Lagrangian Spin-Wave Theory of Frustrated Antiferromagnets: Application to ABX_3 Compounds

M. L. Plumer and A. Caillé
Centre de Recherche en Physique du Solide et Département de Physique, Université de Sherbrooke, Sherbrooke, Québec, Canada J1K 2R1
(Received 26 July 1991)

A theory of magnetic excitations formulated within the framework of the Lagrange equations of motion is used to study spin waves in stacked triangular antiferromagnets. As a consequence of geometrical frustration, a longitudinal mode parasitically coupled to transverse excitations is predicted. The model demonstrates good agreement with inelastic neutron scattering data for spin-1 $CsNiCl_3$ and $RbNiCl_3$, as well as spin-$\frac{5}{2}$ $CsMnI_3$, in contrast with results based on the Haldane conjecture and with standard spin-wave theory. This formalism has general relevance to all frustrated spin systems.

PACS numbers: 75.30.Ds, 75.50.Ee, 76.50.+g

Discrepancies between existing theory and experimental results for spin excitations in a class of quasi-one-dimensional hexagonal ABX_3 antiferromagnets are becoming increasingly clear. Of particular interest is the possibility to observe effects associated with the integer-spin Haldane conjecture in spin-1 $CsNiCl_3$ and $RbNiCl_3$. It is now evident that the field-theory model proposed by Affleck [1,2] does not provide convincing agreement with experimental results [3–6] to support the existence of this novel quantum state at temperatures well below the onset of long-range magnetic order, T_N. It may be that other manifestations of the Haldane proposal are responsible for the failure of conventional spin-wave theory to explain the inelastic neutron diffraction data of these Ni compounds; however, related discrepancies also exist for the non-Haldane spin-$\frac{5}{2}$ system $CsMnI_3$ [7]. In the present work we suggest that standard spin-wave theory fails to fully account for effects associated with the geometrical frustration inherent to triangular antiferromagnets. A theory of magnetic excitations based on the Lagrangian approach introduced by Dzyaloshinskii and Kukharenko [8] (DK) is shown to give a satisfactory explanation of the observed magnon dispersions for $CsNiCl_3$, $RbNiCl_3$, and $CsMnI_3$. The proposed model has general relevance for all frustrated spin systems.

Much of the general formalism relevant to this work has already been presented in our study of a phenomenological Lagrangian model of longitudinal spin fluctuations in $CsNiCl_3$ [9,10], which reproduced the results of Affleck [1,2]. Conventional spin dynamics are determined by the familiar torque equations [11], as introduced by Landau and Lifshitz [12], which exclude longitudinal (magnitude) fluctuations. In an alternate approach, DK give symmetry arguments to support a Lagrangian formalism based on a kinetic part of the form

$$T=\tfrac{1}{2}\sum_{ij}\int d\mathbf{r}(\mu_{ij}\dot{s}_i\dot{s}_j+\rho_{ij}s_i\dot{s}_j)\,, \tag{1}$$

where $\dot{s}_i=ds_i/dt$ and ρ_{ij} must be antisymmetric and an odd function of the spin density $\mathbf{s}(\mathbf{r})$. DK remark that although the coefficient $\underline{\mu}$ has no obvious physical interpretation, rigorous arguments demonstrate that $\underline{\rho}$ is related to the gyromatic ratio γ and equilibrium spin density. In addition, these authors make the important observation that the equations of motion resulting from (1) with $\mu=0$ are very similar, but not necessarily identical, to the conventional torque equations (see below). We examine here the results of a model Lagrangian formulation applied to triangular antiferromagnets with *only* the linear time-derivative term retained in the kinetic energy. We justify this approach by the physical motivation of DK and by its close resemblance to standard spin-wave theory; in fact, *the two formalisms differ only for frustrated spin systems*. With inspiration from example cases considered by DK, as well as by Izyumov and Laptev [13], we take the kinetic part to be of a general form

