

# Low-temperature properties of a classical zigzag spin chain near the ferromagnet-helimagnet transition point

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Low-temperature thermodynamics of the classical frustrated ferromagnetic spin chain near the ferromagnet-helimagnet transition point is studied by means of mapping to the continuum limit. The calculation of the partition function and spin-correlation function is reduced to quantum problem of a particle in potential well. It is shown that exactly at the transition point the correlation length behaves as  $T^{-1/3}$  and the magnetic susceptibility diverges as  $T^{-4/3}$  in the low-temperature limit. Corresponding numerical factors for the correlation length and the susceptibility is calculated. It is shown that the low-temperature susceptibility in the helical phase near the transition point has a maximum at some temperature. Such behavior as well as the location and the magnitude of the maximum as a function of deviation from the transition point are in agreement with that observed in several materials described by the quantum  $s=1/2$  version of this model.

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## I. INTRODUCTION

Lately, there has been considerable interest in low-dimensional spin models that exhibit frustration.<sup>1</sup> A very interesting class of such systems with unique physical properties is chain compounds consisting of edge-sharing  $\text{CuO}_4$  units.<sup>2-7</sup> The frustration in these compounds arises from the competition of the ferromagnetic (F) interaction  $J_1$  of nearest-neighbor (NN) spins and the antiferromagnetic (AF) next-nearest-neighbor (NNN) interaction  $J_2$ . An appropriate model describing the magnetic properties of such copper oxides is so-called F-AF spin chain model, the Hamiltonian of which has a form

$$H = J_1 \sum \mathbf{S}_n \cdot \mathbf{S}_{n+1} + J_2 \sum \mathbf{S}_n \cdot \mathbf{S}_{n+2}, \quad (1)$$

where  $J_1 < 0$  and  $J_2 > 0$ .

This model is characterized by the frustration parameter  $\alpha = J_2/|J_1|$ . The ground-state properties of the quantum  $s=1/2$  F-AF chain have been intensively studied last years.<sup>8-16</sup> It is known that the ground state of model (1) is ferromagnetic for  $\alpha < 1/4$ . At  $\alpha = 1/4$  the quantum phase transition to the incommensurate singlet phase with helical spin correlations takes place. Remarkably, this transition point does not depend on a spin value, including the classical limit  $s \rightarrow \infty$ .

However, the influence of the frustration on low-temperature thermodynamics is less studied, especially in the vicinity of the ferromagnet-helimagnet transition point. It is of a particular importance to study this problem because edge-sharing cuprates with  $\alpha \approx 1/4$  (for example,  $\text{Li}_2\text{CuZrO}_4$  and  $\text{Rb}_2\text{Cu}_2\text{Mo}_3\text{O}_{12}$ ) are of special interest.<sup>17</sup> Unfortunately, at present the low-temperature thermodynamics of quantum  $s=1/2$  model (1) at  $\alpha \neq 0$  can be studied only either by using of numerical calculations of finite chains or by approximate methods. On the other hand, the classical version of model (1) can be studied by analytical methods giving exact results at  $T \rightarrow 0$ . Of course, the question arises about the relation of these results (in particular, for the susceptibility) to those of the quantum model. It is known<sup>18-20</sup> that quantum and classical ferromagnetic chains ( $\alpha=0$ ) have universal low-

temperature behavior. As was noted in Ref. 19 the physical reason of this universality is the consequence of the fact that the correlation length at  $T \rightarrow 0$  is larger than de Broglie wavelength of the spin waves. This property is inherent in the frustrated ferromagnet too. Though such universality for the frustrated ferromagnetic chains is not strictly checked at present, one can expect that the universality holds on for the F-AF chain as well. Therefore, the study of classical model (1) can be useful for the understanding of the low-temperature properties of the quantum F-AF chains.

At zero-temperature classical model (1) has long-range order (LRO) for all values of  $\alpha$ : the ferromagnetic LRO at  $\alpha \leq 1/4$  and the helical one at  $\alpha > 1/4$ . At finite temperature the LRO is destroyed by thermal fluctuations and thermodynamic quantities have singular behavior at  $T \rightarrow 0$ . In particular, the zero-field magnetic susceptibility  $\chi$  diverges. For the one-dimensional Heisenberg ferromagnet (HF)  $\chi = 2|J_1|/3T^2$ .<sup>21</sup> At  $0 < \alpha < 1/4$  the susceptibility is  $\chi = 2(1-4\alpha)|J_1|/3T^2$ . This behavior of  $\chi$  is similar to that for the quantum  $s=1/2$  F-AF model.<sup>22</sup> The value  $\chi T^2$  vanishes at the transition point. As it was noted in Ref. 22 this fact indicates the change in the critical exponent.

In this paper we focus on the low-temperature behavior of the classical F-AF chain near the ferromagnet-helimagnet transition point. At first we consider the case  $\alpha = 1/4$ , i.e., the F-AF model exactly at the transition point. This problem is interesting on its own account because the spectrum of low-energy excitations is proportional to  $k^4$  rather than  $k^2$  as for the HF model. It means that the critical exponents characterizing the low-temperature behavior of thermodynamic quantities at  $\alpha = 1/4$  can be different from those for the HF chain. Besides, the method developed for the study of the transition point can be generalized to investigate the vicinity of the transition point.

At the ferromagnet-helimagnet transition point  $\alpha = 1/4$  it is convenient to rewrite Hamiltonian (1) in the form<sup>23</sup>

$$H = \frac{1}{8} \sum (\mathbf{S}_{n+1} - 2\mathbf{S}_n + \mathbf{S}_{n-1})^2. \quad (2)$$

In Eq. (2) we put  $|J_1|=1$  and omit unessential constant.

In the classical approximation the spin operators  $\mathbf{S}_n$  are replaced by the classical vectors  $\vec{S}_n$  of the unit length. In what follows we use the continuum approach replacing  $\vec{S}_n$  by the classical vector field  $\vec{s}(x)$  with slowly varying orientations so that

$$\vec{S}_{n+1} - 2\vec{S}_n + \vec{S}_{n-1} \approx \frac{\partial^2 \vec{s}(x_n)}{\partial x^2}, \quad (3)$$

where the lattice constant is chosen as unit length.

