Heisenberg magnet with modulated exchange

I. A. Zaliznyak
Brookhaven National Laboratory, Upton, New York 11973-5000, USA
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A modification of the ground state of the classical-spin Heisenberg Hamiltonian in the presence of a weak superstructural distortion of an otherwise Bravais lattice is examined. It is shown that a slight modulation of the crystal lattice with wave vector $\mathbf{Q}_c$ results in a corresponding modulation of the exchange interaction which, in the leading order, is parametrized by no more than two constants per bond, and perturbs the spin Hamiltonian by adding the ''umklapp'' terms $\sim S^{\alpha}_{\mathbf{q}} S^{\alpha}_{\mathbf{q}\pm\mathbf{Q}_c}$. As a result, for a general spin-spiral ground state of the nonperturbed exchange Hamiltonian, an incommensurate shift of the propagation vector $\mathbf{Q}$ and additional new magnetic Bragg peaks, at $\mathbf{Q}\pm n\mathbf{Q}_c$, $n=1,2,...$, appear, and its energy is lowered as it adapts to the exchange modulation. Consequently, the lattice distortion may open a region of stability of the incommensurate spiral phase which otherwise does not win the competition with the collinear Néel state. Such is the case for the frustrated square-lattice antiferromagnet. In addition, the umklapp terms provide a commensuration mechanism, which may lock the spin structure to the lattice modulation vector $\mathbf{Q}_c$, if there is sufficient easy-axis anisotropy, or a magnetic field in an easy plane.

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I. INTRODUCTION

An interplay between the distortion of a crystal lattice and the magnetic properties of the material has recently become a subject of renewed interest. One problem which provides strong motivation for studying the effect of a weak superstructural modulation on the spin system is that of striped phases.[1,2] These charge-ordered states are found in lightly doped high-T_c cuprates $La_{2-x}Sr_xCuO_{4+y}$ (LSCO) and in related nickelates, and are always associated with a weak superstructural distortion of the original ''stacked square lattice'' structure of the undoped parent material. Incommensurate magnetism in these compounds is usually interpreted in terms of the segregation of the doped charges into lines which separate the antiferromagnetic domains (''stripes''), characteristic of the undoped material. There is also a modulation of the crystal structure induced by the charge-stripe segregation, but it is often too small to be observed in experiment.[1] It is clear that the essential effect of the stripe order on the spin system of cuprates is that of a periodic modulation of the exchange coupling in the Heisenberg spin Hamiltonian which describes their magnetic properties.[3] However, only the simplest ''average'' consequence of stripe superstructure, in the form of the effective weakening of the exchange coupling in the direction perpendicular to stripes, has been considered so far.[4] A similar problem, of an interplay between the spin order and a cooperative Jahn-Teller distortion accompanying the charge order, arises in the context of the charge-ordered phases in doped manganites.[5]

A number of examples not related to charge ordering but no less interesting involve an intriguing interplay between small superlattice modulation and spin structure in spin-frustrated antiferromagnetic dielectrics. In the square-lattice antiferromagnet, a distortion could actually be the source of the frustration. For example, it may generate a well-known generalized Villain model.[6] In the triangular-lattice antiferromagnet (TLA), a weak distortion may partially release frustration, and result in complicated spiral phases. Among the simple realizations of the distorted TLA are the so-called ''row models'' which were extensively studied in the past.[7–10] However, previous studies were mainly restricted to a specialized analysis of a few particular models; no general approach that would allow a unified treatment of the effect of a small lattice distortion on a spin system has been developed so far.

A number of interesting experimental examples of distorted triangular lattice antiferromagnets (DTLA's), which instigated this study, are found among the $CsNiCl_3$-type compounds with the general chemical formula ABX_3. In the (anti)ferroelectric phases that are realized in some of these materials at low temperatures relevant for the magnetic order, the $P6_3/mmc$ hexagonal symmetry in which they crystallize is lowered, and a fully frustrated triangular lattice inherent in the original $CsNiCl_3$-type ''stacked triangular lattice'' crystal structure is slightly distorted. Typically, the high-temperature hexagonal structure with a Bravais lattice of equivalent magnetic B^{2+} sites is changed to either a hexagonal $P6_3/cm$ structure with a three times larger unit cell, or to a large-cell orthorhombic structure. These superstructures are characterized by the appearance of the superlattice Bragg reflections at $\mathbf{Q}_c=(\eta,\eta,0)$,[11] with $\eta=\frac{1}{3}$, or $\eta=\frac{1}{4},\frac{1}{8}$, ..., respectively. In some cases, as in $KNiCl_3$, both phases are found to coexist at low temperature.[12] Perhaps the most intriguing is the case of $RbMnBr_3$, in which most experiments find the orthorhombic low-T phase,[13–15] and an incommensurate spiral spin structure with propagation vector $\mathbf{Q}=(\frac{1}{3}+q, \frac{1}{3}+q, 1)$,[13–16] in place of the commensurate ''triangular'' antiferromagnetic order with $\mathbf{Q}=(\frac{1}{3}, \frac{1}{3}, 1)$, which is characteristic of the nondistorted hexagonal materials $CsMnBr_3$, $CsNiCl_3$, etc.[17–19] In a magnetic field of about 3 T applied in the easy plane the spin structure becomes commensurate, with $\mathbf{Q}=(\frac{1}{8}, \frac{1}{8}, 1)$. In the similar orthorhombic modification of $KNiCl_3$, which is a related material but with

an easy-axis spin anisotropy, this latter structure is realized already at $H=0$.

Until now, these experimental findings remained to a large extent unexplained. One reason for this is that traditionally, the effect of each particular lattice distortion on the spin Hamiltonian was considered separately, by devising a specific, generally multisublattice spin model (e.g., the row models), where the distortion simply defines the particular setup of the near-neighbor exchange interactions. For long-period structural modulations, this approach leads to models with a large number of inequivalent spin sites [e.g., up to 8 for $\mathbf{Q}_c=(\frac{1}{8},\frac{1}{8},0)$], resulting in tremendously complicated spin Hamiltonians, and therefore the analysis has never been carried through. The same problem is outstanding for the stripe phases in LSCO cuprates, where the most stable superstructure has a pitch of about $\frac{1}{8}$ [curiously, it is the same as that of the antiferroelectric lattice distortion in $RbMnBr_3$ (Ref. 13)]. In addition, the modulation has an even longer period at small doping, and, in general, can also be incommensurate. Here we devise an alternative approach, which lays grounds for the consistent and general explanation of spin incommensurability, commensuration transition, and other phenomena arising from the lattice distortion, that were mentioned above. We treat the effect of an *arbitrary* but *small* lattice distortion on the microscopic spin Hamiltonian in the perturbation framework. The analysis in this paper most directly applies in the case of the dielectrics with localized spins, although we expect it to hold also for the doped perovskites, to the extent that the itineracy effects can be neglected.

Consider a system of N equivalent spins on a simple Bravais lattice, coupled by Heisenberg exchange interactions. The model Hamiltonian, which allows also for a uniaxial spin anisotropy and a Zeeman energy, is

$$\mathcal{H}=\sum_{i,j} J_{ij}\mathbf{S}_i\cdot\mathbf{S}_j+D\sum_i (S_i^z)^2-\sum_i \mathbf{h}\cdot\mathbf{S}_i, \tag{1}$$

where $J_{ij}=J_{ji}$ parametrize the exchange coupling between the spins at lattice sites i and j, D is the anisotropy constant, and $\mathbf{h}=g\mu_B\mathbf{H}$ is the magnetic field. Without the anisotropy and magnetic field, the classical ground state of Eq. (1) is a planar spin spiral,[20–24] $\mathbf{S}_j=(S\cos\theta_j, S\sin\theta_j, 0)$, $\theta_j=(\mathbf{Q}\cdot\mathbf{r}_j)$. The ordering wave vector $\mathbf{Q}$ corresponds to the minimum of the lattice Fourier transform of the exchange interaction, $J_{\mathbf{q}}=\Sigma_{\mathbf{r}_{ij}}J_{ij}\exp(-i\mathbf{q}\cdot\mathbf{r}_{ij})$, $\mathbf{r}_{ij}=\mathbf{r}_j-\mathbf{r}_i$ [this includes ferro- and antiferromagnetism, corresponding to $\mathbf{Q}=0$ and, e.g., $\mathbf{Q}=(\pi,\pi,\pi)$, respectively]. For nonzero D and $\mathbf{h}$, the spin structure is, in general, modified by the appearance of higher-order harmonics in the lattice Fourier transform of the spin distribution $\mathbf{S}_{\mathbf{q}}=\Sigma_{\mathbf{r}_j}\mathbf{S}_j\exp(-i\mathbf{q}\cdot\mathbf{r}_j)$, with the wave vectors $\mathbf{Q}_n=n\mathbf{Q}$, in addition to $\mathbf{S}_{\mathbf{Q}}$.[25–27] For small D and h the corresponding terms in the Hamiltonian (1) can be treated as a perturbation. The perturbation expansion for the correction to the spiral winding angle has the form $\delta\theta_j=\Sigma_n[\alpha_n\cos(n\mathbf{Q}\cdot\mathbf{r}_j)+\beta_n\sin(n\mathbf{Q}\cdot\mathbf{r}_j)]$, where the coefficients α_n and β_n are of the order $O[(|D|/J)^{n/2},(h/J)^n]$.[26]

In this paper we consider what happens to the classical spiral ground state of the Hamiltonian (1) if a weak superstructural modulation of the original Bravais lattice appears in the crystal whose spin system it describes. It is clear that results obtained for classical spins are subject to corrections from quantum and thermal fluctuations, and these are often crucial. Nevertheless, if we find the spiral state that has the lowest energy already on the classical level, inclusion of the $1/S$ corrections may still result in this state being the ground state (clearly, this will always be the case for large enough spin S). In fact, this may appear to be the case even for small spins, so long as the spin-wave theory holds. While accounting for the fluctuations is important, it falls beyond the scope of this paper and is deferred to further studies.

