Internal oscillations of solitons in $(CH_3)_4NMnCl_3$ above and below T_N

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The problem of internal soliton oscillations or soliton-magnon bound states occurring in the quasi-one-dimensional antiferromagnet $(CH_3)_4NMnCL_3$ (TMMC) is studied by approximated analytical methods. Above T_N, besides the well-known Goldstone mode, we found a second bound state which is due to the coupling between in-plane and out-of-plane components of the spins when going beyond the sine-Gordon limit. For typical experimental conditions the frequency of this second bound state is very close to the bottom of the magnon band. Below T_N using an interchain mean-field approach we also found two bound states: the Goldstone mode and a second state which originates from the pairing of π kinks described by a double-sine-Gordon equation. Out-of-plane effects for this mode are also included by means of perturbation methods. In contrast to the situation above T_N the frequency of this mode can be located in the whole gap between zero and the continuum. Both above and below T_N the polarization of these second bound-state modes has the same orientation, which differs significantly from that of usual magnon modes.

I. INTRODUCTION

During the last 15 years there has been great theoretical and experimental interest in solitons in magnetic systems.[1,2] A thermal gas of these excitations has been observed in several quasi-one-dimensional magnets by means of neutron-scattering experiments,[3] nuclear magnetic resonance[4,5] (NMR), electron spin resonance[5] (ESR), and in other experiments.[6] Corresponding theoretical models usually consider systems of weakly coupled classical spins[1,2] and are based on the Landau-Lifshitz equation[7] or its approximations such as sine-Gordon[8] or double-sine-Gordon (DSG) equations.[9,10] Although, the sine-Gordon equation and several special cases of the Landau-Lifshitz equation belong to completely integrable systems in the sense of the inverse scattering method,[11] there exists also a class of magnetic models leading to equations which are not completely integrable.[9,10] The remarkable feature of these models is the presence of non-Goldstone soliton-magnon bound states[12,13] that have never been observed in completely integrable systems. These bound states can be interpreted as internal oscillations of solitons and the frequency $\Omega_{(b)}$ of these oscillations is in the range between zero and the frequency of the uniform spin precession $\Omega_{(k=0)}$. The existence of such bound states can influence the thermodynamic properties of the collective soliton-magnon gas.[13]

In this work we consider the problem of bound states in the well-known soliton-bearing antiferromagnet $(CH_3)_4NMnCl_3$ (TMMC). This material has the remarkable property that a soliton gas occurs both *above*[3,4] *and below* its Néel temperature T_N.[10,14–16] We show that non-Goldstone bound states also exist above and below T_N, but their properties are completely different.

The paper is organized as follows. In Sec. II we introduce the Hamiltonian describing TMMC above and below T_N and present the corresponding equations of motion. Section III is devoted to the problem of the internal oscillations of π kinks existing in TMMC above T_N, while Sec. IV presents results on internal oscillations of 2π kinks existing below T_N.

II. THE MODEL

The quasi-one-dimensional antiferromagnet $(CH_3)_4NMnCl_3$ is well described by a Hamiltonian of weakly coupled antiferromagnetic spin chains[10,14,15]

$$H^{3D}=\sum_j H_j^{1D}-\tfrac{1}{2}J_\perp \sum_{j\neq j'}{}' \sum_m \mathbf{S}_{j,m}\cdot\mathbf{S}_{j',m} \tag{1}$$

with

$$H_j^{1D}=\sum_m [J_\parallel \mathbf{S}_{j,m}\cdot\mathbf{S}_{j,m+1}+A(S_{j,m}^Z)^2-D(S_{j,m}^X)^2 -g\mu_B B^X S_{j,m}^X-g\mu_B B^Y S_{j,m}^Y] , \tag{2}$$

where H_j^{1D} describes the Hamiltonian of a single chain labeled by $j=(j_1,j_2)$ and the spins $\mathbf{S}_{j,m}$ are classical vectors localized at lattice sites (j_1,j_2,m). For TMMC we have the following characteristic material parameters:[15] an antiferromagnetic intrachain exchange constant $J_\parallel/k_B=13.4$ K, a ferromagnetic interchain constant $J_\perp/k_B\simeq 0.9$ mK, a planar anisotropy $A/k_B=0.3$ K, an in-plane anisotropy $D/k_B=0.05$ K, the spin quantum number $S=\frac{5}{2}$, and the gyromagnetic ratio $g=2.01$. **B**