$$T=\tfrac{1}{2}\sigma\int d\mathbf{r}\,\mathbf{s}^0\cdot(\tilde{\mathbf{s}}\times\dot{\tilde{\mathbf{s}}})\,, \tag{2}$$

where $\mathbf{s}^0$ and $\tilde{\mathbf{s}}$ are the equilibrium and perturbed contributions to the spin density [9], respectively, and $\sigma=(2\gamma S^2)^{-1}$, where the magnitude of $\mathbf{s}^0$ is given by $\sqrt{2}S$. [Comparison with (1) demonstrates that $\rho_{ij}=\sigma\sum_k\varepsilon_{ijk}s_k^0$. In the torque equations [9,10], $\dot{\tilde{s}}_i=-\sum_j\Gamma_{ij}\partial F/\partial\tilde{s}_j$, one has $\Gamma_{ij}=-\gamma\sum_k\varepsilon_{ijk}s_k^0$. The inverses of the matrices $\underline{\rho}$ and $\underline{\Gamma}$ are not defined unless irrelevant diagonal components [8] are added; if this is done, the torque and Lagrange equations of motion can then be shown to be formally equivalent *only* for collinear, i.e., unfrustrated, spin systems.] Our previous work demonstrated the equivalence of Affleck's field-theory model and the Lagrangian formalism for the *opposite* case (with no physical justification from classical magnetism) where only the μ term is retained in T.

The method used in Ref. [9] is based on a knowledge of the inverse static susceptibility and a crucial feature of the present study is the contribution made by off-diagonal elements (which were incorrectly set to zero [2,10] in our previous work [9]). With $\mu\equiv0$, these terms provide a mechanism for longitudinal excitations, parasitically coupled to transverse modes, which gives rise to important modifications to the conventionally derived magnon spectrum. Such off-diagonal elements of the inverse susceptibility are zero in the case of nonfrustrated (bipartite) lattices and the standard spin-wave dispersions are recovered.

It is demonstrated here that several outstanding discrepancies between existing theory and experimental results on ABX_3 compounds are resolved by this $\mu=0$ Lagrangian formalism. Standard spin-wave theory [3] fails to explain the energy (an unreasonably large value of the anisotropy must be assumed) or polarization of the "gap mode" at the ordering wave vector $(\frac{1}{3},\frac{1}{3},1)$, and also predicts a low-energy mode at (0,0,1) which is not observed, whereas the Haldane-inspired field-theory model yields many modes with significant predicted intensity which are not observed. (Note that only Refs. [2] and [6] contain comparisons of experimental data with the revised predictions of the field-theory model.) The theory presented here is applicable only at temperatures below T_N and has no relevance to experimentally observed manifestations of the Haldane conjecture at higher temperatures.

A brief outline of the method and some results as described in Ref. [9] is useful here. The hexagonal antiferromagnets of interest are characterized by a Hamiltonian of the form

$$\mathcal{H}=J_{\parallel}\sum_{\langle ij\rangle}\mathbf{S}_i\cdot\mathbf{S}_j+J_{\perp}\sum_{\langle kl\rangle}\mathbf{S}_k\cdot\mathbf{S}_l+D\sum_i(S_i^z)^2, \tag{3}$$

where $J_{\parallel}>0$, $J_{\perp}>0$ represent nearest-neighbor exchange along the c axis and in the basal plane, respectively, $D<0$ is the single-ion anisotropy, and [14] $J_{\parallel}\gg J_{\perp},|D|$. At the very low temperatures of interest here ($T\ll T_N$), effects from the D term are small (again, we take $D\equiv 0$) and the equilibrium magnetic structure is described by a helically polarized 120° spin density with moments lying in the x-z plane. Within a molecular-field treatment of the Hamiltonian (3), a Landau-type free energy F expanded to fourth order in the spin density gives an approximation for the potential-energy contribution to the Lagrangian, $\mathcal{L}=T-F$. As with all helical spin structures, it is convenient to use a rotating (orthonormal) basis $\boldsymbol{\varepsilon}_+(\mathbf{r})$, $\boldsymbol{\varepsilon}_-(\mathbf{r})$, $\boldsymbol{\varepsilon}_0$ [see Eqs. (18)-(21) in Ref. [9]] so that $\mathbf{s}^0(\mathbf{r})=\sqrt{2}S\boldsymbol{\varepsilon}_+(\mathbf{r})$ and $\hat{\mathbf{y}}=\boldsymbol{\varepsilon}_0$. Within this coordinate system, the inverse static susceptibility $R_{\mu\nu}(\mathbf{q},\mathbf{q}')$ is diagonal in $\mathbf{q}$ and $\mathbf{q}'$, with nonzero elements $R_{\mu\mu}$ as well as [10]