In the low-temperature limit the thermal fluctuations are weak so that neighbor spins are directed almost parallel and continuum approach [Eq. (3)] is justified. Using the continuum approximation, Hamiltonian (2) goes over into the energy functional of the vector field  $\vec{s}(x)$ ,

$$E = \frac{1}{8} \int dx \left( \frac{\partial^2 \vec{s}}{\partial x^2} \right)^2. \quad (4)$$

This energy functional is a starting point of the investigations of model (1) at  $\alpha=1/4$ . The partition function is a functional integral over all configurations of the vector field on a ring of length  $L$ ,

$$Z = \int D\vec{s}(x) \exp \left\{ -\frac{1}{8T} \int_0^L dx \left( \frac{d^2 \vec{s}}{dx^2} \right)^2 \right\}. \quad (5)$$

It is useful to scale the spatial variable as

$$\xi = 2T^{1/3} x. \quad (6)$$

Then, the partition function takes the dimensionless form

$$Z = \int D\vec{s}(\xi) \exp \left\{ -\int_0^\lambda d\xi \left( \frac{d^2 \vec{s}}{d\xi^2} \right)^2 \right\}, \quad (7)$$

where the rescaled system length is  $\lambda=2T^{1/3}L$ . The partition function [Eq. (7)] and the correlation function  $\langle \vec{s}(l) \cdot \vec{s}(0) \rangle$  are the objects of the current study.

The paper is organized as follows. In Sec. II we consider the planar version of spin model (4) at  $\alpha=1/4$ . For this more simple model we demonstrate the technique of the calculation of the correlation function. We show that the thermodynamics of this classical one-dimensional model reduces to the zero-dimensional quantum problem of a particle in a potential well. In Sec. III the classical continuum F-AF model at the transition point is studied. In this case the partition function describes a quantum particle in an axially symmetrical potential well. We obtain the exact expressions for the susceptibility and the structure factor. In Sec. IV the behavior of the uniform susceptibility in the helical phase at  $\alpha \geq 1/4$  is studied and compared with the experimental data for the edge-shared compounds and with the results for the quantum  $s=1/2$  model. The conclusions are summarized in Sec. V. and the Appendix contains some technical aspects of the calculation of the correlation function.

## II. PLANAR SPIN CASE AT $\alpha=1/4$

### A. Partition function

We begin our investigation of the thermodynamics in the transition point with a more simple planar spin version of model (4), when all spin vectors lie in one plane and have only two components,

$$\vec{s}(\xi) = [\sin \theta(\xi), \cos \theta(\xi)]. \quad (8)$$

Such order of study is methodically justified because the technique of the correlation function calculation is similar for both planar and original three-component spin models but it is easier to demonstrate on the simple planar model.

In terms of  $\theta(\xi)$  the Hamiltonian transforms to

$$\left( \frac{d^2 \vec{s}}{d\xi^2} \right)^2 = \left( \frac{d^2 \theta}{d\xi^2} \right)^2 + \left( \frac{d\theta}{d\xi} \right)^4 \quad (9)$$

and the partition function becomes

$$Z = \int D\theta(\xi) \exp \left\{ -\int_0^\lambda d\xi (\theta'^2 + \theta'^4) \right\}, \quad (10)$$

where the prime denotes the space derivatives  $d/d\xi$ .

In general, when one deals with the field theory containing the second or higher order derivatives one has to follow the Ostrogradski prescription.<sup>24</sup> However, as will be demonstrated below, in our case we can avoid such complications and calculate the partition function and correlation functions in a more simple way.

Since the Hamiltonian contains only derivatives of the field  $\theta(\xi)$ , the partition function can be rewritten in terms of a new field

$$Z = \int Dq(\xi) \exp \left\{ -\int_0^\lambda d\xi (q'^2 + q^4) \right\}, \quad (11)$$

where

$$q(\xi) = \frac{d\theta(\xi)}{d\xi}. \quad (12)$$

To calculate the partition function we utilize well-known equivalence of the  $n$ -dimensional statistical field theory with the  $(n-1)$ -dimensional quantum field theory. It is obvious in advance that partition function [Eq. (11)] describes a quantum particle in a potential well  $U(q)=q^4$  at "temperature"  $1/\lambda$ . However, we will follow all intermediate steps because we will need them in the subsequent calculations of the correlation function.

The transition amplitude (or propagator) of a particle located initially at  $q(0)=q_i$  and finally at  $q(t)=q_f$  takes the form of a path integral

$$\langle q_f | e^{-it\hat{H}} | q_i \rangle \propto \int_{q_i}^{q_f} Dq(t) \exp \left\{ i \int_0^t dt L(\dot{q}, q) \right\}. \quad (13)$$

Then, imposing the periodic boundary conditions  $q_f=q_i=q$  and integrating over  $q$ , we obtain the partition function in a form

$$Z \propto \int dq \langle q | e^{-it\hat{H}} | q \rangle. \quad (14)$$

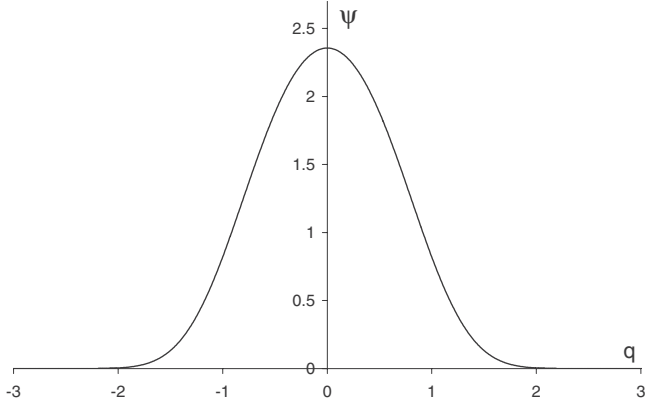


FIG. 1. Wave function  $\psi_0(q)$  for the lowest level  $\varepsilon_0=0.4208$  of planar spin model.

In our case we replace  $\xi$  by an imaginary time  $\xi \rightarrow it$  and partition function [Eq. (11)] takes the form of a path integral of a quantum particle in a potential well,

$$Z = \int Dq(t) \exp \left\{ i \int_0^{-i\lambda} dt L_0(\dot{q}, q) \right\}, \quad (15)$$

where the Lagrangian is

$$L_0 = \dot{q}^2 - q^4. \quad (16)$$

The momentum  $p$  is  $p=2\dot{q}$  and the Hamiltonian is

$$H_0 = \frac{1}{4}p^2 + q^4. \quad (17)$$

The corresponding Schrödinger equation describes a quantum anharmonic oscillator,

$$-\frac{1}{4} \frac{d^2 \psi}{dq^2} + q^4 \psi = \varepsilon \psi. \quad (18)$$

The spectrum of Eq. (18) is calculated numerically

$$\varepsilon_\alpha = 0.4208; 1.508; 2.96\dots \quad (19)$$

and the wave function  $\psi_0(q)$  for the lowest level  $\varepsilon_0=0.4208$  is shown in Fig. 1.