II. SUPERLATTICE DISTORTION AND THE SPIN HAMILTONIAN

Consider a slight distortion of the crystal structure which is characterized by the appearance of additional, weak superlattice Bragg reflections at wave vectors $\pm\mathbf{Q}_c$ in the Brillouin zone of the nondistorted Bravais lattice. Most generally, such a superstructure corresponds to a small harmonic modulation of the positions $\mathbf{r}_j^\mu$ of the ions and the characteristic symmetry points of the local electron-density distribution (orbitals) in the lattice,

$$(\mathbf{r}_j^\mu)'=\mathbf{r}_j^\mu+\boldsymbol{\epsilon}_j^\mu,\quad \boldsymbol{\epsilon}_j^\mu=\boldsymbol{\epsilon}_1^\mu\cos(\mathbf{Q}_c\cdot\mathbf{r}_j^\mu)+\boldsymbol{\epsilon}_2^\mu\sin(\mathbf{Q}_c\cdot\mathbf{r}_j^\mu). \tag{2}$$

Here j numbers the sites of the original Bravais lattice of magnetic ions,[28] and μ indexes positions of ligands and symmetry points of the magnetic orbitals within the unit cell. It is shown in the Appendix that a leading correction to the exchange coupling in this case is most generally expressed as

$$J'_{ij}=\tilde{J}_{ij}+j'_{ij}\cos(\tilde{\mathbf{Q}}_c\cdot\mathbf{R}_{ij})-j''_{ij}\sin(\tilde{\mathbf{Q}}_c\cdot\mathbf{R}_{ij}), \tag{3}$$

where $\mathbf{R}_{ij}=\frac{1}{2}(\mathbf{R}_i+\mathbf{R}_j)$ is the middle of the ij bond, $j'_{ij}=j'_{ji}$, $j''_{ij}=j''_{ji}$ are real and symmetric, and, most importantly, satisfy all symmetries of the original lattice which leave $\mathbf{Q}_c$ and the polarizations $\boldsymbol{\epsilon}_1^\mu$, $\boldsymbol{\epsilon}_2^\mu$ invariant; the same is true for $\tilde{J}_{ij}$. In particular, $\tilde{J}_{ij}$, j'_{ij}, and j''_{ij} are invariant under the translation group of the nondistorted lattice. Finally, $\tilde{\mathbf{Q}}_c=n\mathbf{Q}_c$, where $n=1,2$ is the order of the leading correction to J_{ij} of Eq. (3) in terms of the small parameter $\epsilon\sim(\epsilon_{1,2}^\mu/r_{ij})\ll 1$ $(j'_{ij},j''_{ij}\sim\epsilon^n J_{ij})$.[29]

In the first order in ϵ, lattice modulation (2) results in the modulation of the exchange coupling J'_{ij} with the same wave vector $\mathbf{Q}_c$. It is described by Eq. (3), with $\tilde{\mathbf{Q}}_c=\mathbf{Q}_c$, $\tilde{J}_{ij}=J_{ij}$, and j'_{ij}, j''_{ij}, defined by Eqs. (A16) and (A17) of the Appendix. The structure of the expressions for j'_{ij}, j''_{ij} is quite illuminating. There are two contributions, one of which is $\sim\sin(\mathbf{Q}_c\cdot\mathbf{r}_{ij}/2)$, and therefore antisymmetric with respect to $\mathbf{Q}_c\to-\mathbf{Q}_c$, and the other depends on the relative alignment of $\mathbf{Q}_c$ and the distortion polarizations with respect to the bond geometry. Only the first contribution survives in the simplest case when exchange depends on the bond length

alone, $J_{ij}=J(r_{ij})$; in this case the bonds that are perpendicular to $\mathbf{Q}_c$ are not changed by the lattice distortion.

In many important cases the first-order corrections vanish, and the leading correction in Eq. (3) is $\sim\epsilon^2 J_{ij}$, in which case $\tilde{\mathbf{Q}}_c=2\mathbf{Q}_c$, and j'_{ij}, j''_{ij} are given by Eqs. (A9)–(A12). In general, the latter have to be amended in accordance with Eqs. (A14) and (A15). Importantly, a $\mathbf{Q}_c$-independent contribution which determines the leading correction to the bond strength J_{ij},

$$\tilde{J}_{ij}=J_{ij}+\delta J_{ij}, \quad \delta J_{ij}\sim\epsilon^2, \tag{4}$$

always appears in this order. Because the correction to the bond strength, δJ_{ij}, arises from replacing $2\cos^2(\mathbf{Q}_c\cdot\mathbf{R}_{ij})$, $2\sin^2(\mathbf{Q}_c\cdot\mathbf{R}_{ij})$, with $\cos(2\mathbf{Q}_c\cdot\mathbf{R}_{ij})\pm 1$, it is directly related with the amplitudes of the exchange modulation. Each second-order term that contributes to j'_{ij}, j''_{ij}, also adds to δJ_{ij}, this is explicit in Eq. (A8). As before, there are two contributions to j'_{ij}, j''_{ij}; the one that survives for $J_{ij}=J(r_{ij})$ does not affect the bonds that are perpendicular to $\mathbf{Q}_c$, but is now symmetric with respect to $\mathbf{Q}_c\rightarrow-\mathbf{Q}_c$.

Accounting for the corrections to the bond strength, Eq. (4), is straightforward and does not require any additional consideration. It simply amounts to a change in the Fourier-transformed exchange coupling $J_{\mathbf{q}}$, which determines the ground-state energy and the spin-wave spectrum of the Hamiltonian (1).[20–26] The significance of this correction, however, is in that a change in $J_{\mathbf{q}}$ applies directly to the ground-state energy, which therefore is corrected in the same order, $O(\epsilon^2)$. As we shall see below, the first-order contribution to the exchange modulation in Eq. (3), $O(\epsilon)$, corrects the ground-state energy of the spin Hamiltonian (1) only in the second order of perturbation. Therefore, except for special cases,[30] these two contributions have to be treated equally.

To summarize, a superlattice distortion (2) leads to a harmonic modulation of the exchange coupling, with either the same wave vector $\mathbf{Q}_c$, if it appears as a first-order correction to J_{ij}, or with the wave vector $2\mathbf{Q}_c$, if it appears in the second order, $\sim\epsilon^2$. There is also a second-order correction to the bond strength. In the most general case these are described by Eq. (3). In what follows, we study the effect of the exchange modulation on the ground state of the exchange part of the spin Hamiltonian (1) (i.e., the case $D=H=0$), which now reads

$$\mathcal{H}=\sum_{i,j}\,(J_{ij}+j_{ij}e^{i\mathbf{Q}_c\cdot\mathbf{R}_{ij}}+j^*_{ij}e^{-i\mathbf{Q}_c\cdot\mathbf{R}_{ij}})\mathbf{S}_i\cdot\mathbf{S}_j\,. \tag{5}$$

Here we have introduced a complex $j_{ij}=j'_{ij}+ij''_{ij}$, and omitted the tildes, keeping implicit that $\mathbf{Q}_c$, J_{ij}, j'_{ij}, j''_{ij} are all appropriately chosen in accordance with the situation, as discussed above. While in the absence of the distortion J_{ij} would satisfy all symmetries of the lattice, exchange constants in Eq. (5) possess only those symmetries of the undistorted lattice which preserve $\mathbf{Q}_c$ and the polarizations ϵ_1^μ, ϵ_2^μ (this includes all translations).

III. CLASSICAL GROUND STATE OF THE MODULATED-EXCHANGE HAMILTONIAN

To proceed with finding the ground state of the modulated-exchange Hamiltonian (5), we first rewrite it in terms of the lattice Fourier transforms, taking advantage of the lattice translational symmetry,

$$\frac{\mathcal{H}}{N}=\sum_{\mathbf{q}}\,\{J_{\mathbf{q}}\mathbf{S}_{\mathbf{q}}\cdot\mathbf{S}_{-\mathbf{q}}+j_{\mathbf{q}+\mathbf{Q}_c/2}\mathbf{S}_{\mathbf{q}}\cdot\mathbf{S}_{-\mathbf{q}-\mathbf{Q}_c} +j^*_{\mathbf{q}-\mathbf{Q}_c/2}\,\mathbf{S}_{\mathbf{q}}\cdot\mathbf{S}_{-\mathbf{q}+\mathbf{Q}_c}\}. \tag{6}$$

Here $j_{\mathbf{q}}=\Sigma_{\mathbf{r}_{ij}}j_{ij}\exp(-i\mathbf{q}\cdot\mathbf{r}_{ij})=j_{-\mathbf{q}}$, similar with the Fourier transforms $J_{\mathbf{q}}$ and $S_{\mathbf{q}}$ introduced above. We note, however, that unlike $J_{\mathbf{q}}$ which is real, $j_{\mathbf{q}}$ is, in general, complex, so $j^*_{\mathbf{q}}\neq j_{-\mathbf{q}}$.

A. General approach

A general approach to finding the classical ground state for a system of N equal spins on a simple Bravais lattice that are coupled by isotropic Heisenberg exchange interaction was developed in Refs. 20–22, and recently discussed in Ref. 26. We need to solve the mathematical problem of finding the absolute minimum of a function (6), which depends on N classical vector variables $\mathbf{S}_{\mathbf{q}}$ under N constraints,

$$\mathbf{S}_j^2=S^2, \quad \forall j, \tag{7}$$

that are imposed on the length of classical spins. In terms of the Fourier components these constraints become

$$\sum_{\mathbf{q}'}\,\mathbf{S}_{\mathbf{q}'}\cdot\mathbf{S}_{\mathbf{q}-\mathbf{q}'}=S^2\delta_{\mathbf{q},0}, \quad \forall\mathbf{q}, \tag{8}$$

where $\delta_{\mathbf{q},\mathbf{q}'}$ is the three-dimensional (3D) Kronekker symbol. Upon introducing N Lagrange multipliers $\lambda_{\mathbf{q}}$ a straightforward variation leads to the following equations for the spin structure that minimizes the Hamiltonian (6),

$$J_{\mathbf{q}}\mathbf{S}_{\mathbf{q}}+j_{\mathbf{q}-\mathbf{Q}_c/2}\mathbf{S}_{\mathbf{q}-\mathbf{Q}_c}+j^*_{\mathbf{q}+\mathbf{Q}_c/2}\mathbf{S}_{\mathbf{q}+\mathbf{Q}_c}-\sum_{\mathbf{q}'}\,\lambda_{\mathbf{q}'}\mathbf{S}_{\mathbf{q}-\mathbf{q}'}=0, \tag{9}$$

$\forall\,\mathbf{q}$, which have to be solved together with Eq. (8). Substituting these into Eq. (6), we obtain the ground-state (GS) energy per site,

$$\frac{E_{GS}}{N}=\lambda_0 S^2, \tag{10}$$

which depends only on $\lambda_{\mathbf{q}}$ with $\mathbf{q}=0$. While this suggests searching for the solution with $\lambda_{\mathbf{q}}=\lambda_0\delta_{\mathbf{q},0}$, it is easily verified by direct substitution that Eq. (9) does not allow any nontrivial solutions of this type for $j_{\mathbf{q}}\neq 0$. In fact, using such an ''ansatz'' for $\lambda_{\mathbf{q}}$ is equivalent to replacing Eq. (7) with a single ''weak'' condition, $\Sigma_j\mathbf{S}_j^2=NS^2$. This ''weak'' condition approach, also known as the Luttinger-Tisza method, was widely employed throughout the early studies of complex spin structures,[20–24] as it does lead to the correct solution in several important particular cases, including the case

of a simple exchange spiral. General reasons for the failure of this approach, which, in particular, occurs for Hamiltonian (1) with $D\neq 0$, $H\neq 0$, were discussed in Ref. 26. The current situation, in the form of Eqs. (6) and (9), presents another example of such failure.