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denotes the external magnetic field which is applied perpendicular to the chain direction Z. We assume that temperature and material parameters fulfill the conditions $k_B T \ll g\mu_B BS \ll AS^2$ and $J_\perp S^2 \ll (g\mu_B B)^2/(4J_\parallel)$. Moreover, we have $D \ll A \ll J_\parallel$. Above the Néel temperature T_N interchain interactions are negligible, and the system can be described by the one-dimensional (1D) Hamiltonian (2). Below T_N, it seems reasonable[10,14] to describe the dynamics of the system in terms of an interchain mean-field approach,[16] and as a result, instead of the three-dimensional Hamiltonian (1), we obtain the effective one-dimensional Hamiltonian[14] with an interchain mean field $\mathbf{B}_{\perp_{\rm MF}}$

$$H_{\rm eff}^{\rm 1D} = \sum_m [J_\parallel \mathbf{S}_m \cdot \mathbf{S}_{m+1} + A(S_m^Z)^2 - D(S_m^X)^2 - g\mu_B B^X S_m^X - g\mu_B B^Y S_m^Y - g\mu_B B^X_{\perp_{\rm MF}}(-1)^m S_m^X - g\mu_B B^Y_{\perp_{\rm MF}}(-1)^m S_m^Y] \;. \quad (3)$$

Here we assumed saturation of the interchain mean field $|\mathbf{B}_{\perp_{\rm MF}}| = 6J_\perp S/(g\mu_B)$. Its direction depends on the amount and orientation of the external field $\mathbf{B}=(B^X,B^Y)$ with respect to the in-plane anisotropy D, i.e., the field $\mathbf{B}_{\perp_{\rm MF}}$ is directed along the easy axis of the effective anisotropy. Using the standard parametrization[17] for odd and even spins,

$$\left.\begin{matrix}\{\mathbf{S}_{2m}\} \\ \{\mathbf{S}_{2m+1}\}\end{matrix}\right\} = \pm S[\sin(\theta_m \pm \vartheta_m)\cos(\phi_m \pm \varphi_m) \,, \sin(\theta_m \pm \vartheta_m)\sin(\phi_m \pm \varphi_m) \,, \cos(\theta_m \pm \vartheta_m)] \;, \quad (4)$$

where θ_m and ϕ_m denote the mainly antiferromagnetic order of the two spin sublattices while ϑ_m and φ_m describe a slight canting of the staggered spins towards the direction of the magnetic field ($|\vartheta_m|, |\varphi_m| \ll 1$), we get four equations for the evolution of the angles $\vartheta_m, \varphi_m, \theta_m$, and ϕ_m. After performing the continuum limit, making use of the relations between the material parameters, and eliminating the small variables ϑ and φ, we obtain in leading order the following coupled equations for $\theta(z,t)$ and $\phi(z,t)$:

$$J_\parallel \theta_{zz} - \frac{\theta_{tt}}{4J_\parallel S^2} = -6J_\perp \cos\theta\cos(\phi-\gamma_\perp) - \frac{g\mu_B B}{2J_\parallel S^2}\sin^2\theta\cos(\phi-\gamma)\phi_t + \sin\theta\cos\theta\left[J_\parallel(\phi_z)^2 - \frac{(\phi_t)^2}{4J_\parallel S^2} - 2A + \frac{(g\mu_B B)^2}{4J_\parallel S^2}\cos^2(\phi-\gamma) - 2D\cos^2\phi\right] , \quad (5)$$

$$J_\parallel \phi_{zz} - \frac{\phi_{tt}}{4J_\parallel S^2} = \frac{g\mu_B B}{2J_\parallel S^2}\cos(\phi-\gamma)\theta_t + D\sin(2\phi) - 2\cot\theta\left[J_\parallel\theta_z\phi_z - \frac{\theta_t\phi_t}{4J_\parallel S^2}\right] + 6J_\perp\frac{\sin(\phi-\gamma_\perp)}{\sin\theta} - \frac{(g\mu_B B)^2}{8J_\parallel S^2}\sin(2\phi-2\gamma) \;. \quad (6)$$

Here, the distance z along the chain is measured in lattice constants, and the parameter γ ($\gamma_\perp$) describes the angle between the field $\mathbf{B}$ ($\mathbf{B}_{\perp_{\rm MF}}$) and the X axis.