Now the exponent of  $\hat{H}_0$  can be represented as follows:

$$e^{-\lambda \hat{H}_0} = \sum_\alpha |\psi_\alpha\rangle e^{-\lambda \varepsilon_\alpha} \langle \psi_\alpha| \quad (20)$$

and the partition function becomes

$$Z \propto \int dq \langle q | e^{-\lambda \hat{H}_0} | q \rangle = \sum_\alpha e^{-\lambda \varepsilon_\alpha}. \quad (21)$$

As expected, we obtain the partition function of a quantum anharmonic oscillator at temperature  $1/\lambda$ . In the thermodynamic limit  $\lambda = (2T^{1/3}L) \rightarrow \infty$  only the lowest eigenvalue  $\varepsilon_0=0.4208$  gives contribution to the partition function,

$$Z \rightarrow e^{-\lambda \varepsilon_0}. \quad (22)$$

## B. Correlation function

The scalar product of vector fields located on distance  $l$  can be written as

$$\begin{aligned} \vec{s}(l) \cdot \vec{s}(0) &= \cos[\theta(l) - \theta(0)] = \cos \left[ \int_0^l \theta'(x) dx \right] \\ &= \Re \left[ \exp \left( i \int_0^\mu q(\xi) d\xi \right) \right], \end{aligned} \quad (23)$$

where  $\mu = 2T^{1/3}l$ .

Then, the correlation function can be represented as a ratio of two functional integrals,

$$\langle \vec{s}(l) \cdot \vec{s}(0) \rangle = \frac{1}{Z} \Re[Z_c], \quad (24)$$

where denominator  $Z$  is already calculated [see Eq. (21)] and

$$Z_c = \int Dq \exp \left\{ - \int_0^\lambda d\xi (q'^2 + q^4) + i \int_0^\mu q d\xi \right\}. \quad (25)$$

The latter path integral is clearly divided on two parts  $[0, \mu]$  and  $[\mu, \lambda]$  and  $Z_c$  can be represented as

$$Z_c = \int dq_0 dq_\mu Z_1(q_0, q_\mu) Z_2(q_\mu, q_0), \quad (26)$$

where the propagators  $Z_1$  and  $Z_2$  are

$$Z_1(q_0, q_\mu) = \int_{q_0}^{q_\mu} Dq \exp \left[ - \int_0^{q_\mu} dt L_1(\dot{q}, q) \right], \quad (27)$$

$$Z_2(q_\mu, q_\lambda) = \int_{q_\mu}^{q_\lambda} Dq \exp \left[ - \int_{-i\mu}^{-i\lambda} dt L_0(\dot{q}, q) \right] \quad (28)$$

and periodic boundary condition  $q_\lambda = q_0$  is applied.

The Lagrangian  $L_0$  is given by Eq. (16) and

$$L_1 = \dot{q}^2 - q^4 + iq. \quad (29)$$

The propagator  $Z_2$  is calculated straightforward using Eqs. (13) and (20),

$$Z_2 = \sum_\alpha e^{-(\lambda-\mu)\varepsilon_\alpha} \langle q_\mu | \psi_\alpha \rangle \langle \psi_\alpha | q_0 \rangle. \quad (30)$$

But the propagator  $Z_1$  requires special treatment because  $L_1$  and the corresponding quantum Hamiltonian  $H_1$  are non-Hermitian,

$$\hat{H}_1 = -\frac{1}{4} \frac{d^2}{dq^2} + q^4 - iq. \quad (31)$$

Non-Hermitian operator  $\hat{H}_1$  can be represented as

$$\hat{H}_1 = \sum_\alpha \eta_\alpha |u_\alpha\rangle \langle v_\alpha|, \quad (32)$$

where  $|u_\alpha\rangle$  and  $|v_\alpha\rangle$  are eigenfunctions of direct and conjugate eigenvalue equations,

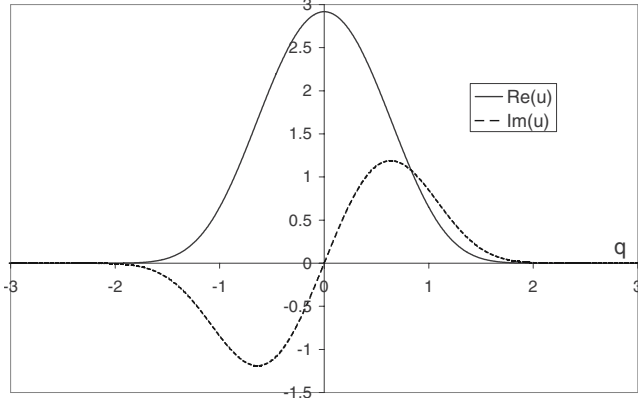


FIG. 2. Real and imaginary parts of the wave function  $u_0(q)$  for the lowest level  $\eta_0=0.6472$  of planar spin model.

$$\hat{H}_1|u_\alpha\rangle = \eta_\alpha|u_\alpha\rangle,$$

$$\hat{H}_1^\dagger|v_\alpha\rangle = \eta_\alpha^*|v_\alpha\rangle. \quad (33)$$

The normalization conditions for  $|u_\alpha\rangle$  and  $|v_\alpha\rangle$  are

$$\langle v_\alpha|u_\beta\rangle = \langle u_\alpha|v_\beta\rangle = \delta_{\alpha,\beta}. \quad (34)$$

Equation (33) for Hamiltonian (31) transform to each other by complex conjugation operation. This implies that eigenfunctions in Eq. (33) satisfy the relation  $|v_\alpha\rangle = |u_\alpha^*\rangle$ . Thus, we need to solve only one of the differential equations [Eq. (33)]. The numerical calculations show that all eigenvalues of Eq. (33) are real and positive. A few lowest eigenvalues are presented in Eq. (35),

$$\eta_\alpha = 0.6472; 1.517; 2.99\dots \quad (35)$$

Real and imaginary parts of  $u_0(q)$  for the lowest level  $\eta_0=0.6472$  are shown in Fig. 2.