$$R_{+-}(\mathbf{q})=-R_{-+}(\mathbf{q})=\tfrac{1}{2}i[J_{\mathbf{q}+\mathbf{Q}}-J_{\mathbf{q}-\mathbf{Q}}]=iJ_{\perp}g_{\mathbf{q}}, \tag{4}$$

where $J_{\mathbf{q}}$ is the Fourier transform of the exchange integral $J(\mathbf{r})$, and

$$g_{\mathbf{q}}=\pm\sqrt{3}[\sin(a q_x)-2\sin(\tfrac{1}{2}a q_x)\cos(\sqrt{3}q_y)]. \tag{5}$$

The equilibrium magnetic structure is characterized by a wave vector $\mathbf{Q}=\pm(4\pi/3a)\hat{\mathbf{x}}+(\pi/c)\hat{\mathbf{z}}=\frac{1}{3}\mathbf{G}_{\perp}+\frac{1}{2}\mathbf{G}_{\parallel}$ (where $\mathbf{G}$ is a reciprocal-lattice vector). From expression (4) it is evident that off-diagonal elements of $\underline{R}$ are zero in the case of bipartite lattices where $\mathbf{Q}=\frac{1}{2}\mathbf{G}$.

Within the framework of a free energy approximated by a Landau-type expansion to only fourth order, $R_{++}=R_{--}-2A_Q$ with $A_Q=a(T-T_N)$, where a depends on the ionic angular momentum j through the Brillouin function [15]. Such a relation is unreliable at the low temperatures of interest here so that we take (as in Ref. [9]) $A_Q=\Delta^2/J_{\parallel}-3J_{\perp}$ and, following the procedure of Ref. [6], treat Δ as an adjustable parameter. Within the framework of Affleck's field-theory model, Δ would represent the single-chain Haldane gap; no such interpretation is made here.

The equations of motion which result from the Lagrangian as described above can be written as [cf., Eqs. (37)-(39) of Ref. [9]]

$$R_+\tilde{S}_+ + R_{+-}\tilde{S}_- = 0, \tag{6}$$

$$R_-\tilde{S}_- + R_{-+}\tilde{S}_+ + \beta\dot{\tilde{S}}_0 = 0, \tag{7}$$

$$R_0\tilde{S}_0 - \beta\dot{\tilde{S}}_- = 0, \tag{8}$$

where $R_{\mu\mu}\equiv R_\mu$ and $\beta=\sqrt{2}\sigma S=(\sqrt{2}S)^{-1}$, in frequency units where $\gamma=1$. Note that the longitudinal mode given by $\tilde{S}_+=-(R_{+-}/R_+)\tilde{S}_-$ is zero in the absence of the off-diagonal elements of $\underline{R}$ and that this amplitude excitation is induced by the (xz) transverse mode $\tilde{S}_-$. (This is in contrast to the independent longitudinal mode which results from a model with $\mu\neq 0$.) All excitations (xz longitudinal, xz transverse, and y transverse) are coupled with a characteristic frequency

