0}\frac{\mu}{\sinh\kappa_0}\,\frac{dB_0(\mu)}{d\mu}d\mu\right],\end{aligned} \tag{D21}$$

while from the left-hand part we get

$$\Gamma_{02}=2\overline{J}\int_0^{\mu_1}\kappa_0 d\mu=2\overline{J}\kappa_0(\mu_1)\mu_1, \tag{D22}$$

as κ_0 is a constant in this range. Since $\kappa_0(\mu_0)=0$,Γ_{02} cancels the first term in Eq. (D21), leaving us only with the second for Γ_0. Using Eqs. (D13) and (D14), we find

$$\Gamma_0=2\overline{J}(1-\mu_1^2)^{1/2}\int_{\mu_1}^{\mu_0}\frac{d\mu}{(1-\mu^2)(\mu^2-\mu_1^2)^{1/2}}. \tag{D23}$$

The integration is now elementary, and the result, expressed back in terms of λ and h_x is Eq. (5.4).

The second integral to be evaluated is Λ_n [Eq. (4.30)]. For the first term, we integrate by parts, and use Eqs. (D18) and (D19):

$$\begin{aligned}2\overline{J}\int_0^{\mu_1}\chi_0(\mu)d\mu&=2\overline{J}\int_0^{\mu_1}\frac{\mu}{\sin\chi_0}\,\frac{d}{d\mu}\cos\chi_0 d\mu\\ &=2\overline{J}\int_0^{\mu_1}\frac{\mu^2}{(1-\mu^2)(\mu_1^2-\mu^2)^{1/2}}d\mu\\ &=\pi\overline{J}[1-(1-\mu_1^2)^{1/2}].\end{aligned} \tag{D24}$$

For the second term in Eq. (4.30), we have, with $\epsilon=(n+\frac{1}{2})\omega_0$, and Eqs. (D12) and (D20),

$$2\epsilon\overline{J}\int_0^{\mu_1}\chi_0' d\mu = -(2n+1)\mu_0(\mu_0^2-\mu_1^2)^{1/2} \times \int_0^{\mu_1}\frac{d\mu}{(\mu_0^2-\mu^2)(\mu_1^2-\mu^2)^{1/2}} = -(n+\tfrac{1}{2})\pi. \quad \text{(D25)}$$

Adding together the parts, and rewriting the result in terms of H_x and λ, we get Eq. (5.5). The restriction that Λ_n be positive follows from the fact that we chose $q(m)$ to have a positive real part in Eq. (4.27). Thus Λ is necessarily positive as defined in Eq. (4.30). If H_x is so large as to yield a negative value for the function of H_x that results after doing the integral, that means that in fact there are no irregular turning points in the problem. Both terms in Eq. (4.27) are then equal, and the formula reduces to the expected one when there are only regular turning points.

The third integral we need is that of κ_0' from 0 to μ_1. Using Eqs. (D12) and (D20), we get

$$2\omega_0\overline{J}\int_0^{\mu_1}\kappa_0' d\mu = -2\mu_0\int_0^{\mu_1}\frac{d\mu}{\mu_0^2-\mu^2} = \ln\frac{\mu_0-\mu_1}{\mu_0+\mu_1}. \quad \text{(D26)}$$

The fourth and last integral needed is Q_1. Substituting Eq. (D16) in Eq. (4.32), we obtain

$$Q_1 = -\int_{\mu_0}^{\mu_1}\left[\frac{\mu_0}{\sqrt{\mu^2-\mu_1^2}(\sqrt{\mu_0^2-\mu_1^2}-\sqrt{\mu^2-\mu_1^2})} - \frac{1}{\mu_0-\mu}\right]d\mu. \quad \text{(D27)}$$

The integrand is now nonsingular at $\mu=\mu_0$. We can make this manifest by rationalizing the difference of square roots in the first term. Some simple algebra yields

$$Q_1 = -\int_{\mu_0}^{\mu_1}\frac{1}{\sqrt{\mu^2-\mu_1^2}}\frac{\mu_0^2+\mu^2-\mu_1^2}{\mu_0\sqrt{\mu_0^2-\mu_1^2}+\mu\sqrt{\mu^2-\mu_1^2}}d\mu. \quad \text{(D28)}$$

We now make the substitution $\mu=\mu_1\cosh z$, and define

$$\cosh z_0 = \mu_0/\mu_1. \quad \text{(D29)}$$

This yields

$$Q_1 = -\int_0^{z_0}\frac{\cosh 2z_0+\cosh 2z}{\sinh 2z_0+\sinh 2z}dz = -\int_0^{z_0}\frac{\cosh(z+z_0)}{\sinh(z+z_0)}dz = -\ln(2\cosh z_0) = -\ln\frac{2\mu_0}{\mu_1}. \quad \text{(D30)}$$

We now have all the ingredients needed to calculate the quantity F. Substituting Eqs. (D11), (D12), (D26), and (D30) in Eq. (4.31), and writing the result in terms of λ and h_x, we obtain Eq. (5.3). Note that in writing down the final answer, we have replaced $\overline{J}$ by J and h_x by h_{x0} in this formula. This is because F is part of the pre-exponential factor in Δ_n, which is determined only to leading order in $1/J$. Keeping higher-order corrections by distinguishing between $\overline{J}$ and J or h_x and h_{x0} is not justified.

The final answer (5.1) for Δ_n is obtained by substituting Eqs. (5.4), (5.5), and (5.3) in Eq. (4.28).

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