Now, the propagator  $Z_1$  can be expressed through the solutions of Eq. (33),

$$Z_1 = \langle q_0|e^{-\mu\hat{H}_1}|q_\mu\rangle = \sum_\alpha e^{-\mu\eta_\alpha}\langle q_0|u_\alpha\rangle\langle v_\alpha|q_\mu\rangle, \quad (36)$$

where we used the identity

$$e^{-\mu\hat{H}_1} = \sum_\alpha |u_\alpha\rangle e^{-\mu\eta_\alpha}\langle v_\alpha|. \quad (37)$$

Then, substituting Eqs. (30) and (36) into Eq. (26) and integrating over  $q_0$  and  $q_\mu$  we obtain

$$Z_c = \sum_{\alpha,\beta} e^{-\mu(\eta_\alpha-\varepsilon_\beta)} e^{-\lambda\varepsilon_\beta}\langle\psi_\beta|u_\alpha\rangle^2. \quad (38)$$

The main contribution in the thermodynamic limit  $\lambda \rightarrow \infty$  is given by the lowest value  $\varepsilon_0$  and  $Z_c$  reduces to

$$Z_c \rightarrow e^{-\lambda\varepsilon_0} \sum_\alpha e^{-\mu(\eta_\alpha-\varepsilon_0)}\langle\psi_0|u_\alpha\rangle^2. \quad (39)$$

Substituting  $\mu=2T^{1/3}l$  into Eq. (39), we find from Eq. (24) the correlation function

$$\langle\vec{s}(l) \cdot \vec{s}(0)\rangle = \Re \sum_\alpha \langle\psi_0|u_\alpha\rangle^2 e^{-2T^{1/3}(\eta_\alpha-\varepsilon_0)l}. \quad (40)$$

The correlation length is governed by the lowest eigenvalue  $\eta_0$  and equals to

$$l_c = \frac{1}{2T^{1/3}(\eta_0 - \varepsilon_0)} = 2.2T^{-1/3}. \quad (41)$$

So, the low-temperature behavior of the correlation length is different from the HF model, where  $l_c \sim T^{-1}$ .

Now, the structure factor can be also calculated

$$\begin{aligned} S(k) &= 2\Re \int_0^\infty dl e^{ikl} \langle\vec{s}(l) \cdot \vec{s}(0)\rangle \\ &= 2 \sum_\alpha \langle\psi_0|u_\alpha\rangle^2 \frac{2T^{1/3}(\eta_\alpha - \varepsilon_0)}{4T^{2/3}(\eta_\alpha - \varepsilon_0)^2 + k^2}. \end{aligned} \quad (42)$$

In the low-temperature limit, the expansion of the structure factor for any  $k \gg T^{1/3}$  has the form

$$\begin{aligned} S(k) &= \frac{4T^{1/3}}{k^2} \sum_\alpha (\eta_\alpha - \varepsilon_0) \langle\psi_0|u_\alpha\rangle^2 \\ &\quad - \frac{16T}{k^4} \sum_\alpha \langle\psi_0|u_\alpha\rangle^2 (\eta_\alpha - \varepsilon_0)^3 + \dots \end{aligned} \quad (43)$$

The first term in Eq. (43) is zero because

$$\sum_\alpha (\eta_\alpha - \varepsilon_0) \langle\psi_0|u_\alpha\rangle^2 = \langle\psi_0|\hat{H}_1 - \hat{H}_0|\psi_0\rangle = -i\langle\psi_0|q|\psi_0\rangle = 0 \quad (44)$$

and  $|\psi_0\rangle$  is even function of  $q$ .

Therefore, the structure factor is given by the second term in Eq. (43), which can be calculated exactly

$$\begin{aligned} \sum_\alpha (\eta_\alpha - \varepsilon_0)^3 \langle\psi_0|u_\alpha\rangle^2 &= \langle\psi_0|(\hat{H}_1 - \hat{H}_0)^3 + \frac{1}{2}[(\hat{H}_0, \hat{H}_1), \hat{H}_1]|\psi_0\rangle \\ &= -\frac{1}{4}. \end{aligned} \quad (45)$$

Therefore, the low-temperature asymptotic of the structure factor is

$$S(k) = \frac{4T}{k^4}. \quad (46)$$

Hence, the susceptibility  $\chi(k)$  for  $k \gg T^{1/3}$  remains finite in the low-temperature limit,

$$\chi(k) = \frac{S(k)}{2T} = \frac{2}{k^4}. \quad (47)$$

The fact that  $\chi(k) \sim k^{-4}$  (instead of  $k^{-2}$  for HF chain) is a consequence of the fact that the excitation spectrum becomes  $\sim k^4$  at the transition point.

For  $k=0$  the structure factor [Eq. (42)] diverges at  $T \rightarrow 0$  as

$$S(0) = T^{-1/3} \sum_\alpha \frac{\langle\psi_0|u_\alpha\rangle^2}{\eta_\alpha - \varepsilon_0}. \quad (48)$$

The sum in Eq. (48) is calculated numerically and gives the factor  $\approx 5.36$ . Therefore, the magnetic susceptibility behaves as  $T^{-4/3}$ ,

$$\chi(0) = \frac{S(0)}{2T} = \frac{2.68}{T^{4/3}}. \quad (49)$$

In conclusion of this section we emphasize that the exact calculation of the correlation function for the planar spin model demonstrates that the critical exponents at the transition point of the F-AF chain can differ from that for the HF chain.

### III. CLASSICAL SPIN MODEL AT THE TRANSITION POINT

#### A. Partition function

The calculation of the correlation functions for the classical three-component spin model (2) is to a large extent similar to the planar spin case and to avoid duplications we will often refer to the previous section. So, in this section we consider the continuous model described by energy functional [Eq. (4)] where three-component vector field  $\vec{s}(\xi)$  has the constraint  $\vec{s}^2(\xi)=1$ .

Since in the partition function the integration occurs over all possible spin configurations, we are free to choose any local coordinate system. It is convenient to choose it so that the  $Z$  axis at the point  $\xi$  is directed along the spin vector  $\vec{s}(\xi)$  so that the spin vector  $\vec{s}(\xi)=(0,0,1)$ .

Let us introduce a new vector field

$$\vec{q}(\xi) = \frac{d\vec{s}}{d\xi} = (q_x, q_y, q_z). \quad (50)$$

The constraint  $\vec{s}^2(\xi)=1$  converts to the relations for  $\vec{q}(\xi)$ ,

$$q_z = 0,$$

$$q'_z = -q_x^2 - q_y^2, \quad (51)$$

where the prime denotes the space derivatives  $d/d\xi$ .