$$\omega_{\mathbf{q}}^2=R_0(R_- - |R_{+-}|^2/R_+)/\beta^2. \tag{9}$$

The standard spin-wave dispersion [3,9] $\omega_{\mathbf{q}}^2=R_0R_-/\beta^2$ (with $2\sqrt{2}S=j$ at zero temperature) is thus recovered for nonfrustrated systems, and also at $\mathbf{q}_{\perp}=0$ in the present case [from (4) and (5)]. Following the method of Ref. [16], a calculation of the neutron scattering cross section reveals that it is proportional to

$$I=(1-\hat{\mathbf{q}}_y^2)(R_- - |R_{+-}|^2/R_+)_{\mathbf{q}}(1/\omega_{\mathbf{q}})\delta(\omega-\omega_{\mathbf{q}}) + \tfrac{1}{4}(1+\hat{\mathbf{q}}_y^2)\sum_{\pm}\{[R_0(|R_{+-}|/R_+\pm 1)^2]_{\mathbf{q}\pm\mathbf{Q}}(1/\omega_{\mathbf{q}\pm\mathbf{Q}})\delta(\omega-\omega_{\mathbf{q}\pm\mathbf{Q}})\}, \tag{10}$$

so that three modes can occur for a given wave vector $\mathbf{q}$, which are labeled here by $\omega_0=\omega_{\mathbf{q}}$ and $\omega_{\pm}=\omega_{\mathbf{q}\pm\mathbf{Q}}$. Note that the 0 mode corresponds to the y mode discussed in previous work [1-7].

Comparison of the above results with observed data for spin-excitation dispersions along high-symmetry directions was made using values for $J_{\parallel}$ and $J_{\perp}$ (in units of THz) as estimated in Refs. [3,6,7]: $CsNiCl_3$, $J_{\parallel}=1.38$, $J_{\perp}=0.024$; $RbNiCl_3$, $J_{\parallel}=1.94$, $J_{\perp}=0.057$; $CsMnI_3$, $J_{\parallel}=0.792$, $J_{\perp}=0.004$. The unknown parameter Δ was then estimated as in Ref. [6] so that the predicted gap mode, in this case ω_+ at $\mathbf{q}=\mathbf{Q}$, agrees with experimental results. In accord with experiment, this mode is predicted

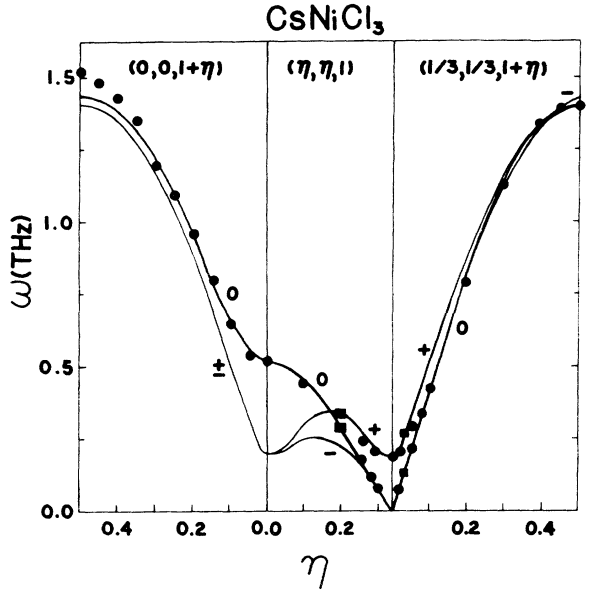

FIG. 1. Dispersion curves (see [10]) $\omega_0=\omega_q$, $\omega_+=\omega_{q+Q}$, $\omega_-=\omega_{q-Q}$ (labeled 0, +, and −, respectively) from (9) with $\Delta=0.255$ THz and data from Ref. [3] (circles) and Ref. [4] (squares) as a function of reduced-wave-vector component. Thicker lines indicate higher intensity (see Fig. 2).