An alternative approach to finding the ground state and the spin-wave spectrum of the Hamiltonian (1) for small D and h was used in Refs. 26 and 27. It is based on a perturbative solution for the real-space spin structure that is stationary with respect to small deviations in the form of a slightly distorted flat spiral. The correction to the spiral winding angle, θ_j, was obtained in the form of the expansion, $\delta\theta_j=\Sigma_n\ [\alpha_n\cos(n\mathbf{Q}\cdot\mathbf{r}_j)+\beta_n\sin(n\mathbf{Q}\cdot\mathbf{r}_j)]$, where the coefficients α_n and β_n are of the order $O((|D|/J)^{n/2},(h/J)^n)$. As a result, additional harmonics in the Fourier transform of the spin structure, at wave vectors $\pm n\mathbf{Q}$, $n=2,3,...$, and a magnetization $\mathbf{S}_0$, appear. In fact, the same result can be obtained from the conditional minimization of the Hamiltonian (1) outlined above, if a perturbative solution is searched in the form of a *harmonic expansion*, $\lambda_{\mathbf{q}}=\Sigma_n\lambda_n\delta_{\mathbf{q},n\mathbf{Q}}$, where $\lambda_{n\neq 0}\sim O((|D|/J)^{1/2},h/J)\cdot\lambda_{|n|-1}$, and $\mathbf{S}_{\mathbf{q}}=\Sigma_n\mathbf{S}_{n\mathbf{Q}}\delta_{\mathbf{q},n\mathbf{Q}}$, $|\mathbf{S}_{n\mathbf{Q}}|\sim O(\lambda_{|n|-1})$. It is this approach, which is both natural and straightforward to apply to the Hamiltonian (6) in order to obtain the spin-wave expansion upon expressing spins through magnon creation/annihilation operators,[31] that we shall employ here.

B. Harmonic expansion for modulated exchange

Because the modulated-exchange terms in the spin Hamiltonian allow the umklapp processes which couple $\mathbf{S}_{\mathbf{q}}$ and $\mathbf{S}_{\mathbf{q}\pm\mathbf{Q}_c}$ and, consequently, couple these Fourier components in Eq. (9), we search for the solution of the Eqs. (8) and (9) in the form of the expansion

$$\mathbf{S}_{\mathbf{q}}=\sum_n\ \{\mathbf{S}_{\mathbf{Q}+n\mathbf{Q}_c}\delta_{\mathbf{q},\mathbf{Q}+n\mathbf{Q}_c}+\mathbf{S}^*_{\mathbf{Q}+n\mathbf{Q}_c}\delta_{-\mathbf{q},\mathbf{Q}+n\mathbf{Q}_c}\},\quad(11)$$

where $S_{\mathbf{Q}+n\mathbf{Q}_c}\sim O(\epsilon^n)$. Substituting this into Eq. (9) it is easy to see that a nontrivial solution requires that $\lambda_{\mathbf{q}}=0$ unless $\mathbf{q}\pm(\mathbf{Q}+n\mathbf{Q}_c)=\pm(\mathbf{Q}+n'\mathbf{Q}_c)$ is satisfied for some n and n'. Therefore the general solution for $\lambda_{\mathbf{q}}$ can be written as

$$\lambda_{\mathbf{q}}=\sum_n\ \lambda_n\delta_{\mathbf{q},n\mathbf{Q}_c},\quad(12)$$

where $\lambda_n\sim O(\epsilon^{|n|})$. Here and below, if the limits are not specified explicitly, it is implied that the summation extends over all integers (this, in particular, implies taking the thermodynamic limit, $N\to\infty$).[32] With Eqs. (11) and (12) the energy-minimum conditions of Eqs. (9) become

$$J_{\mathbf{Q}+n\mathbf{Q}_c}\mathbf{S}_{\mathbf{Q}+n\mathbf{Q}_c}+j_{\mathbf{Q}+(n-1/2)\mathbf{Q}_c}\mathbf{S}_{\mathbf{Q}+(n-1)\mathbf{Q}_c}$$
$$+j^*_{\mathbf{Q}+(n+1/2)\mathbf{Q}_c}\mathbf{S}_{\mathbf{Q}+(n+1)\mathbf{Q}_c}-\sum_{n'}\ \lambda_{n'}\mathbf{S}_{\mathbf{Q}+(n-n')\mathbf{Q}_c}=0,\quad(13)$$

$\forall\ n$. Subsequently, upon substituting Eq. (11) into Eq. (8), the equal-spin constraint is rewritten as

$$\sum_{n'}\ (\mathbf{S}_{\mathbf{Q}+n'\mathbf{Q}_c}\cdot\mathbf{S}_{\mathbf{Q}+(n-n')\mathbf{Q}_c})=0,\quad(14)$$

$$2\sum_{n'}\ (\mathbf{S}_{\mathbf{Q}+(n+n')\mathbf{Q}_c}\cdot\mathbf{S}^*_{\mathbf{Q}+n'\mathbf{Q}_c})=S^2\delta_{n,0},\quad(15)$$

$\forall\ n$. At this point, Eqs. (13)–(15) are still a complicated nonlinear system of equations, and remains such even if we retain only the terms $\sim O(\epsilon)$ which determine the lowest-order corrections to the simple exchange spiral.

C. Exchange symmetry

Further progress into finding the perturbative solution to Eqs. (13)–(15) which would describe a weakly distorted exchange spiral, $\{\mathbf{S}_{\mathbf{Q}+n\mathbf{Q}_c};\lambda_n\}$, is made by employing a powerful ''exchange symmetry'' argument, which relates back to the Landau theory of phase transitions. It was developed in Ref. 33 as a basis for the unified Lagrangian description of the long-wavelength, macroscopic dynamics of the complicated spin systems with complex order parameters, including spin glasses. Subsequently, this approach was used with great success, in particular, for calculating the low-energy spin dynamics in a variety of situations encountered in the noncollinear ground states of $CsNiCl_3$-type triangular-lattice antiferromagnets.[8,34–36] It is based on a very simple observation, that a macroscopic Lagrangian (or a Hamiltonian) of a spin system in a state which is described by an order parameter (at $T\approx 0$), when expressed in terms of the canonical variables that parametrize the long-wavelength dynamics of this order parameter, has to satisfy all remaining symmetries of the ground state (order parameter). Practically, this works as follows. In exchange approximation, possible ground states are few, easily classified, and Lagrangians are relatively easy to write. Perturbation account for the anisotropy, magnetic field, etc., adds terms to the Lagrangian (Hamiltonian), which are expansions in powers of the order parameter, and whose general form is essentially determined by the above symmetry requirement.

We extend the exchange symmetry argument to the microscopic description of the present paper by nothing that, so long as the solution of Eqs. (13)–(15) is a weakly distorted simple exchange spiral $\mathbf{S}_j=\mathbf{S}_{\mathbf{Q}}\exp(i\mathbf{Q}\cdot\mathbf{r}_j)+\mathbf{S}^*_{\mathbf{Q}}\exp(-i\mathbf{Q}\cdot\mathbf{r}_j)$ (i.e., no independent order parameter appears in addition to $\mathbf{S}_{\mathbf{Q}}$), and if the perturbation does not violate the O(3) spin symmetry of the initial exchange Hamiltonian, all vectors in spin space $\mathbf{S}_{\mathbf{q}}$ that define the corrections to the initial exchange structure, have to be proportional to $\mathbf{S}_{\mathbf{Q}}$. In other words, the only ''selected'' directions in spin space which can determine direction of spin vectors in the perturbation series are those resulting from the spontaneous breaking of the spin symmetry that already exists in the nonperturbed system, i.e., those defined by $\mathbf{S}_{\mathbf{Q}}$. This holds for the Hamiltonian (5), because the modulated-exchange terms preserve the O(3) spin symmetry. Consequently, we write

$$\mathbf{S}_{\mathbf{Q}+n\mathbf{Q}_c}=\varepsilon_n\mathbf{S}_{\mathbf{Q}},\quad \mathbf{S}_{-\mathbf{Q}+n\mathbf{Q}_c}=\varepsilon^*_{-n}\mathbf{S}_{-\mathbf{Q}}, \tag{16}$$

where $\varepsilon_n\sim O(\epsilon^n)$, and $\varepsilon_0\equiv 1$. This simplifies Eqs. (13)–(15) tremendously, as they shall now involve only scalar variables ε_n, λ_n. In addition, Eq. (14) is automatically satisfied for $(\mathbf{S}_{\mathbf{Q}})^2=0$, which holds in the case of a simple exchange spiral. It requires that spin vectors of real and imaginary parts of $\mathbf{S}_{\mathbf{Q}}=\mathbf{S}'+i\mathbf{S}''$ are mutually perpendicular, $(\mathbf{S}'\cdot\mathbf{S}'')=0$, and have equal length. This length is determined from Eq. (15),

$$|\mathbf{S}_{\mathbf{Q}}|^2=|\mathbf{S}'|^2+|\mathbf{S}''|^2=\frac{S^2}{2\Sigma_n|\varepsilon_n|^2}, \tag{17}$$

along with the following set of conditions on ε_n:

$$\sum_{n'=0}^{n}(\varepsilon_{n-n'}\varepsilon^*_{-n'})+\sum_{n'=1}^{\infty}(\varepsilon_{n+n'}\varepsilon^*_{n'}+\varepsilon_{-n'}\varepsilon^*_{-n-n'})=0, \tag{18}$$

which have to be satisfied $\forall\ n>0$. The energy minimum conditions of Eqs. (9) and (13) become, on account of Eq. (16),

$$\begin{aligned}J_{\mathbf{Q}+n\mathbf{Q}_c}\varepsilon_n&-\sum_{n'=0}^{n}\lambda_{n'}\varepsilon_{n-n'}\\&+j_{\mathbf{Q}+(n-1/2)\mathbf{Q}_c}\varepsilon_{n-1}+j^*_{\mathbf{Q}+(n+1/2)\mathbf{Q}_c}\varepsilon_{n+1}\\&-\sum_{n'=1}^{\infty}(\lambda_{n+n'}\varepsilon_{-n'}+\lambda^*_{n'}\varepsilon_{n+n'})=0,\end{aligned} \tag{19}$$

$\forall\ n\geqslant 0$. Similar equations for $n<0$ are solved simultaneously with the above, provided that

$$\lambda_{-|n|}=\lambda^*_{|n|}, \tag{20}$$

in which case they are simply complex conjugates of Eqs. (13) and (19). We note that $\mathbf{S}_{-\mathbf{q}}=\mathbf{S}^*_{\mathbf{q}}$ because $\mathbf{S}_j$ are real, so Eqs. (20) just require that Lagrange multipliers used to account for the conditions (7) are also real. On the other hand, $\varepsilon_{-n}\neq\varepsilon^*_n$. The solution $\{\varepsilon_n;\lambda_n\}$ of Eqs. (18) and (19) determines the minimum-energy configuration of the equal-length spins through Eqs. (11), (16), and (17).