Then, the Hamilton function in Eq. (7) transforms to

$$\left(\frac{d^2\vec{s}}{d\xi^2}\right)^2 = \left(\frac{d\vec{q}}{d\xi}\right)^2 = q_x'^2 + q_y'^2 + (q_x^2 + q_y^2)^2. \quad (52)$$

Here we see that the constraint  $\vec{s}^2=1$  effectively eliminates the  $Z$  component of  $\vec{q}$  from the Hamilton function. Therefore, henceforth we deal with the  $q_x$  and  $q_y$  components of the vector field  $\vec{q}$  only, and we denote a two-component vector field by  $\mathbf{q}(\xi)=(q_x, q_y)$ .

The partition function in terms of  $\mathbf{q}(\xi)$  takes the form

$$Z = \int D\mathbf{q} \exp\left\{-\int_0^\lambda d\xi(\mathbf{q}'^2 + \mathbf{q}^4)\right\}. \quad (53)$$

Similar to the planar spin case, we treat the partition function as path integral [Eq. (15)] for the quantum mechanics of a single particle with the Hamiltonian

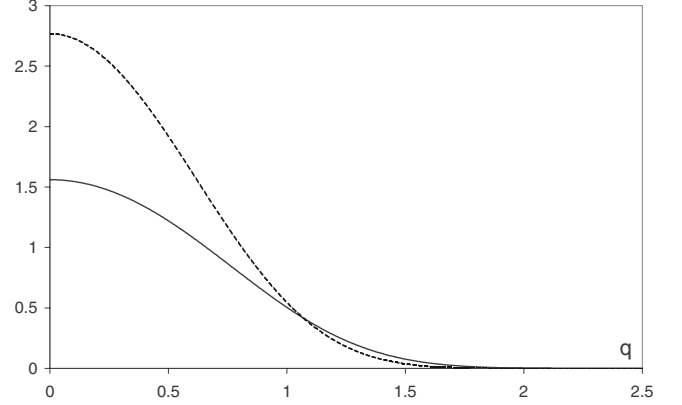


FIG. 3. Wave functions  $\psi(q)$  (solid line) for the lowest level  $\varepsilon_0=0.9305$  and  $u_3(q)$  (dashed line) for the level  $\eta_0=1.4113$ .

$$\hat{H}_0 = -\frac{1}{4}\Delta + \mathbf{q}^4, \quad (54)$$

where  $\Delta = \partial_x^2 + \partial_y^2$  is two-dimensional (2D) Laplace operator. Hamiltonian (54) commutes with the  $z$  component of the angular momentum  $\hat{l}_z$  and eigenstates  $\psi(\mathbf{q})$  of the corresponding Schrodinger equation

$$\hat{H}_0\psi = \varepsilon\psi \quad (55)$$

are divided to subspaces of azimuthal quantum numbers  $l_z=0, \pm 1, \pm 2, \dots$

Thus, the wave function  $\psi_{l_z}(\mathbf{q})$  describes a particle with the azimuthal quantum number  $l_z$  in 2D axially symmetrical potential well  $U(q)=q^4$ . Numerical solution of Eq. (55) gives the lowest levels for  $l_z=0, \pm 1, \pm 2$ ,

$$\varepsilon_\alpha(l_z=0) = 0.9305; 3.78; 7.44\dots$$

$$\varepsilon_\alpha(l_z = \pm 1) = 2.14; 5.48; 9.44\dots$$

$$\varepsilon_\alpha(l_z = \pm 2) = 3.54; 7.27; 11.5\dots \quad (56)$$

The wave function  $\psi_0(q)$  for the lowest eigenvalue  $\varepsilon_0=0.9305$  is shown in Fig. 3.

Now, by the analogy with the planar spin case, Eqs. (20) and (21), we obtain the partition function as a sum of exponents over all quantum numbers  $\alpha$  and  $l_z$ ,

$$Z = \sum_{\alpha, l_z} e^{-\lambda \varepsilon_{\alpha, l_z}}. \quad (57)$$

In the thermodynamic limit  $\lambda \rightarrow \infty$  only the lowest level  $\varepsilon_0=0.9305$  survives and the partition function is

$$Z = e^{-\lambda \varepsilon_0}. \quad (58)$$

#### B. Correlation function

Since we work with the local coordinate system directed so that the spin vector is directed along the  $Z$  axis, in order to find the scalar product of spin vectors  $\vec{s}(l) \cdot \vec{s}(0)$  we need to express the vector  $\vec{s}(0)$  in the local coordinate system located

at the point  $x=l$ . This can be represented as a chain of successive rotations describing the trajectory  $\vec{s}(x)$  (see the Appendix),

$$\vec{s}_{x=l}(0) = \exp\left(i \int_0^l \Omega dx\right) \vec{s}_{x=0}(0), \quad (59)$$

where  $\Omega = [\vec{\sigma} \cdot \vec{\omega}(x)]$  can be expressed through the vector  $\vec{q}(x)$  as

$$\Omega = \frac{(\vec{\sigma} \cdot [\vec{q} \times \dot{\vec{q}}])}{q^2} = \sigma_y q_x - \sigma_x q_y + \sigma_z \frac{q_x q'_y - q_y q'_x}{q_x^2 + q_y^2}. \quad (60)$$

In this equation we used relations (51) for the  $Z$  component of  $\vec{q}$  and  $\dot{\vec{q}}$ .

Then, taking into account that the spin vectors are directed along the  $Z$  axis of local coordinate system  $\vec{s}_{x=0}(0) = \vec{s}_{x=l}(l) = (0, 0, 1)$ , the scalar product of spin vectors  $\vec{s}(l) \cdot \vec{s}(0)$  becomes

$$\vec{s}(l) \cdot \vec{s}(0) = \begin{pmatrix} 0 & 0 & 1 \end{pmatrix} \exp\left[i \int_0^l \Omega(\mathbf{q}, \mathbf{q}') dx\right] \begin{pmatrix} 0 \\ 0 \\ 1 \end{pmatrix}. \quad (61)$$

Similar to the planar case, the correlation function can be written as a ratio of two path integrals [Eq. (24)], where the denominator  $Z$  is given by Eq. (58) and the numerator represents the following path integral:

$$\int D\mathbf{q} \exp\left\{-\int_0^\lambda d\xi(\mathbf{q}'^2 + \mathbf{q}^4) + i \int_0^\mu \Omega(\mathbf{q}, \mathbf{q}') d\xi\right\}. \quad (62)$$