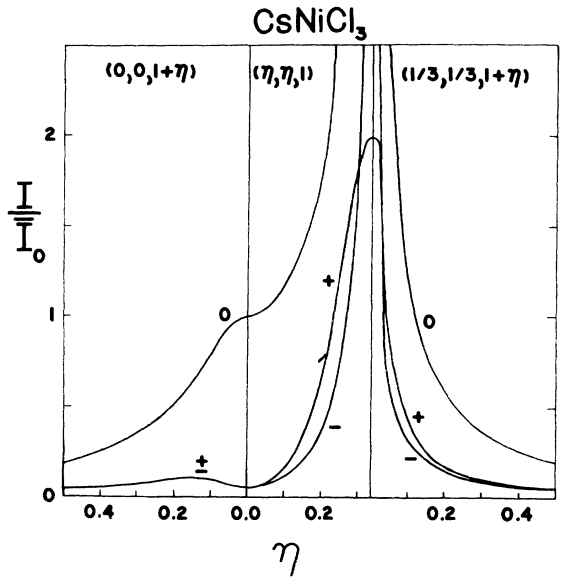

FIG. 2. Single-domain [6] intensities from (10) relative to the 0 mode at (0,0,1), $\bar{I}_0$. Qualitatively similar results occur for $RbNiCl_3$ and $CsMnI_3$.

here to be mainly associated with xz fluctuations. As emphasized in previous comparisons between theory and experiment, the predicted relative intensities of modes must be considered.

Dispersion curves for $CsNiCl_3$ shown in Fig. 1 using the value $\Delta=0.255$ THz demonstrate good agreement with experiment [except for the 0 mode close to $(0,0,\frac{3}{2})$, possibly due to the omission of single-ion anisotropy [3]]. The absence of clearly observed $+/-$ modes near (0,0,1), at which $\omega_\pm\cong 0.194$ THz, can be explained by their relatively low predicted intensities, as shown in Fig. 2, where the ratio $I_\pm/I_0$ is 0.051. These results can be compared with values from standard ($D=0$) spin-wave theory, $\omega_\pm=\omega_0/\sqrt{2}\cong 0.370$ THz and $I_\pm/I_0=\sqrt{2}/4\cong 0.35$. Observed and predicted intensity ratios at some specific wave vectors examined in the polarized (single-domain [6]) neutron experiments of Ref. [4] are in reasonably good agreement: At $(-\frac{1}{3},-\frac{1}{3},1.03)$ and (0.2,0.2,1) (see squares in Fig. 1), relation (10) yields the values 0.36 and 0.33, respectively, whereas (crude) experimental estimates are 0.3 and 0.4. At (0.9,0.9,0.97), a peak in the spin-flip scattering [Fig. 2(b) of Ref. [4]] occurs near 0.45 THz, in contrast with the predicted energies $\omega_+=0.29$ and $\omega_-=0.33$ THz; however, the smaller anomaly observed near 0.3 THz in the higher-resolution unpolarized scan [Fig. 2(a) of Ref. [4]] may be a signature of these predicted low-intensity ($I_\pm/I_0\cong 0.2$) modes. It is also of interest to examine a measure of the relative content of longitudinal polarization, as determined by $|\tilde{S}_+/\tilde{S}_-|_{q+Q}$ from (6). This ratio has equal maxima at wave vectors (0,0,1) and **Q**, where it attains a value of 0.72. Although clearly an important test of current theories, there have been no experimental estimates of this or related quantities.

Similarly good agreement between theory and observed results for $RbNiCl_3$ is seen in Fig. 3 where $\Delta=0.245$ THz has been used. The relative intensity for this material is similar to that shown in Fig. 2, except that $I_+/\bar{I}_0$ does not have such a narrow peak around **Q** (where its maximum value is 1.07). Of particular interest is the relatively large predicted value $I_\pm/I_0=0.13$ at (0,0,1) and $I_\pm/I_0=0.29$ at (1,1,1). The data of Fig. 2 in Ref. [6] suggest the possibility of a lower-energy mode at these wave vec-