D. Recursion for the perturbation series and the leading-order solution

Although they superficially look cumbersome, Eqs. (18) and (19) are well suited for the perturbation treatment. Indeed, because

$$\lambda_n\mathbf{S}_{\mathbf{Q}+n'\mathbf{Q}_c}\sim O(\epsilon^{|n|+|n'|}), \tag{21}$$

it is easy to see that the first line in Eq. (19) is $\sim\epsilon^n$, the second line, except for $n=0$, when both terms in it are $\sim\epsilon^2$, contains a term $\sim\epsilon^n$, and a term $\sim\epsilon^{n+2}$, and the last line, like the last sum in Eq. (18), sums up contributions $\sim\epsilon^{n+2n'}$, $n'=1,2,...$, i.e., overall is $O(\epsilon^{n+2})$. Starting with $\varepsilon_0=1$, this defines a set of recursion relations which determine $\{\varepsilon_n;\lambda_n\}$ for any given n through their values for $1,2,...,n-1$. In particular, the leading-order, $\sim O(\epsilon^n)$, condition on ε_n is

$$\varepsilon_n+\varepsilon^*_{-n}=-\sum_{n'=1}^{n-1}(\varepsilon_{n-n'}\varepsilon^*_{-n'})+O(\epsilon^{n+2}), \tag{22}$$

or, explicitly for the first few orders,

$$\varepsilon_1+\varepsilon^*_{-1}=O(\epsilon^3), \tag{23}$$

$$\varepsilon_2+\varepsilon^*_{-2}=\varepsilon_1^2+O(\epsilon^4), \tag{24}$$

$$\varepsilon_3+\varepsilon^*_{-3}=\varepsilon_1(\varepsilon_2-\varepsilon^*_{-2})+O(\epsilon^5), \tag{25}$$

$$\cdots.$$

The leading-order correction to λ_0, which determines the ground-state energy, appears in the second-order perturbation, $\sim\epsilon^2$. In the same order appears the intensity of the new magnetic Bragg peaks, $\sim|\mathbf{S}_{\mathbf{Q}\pm\mathbf{Q}_c}|^2$, that are induced by the exchange modulation. Up to this order, we obtain from Eq. (19)

$$\lambda_0=J_{\mathbf{Q}}+(j_{\mathbf{Q}-\mathbf{Q}_c/2}-\lambda_1)\varepsilon_{-1}+(j^*_{\mathbf{Q}+\mathbf{Q}_c/2}-\lambda^*_1)\varepsilon_1+O(\epsilon^4), \tag{26}$$

$$\lambda_1=j_{\mathbf{Q}+\mathbf{Q}_c/2}+(J_{\mathbf{Q}+\mathbf{Q}_c}-\lambda_0)\varepsilon_1+O(\epsilon^3), \tag{27}$$

$$\lambda^*_1=j^*_{\mathbf{Q}-\mathbf{Q}_c/2}+(J_{\mathbf{Q}-\mathbf{Q}_c}-\lambda_0)\varepsilon_{-1}+O(\epsilon^3), \tag{28}$$

which, on account of Eq. (23), have the solution

$$\varepsilon_1=\frac{j_{\mathbf{Q}-\mathbf{Q}_c/2}-j_{\mathbf{Q}+\mathbf{Q}_c/2}}{J_{\mathbf{Q}+\mathbf{Q}_c}+J_{\mathbf{Q}-\mathbf{Q}_c}-2J_{\mathbf{Q}}}+O(\epsilon^3), \tag{29}$$

with $\varepsilon_{-1}=-\varepsilon^*_1+O(\epsilon^3)$ (for now, we exclude from consideration a singular case, $J_{\mathbf{Q}+\mathbf{Q}_c}+J_{\mathbf{Q}-\mathbf{Q}_c}=2J_{\mathbf{Q}}$). The ground-state energy is then determined by

$$\lambda_0=J_{\mathbf{Q}}-\frac{|j_{\mathbf{Q}-\mathbf{Q}_c/2}-j_{\mathbf{Q}+\mathbf{Q}_c/2}|^2}{J_{\mathbf{Q}+\mathbf{Q}_c}+J_{\mathbf{Q}-\mathbf{Q}_c}-2J_{\mathbf{Q}}}+O(\epsilon^4), \tag{30}$$

and, unless $j_{\mathbf{Q}-\mathbf{Q}_c/2}=j_{\mathbf{Q}+\mathbf{Q}_c/2}$ and the correction vanishes, it is lower than that of the initial, nondistorted exchange spiral, because $J_{\mathbf{Q}}$ is a minimum value of $J_{\mathbf{q}}$, and therefore $J_{\mathbf{Q}+\mathbf{Q}_c}+J_{\mathbf{Q}-\mathbf{Q}_c}-2J_{\mathbf{Q}}\geqslant 0$. This is in agreement with a very general argument, that a nonvanishing second-order perturbation correction always lowers the ground-state energy.

E. Some remarks

It is useful to express the results obtained in the previous section in terms of the spin-wave spectrum,

$$\omega_{\mathbf{q}}=S\sqrt{2(J_{\mathbf{q}}-J_{\mathbf{Q}})(J_{\mathbf{q}+\mathbf{Q}}+J_{\mathbf{q}-\mathbf{Q}}-2J_{\mathbf{Q}})}, \tag{31}$$

and the $\mathbf{q}$-dependent transverse (perpendicular to the spin plane) classical static staggered spin susceptibility,[37]

$$\chi_\perp(\mathbf{q}) = \frac{1}{2(J_\mathbf{q} - J_\mathbf{Q})}, \tag{32}$$

of the initial, nondistorted, single-$\mathbf{Q}$ exchange spiral. The leading new Fourier components of the spin density, $\mathbf{S}_{\mathbf{Q}\pm\mathbf{Q}_c}$ of Eqs. (16), (29), are

$$\mathbf{S}_{\mathbf{Q}+\mathbf{Q}_c} = \left(\frac{j_{\mathbf{Q}-\mathbf{Q}_c/2} - j_{\mathbf{Q}+\mathbf{Q}_c/2}}{\chi_\perp(\mathbf{Q}_c)\omega^2_{\mathbf{Q}_c} S^{-2}} \right) \mathbf{S}_\mathbf{Q} + O(\epsilon^3), \tag{33}$$

$$\mathbf{S}_{\mathbf{Q}-\mathbf{Q}_c} = \left(\frac{-j^*_{\mathbf{Q}-\mathbf{Q}_c/2} + j^*_{\mathbf{Q}+\mathbf{Q}_c/2}}{\chi_\perp(\mathbf{Q}_c)\omega^2_{\mathbf{Q}_c} S^{-2}} \right) \mathbf{S}_\mathbf{Q} + O(\epsilon^3). \tag{34}$$

It is clear now that the singular case mentioned in the previous section corresponds to the exchange modulation with the wave vector at which the spin-wave energy vanishes. In this case, unless the numerator in Eqs. (33) and (34) is also zero, the leading corrections diverge, and the perturbation approach fails. Generally, there are two soft spots in the spin-wave spectrum of the exchange spiral. They correspond to the Goldstone modes at $\mathbf{q}=0$ and at the magnetic ordering wave vector, $\mathbf{q}=\mathbf{Q}$. For $\mathbf{Q}_c=\mathbf{Q}$, the numerator, $(j_{\mathbf{Q}/2} - j_{3\mathbf{Q}/2})$, is, in general, nonzero and the corrections diverge. Unless $\mathbf{q}=0$ is a special (extremum) point of $j_\mathbf{q}$, the numerators in Eqs. (33) and (34) vanish $\sim Q_c$ in the limit $\mathbf{Q}_c \to 0$, while $\omega^2_{\mathbf{Q}_c}$ in the denominator is $\lesssim Q_c^2$. Therefore for a sufficiently long-wavelength distortion the corrections may become arbitrarily large. It is not at all unexpected, though, that the perturbation approach fails extrapolation to $\mathbf{Q}_c=0$, where the modulation is absent, and $\mathbf{S}_{\mathbf{Q}\pm\mathbf{Q}_c} \equiv \mathbf{S}_\mathbf{Q}$.

Additional soft regions, such as the lines of soft modes, often appear in frustrated spin systems, due to the accidental cancellation of the interactions. In such cases the spin system is extremely sensitive to structural modulation with the wave vector that is close to the soft region(s) of the dispersion. The same is true for distortions that propagate along the direction(s) of weak interaction (and weak magnon dispersion) in quasi-low-dimensional spin systems.