Then, repeating the arguments presented in Eqs. (25)–(30), we arrive at the problem of the calculation of the propagator,

$$\int_{q_0}^{q_\mu} D\mathbf{q} \exp\left\{i \int_0^{-i\mu} dt L_1(\dot{\mathbf{q}}, \mathbf{q})\right\} = \langle q_0 | e^{-\mu \hat{H}_1} | q_\mu \rangle, \quad (63)$$

where the Lagrangian and the corresponding quantum Hamiltonian are

$$L_1 = \dot{\mathbf{q}}^2 - \mathbf{q}^4 - i\sigma_y q_x + i\sigma_x q_y - \sigma_z \frac{q_x \dot{q}_y - q_y \dot{q}_x}{q_x^2 + q_y^2}, \quad (64)$$

$$\hat{H}_1 = -\frac{1}{4}\Delta + \mathbf{q}^4 + \frac{\sigma_z^2 - 2\sigma_z \hat{l}_z}{4\mathbf{q}^2} + i\sigma_y q_x - i\sigma_x q_y. \quad (65)$$

Substituting  $\sigma_\alpha$  from Eq. (A5), the Hamiltonian takes the matrix form

$$\hat{H}_1 = \begin{pmatrix} \hat{H}_0 + \frac{1}{4\mathbf{q}^2} & \frac{1}{2\mathbf{q}^2} i \hat{l}_z & q_x \\ -\frac{1}{2\mathbf{q}^2} i \hat{l}_z & \hat{H}_0 + \frac{1}{4\mathbf{q}^2} & q_y \\ -q_x & -q_y & \hat{H}_0 \end{pmatrix}, \quad (66)$$

where  $\hat{H}_0$  is defined by Eq. (54).

Operator  $\hat{H}_1$  is non-Hermitian and the exponent of  $\hat{H}_1$  can be represented as

$$e^{-\mu \hat{H}_1} = \sum_\alpha |\vec{u}_\alpha\rangle e^{-\mu \eta_\alpha} \langle \vec{v}_\alpha|, \quad (67)$$

where three-component eigenvectors  $\vec{u}(q_x, q_y) = (u_1, u_2, u_3)$  and  $\vec{v}(q_x, q_y) = (v_1, v_2, v_3)$  satisfy the corresponding eigenvalue equations

$$\begin{aligned} \hat{H}_1 \vec{u} &= \eta \vec{u}, \\ \hat{H}_1^\dagger \vec{v} &= \eta^* \vec{v}. \end{aligned} \quad (68)$$

The normalization conditions are

$$\langle \vec{v}_\alpha | \vec{u}_\beta \rangle = \langle \vec{u}_\alpha | \vec{v}_\beta \rangle = \delta_{\alpha, \beta}. \quad (69)$$

Making the same procedure for non-Hermitian operators as for the planar spin case we obtain the correlation function in a form

$$\langle \vec{s}(l) \cdot \vec{s}(0) \rangle = \Re \sum_\alpha \langle \psi_0 | u_{3, \alpha} \rangle \langle v_{3, \alpha} | \psi_0 \rangle e^{-\mu(\eta_\alpha - \varepsilon_0)}. \quad (70)$$

Only the eigenfunctions  $|u_{3, \alpha}\rangle$  and  $|v_{3, \alpha}\rangle$  are present in the above equation, because according to Eq. (61) we need only the element (3,3) of the resultant matrix.

Since the wave function  $\psi_0$  has zero angular momentum, then only the sector  $l_z=0$  of Eq. (68) gives the contribution to the correlation function. In this sector the wave functions depend only on  $q=|\mathbf{q}|$ , the Hamiltonian is simplified so that we have to solve a pair (instead of three) differential equations for  $u_3(q)$  and  $\phi(q) = (q_x u_1 + q_y u_2)/q$ ,

$$\begin{aligned} -\frac{1}{4} \frac{d^2 \phi}{dq^2} - \frac{1}{4q} \frac{d\phi}{dq} + \frac{1}{4q^2} \phi + q^4 \phi + qu_3 &= \eta \phi, \\ -\frac{1}{4} \frac{d^2 u_3}{dq^2} - \frac{1}{4q} \frac{du_3}{dq} + q^4 u_3 - q\phi &= \eta u_3. \end{aligned} \quad (71)$$

The conjugate eigenvalue problem for  $\vec{v}$  in Eq. (68) transforms to exactly the same differential equations [Eq. (71)] for the functions  $v_3^*(q)$  and  $\chi^*(q) = -(q_x v_1^* + q_y v_2^*)/q$ . Therefore, the function  $v_3(q)$  is found from the solution of Eq. (71) by the relation  $v_3(q) = u_3^*(q)$  and the normalization conditions [Eq. (69)] transform to

$$\langle u_{3, \alpha}^* | u_{3, \beta} \rangle - \langle \phi_\alpha^* | \phi_\beta \rangle = \delta_{\alpha, \beta}. \quad (72)$$

One can see that Eq. (71) describes a two-level system in an axially symmetric potential well  $U(q) = q^4$ , where two levels with angular momenta  $l_z=0$  and  $l_z=1$  are coupled by non-Hermitian transition operator. The spectrum of this system of equations turns out to be real and positive as for the planar case and a few lowest levels are

$$\eta_\alpha = 1.4113; 1.83; 3.98 \dots \quad (73)$$

The reality of the spectrum  $\eta_\alpha$  has an important consequence: the correlation function [Eq. (70)] decays on large distances without oscillations.

The wave function  $u_3(q)$  for the lowest level  $\eta_0 = 1.4113$  is shown in Fig. 3. As follows from Fig. 3 the behavior of the

functions  $\psi_0(q)$  and  $u_3(q)$  are similar. Therefore, the main contribution to the correlation function, the structure factor, and the susceptibility is given by the level  $\eta_0$ .

The correlation function and the structure factor are given by Eqs. (40) and (42) with the substitution  $u_{3,\alpha}$  for  $u_\alpha$  and eigenvalues presented in Eqs. (56) and (73). Therefore, the correlation length defined by the lowest eigenvalue  $\eta_0$  behaves similar to the planar spin case  $\sim T^{-1/3}$  but the numerical factor is different,

$$l_c = 1.04T^{-1/3}. \quad (74)$$

The low-temperature asymptotic of static structure factor  $S(k)$  and the susceptibility  $\chi(k)$  for  $k \gg T^{1/3}$  is calculated in a similar way as for the planar spin case [see Eq. (43)] resulting in

$$\begin{aligned} S(k) &= \frac{8T}{k^4}, \\ \chi(k) &= \frac{8}{3k^4}. \end{aligned} \quad (75)$$

For  $k=0$  the structure factor  $S(0)$  is defined by Eq. (48) with the corresponding eigenfunctions and eigenvalues. The numerical calculation of the sum gives

$$S(0) = T^{-1/3} \sum_{\alpha} \frac{\langle \psi_0 | u_{3,\alpha} \rangle^2}{\eta_{\alpha} - \varepsilon_0} = \frac{3.21}{T^{1/3}}. \quad (76)$$

Thus, we have arrived at the final result for the magnetic susceptibility of the classical spin model at the transition point,

$$\chi(0) = \frac{S(0)}{3T} = \frac{1.07}{T^{4/3}}. \quad (77)$$

We see that the planar spin model gives correct critical exponent for the magnetic susceptibility.