III. SOLITON-MAGNON BOUND STATES ABOVE T_N

A. Eigenvalue problem

Above T_N the effect of interchain interactions can be neglected, i.e., we put $J_\perp = 0$, and the exact static solution of (5) and (6) is obtained in the form of a static π kink: $\theta_\pi = \pi/2$,

$$\sin[\phi_\pi(z) - \beta] = \tanh[g\mu_B B_{\rm eff} z/(2J_\parallel S)] \;,$$

which corresponds to a π rotation of spin vectors in the XY plane. Here, the parameter $B_{\rm eff}$ (>0) is the value of the "effective" magnetic field defined by

$$B_{\rm eff}^4 \equiv B^4 - [16S^2 DJ_\parallel B^2/(g\mu_B)^2]\cos(2\gamma) + 64S^4D^2J_\parallel^2/(g\mu_B)^4 \;,$$

while β denotes (see Fig. 1) the angle between the direction of this effective field in the XY plane and the X axis and is given by

$$\sin(2\beta) = (B/B_{\rm eff})^2 \sin(2\gamma) \;,$$

$$\cos(2\beta) = (B/B_{\rm eff})^2\cos(2\gamma) - 8S^2DJ_\parallel/(g\mu_B B_{\rm eff})^2 \;.$$

Linearizing (5) and (6) around the π kink, i.e., putting $\phi(z,t) = \phi_\pi(z) + \delta\phi(z,t)$ and $\theta(z,t) = \theta_\pi + \delta\theta(z,t)$ with

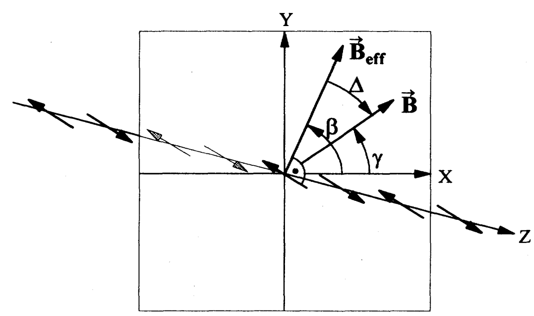

FIG. 1. Orientation of antiferromagnetic spin chain (Z), easy axis of anisotropy (X), and resulting effective field $\mathbf{B}_{\rm eff}$. Ground-state orientation of spins is perpendicular to $\mathbf{B}_{\rm eff}$.

$|\delta\phi|,|\delta\theta| \ll 1$ and assuming for $\delta\phi$ and $\delta\theta$ a harmonic time dependence with frequency Ω, we obtain the two-dimensional eigenvalue problem

$$(\delta\theta)_{zz}+\frac{\Omega^2}{4(J_\parallel S)^2}\delta\theta = -\frac{ig\mu_B B\Omega}{2(J_\parallel S)^2}(\operatorname{sech}\zeta\cos\Delta+\tanh\zeta\sin\Delta)\delta\phi -\delta\theta\left[f(z)-\frac{2A}{J_\parallel}\right], \quad (7)$$

$$(\delta\phi)_{zz}+\frac{\Omega^2}{4(J_\parallel S)^2}\delta\phi = -\frac{(g\mu_B B_{\text{eff}})^2}{4(J_\parallel S)^2}(2\operatorname{sech}^2\zeta-1)\delta\phi +\frac{ig\mu_B B\Omega}{2(J_\parallel S)^2}(\operatorname{sech}\zeta\cos\Delta+\tanh\zeta\sin\Delta)\delta\theta, \quad (8)$$

where $\zeta \equiv g\mu_B B_{\text{eff}} z/(2J_\parallel S)$, $\Delta\equiv\gamma-\beta$, and the function $f(z)$ reads

$$f(z)\equiv\left[\frac{(g\mu_B B)^2}{4(J_\parallel S)^2}\cos(2\Delta)-\frac{2D}{J_\parallel}\cos(2\beta)+\frac{(g\mu_B B_{\text{eff}})^2}{4(J_\parallel S)^2}\right]\operatorname{sech}^2\zeta +\left[\frac{(g\mu_B B)^2}{4(J_\parallel S)^2}\sin(2\Delta)+\frac{2D}{J_\parallel}\sin(2\beta)\right]\operatorname{sech}\zeta\tanh\zeta+\frac{(g\mu_B B)^2}{4(J_\parallel S)^2}\sin^2\Delta-\frac{2D}{J_\parallel}\sin^2\beta . \quad (9)$$

Usually[18] this type of equation is decoupled by setting either $\delta\theta$ or $\delta\phi$ equal to zero, i.e., by considering in-plane and out-of-plane oscillations separately. However, for the corresponding problem of a soliton-bearing ferromagnet[19] the coupling between in- and out-of-plane spin components essentially influences the eigenvalue spectrum. We consider the influence of this coupling on the occurrence of soliton-magnon bound states in terms of a perturbation method and focus on the properties of low-lying ($\Omega \le g\mu_B B_{\text{eff}}$) weakly dispersive eigenmodes of (7) and (8). Thus the leading order of (7) reads

$$\delta\theta(\zeta)\approx\frac{ig\mu_B B\Omega}{4AJ_\parallel S^2}\cos[\phi_\pi(\zeta)-\gamma]\delta\phi(\zeta) . \quad (10)$$