From Eqs. (10) and (30), the ground-state energy is

$$\frac{E_{GS}}{N} = J_\mathbf{Q} S^2 - \frac{|j_{\mathbf{Q}-\mathbf{Q}_c/2} - j_{\mathbf{Q}+\mathbf{Q}_c/2}|^2 S^4}{\chi_\perp(\mathbf{Q}_c)\omega^2_{\mathbf{Q}_c}} + O(\epsilon^4). \tag{35}$$

In the general case, it is lowered in response to the exchange modulation. This occurs as a result of the appropriate adjustment (bunching) of the initial, single-$\mathbf{Q}$ spiral spin structure through the appearance of the additional Fourier harmonics, $\mathbf{S}_{\mathbf{Q}+n\mathbf{Q}_c}$, $n=\pm1,\pm2,\ldots$. In addition, the pitch of the primary spiral component, $\mathbf{S}_\mathbf{Q}$, may also change, $\mathbf{Q}\to\tilde{\mathbf{Q}}$, because the spiral propagation vector $\tilde{\mathbf{Q}}$ is now defined by the minimum of the corrected energy, Eq. (30) or Eq. (35). In the case where the modulation of the crystal structure is long periodic, i.e., for small $Q_c \ll 1$, and assuming that correction to the propagation vector, $\delta\mathbf{Q} = \tilde{\mathbf{Q}} - \mathbf{Q}$, is small, $|\delta Q| \ll Q$, we can expand $J_{\tilde{\mathbf{Q}}\pm\mathbf{Q}_c}$ and $j_{\tilde{\mathbf{Q}}\pm\mathbf{Q}_c/2}$ in Taylor series and obtain

$$\lambda_0 = J_\mathbf{Q} + \frac{1}{2}(\delta\mathbf{Q}\cdot\boldsymbol{\nabla})^2 J_\mathbf{Q} - \frac{3|(\mathbf{Q}_c\cdot\boldsymbol{\nabla}) j_\mathbf{Q}|^2}{(\delta\mathbf{Q}\cdot\boldsymbol{\nabla})[(\mathbf{Q}_c\cdot\boldsymbol{\nabla})^2 J_\mathbf{Q}] + 3(\mathbf{Q}_c\cdot\boldsymbol{\nabla})^2 J_\mathbf{Q}} + O(\epsilon^3), \tag{36}$$

where $\boldsymbol{\nabla} = \partial/\partial\mathbf{q}$. Note that unlike Eqs. (33) and (34) this, generally, does not diverge for $\mathbf{Q}_c \to 0$. If, in the correction term, we cancel Q_c^2 and then expand the denominator keeping only the leading term in δQ, Eq. (36) takes the form $\lambda_0 = J_\mathbf{Q} + c_0 + c_1(\delta Q) + c_2(\delta Q)^2$, with $c_1 = O(\epsilon^2)$ and $c_2 = O(\epsilon)$. Clearly, minimum of this expression occurs, in the general case, for nonzero $\delta Q = O(\epsilon)$. However, if the linear term $\sim \delta Q$ in the denominator of Eq. (36) vanishes, then $\delta\mathbf{Q}=0$. Also, in many important cases such as, for example, the nearest-neighbor nonfrustrated antiferromagnet, $\mathbf{Q}$ is a special symmetry point of $j_\mathbf{Q}$ ($j_\mathbf{Q} \sim \epsilon J_\mathbf{Q}$), and the correction term identically vanishes by symmetry. Therefore simple exchange structures, such as antiferromagnets, are usually insensitive to small long-periodic modulations of the exchange coupling in the spin Hamiltonian (5).

IV. SOME EXAMPLES

Now we shall apply the formalism developed above to several representative one- and two-dimensional (1D and 2D) systems. The fact that the ordered mean-field (MF) ground state, such as that analyzed in this paper, in low dimension is unstable against the fluctuations[38] will not be a concern here. First, a single-$\mathbf{Q}_c$ structural distortion considered in this paper is homogeneous within the crystallographic planes that are perpendicular to $\mathbf{Q}_c$. In many cases these planes contain one or two unit-cell directions, so the modulation only exists along the remaining direction(s), and the distortion is explicitly 2D, or 1D, respectively (cf. Fig. 1). The bonds that are not changed by the distortion cancel out in the resulting expressions for the corrections to the spin structure and to the ground-state energy, Eqs. (29) and (30). Therefore, even though the Hamiltonian (1) may be on a 3D lattice, the distortion corrections will be the same as for the lower-dimensional system. Also, in many quasi-low-dimensional materials the essential physics is 1D or 2D, while the MF order is stabilized by weak interaction in the remaining direction(s). Such is the case of high-T_c cuprates which are made of two-dimensional square-lattice layers. In the presence of a charge order the layers are modulated, as illustrated in Fig. 1(b). In fact, the MF analysis has certain value even for purely low-dimensional systems, because it highlights possible phases and instabilities, and guides the behavior of the critical points, at least asymptotic ($S\to\infty$).

A. *n*-merized 1D antiferromagnetic chain

As a simple example, consider the antiferromagnetic spin chain with *n*-periodic nearest-neighbor exchange coupling (the ferromagnetic case, $J<0$, is trivial, because small bond modulation has no effect on the ground state). It corresponds

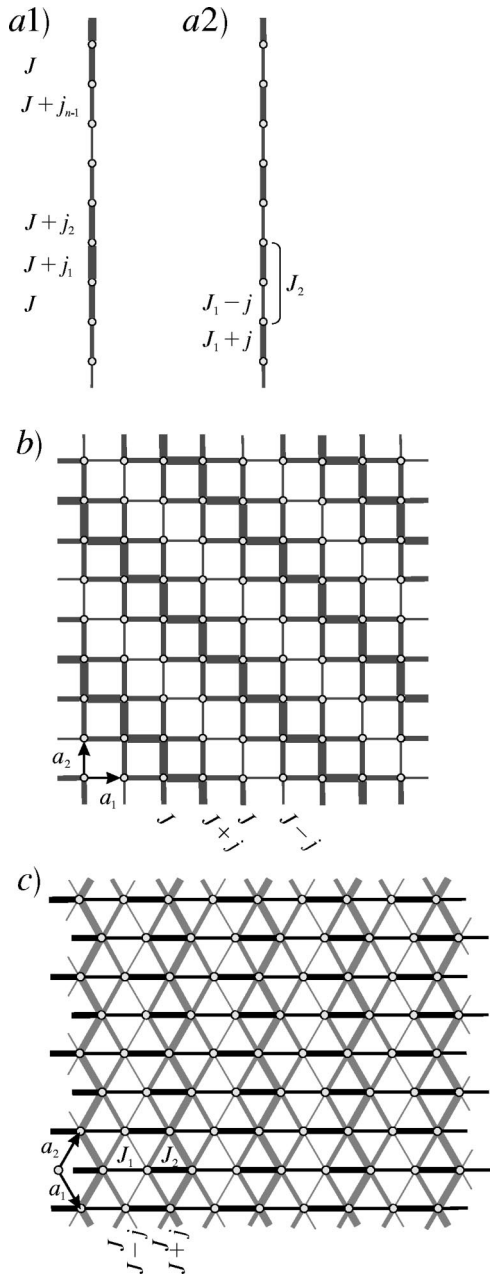

FIG. 1. Examples of the modulated exchange patterns discussed in the text. Different bond thicknesses illustrate different coupling strengths. (a1) *n*-merized 1D chain with modulated nearest-neighbor coupling, $Q_c=2\pi/n$; (a2) dimerized ($n=2$) antiferromagnetic chain with frustrating next-nearest neighbor coupling, $4J_2 \geqslant J_1$. (b) "Stripes" on a square lattice with diagonal modulation, $Q_c=(2\pi/n,2\pi/n)$; $n=4$ case is shown. (c) Generalized "staggered row model" on a triangular lattice, obtained with $Q_c=(2\pi/n,2\pi/n)$. Shown is the case of $n=4$; n equals 8 in the practical case of $RbMnBr_3$.

to a modulation with $Q_c=2\pi/n$, Fig. 1(a1). In this case, $J_{\mathbf{q}}=2J\cos q$, $j_{\mathbf{q}}=2j\cos q$, the modified GS energy is determined by

$$\lambda_0=2J\cos Q(1+|j/J|^2\tan^2 Q), \tag{37}$$

and does not depend on n. Clearly, there are new local extrema at $\cos^2 Q\approx|j/J|^2$, corresponding to a spiral with pitch of $\approx\pi/2$, which appear on account of the exchange modulation. However, the new minimum is very shallow, $\lambda_0\approx-4|j|$, much higher in energy than the global minimum, $\lambda_0=-2J$, which remains at $Q=\pi$.

A more interesting 1D example is illustrated in Fig. 1(a2), where in addition to the nearest-neighbor coupling J_1 there is a frustrating next-nearest-neighbor exchange J_2. In this case $J_{\mathbf{q}}=2J_1\cos q+2J_2\cos 2q$ and, for $4J_2\geqslant J_1$, frustration (for classical spins) results in a spiral MF ground state, with a pitch defined from the condition $\cos Q=-J_1/4J_2$. For the sake of simplicity we consider only modulation of the nearest-neighbor coupling, i.e., $j_2=0$. In this case, as before, $j_{\mathbf{q}}=2j_1\cos q$, and

$$\lambda_0=J_Q+\frac{2|j_1|^2\sin^2 Q}{J_1\cos Q+4J_2\cos^2\dfrac{\pi}{n}\cos 2Q}. \tag{38}$$

The minimum of this expression is achieved for $\tilde{Q}=Q+\delta Q$ which differs from $Q=\cos^{-1}(-J_1/4J_2)$. For small δQ the leading correction is

$$\delta Q=\left|\frac{j_1}{2J_2}\right|^2\frac{\cos^2 Q-\cos\dfrac{2\pi}{n}}{\tan Q\left(1-\cos\dfrac{2\pi}{n}\cos 2Q\right)^2}, \tag{39}$$

and diverges for $Q\to\pi$, i.e., for a frustrated 1D antiferromagnet with $J_2=0.25J_1$. Perhaps this interesting finding is an indication of an instability towards a spontaneous *n*-merization and a spin gap formation in the vicinity of this point. In fact, for $S=\frac{1}{2}$ quantum spin chain the whole region $J_2>J_{2c}\approx 0.25J_1$ is believed to belong to a spin-gap phase,[39–41] including the special point, $J_2=0.5J_1$, where the spontaneously dimerized ground state is known exactly.[42] While the most recent numerical estimate for $S=\frac{1}{2}$ chain is $J_{2c}\approx 0.24J_1$,[41] a $J_{2c}=0.25J_1$ for $S\to\infty$ coincides with the earlier result obtained from the semiclassical mapping on the nonlinear sigma model.[39] Clearly, studying the susceptibility of spin system towards bond modulation is a proper way to investigate its instability towards *n*-merization (plaquette formation) and to characterize the corresponding phase diagram. To this end it seems possible to develop a perturbation approach similar to the one presented here and starting with the Hamiltonian (5) also for quantum spins, but this goes far beyond the scope of this paper.