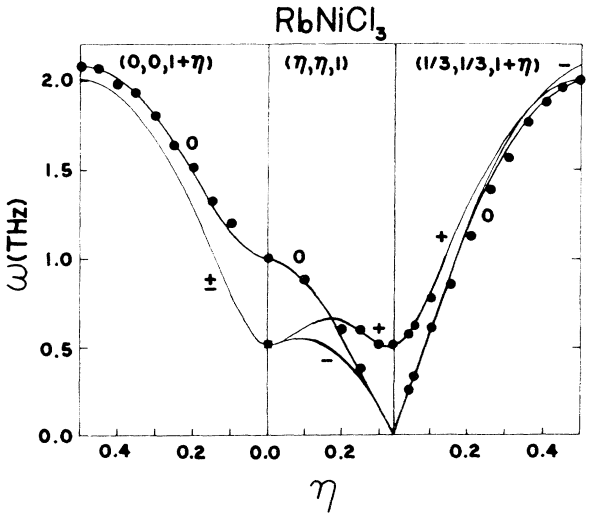

FIG. 3. As in Fig. 1 using $\Delta=0.245$ THz with data from Ref. [6].

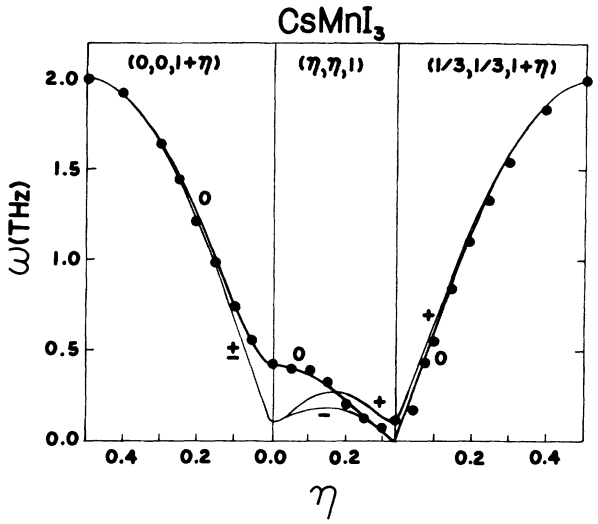

FIG. 4. As in Fig. 1 using $\Delta=0.092$ THz with data from Ref. [7].

tors corresponding to the predicted value $\omega_{\pm}=0.51$ THz. The predicted frequency and (domain-averaged) intensity ratio for the + mode at $(-\frac{1}{3},\frac{1}{3},0.96)$ are 0.58 THz and 0.13, respectively, in good agreement with observed results [6]. Note also from Fig. 3 the good agreement between the data and theory for the 0 mode at all the wave vectors considered. (The field-theory model of Affleck is restricted to values near $|q_z|=1$ even for this mode; the corresponding y-mode curve shown in Fig. 3 of Ref. [6] at values $|q_z|\gtrsim 1.1$ is from $D=0$ spin-wave theory [17].) A somewhat smaller relative content of longitudinal polarization is predicted for this material, where the maximum value of $|\tilde{S}_+/\tilde{S}_-|_{q+Q}$ is 0.48.

Corresponding results are shown in Fig. 4 for the non-Haldane, nearly one-dimensional $CsMnI_3$ using $\Delta=0.092$ THz. The gap mode is small in this material, $\omega_+=0.10$ THz, due to the relatively low value of $J_\perp/J_\parallel\cong 0.005$. It is for this reason that the predicted intensity ratio $I_\pm/I_0$ at (0,0,1) is also very small, 0.015, so that the failure to observe more than one mode as reported in Ref. [7] near this wave vector is explained by the present theory. The relative intensities corresponding to Fig. 2 are more sharply peaked near $(\frac{1}{3},\frac{1}{3},1)$ (with $I_+/I_0=3.5$), as is the relative content of longitudinal fluctuations ($|\tilde{S}_+/\tilde{S}_-|_{q+Q}=0.88$).