Inserting (10) into (8) we get

$$-(\delta\phi)_{\zeta\zeta}(\zeta)+\{1-[2+\alpha\omega^2\cos(2\Delta)]\operatorname{sech}^2\zeta+V_1(\zeta)\}\delta\phi(\zeta) =\omega^2(1+\alpha\sin^2\Delta)\delta\phi(\zeta) , \quad (11)$$

where we introduced the reduced frequency $\omega\equiv\Omega/(g\mu_B B_{\text{eff}})$, the perturbation potential

$$V_1(\zeta)\equiv-\alpha\omega^2\operatorname{sech}\zeta\tanh\zeta\sin(2\Delta) ,$$

and besides the angle Δ there is only one parameter $\alpha\equiv(g\mu_B B)^2/(2AJ_\parallel S^2)\ll 1$ which determines the properties of (11). First we solve (11) for $V_1\approx 0$ and then we will show that the influence of the "potential" V_1 is negligible. In this case the limit $\alpha\to 0$ corresponds to the eigenvalue problem appearing in the stability analysis of a sine-Gordon equation while for nonvanishing α the resulting equation is related to a modified Pöschel-Teller eigenvalue problem which can be solved exactly.[20] In fact, we find a continuum of delocalized (scattering) states $\delta\phi_{(k)}(\zeta)$ with frequencies starting from $\omega^2_{(k=0)}=1/(1+\alpha\sin^2\Delta)$, and a number of bound states with $\omega^2_{(b)}<1/(1+\alpha\sin^2\Delta)$.

B. Number of bound states

Since we have assumed that $\alpha\ll 1$, we get $\alpha\omega^2_{(b)}\ll 1$ and in this case the Pöschell-Teller potential has only one bound state for $\cos(2\Delta)<0$ and two bound states for $\cos(2\Delta)>0$. It is easy to check that the sign of $\cos(2\Delta)$ is equal to the sign of

$$(g\mu_B B)^2/(J_\parallel S^2)-8D\cos(2\gamma) .$$

Thus the number of bound states is determined by both the magnitude and the direction of the external field **B** (Fig. 2). The single bound state existing for $\cos(2\Delta)<0$ and the lower bound state occurring for $\cos(2\Delta)>0$ correspond with the Goldstone mode of a π-kink soliton, and their existence is related to the translational invariance of (5) and (6). Their frequency and shape are given by $\omega_{(1)}=0$ and by

$$\delta\phi_{(1)}(\zeta)=\tfrac{1}{2}\sqrt{g\mu_B B_{\text{eff}}/(J_\parallel S)}\operatorname{sech}\zeta .$$

C. Frequency of the second bound state

In order to find the second bound state existing for $\cos(2\Delta)>0$, it is convenient to introduce the parameter $\kappa=\alpha\cos(2\Delta)/(1+\alpha\sin^2\Delta)$. After some algebra the frequency and shape of this mode can be written as

$$\omega^2_{(2)}=\left\{1-\left[\frac{\sqrt{(2\kappa+1)^2+8}-3}{2\kappa+2}\right]^2\right\}\Big/(1+\alpha\sin^2\Delta) \approx[1-\tfrac{1}{9}\alpha^2\cos^2(2\Delta)]/(1+\alpha\sin^2\Delta) , \quad (12)$$

$$\delta\phi_{(2)}(\zeta)=\sqrt{K}\,\sinh\zeta(\cosh\zeta)^{-1-\epsilon} , \quad (13)$$

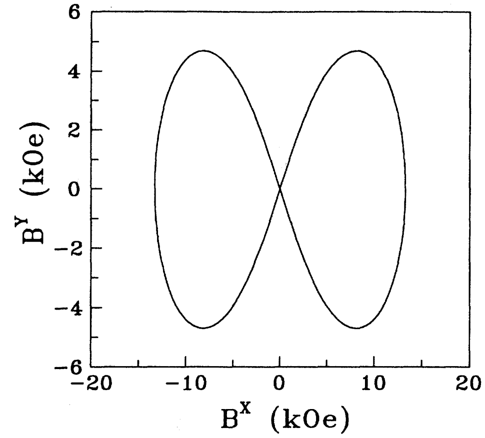

FIG. 2. Diagram of the numbers of bound states in TMMC above T_N. When the external magnetic field **B** is inside the ∞-like figure there is only one bound state, the Goldstone mode. Otherwise there are two bound states.

where

$$\epsilon \equiv \frac{\sqrt{(2\kappa+1)^2+8}-3}{2\kappa+2} \approx \frac{\alpha\cos(2\Delta)}{3} \tag{14}$$

and the constant

$$K \equiv [2^{2\epsilon-1}B(\epsilon,\epsilon) - 2^{2\epsilon+1}B(\epsilon+1,\epsilon+1)]^{-1} \times g\mu_B B_{\text{eff}}/(2J_{\parallel}S)$$

[$B(x,y)$ denoting the beta function] is to ensure the normalizing condition for $\delta\phi_{(2)}(\xi)$.