B. Square-lattice antiferromagnet with diagonal modulation

A square-lattice, nearest-neighbor antiferromagnet with diagonal modulation corresponds to $\mathbf{Q}_c=(2\pi/n,2\pi/n)$, Fig. 1(b), and may be of direct relevance for the charge-ordered stripe phases in doped LSCO cuprates and related perovskites. In the isotropic case the bond strengths and the modulation amplitudes are equal in two directions, $J_{\mathbf{q}}=2J[\cos(\mathbf{q}\cdot\mathbf{a}_1)+\cos(\mathbf{q}\cdot\mathbf{a}_2)]$, $j_{\mathbf{q}}=\epsilon J_{\mathbf{q}}$, and, upon switching to $Q=\mathbf{q}\cdot[(\mathbf{a}_1+\mathbf{a}_2)/2]$ and $Q'=\mathbf{q}\cdot[(\mathbf{a}_1-\mathbf{a}_2)/2]$, the problem is fac-

torized and reduced to the one-dimensional one considered above. The distortion-corrected spin GS energy is determined by λ_0 of Eq. (37), which is simply multiplied by $\cos Q'$. As before, we find that nearest-neighbor antiferromagnetism is stable with respect to the bond modulation. In fact, the same conclusion is reached even if the amplitude of the bond modulation is different in two directions, so that $j_{\mathbf{q}}=2j_1\cos(\mathbf{q}\cdot\mathbf{a}_1)+2j_2\cos(\mathbf{q}\cdot\mathbf{a}_2)$.

Again, an interesting situation occurs if there is frustration. In the case of the square-lattice antiferromagnet it is introduced by the diagonal, next-nearest-neighbor coupling, $J'>0$. This model has been extensively studied in recent years,[43–48] since it was predicted that in a region of parameters in the vicinity of $J/(2J')=1$ it has a disordered, spin-liquid ground state.[43,44] For $S=\frac{1}{2}$ quantum spins this was proposed as a possible candidate for the resonating-valence-bond state.[49,50] In this case, $J_{\mathbf{q}}=4J\cos Q\cos Q'+2J'(\cos 2Q+\cos 2Q')$, and, if both side and diagonal bonds are modulated, $j_{\mathbf{q}}=4j\cos Q\cos Q'+2j'(\cos 2Q+\cos 2Q')$. Consequently, we obtain from Eq. (30)

$$\lambda_0=J_{\mathbf{Q}}+\frac{4\sin^2 Q\left|j\cos Q'+2j'\cos\frac{\pi}{n}\cos Q\right|^2}{J\cos Q\cos Q'+2J'\cos^2\frac{\pi}{n}\cos 2Q}. \quad (40)$$

In the absence of bond modulation the ground state is determined by the hierarchy of the local minima of $J_{\mathbf{Q}}$. It depends on the relative strength of the nearest-neighbor coupling J, and the next-nearest-neighbor, diagonal coupling J', which is parametrized by $\alpha=J/2J'$. For weak frustration, $\alpha>1$, the global minimum is that with $\sin Q=\sin Q'=0$. It corresponds to the conventional, collinear Néel antiferromagnetic order with a single propagation vector $\mathbf{Q}=(\pi,\pi)$, and the ground-state energy is $(1/N)E_{(\pi,\pi)}=-4JS^2(1-1/2\alpha)$. Although there are four equivalent $\mathbf{Q}$ points in the Brillouin zone (BZ), $(\pm\pi,\pm\pi)$, $(\mp\pi,\pm\pi)$, which restore the lattice C_4 rotational symmetry, they are related through addition of the appropriate reciprocal-lattice vectors τ, so there is no true GS degeneracy in the $\mathbf{Q}$ space. The only GS degeneracy is the rotational symmetry in spin space which corresponds to the O(3) symmetry of the Heisenberg spin Hamiltonian.

For strong frustration, $\alpha\leqslant 1$, additional ground-state degeneracy occurs on the MF level. For $\alpha<1$ there are two nonequivalent lowest-energy minima of $J_{\mathbf{Q}}$, satisfying $\cos Q=\cos Q'=0$. They correspond to two pairs of equivalent $\mathbf{Q}$ points in the BZ $(\pm\pi,0)$ and $(0,\pm\pi)$, which represent the antiferromagnetic order propagating along the x and y axis, respectively. The GS energy is $(1/N)E_{(\pi,0)}=-4J'S^2=-4JS^2(1/2\alpha)$. This double degeneracy in $\mathbf{Q}$ space can be used to construct a continuum of states which are the linear combinations of the above two. This continuous GS degeneracy is usually described in terms of two decoupled $\sqrt{2}\times\sqrt{2}$ sublattices based on the diagonals of the original square lattice, which is transparent in the limit $J'\gg J$. Each sublattice has an antiferromagnetic order, but there may be an arbitrary angle between the two, because the mean field from one sublattice cancels on the sites of the other. This continuous degeneracy is lifted by zero-point or thermal fluctuations which prefer collinear arrangements of the two sublattices in the GS. This is a famous example of the order by disorder phenomenon in a frustrated magnet.[51,52]

The most interesting situation occurs for $\alpha=1$, when, on the MF level, there is a continuous GS degeneracy even in the $\mathbf{Q}$ space. The minimum condition for $J_{\mathbf{Q}}$ becomes $\cos Q=\cos Q'$, and is satisfied for any spiral with the propagation vector $\mathbf{Q}$ that belongs to the square with the vertices at $(\pm\pi,\pm\pi)$, $(\mp\pi,\pm\pi)$. All these states belong to the global minimum, and have the same energy, $(1/N)E_{\alpha=1}=-2JS^2=-4J'S^2$. It is this continuous $\mathbf{Q}$ space degeneracy which is at the origin of the spin-liquid phase conjectured for the values of α in a finite region around the special point $\alpha=1$.[43–47]

Importantly, for $\alpha\leqslant 1$ the spiral states with $\mathbf{Q}\approx(\pi,\pi)$ are in close competition with the collinear states. In particular, consider an extremum of $J_{\mathbf{Q}}$ which is a local minimum along the diagonal direction, parallel to the lattice modulation wave vector $\mathbf{Q}_c$, whose energy in the absence of the modulation is $(1/N)E_Q=-2\alpha JS^2$. It corresponds to a spiral with the propagation vector defined by $Q'=0$, $\cos Q=-J/2J'$, i.e., $\mathbf{Q}=(\cos^{-1}(-J/2J'),\cos^{-1}(-J/2J'))$ (there is also a degenerate state with $\mathbf{Q}$ at 90°, respecting the $Q\leftrightarrow Q'$ symmetry of the square lattice). Except for $\alpha=1$ the energy of this extremum is higher than that for the decoupled antiferromagnetic sublattices $E_{(\pi,0)}$. It is clear, however, from Eq. (40) that, while the energy of the collinear antiferromagnetic states is insensitive to bond modulation, the energy of the spiral GS can be lowered as it adapts to distortion! Therefore, at least on the MF level, the spiral may become the lowest energy state (i.e., the ground state) for some range of the parameter α in the vicinity of 1 [whose width is $\sim O(\epsilon^2)$]. For the long-periodic modulations, $Q_c\ll 1$, and for $j'=0$, it is easy to find that spiral phase is stable for $1-|j/J|^2\leqslant\alpha<1$. The ''nominal'' spiral propagation vector $\mathbf{Q}$ is obtained by minimizing Eq. (40), similarly with the case of the frustrated 1D chain. Again, our finding clearly indicates the instability of the frustrated square-lattice antiferromagnet with $J/(2J')$ close to 1 with respect to the bond-modulated states. By selecting spiral spin GS the ''order by distortion'' mechanism proposed here competes with the ''order by disorder'' phenomenon, which prefers collinear states. At least for large enough S spiral always wins in some vicinity of $\alpha=1$.

C. Generalized row models on triangular lattice

Because of inherent frustration, the triangular-lattice antiferromagnet, Fig. 1(c), is a very interesting case to consider. Without modulation, a minimum of $J_{\mathbf{q}}=2J\{\cos(\mathbf{q}\cdot\mathbf{a}_1)+\cos(\mathbf{q}\cdot\mathbf{a}_2)+\cos[\mathbf{q}\cdot(\mathbf{a}_1+\mathbf{a}_2)]\}$ is achieved for a commensurate spiral GS with propagation vector $\mathbf{Q}=(2\pi/3,2\pi/3)$. It corresponds to a noncollinear GS structure where spins are aligned along one of the three directions, at 120° with each other. Structural distortion may result in a variety of coupling patterns where equivalent bonds are related by translations perpendicular to $\mathbf{Q}_c$. These can be classified as ''generalized row models,'' the simplest of which is the original row model

of Ref. 7. It can arise, for example, as a second-order effect from the distortion with $\mathbf{Q}_c=(\pm\pi,\pi)$. Because $2\mathbf{Q}_c=\boldsymbol{\tau}$ ($\boldsymbol{\tau}$ is a reciprocal-lattice vector), it is equivalent to a homogeneous uniaxial compression, i.e., it simply changes $J_{\mathbf{q}}$ and does not result in a bond modulation. Nevertheless, the ground state becomes incommensurate, with pitch of the spiral determined by the coupling anisotropy.[7–9]

A general modulation of the triangular lattice with $\mathbf{Q}_c=(2\pi/n,-2\pi/n)$ results in a "staggered row model," where the horizontal rows have equal coupling. On the other hand, modulation with $\mathbf{Q}_c=(2\pi/n,2\pi/n)$ leads to a "zigzag row model," an example of which with $n=4$ is shown in Fig. 1(c), and the one with $n=8$ is relevant for the phases realized in $RbMnBr_3$ and $KNiCl_3$.[12–14] In this case $j_{\mathbf{q}}=4j\cos Q\cos Q'+2j\cos 2Q$ and it is easy to see that the energy of the modulated state is given by the same expression as for the frustrated square lattice, Eq. (40), but with $j'=j$ and $J'=J$. As in the previous example, we use $Q=\mathbf{q}\cdot[(\mathbf{a}_1+\mathbf{a}_2)/2]$ and $Q'=\mathbf{q}\cdot[(\mathbf{a}_1-\mathbf{a}_2)/2]$. Exchange modulation leads to a deviation from the commensurate 120° triangular spin structure. The leading correction to the propagation vector is determined from $\cos\tilde{Q}=-\frac{1}{2}+\delta$, where

$$\delta=\left|\frac{j}{J}\right|^2\frac{\left(\cos\frac{\pi}{n}-\cos\frac{2\pi}{n}\right)\left(\frac{5}{4}-\frac{9}{4}\cos\frac{\pi}{n}+\frac{5}{2}\cos^2\frac{\pi}{n}\right)}{2\left(\frac{1}{2}+\cos^2\frac{\pi}{n}\right)^2}. \tag{41}$$

In the two limiting cases, $n=2$ and $n\gg 1$, we find $\delta=\frac{5}{2}|j/J|^2$ and $\delta\approx(\pi/2n)^2|j/J|^2$, respectively. It is clear that δ is small for all n, so there is no evidence for an instability towards n-merization in the case of ideal triangular lattice. Perhaps, such evidence can be found in the anisotropic, quasi-1D case, where exchange in zigzag rows is much smaller than that in straight rows, or vice versa.