#### IV. LOW-TEMPERATURE SUSCEPTIBILITY IN THE HELICAL PHASE

In the preceding sections we have considered the low-temperature thermodynamics at the transition point  $\alpha=1/4$ . Likewise it is possible to extend the developed method for the study of the vicinity of the transition point. In this case an additional term  $2(\alpha-1/4)\tilde{s}'^2$  appears in energy functional [Eq. (4)], which after rescaling of spatial variable [Eq. (6)] forms the scaling variable

$$\gamma = \frac{\alpha - 1/4}{T^{2/3}}. \quad (78)$$

Especially interesting is to study the vicinity of the transition point when values  $(\alpha-1/4)$  and  $T$  are small but the parameter  $\gamma$  is fixed. All the steps in derivation of the expressions for the correlation function are exactly the same as was done for the transition point and only the form of the potential well in the corresponding differential equations [Eq. (71)] is modified:  $U(q) = q^4 - 4\gamma q^2$ . Numerical solution of the

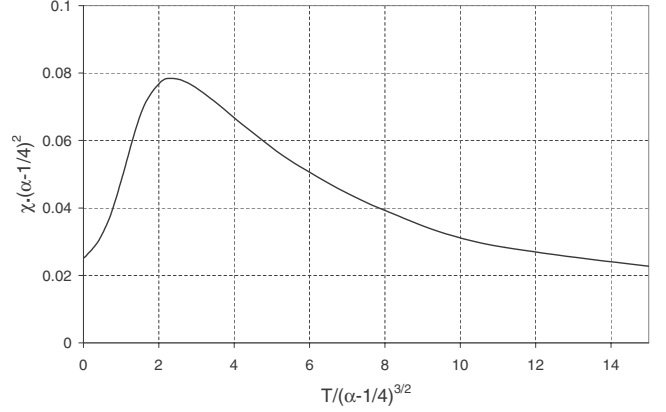


FIG. 4. Normalized susceptibility  $\chi(\alpha-1/4)^2$  as a function of normalized temperature  $T/(\alpha-1/4)^{3/2}$ .

corresponding differential equations allows to find the correlation function, the static structure factor  $S(k)$ , and the susceptibility as a function of  $\gamma$ . As follows from Eqs. (76) and (77) the susceptibility can be rewritten as

$$\chi = \frac{f(\gamma)}{T^{4/3}} = \frac{\gamma^2 f(\gamma)}{(\alpha-1/4)^2}. \quad (79)$$

Thus, the normalized susceptibility  $\tilde{\chi} = \chi(\alpha-1/4)^2$  is a universal function of  $\gamma$ .

On the ferromagnetic side of the transition point ( $\alpha < 1/4$ ) the susceptibility diverges at  $T \rightarrow 0$ , but the exponent changes from  $4/3$  to  $2$ , so that the susceptibility becomes  $\chi \sim (1/4 - \alpha)/T^2$ .

The behavior of the susceptibility in the helical phase is more interesting. The dependence of the uniform susceptibility  $\tilde{\chi}$  on the normalized temperature  $x = \gamma^{-3/2} = T/(\alpha-1/4)^{3/2}$  in the helical phase is shown in Fig. 4. The characteristic features of this dependence are the maximum of  $\tilde{\chi}$  at  $x = x_m$  and the finite value of  $\tilde{\chi}$  at  $T \rightarrow 0$ . The latter fact is a classical effect and can be destroyed by quantum fluctuations. In the quantum  $s=1/2$  F-AF model the ground state is believed to be gapped (though the gap can be extremely small<sup>9</sup>) and so  $\chi \rightarrow 0$  at  $T \rightarrow 0$ . In real materials interchain interactions cause the three-dimensional spiral LRO and the susceptibility can remain finite at  $T=0$ .

The obtained dependence  $\chi(T)$  is in a qualitative agreement with those observed in the edge-shared compounds with  $\alpha$  close to  $1/4$  [ $\text{Li}_2\text{CuO}_2$ ,<sup>2</sup>  $\text{Rb}_2\text{Cu}_2\text{Mo}_3\text{O}_{12}$ ,<sup>4</sup> and  $\text{Li}_2\text{CuZrO}_4$  (Ref. 17)]. As follows from Eq. (79) the location of maximum of  $\chi(T)$  is at  $T_m \sim (\alpha-1/4)^{3/2}$  and  $\chi_m \sim (\alpha-1/4)^{-2}$ , i.e., with the increase in  $\alpha$  the maximum of  $\chi$  shifts to higher temperatures and the magnitude of the maximum  $\chi_m$  decreases. These dependencies of  $T_m$  and  $\chi_m$  on the frustration parameter  $\alpha$  are also in accord with the experimental observations.<sup>17</sup>

The dependence  $\tilde{\chi}(x)$  agrees also with the numerical results for the uniform susceptibility obtained by transfer matrix renormalization group (TMRG) and exact diagonalization methods for the quantum F-AF chain with  $s=1/2$ .<sup>12,17</sup> The dependence  $T_m(\alpha)$  is similar to that obtained by the TMRG calculations in Ref. 12. Thus, the classical model

catches the physics of quantum spin systems in the helical phase and, therefore, the developed method for the classical spin model represents a useful tool for the investigation of the low-temperature behavior of the quantum systems.