For estimating the influence of the perturbation $V_1(\xi)$ on the spectrum of (11) one should consider the fact that the resulting first-order corrections to the eigenvalues $\omega_{(1)}^2$ and $\omega_{(2)}^2$ are vanishing for symmetry reasons. On the other hand, since the perturbation term V_1 is proportional to ω^2, its off-diagonal matrix elements, which include the ground-state function $\delta\phi_{(1)}(\xi)$, also disappear. Thus the results (12) and (13) are exact up to second-order corrections in $V_1(\xi)$.

It is interesting to note that, similarly to the case of out-of-plane effects in the easy-plane ferromagnet,[19] the second bound state ω_2^2 emerges from the continuum threshold $\omega_{(k=0)}^2$. This means, however, that for typical experimental conditions in TMMC this state occurs very close to the broad uniform mode of spin precession $\omega_{(k=0)}$ and can hardly be detected. In fact, using the parameters for TMMC we obtain from (12) that the relative shift of the second bound-state frequency with respect to the bottom of the magnon band is given by

$$\frac{\omega_{(k=0)}-\omega_{(2)}}{\omega_{(k=0)}} \approx \tfrac{1}{18}\alpha^2\cos^2(2\Delta) \approx (0.8\times 10^{-8})\cos^2(2\Delta)\,(B/\text{kOe})^4 \,. \tag{15}$$

D. Polarization

The second quantity characterizing the bound state which can be probed in experiment is its polarization, i.e., the direction of the oscillating part of the magnetization $\langle \delta \mathbf{S} \rangle$ carried by such a mode. To calculate this value we started from the parametrization (4) and found that the mean values of components of magnetization for *any distribution* of angles θ, ϕ, ϑ, and φ can be written as

$$\langle S^X \rangle = S\langle \vartheta\cos\theta\cos\phi - \varphi\sin\theta\sin\phi \rangle \,, \tag{16a}$$

$$\langle S^Y \rangle = S\langle \vartheta\cos\theta\sin\phi + \varphi\sin\theta\cos\phi \rangle \,, \tag{16b}$$

$$\langle S^Z \rangle = -S\langle \vartheta\sin\theta \rangle \,. \tag{16c}$$

In order to calculate the change of net magnetization connected with bound states, we have linearized (16) around the static soliton solution. After some algebra we obtained that the mean values of all components of the magnetization vector $\langle \delta \mathbf{S} \rangle_{(1)}$ carried by the Goldstone mode $\delta\phi_{(1)}$ are equal to zero. This fact can be interpreted in the following way: the Goldstone mode $\delta\phi_{(1)}(z)$ is proportional to the spatial derivative of the soliton shape

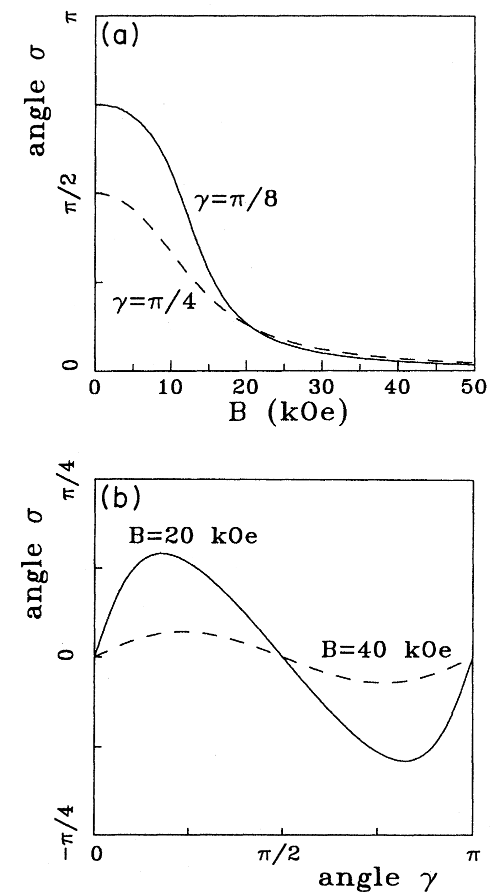

FIG. 3. Dependence of polarization angle σ of the bound state on magnitude and direction of the external magnetic field **B**.