V. SUMMARY AND CONCLUSIONS

In a great variety of important practical cases the complex crystal structure which is at the origin of the intricate magnetic behavior in magnetic material results from a small superstructural distortion of a much simpler structure, in which the magnetic ions form a primitive Bravais lattice. Reduction of the crystal symmetry related to the appearance of even a single, commensurate with the original lattice, superstructural Bragg reflection at a wave vector $\mathbf{Q}_c$ formally requires folding the original Brillouin zone to a much smaller one, many times reducing its volume. Consequently, the magnetic system is usually described in terms of multiple spin sites, and multiple sheets of spin excitations. Not only does this greatly complicate understanding and predicting magnetic properties, such violent modification of the $\mathbf{q}$ space clearly seems an unsatisfactory way to account for a small distortion of the crystal structure. Moreover, BZ folding is not an option for the incommensurate structural modulations, such as arise in various charge-density-wave ordered states.

In this paper the effect of a small lattice modulation with single propagation vector $\mathbf{Q}_c$ on the system of localized spins, coupled by Heisenberg exchange interaction (1), was considered. It was found that lattice distortion results in a modulation of the exchange coupling which, to the leading order, is parametrized by no more than two constants per bond, Eq. (5) (this is valid irrespective of whether the spins are quantum or classical). There are also corrections of the order $O(\epsilon^2)$ (ϵ is a small parameter that parametrizes the lattice distortion) to the exchange constants J_{ij} in the covariant part of the Hamiltonian. It should be noted here that, although the distortion considered is small, the resulting corrections to the coupling constants need not be small compared to the initial values of the couplings in the spin Hamiltonian, which may be small themselves. Therefore the result expressed by Eq. (5) is quite general and does not automatically imply the condition $|j_{ij}|\ll|J_{ij}|$. In other words, the umklapp terms $\sim S^{\alpha}_{\mathbf{q}}S^{\alpha}_{\mathbf{q}\pm\mathbf{Q}_c}$ added to the spin Hamiltonian by small lattice distortion may be relatively large, even larger than the original exchange interactions.

While it would be interesting to study the modulated-exchange Hamiltonian for quantum spins and for the arbitrary values of $|j_{ij}/J_{ij}|$, it is a formidable task which is beyond the scope of this paper. Here we developed a perturbative scheme for finding the mean-field ground state of the Hamiltonian (5) which is valid for classical spins, $S\gg 1$, and in the case of small exchange modulation, $|j_{ij}/J_{ij}|\sim\epsilon\ll 1$. One of the initial motivations here was to develop a basis for the spin-wave theory in weakly distorted crystal structures in terms of the modification of the magnon spectrum in the original, large Brillouin zone of the nondistorted Bravais lattice. The other, no less important motivation, was to see whether it would be possible to understand, already on the mean-field level, the incommensurate phases observed in the distorted triangular-lattice antiferromagnets $RbMnBr_3$ and $KNiCl_3$, and in the doped, distorted square-lattice antiferromagnets, such as LSCO or related Ni, Mn, and Co materials, such as $La_{1.5}Sr_{0.5}CoO_4$.[53]

The essential results of this paper are expressed by Eqs. (29) and (30), or, equivalently, by Eqs. (33)–(35). They show that a transverse, equal-spin spiral structure, which is the ground state of the initial Heisenberg Hamiltonian, adapts to the exchange modulation through appearance of the additional Fourier harmonics, $\mathbf{S}_{\mathbf{Q}+n\mathbf{Q}_c}$, $n=\pm 1,\pm 2,...$ (bunching). As a result, in a general case the GS energy is lowered by exchange modulation. In addition, the pitch of the primary spiral component $\mathbf{S}_{\mathbf{Q}}$ may also change, $\mathbf{Q}\to\tilde{\mathbf{Q}}$, because it is now defined by the minimum of modulation-corrected energy, Eq. (35).

Applying these results to several particular examples of the topical frustrated spin systems appears quite revealing. We find that in the case of the frustrated square-lattice antiferromagnet with diagonal coupling J', such that $\alpha=J/(2J')$ is close to 1, lattice modulation opens a region of stability of the incommensurate spiral phase. This "order by distortion" phenomenon[54] competes with "order by disorder," which prefers collinear arrangements of two antiferromagnetic sublattices. The incommensurate spiral phase with the propagation vector $\tilde{\mathbf{Q}}=(\pi\pm\delta,\pi\pm\delta)$ close to (π,π) wins for the range $O(\epsilon^2)$ of the parameter α in the vicinity of α

$=1$. This provides a plausible explanation for the incommensurate spin order observed in $La_{1.5}Sr_{0.5}CoO_4$,[53] and in a number of other doped perovskites, and may also be of a direct relevance for the doped LSCO materials. It is important to mention here that incommensurate spin-ordered phases are among the most interesting and puzzling features of doped layered perovskites. In the absence of distortion one needs at least a third-neighbor coupling in order to stabilize spiral MF ground state for the Heisenberg spin Hamiltonian on square lattice.

Exchange modulation in Heisenberg antiferromagnet on a distorted triangular-lattice leads to an incommensurate shift of the spiral propagation vector, in qualitative agreement with what is observed in $RbMnBr_3$. However, Eq. (41) implies that $\tilde{\mathbf{Q}}$ is decreased compared to $\mathbf{Q}=(2\pi/3,2\pi/3)$ for the ideal triangular lattice, while $\tilde{\mathbf{Q}}\approx 2\pi\cdot(0.357,0.357)$ is observed in experiments.[13,14] Therefore it is likely that the shift in $RbMnBr_3$ is mainly due to the anisotropic corrections to the nearest-neighbor coupling, which are captured already in the simplest row model.[8,9] Nevertheless, correction of Eq. (41) is not unimportant. In the case of $n=4$, which may be relevant for $RbMnBr_3$, it gives the same magnitude shift of the ordering wave vector, δQ, as measured in experiment, for $|j/J|^2\approx 0.2$ (for $n=8$, $|j/J|^2\approx 1$ is needed).

More importantly, bunching of the spin spiral as a result of the lattice distortion provides, already on the mean-field level, a possible explanation for the commensuration transition in $RbMnBr_3$ and for the long-periodic lattice-commensurate structure in the related phase of $KNiCl_3$. Indeed, an easy-axis anisotropy, such as in $KNiCl_3$, or a magnetic field applied within the easy plane, as in $RbMnBr_3$, also lead to bunching of the exchange spiral, generating additional Fourier harmonics, $\mathbf{S_q}$, at $\mathbf{q}=n\tilde{\mathbf{Q}}$, $n=\pm 2,\pm 3,\ldots$.[26] Appearance of these Fourier components in the spin distribution lowers the spin anisotropy and the Zeeman energy, but competes with the modulated exchange, which requires additional Fourier components at $\mathbf{q}=\tilde{\mathbf{Q}}+n\mathbf{Q}_c$. Therefore for some finite value of the easy-axis anisotropy, or the in-plane magnetic field, a commensuration transition may be expected, where $\mathbf{Q}$ becomes equal to $m\mathbf{Q}_c$ with some integer m. In the lattice-commensurate phase both sets of additional harmonics coincide, and both the modulated exchange energy, and the spin anisotropy and Zeeman energy, can be lowered simultaneously (this, of course, should offset the increase in the unmodulated exchange energy caused by the shift in $\mathbf{Q}$). Extending the results of this paper and those of Ref. 26 to the Hamiltonian (1) with modulated exchange of the Eq. (5) and D, $H\neq 0$ in order to map out such phase diagram is one of the most obvious directions for further studies.

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APPENDIX: EFFECT OF LATTICE MODULATION ON EXCHANGE COUPLING

The simplest assumption, and the one which is most often employed in literature,[55] is that the exchange coupling J_{ij} between the magnetic ions (spins) at positions $\mathbf{r}_i$ and $\mathbf{r}_j$ only depends on the distance between the sites, $J_{ij}=J(|\mathbf{r}_{ij}|)$. However, in magnetic dielectrics this coupling most often results from the superexchange and therefore generally also depends on the position(s) of ligand ions which bridge the superexchange path,

$$J_{ij}=J(|\mathbf{r}_{ij}|,\{\mathbf{r}^{\mu_{ij}}\})=J(r_{ij},\{\mathbf{u}^{\mu_{ij}}\}). \tag{A1}$$

Here μ_{ij} numbers the ligands that participate in the ij bond and $\mathbf{r}^{\mu_{ij}}$ are their positions which are most naturally parametrized in terms of the offsets, $\mathbf{u}^{\mu_{ij}}=\mathbf{r}^{\mu_{ij}}-\mathbf{R}_{ij}$, from the bond center, $\mathbf{R}_{ij}=\frac{1}{2}(\mathbf{R}_i+\mathbf{R}_j)$.

In addition, the superexchange coupling may also depend on the angles of orbital overlaps. This dependence can be parametrized in terms of the positions of some particular symmetry points in the local electron-density distribution, and accounted for in Eq. (A1) by including these points among $\{\mathbf{r}^{\mu_{ij}}\}$. While these additional degrees of freedom do lift some nonessential symmetries which are present in the particular case when the exchange coupling only depends on the bond length, $J_{ij}=J(|\mathbf{r}_{ij}|)$, they do not change the general structure of the corrections to the exchange coupling resulting from the lattice modulation which are summarized by Eq. (3) in the main text. In what follows, we first discuss the particular case of $J_{ij}=J(|\mathbf{r}_{ij}|)$, and then the general case of Eq. (A1).