**V. SUMMARY**

We have obtained the exact results for the low-temperature thermodynamics of the classical F-AF model at the frustration parameter  $\alpha=1/4$ , where the ground-state phase transition from the ferromagnetic to the helical phase occurs. The main result relates to the behavior of the zero-field susceptibility  $\chi$  and the correlation length  $l_c$ . It is shown that the critical exponents of  $\chi$  and  $l_c$  are changed from 2 to 4/3 and from 1 to 1/3 correspondingly, when  $\alpha \rightarrow 1/4$  from the ferromagnetic side. We note that the critical exponents are the same both for the classical spin model and for the planar model. In the present paper we have considered a continuum version of model (2). However, the low-temperature asymptotes of  $\chi$  and  $l_c$  of the continuum and lattice models coincide. In fact, lattice model (2) can be studied on a base of the transfer-matrix method adapted in Ref. 25 to include the NNN interaction. We have shown (Ref. 26) that the corresponding transfer-integral equations are reduced at  $T \rightarrow 0$  to the differential equations [Eqs. (55) and (71)] with the same eigenfunctions and eigenvalues so that the exact low-temperature asymptotic of the susceptibility coincides with Eq. (77). Besides, the structure factor  $S(k)$  for the lattice model is given by Eq. (42) under substitution of  $2(1 - \cos k)$  for  $k^2$  in the dominator of Eq. (42). At the same time, calculations in a frame of path-integral method are essentially simpler and clear in comparison with those in the transfer-integral approach.

It is interesting to compare the exact expression for the susceptibility at the transition point with the results found by approximate methods. One of this methods is the modified spin-wave theory (MSWT) proposed by Takahashi<sup>27</sup> to extend the spin-wave theory to the low-dimensional spin systems without LRO. Another approximate approach is the expansion of the thermodynamics of the  $n$ -vector classical model in powers in  $1/n$  (Refs. 28 and 29) (usually, up to the first order). Remarkably, both methods give the true critical exponent 4/3 for the susceptibility  $\chi = cT^{-4/3}$ . However, the numerical coefficient  $c$  differs from the exact one. For example, the MSWT result is  $c=1.19$ . The  $1/n$  expansion for the classical spin model (2) gives  $c=0.560$  in the zeroth order and  $c=0.897$  in the first order in  $1/n$ . The comparison of these values of  $c$  with the exact one shows that these approximate methods give a satisfactory agreement (within 10–20 %) with the exact coefficient. It is worth to note that the MSWT gives the exact low-temperature asymptotic of  $\chi = 2/3T^2$  for the classical ferromagnetic chain ( $\alpha=0$ ).<sup>21</sup> Moreover, the MSWT gives the exact result for  $\chi$  at  $T \rightarrow 0$  for the quantum ferromagnetic chain with  $s=1/2$  as well. As was noted in Sec. I, the quantum and the classical ferromagnetic chains have universal low-temperature properties. The low-temperature susceptibility of the ferromagnetic chain is described by the scaling function universal for any value of spin  $s$ . For the F-AF model at  $\alpha=1/4$  there is no rigorous

proof of such universality, though MSWT confirms this hypothesis. If this universality holds in the case  $\alpha=1/4$  then we expect that the quantum F-AF chain has the same critical exponent of  $\chi$  as in the classical model.

We have also considered the low-temperature thermodynamics in the vicinity of the transition point. In this case the properties of the system are governed by the scaling parameter  $\gamma = (\alpha - 1/4)/T^{2/3}$ . On the ferromagnetic side of the transition point ( $\alpha < 1/4$ ) the susceptibility transforms smoothly to  $\chi \sim (1/4 - \alpha)/T^2$  at  $\gamma \rightarrow -\infty$ , which describes the change in the exponent at  $T \rightarrow 0$ . The susceptibility in the helical phase for a fixed value of  $\alpha \geq 1/4$  behaves as  $\chi \sim (\alpha - 1/4)^{-2}$  at  $\gamma \rightarrow \infty$ , which means that the uniform susceptibility remains finite at  $T \rightarrow 0$ . Besides, we found that the susceptibility in the helical phase has a maximum at some temperature  $T_m \sim (\alpha - 1/4)^{-3/2}$ . The presence of maximum of the dependence  $\chi(T)$  as well as the location and the magnitude of this maximum as a function of the deviation from the transition point ( $\alpha - 1/4$ ) are in agreement with that observed in several materials described by the quantum  $s=1/2$  version of this model and with the numerical results for the  $s=1/2$  model.

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**APPENDIX**

Let us consider a particle moving along a trajectory  $\vec{r}(t)$ . At any moment the particle motion can be represented as an instant rotation around some instantaneous axis of rotation with definite angular velocity. The radius of the instant rotation  $\rho$  is expressed through the centripetal part  $\ddot{\vec{r}}_c$  of the acceleration vector  $\ddot{\vec{r}}$ ,

$$\rho = \frac{\dot{\vec{r}}^2}{|\ddot{\vec{r}}_c|}. \tag{A1}$$

The angular velocity is

$$\omega = \frac{|\dot{\vec{r}}|}{\rho} = \frac{|\dot{\vec{r}} \times \ddot{\vec{r}}|}{\dot{\vec{r}}^2}, \tag{A2}$$

where we took into account that  $(\ddot{\vec{r}}_c \cdot \dot{\vec{r}}) = 0$ .

Since the rotation takes place in the  $(\dot{\vec{r}}, \ddot{\vec{r}})$  plane, the instantaneous axis of rotation is directed along  $[\dot{\vec{r}} \times \ddot{\vec{r}}]$ . Therefore, if we define the “local” coordinate system associated with the moving particle so that the instant coordinate axes are directed along the vectors  $\dot{\vec{r}}$ ,  $\ddot{\vec{r}}_c$ , and  $[\dot{\vec{r}} \times \ddot{\vec{r}}]$ , then the instant change in the local coordinate system is expressed by the rotation matrix

$$R = \exp[i(\vec{\sigma} \cdot \vec{\omega})], \tag{A3}$$

$$\vec{\omega} = \frac{[\dot{\vec{r}} \times \ddot{\vec{r}}]}{\dot{\vec{r}}^2}, \tag{A4}$$

where



$$\sigma_x = \begin{bmatrix} 0 & 0 & 0 \\ 0 & 0 & i \\ 0 & -i & 0 \end{bmatrix}, \quad \sigma_y = \begin{bmatrix} 0 & 0 & -i \\ 0 & 0 & 0 \\ i & 0 & 0 \end{bmatrix}, \quad \sigma_z = \begin{bmatrix} 0 & i & 0 \\ -i & 0 & 0 \\ 0 & 0 & 0 \end{bmatrix}. \quad (\text{A5})$$

This implies that if a given fixed vector  $\vec{s}$  has components  $\vec{s}_{t_0}$  in the local coordinates corresponding to time  $t_0$ , then the components of this vector in the local coordinates corre-

sponding to time  $t_1$  can be represented as a chain of successive rotations,

$$\vec{s}_{t_1} = \exp \left\{ i \int_{t_0}^{t_1} [\vec{\sigma} \cdot \vec{\omega}(t)] dt \right\} \vec{s}_{t_0}. \quad (\text{A6})$$

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