$\phi_\pi(z)$, and the soliton essentially represents the propagation of the flipping of antiferromagnetic sublattices. It follows that spatial shift of the soliton along the spin chain should not induce any changes of total magnetization of the chain. On the other hand, we found that the components of mean magnetization $\langle \delta \mathbf{S} \rangle_{(2)}$ carried by the normalized second bound state $\delta\phi_{(2)}$ read

$$\langle \delta S^X \rangle_{(2)} = \frac{g\mu_B B}{2J_\parallel S}\cos(2\beta-\gamma)J_\pi \ , \tag{17a}$$

$$\langle \delta S^Y \rangle_{(2)} = \frac{g\mu_B B}{2J_\parallel S}\sin(2\beta-\gamma)J_\pi \ , \tag{17b}$$

$$\langle \delta S^Z \rangle_{(2)} = 0 \ , \tag{17c}$$

where the angles β and γ have been defined above and the constant J_π is given in Appendix A. Equations (17) imply that the polarization of the magnetization connected with the bound state $\delta\phi_{(2)}(z)$ is confined to the XY plane, making an angle $\sigma=2(\beta-\gamma)$ with the direction of the magnetic field $\mathbf{B}$. The dependence of this angle on the value of $\mathbf{B}$ is presented in Fig. 3.

IV. SOLITON-MAGNON BOUND STATES BELOW T_N

Below the Néel temperature T_N neighboring spin chains become correlated and the interchain mean field $\mathbf{B}_{\perp_{\rm MF}}$ has a nonzero value. The direction of this field is defined by the value of the angle $\gamma_\perp$ appearing in (5) and (6) and should be perpendicular to the effective field $\mathbf{B}_{\rm eff}$ resulting from the competition between the external magnetic field $\mathbf{B}$ and the in-plane anisotropy D. This way we get the relation $\gamma_\perp=\beta+\pi/2$. A static solution of (5) and (6) can be written in the form of a static 2π kink which is the solution of a double-sine-Gordon equation.[14] This 2π kink has the shape of a pair of coupled π kinks:[9] $\theta_{2\pi}(\xi)=\pi/2$,

$$\sin[\phi_{2\pi}(\xi)-\beta]=1-2/(1+\eta\sinh^2\xi) \ ,$$

where

$$\eta=[1+(g\mu_B B_{\rm eff})^2/(24J_\parallel J_\perp S^2)]^{-1}$$

and

$$\xi=[6J_\perp/J_\parallel+\tfrac{1}{4}(g\mu_B B_{\rm eff})^2/(J_\parallel S)^2]^{1/2}z \ .$$

Linearizing (5) and (6) around this solution in a similar way as for deriving (10) and (11), we obtain

$$\delta\theta(\xi)\approx\frac{ig\mu_B B\Omega}{4AJ_\parallel S^2}\cos[\phi_{2\pi}(\xi)-\gamma)]\delta\phi(\xi) \ , \tag{18a}$$

$$-(\delta\phi)_{\xi\xi}+\left\{\operatorname{sech}^2R\frac{\sinh^2\xi-\cosh^2R}{\sinh^2\xi+\cosh^2R}+\tanh^2R\left[2\left[\frac{\sinh^2\xi-\cosh^2R}{\sinh^2\xi+\cosh^2R}\right]^2-1\right]\right\}\delta\phi(\xi)=\hat{\omega}^2[1+\alpha V_2(\xi)]\delta\phi(\xi) \ , \tag{18b}$$

where the reduced frequency is given by

$$\hat{\omega}\equiv\Omega[(g\mu_B B_{\rm eff})^2+24J_\parallel J_\perp S^2]^{-1/2} \ ,$$

the parameter R defined by $\sinh R\equiv g\mu_B B_{\rm eff}/(2S\sqrt{6J_\parallel J_\perp})$ denotes half the distance between the centers of both π kinks forming the 2π kink $\phi_{2\pi}(\xi)$, and the perturbation potential $V_2(\xi)$ reads

$$V_2(\xi)\equiv\sin^2\Delta+4\cos(2\Delta)\left[\frac{\cosh R\,\sinh\xi}{\cosh^2R+\sinh^2\xi}\right]^2 -2\sin(2\Delta)\frac{\cosh R\,\sinh\xi(\cosh^2R-\sinh^2\xi)}{(\cosh^2R+\sinh^2\xi)^2} \ . \tag{19}$$