The results presented in this work suggest that a Lagrangian formulation of spin excitations can explain most of the existing experimental data for a variety of hexagonal ABX_3 antiferromagnets, in contrast with standard spin-wave theory and Affleck's model of effects due to the Haldane conjecture. A kinetic part of the Lagrangian with only terms linear in the time derivative appears to be consistent with fundamental results in classical magnetism. The new results presented here (e.g., a parasitic longitudinal mode) are a consequence of the differences between conventional and Lagrangian formulations of spin excitations which occur only in frustrated antiferromagnets. (It is known that standard spin-wave theory fails to describe other types of longitudinal fluctuations [18].) Although clearly of interest, the precise origins of these discrepancies are not known; however, related questions regarding Lagrangian formalisms often occur (as with the field-theory model [1,9] and in a recent study of phase transitions in tripartite systems [19]). The formalism presented here should have general relevance to all frustrated spin systems.

We thank I. Affleck, A. Chubukov, A.-M. S. Tremblay, and Z. Tun for useful discussions and correspondence. This work was supported by the Natural Sciences and Engineering Research Council of Canada and le Fonds FCAR pour l' Aide et le Soutien à la Rechereche du Gouvernement du Québec.

Note added.—A link between triangular frustration and the Haldane conjecture has recently been suggested [20].

[1] I. Affleck, Phys. Rev. Lett. **62**, 474 (1989).
[2] I. Affleck, Phys. Rev. Lett. **65**, 2477(E); **65**, 2835(E) (1990).
[3] W. J. L. Buyers *et al.*, Phys. Rev. Lett. **56**, 371 (1986); R. Morra, W. J. L. Buyers, R. L. Armstrong, and K. Hirakawa, Phys. Rev. B **38**, 543 (1988).
[4] Z. Tun *et al.*, Phys. Rev. B **42**, 4677 (1990).
[5] M. Steiner, J. Appl. Phys. **67**, 5593 (1990).
[6] Z. Tun, W. J. L. Buyers, A. Harrison, and J. A. Rayne, Phys. Rev. B **43**, 13331 (1991).
[7] A. Harrison, M. F. Collins, J. Abu-Dayyeh, and C. V. Stager, Phys. Rev. B **43**, 679 (1991).
[8] I. E. Dzyaloshinskii and V. G. Kukharenko, Zh. Eksp. Teor. Fiz. **70**, 2360 (1976) [Sov. Phys. JETP **43**, 1232 (1976)].
[9] M. L. Plumer and A. Caillé, Phys. Rev. B **42**, 8783 (1990).
[10] M. L. Plumer and A. Caillé, Phys. Rev. B **43**, 11454(E) (1991).
[11] See, e.g., C. Kittel, *Introduction to Solid State Physics* (Wiley, New York, 1976), 5th ed., Chap. 15.
[12] R. Landau and E. Lifshitz, Phys. Z. Sowjet. **8**, 153 (1935) [*Collected Papers of Landau* (Pergamon, New York, 1965)].
[13] Yu. A. Izyumov and V. M. Laptev, Zh. Eksp. Teor. Fiz. **88**, 165 (1985) [Sov. Phys. JETP **61**, 95 (1985)]; J. Magn. Magn. Mater. **51**, 381 (1985).
[14] As emphasized in Ref. [9], our definitions of the exchange constants are related to those of Refs. [1–7] (J,J') by $J_\parallel=4J$ and $J_\perp=4J'$.
[15] See, e.g., M. L. Plumer, A. Caillé, and H. Kawamura, Phys. Rev. B **44**, 4461 (1991).
[16] M. L. Plumer, J. Phys. C **17**, 4663 (1984).
[17] Z. Tun (private communication).
[18] R. S. Fishman and S. H. Liu, J. Phys. Condens. Matter **3**, 8313 (1991).
[19] P. Azaria, B. Delamotte, and T. Jolicoeur, Phys. Rev. Lett. **64**, 3175 (1990).
[20] M. W. Long and S. Siak, J. Phys. Condens. Matter **3**, 4901 (1991).