For $\alpha=0$ the eigenvalue problem (18b) is equivalent to the scattering problem between phonons and a 2π DSG kink, which has been considered in several papers.[21] It was shown that the spectrum consists of a continuum of scattering states and two bound states. The first one again has zero frequency and corresponds to the translational Goldstone mode of the system. As above T_N its shape is given by the spatial derivative of the 2π-kink solution $\phi_{2\pi}(\xi)$. The second bound state corresponds to an internal oscillation of the 2π kink, where the center of mass of the 2π kink does not move but the distance between the centers of both π kinks oscillates in time. The corresponding frequency ranges from zero up to the lower edge of the spin-wave continuum. The first limit corresponds to the situation where the interchain mean field disappears and the gas of 2π kinks dissociates into a gas of π kinks at the ordering temperature T_N.[14–16] The approximate expression for the shape of such a bound state, which is valid for intermediate and large distances ($R\geq 1$) between the centers of π-kink pairs, can be written using results of Ref. 21 as

$$\delta\phi_{(2)}(\xi)\simeq\frac{\operatorname{sech}(\xi-R)-\operatorname{sech}(\xi+R)}{2[1-R\operatorname{csch}(2R)]^{1/2}} \times[6J_\perp/J_\parallel+\tfrac{1}{4}(g\mu_B B_{\rm eff})^2/(J_\parallel S)^2]^{1/4} \ . \tag{20}$$

The influence of the potential $V_2(\xi)$ can be taken into account by means of a perturbation method. The resulting perturbed frequency $\hat{\omega}_{(2)}$ reads in the first-order approximation

$$\hat{\omega}_{(2)}^2 \simeq \frac{3\sinh^{-2}R - [\sinh(2R)+2R]\cosh^{-2}R/[\sinh(2R)-2R]}{1+\alpha[\cos(2\Delta)g(R)+\sin^2\Delta]}, \quad (21)$$

where the nominator of the above expression corresponds to the square of the unperturbed bound-state frequency of the DSG model[21] while the function $g(R)$ is defined in Appendix B and takes its maximal value $\frac{2}{3}$ in the limit $R \to \infty$. The frequency of long-wave magnons, on the other hand, can be written in a similar way as in the paramagnetic phase, i.e., $\hat{\omega}_{(k=0)}^2 = 1/(1+\alpha\sin^2\Delta)$. Below we present plots of the bound-state frequency (in physical units)

$$\Omega_{(2)} = \hat{\omega}_{(2)}[(g\mu_B B_{\text{eff}})^2 + 24J_{\parallel}J_{\perp}S^2]^{1/2}$$

and of the corresponding frequency of the long-wave magnons $\Omega_{(k=0)}$ as functions of the strength and direction of the magnetic field B (Fig. 4). One can easily see that, in contrast to the behavior of $\Omega_{(k=0)}$, the bound-state frequency $\Omega_{(2)}$ is nearly independent of B. The reason for this fact is a balance of two competing effects arising from the increase of B: (i) the increase of the Larmor frequency which is proportional to B; (ii) the softening of the bound-state frequency due to the increasing of the distance R between π kinks in a soliton pair. The third effect, the frequency softening due to out-of-plane effects (21), plays only a minor role.

As above T_N the bound state (20) carries a nonzero value of uniform magnetization $\langle \delta \mathbf{S} \rangle$. Calculations were performed in a similar way as for deriving (17). The final results differ from (17) just by a constant $J_{2\pi}$ which is defined in Appendix A, while the angle σ describing the direction of this magnetization is the same as above T_N.

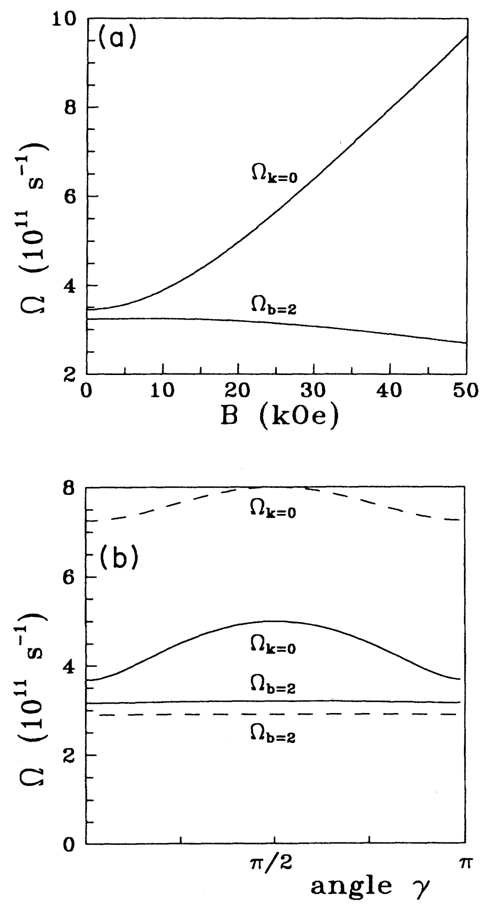

FIG. 4. Dependence of the frequency of long-wave magnons and of the second bound state below T_N (a) on the magnitude of the external magnetic field **B** for the orientation $\gamma = \pi/2$ and (b) on the orientation of **B** for $B = 20$ kOe (solid line) and $B = 40$ kOe (dashed line).

V. CONCLUSIONS

We have discussed the properties of internal oscillations of kink solitons occurring in the quasi-one-dimensional antiferromagnet TMMC. These oscillations can be interpreted as non-Goldstone soliton-magnon bound states. Although these states can exist above and below the three-dimensional ordering temperature T_N their origins and properties are different. Above T_N it is the effect of coupling between in-plane and out-of-plane spin components which causes the appearance of the (π-kink-)magnon bound state. Such a state can exist if the magnitude of the external magnetic field **B** exceeds some critical value which depends on the orientation of the **B** with respect to the easy axis. One can interpret this state as emerging from the bottom of a band of extended magnon states, and for typical experimental conditions its frequency is very close to the bottom of this band.

Below T_N the existence of the (2π-kink-)magnon bound state results from the interaction between the π kinks forming the 2π soliton. In fact, it has been shown for the double-sine-Gordon equation[21] that in the limit when the distance R between the centers of π kinks tends to infinity and their interaction is negligible, the corresponding bound state reduces to an additional Goldstone mode. In contrast to the situation above T_N the frequency of this mode is not close to the bottom of the spin-wave band for typical experimental conditions and is nearly independent of the magnitude and direction of the external magnetic field **B**.

Both bound states considered carry some nonzero net magnetization which can be probed in standard ESR experiments. The polarization of this magnetization depends on the direction and magnitude of the external magnetic field **B** and is the same for both of these states but differs significantly from that of the usual spin-wave modes (especially the uniform ESR modes). Thus, apart from different resonance frequencies, the polarization of the experimentally excitable resonances represents a signature for those types of modes. Taking into account the high frequency resolution and sensitivity of electron spin resonance, which recommends this as an appropriate tool for probing such bound states, we hope to stimulate future experiments confirming these theoretical findings.

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APPENDIX A

The constant appearing in (17) is given by the integral

$$J_\pi \equiv \int_{-\infty}^{+\infty} \sin[\phi_\pi(z)-\beta]\cos[\phi_\pi(z)-\beta]\delta\phi_{(2)}(z)dz = \frac{2J_\parallel S}{g\mu_B B_{\text{eff}}}\sqrt{K}\left[2^\varepsilon B\left[\frac{1+\varepsilon}{2},\frac{1+\epsilon}{2}\right]-2^{2+\epsilon}B\left[\frac{3+\varepsilon}{2},\frac{3+\varepsilon}{2}\right]\right],$$

where we used an explicit form of the solution (13) for the eigenfunction $\delta\phi_2(z)$ and the constant K is given below Eq. (14). Similarly, below T_N the polarization amplitude is determined by the constant

$$\begin{aligned} J_{2\pi} &\equiv \int_{-\infty}^{+\infty} \sin[\phi_{2\pi}(z)-\beta]\cos[\phi_{2\pi}(z)-\beta]\delta\phi_{(2)}(z) \\ &= \frac{-2\sqrt{2}\sinh(2R)}{[1-R\,\text{csch}(2R)]^{1/2}}\left[\frac{24J_\perp}{J_\parallel}+\frac{(g\mu_B B_{\text{eff}})^2}{(J_\parallel S)^2}\right]^{-1/4}\left[-12\rho^2+4\rho^3\frac{6+\rho^{-1}}{\sqrt{4\rho+1}}\ln\left[\rho^{-1}\sqrt{\rho+\tfrac{1}{4}}+1+\frac{1}{2\rho}\right]\right], \end{aligned}$$

where $\rho = 6J_\perp J_\parallel S^2(g\mu_B B_{\text{eff}})^{-2}$ and the expression (20) was used for $\delta\phi_2(z)$.

APPENDIX B

The function $g(R)$ appearing in (21) is defined by

$$g(R) = \int_{-\infty}^{+\infty} [\delta\phi_{(2)}(\xi)]^2\cos^2[\phi_{2\pi}(\xi)-\beta]d\xi ,$$

where the eigenfunction $\delta\phi_{(2)}(\xi)$ is given by (20). The explicit form of $g(R)$ is

$$g(R) = \left\{8\left[1+\frac{1}{4\rho}\right]\left[\tfrac{1}{3}\rho^2-4\rho^3-\frac{4\rho^4}{1+4\rho}+\frac{4\rho^4(3+10\rho)\ln\{[1+2\rho+(1+4\rho)^{1/2}]/2\rho\}}{(1+4\rho)^{3/2}}\right]\right\}\left\{\rho\left[1-\frac{R}{\sinh(2R)}\right]\right\}^{-1}.$$

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