1. Modulation of the bond length only

First, consider the effect of displacement of the magnetic ions alone. In presence of the superstructural modulation (2), the bond lengths become $r'_{ij}=|\mathbf{r}_{ij}+\boldsymbol{\epsilon}_{ij}|$, with $\boldsymbol{\epsilon}_{ij}=\boldsymbol{\epsilon}_j-\boldsymbol{\epsilon}_i$ given by

$$\boldsymbol{\epsilon}_{ij}=2\sin\left(\frac{\mathbf{Q}_c\cdot\mathbf{r}_{ij}}{2}\right)[-\boldsymbol{\epsilon}_1\sin(\mathbf{Q}_c\cdot\mathbf{R}_{ij})+\boldsymbol{\epsilon}_2\cos(\mathbf{Q}_c\cdot\mathbf{R}_{ij})], \tag{A2}$$

where, as usual, $\mathbf{r}_{ij}=\mathbf{r}_j-\mathbf{r}_i$ and $\mathbf{R}_{ij}=\frac{1}{2}(\mathbf{R}_i+\mathbf{R}_j)$.

Expanding the exchange coupling, $J'_{ij}=J(r'_{ij})$, modified by the distortion (A2) in a Taylor series in small displacement, $\boldsymbol{\epsilon}_{ij}\ll r_{ij}$,

$$J'_{ij}=J_{ij}+\sum_n\frac{1}{n!}\left(\boldsymbol{\epsilon}_{ij}\cdot\frac{\partial}{\partial\mathbf{r}_{ij}}\right)^n J(r_{ij}), \tag{A3}$$

we find, up to a second order in $(\boldsymbol{\epsilon}_{ij}/r_{ij})\sim\epsilon\ll 1$,

$$J'_{ij}=J_{ij}+\frac{(\boldsymbol{\epsilon}_{ij}\cdot\mathbf{r}_{ij})}{r_{ij}}\frac{\partial J(r_{ij})}{\partial r_{ij}}+\frac{1}{2}\frac{(\boldsymbol{\epsilon}_{ij}\cdot\mathbf{r}_{ij})^2}{r_{ij}^2}\frac{\partial^2 J(r_{ij})}{\partial r_{ij}^2}+\frac{1}{2}\left(\frac{\boldsymbol{\epsilon}_{ij}^2}{r_{ij}}-\frac{(\boldsymbol{\epsilon}_{ij}\cdot\mathbf{r}_{ij})^2}{r_{ij}^3}\right)\frac{\partial J(r_{ij})}{\partial r_{ij}}+O(\epsilon^3). \tag{A4}$$

For $(\boldsymbol{\epsilon}_{ij}\cdot\mathbf{r}_{ij})\neq 0$ and $\partial J(r_{ij})/\partial r_{ij}\neq 0$ the leading contribution is given by the first-order term, $\sim\boldsymbol{\epsilon}$, and we obtain,

$$J'_{ij}=J_{ij}+j'_{ij}\cos(\mathbf{Q}_c\cdot\mathbf{R}_{ij})-j''_{ij}\sin(\mathbf{Q}_c\cdot\mathbf{R}_{ij})+O(\epsilon^2), \tag{A5}$$

where

$$j'_{ij}=2\sin\left(\frac{\mathbf{Q}_c\cdot\mathbf{r}_{ij}}{2}\right)\frac{\partial J(r_{ij})}{r_{ij}\partial r_{ij}}(\boldsymbol{\epsilon}_2\cdot\mathbf{r}_{ij}), \tag{A6}$$

$$j''_{ij}=2\sin\left(\frac{\mathbf{Q}_c\cdot\mathbf{r}_{ij}}{2}\right)\frac{\partial J(r_{ij})}{r_{ij}\partial r_{ij}}(\boldsymbol{\epsilon}_1\cdot\mathbf{r}_{ij}). \tag{A7}$$

Clearly, to the first order, there is no $\mathbf{Q}_c$-independent correction which would change the bond strength J_{ij}. Such a correction does appear in the second order in $\boldsymbol{\epsilon}$.

If $(\boldsymbol{\epsilon}_{ij}\cdot\mathbf{r}_{ij})=0$, i.e., the displacements are perpendicular to the bonds, the leading correction is given by the second-order term $\sim\boldsymbol{\epsilon}_{ij}^2$. In fact, this important situation is often encountered in practice, in particular, it is the basis for the so-called ''row models'' for triangular-lattice antiferromagnets. These are typically thought to be realized through a modulation of the hexagonal lattice where the displacements of the magnetic ions are parallel to the C_6 symmetry axis, and are perpendicular to the bonds in the hexagonal plane in which the modulation propagates. $RbMnBr_3$ is believed to present an example of such situation. For such case of a transverse structural modulation we obtain

$$J'_{ij}=J_{ij}+\tilde{j}'_{ij}\cos(2\mathbf{Q}_c\cdot\mathbf{R}_{ij})-\tilde{j}''_{ij}\sin(2\mathbf{Q}_c\cdot\mathbf{R}_{ij}) + s_{ij}[(\tilde{j}'_{ij})^2+(\tilde{j}''_{ij})^2]^{1/2}+O((\boldsymbol{\epsilon}_{ij}\cdot\mathbf{r}_{ij})^2,\epsilon^3), \tag{A8}$$

where $s_{ij}=\mathrm{sgn}[\partial J(r_{ij})/\partial r_{ij}]$, and

$$\tilde{j}'_{ij}=\sin^2\left(\frac{\mathbf{Q}_c\cdot\mathbf{r}_{ij}}{2}\right)\frac{\partial J(r_{ij})}{r_{ij}\partial r_{ij}}(\boldsymbol{\epsilon}_2^2-\boldsymbol{\epsilon}_1^2), \tag{A9}$$

$$\tilde{j}''_{ij}=\sin^2\left(\frac{\mathbf{Q}_c\cdot\mathbf{r}_{ij}}{2}\right)\frac{\partial J(r_{ij})}{r_{ij}\partial r_{ij}}2(\boldsymbol{\epsilon}_1\cdot\boldsymbol{\epsilon}_2). \tag{A10}$$

The first-order correction to J'_{ij} also vanish if $\partial J(r_{ij})/\partial r_{ij}=0$. In this case the leading contribution comes from the last second-order term in Eq. (A4), and is also expressed by Eq. (A8), but with $s_{ij}=\mathrm{sgn}[\partial^2 J(r_{ij})/\partial r_{ij}^2]$, and

$$\tilde{j}'_{ij}=\sin^2\left(\frac{\mathbf{Q}_c\cdot\mathbf{r}_{ij}}{2}\right)\frac{\partial^2 J(r_{ij})}{r_{ij}^2\partial r_{ij}^2}[(\boldsymbol{\epsilon}_2\cdot\mathbf{r}_{ij})^2-(\boldsymbol{\epsilon}_1\cdot\mathbf{r}_{ij})^2], \tag{A11}$$

$$\tilde{j}''_{ij}=\sin^2\left(\frac{\mathbf{Q}_c\cdot\mathbf{r}_{ij}}{2}\right)\frac{\partial^2 J(r_{ij})}{r_{ij}^2\partial r_{ij}^2}2(\boldsymbol{\epsilon}_1\cdot\mathbf{r}_{ij})(\boldsymbol{\epsilon}_2\cdot\mathbf{r}_{ij}), \tag{A12}$$

in place of Eqs. (A9) and (A10).

A $\mathbf{Q}_c$-independent correction which changes the bond strength J_{ij} first appears in the second order, Eq. (A8). In general, it is obtained by summing up all $\mathbf{Q}_c$-independent contributions from all second-order terms in Eq. (A4). Their common multiplier, $\sin^2(\mathbf{Q}_c\cdot\mathbf{r}_{ij}/2)$, makes the structure of this correction rather simple. It does not affect the bonds that are perpendicular to the direction of propagation of the lattice distortion, while those bonds that are symmetric with respect to this direction are modified equally. On the other hand, j'_{ji} and j''_{ij} of Eqs. (A6) and (A7) which describe the first-order correction to the exchange coupling resulting from the lattice modulation are $\sim\sin(\mathbf{Q}_c\cdot\mathbf{r}_{ij}/2)$, and therefore are antisymmetric with respect to $\mathbf{Q}\rightarrow-\mathbf{Q}$.

2. Modulation of the ligand positions, etc.

It is also straightforward to account for the dependence of the superexchange coupling, Eq. (A1), on the positions $\mathbf{u}^{\mu_{ij}}$ of the ligand ions and the symmetry points of the local electron density distribution which define the orbital overlaps. In the presence of the superstructural distortion (2),

$$(\mathbf{r}^{\mu_{ij}})'=\mathbf{r}^{\mu_{ij}}+\boldsymbol{\epsilon}_1^{\mu}\cos(\mathbf{Q}_c\cdot\mathbf{r}^{\mu_{ij}})+\boldsymbol{\epsilon}_2^{\mu}\sin(\mathbf{Q}_c\cdot\mathbf{r}^{\mu_{ij}}), \tag{A13}$$

where μ indexes different types of $\mathbf{r}^{\mu_{ij}}$ positions within the unit cell, and the polarization vectors $\boldsymbol{\epsilon}_{1,2}^{\mu}$ which parametrize the displacement for point of type μ are determined by the superlattice Bragg intensities that appear with distortion. This can be rewritten as

$$(\mathbf{u}^{\mu_{ij}})'=\mathbf{u}^{\mu_{ij}}+\boldsymbol{\epsilon}_1^{\mu_{ij}}\cos(\mathbf{Q}_c\cdot\mathbf{R}_{ij})+\boldsymbol{\epsilon}_2^{\mu_{ij}}\sin(\mathbf{Q}_c\cdot\mathbf{R}_{ij}), \tag{A14}$$

where the new polarization vectors $\boldsymbol{\epsilon}_{1,2}^{\mu_{ij}}$ now depend on $\mathbf{Q}_c$. They are obtained by rotating $\boldsymbol{\epsilon}_{1,2}^{\mu}$ through an angle $\phi_{\mu_{ij}}=(\mathbf{Q}_c\cdot\mathbf{u}^{\mu_{ij}})$, and subtracting $\boldsymbol{\epsilon}_{1,2}\cos(\frac{1}{2}\mathbf{Q}_c\cdot\mathbf{r}_{ij})$ (this accounts for change in the bond center position $\mathbf{R}_{ij}$), correspondingly.

Consequently, in the general case of Eq. (A1), the Taylor series (A3) for J'_{ij} has to be amended, by adding

$$J'_{ij}\rightarrow J'_{ij}+\sum_n\frac{1}{n!}\sum_{\mu_{ij}}\left(\boldsymbol{\epsilon}^{\mu_{ij}}\cdot\frac{\partial}{\partial\mathbf{u}^{\mu_{ij}}}\right)^n J(r_{ij},\{\mathbf{u}^{\mu_{ij}}\}), \tag{A15}$$

whose first- and second-order terms are easily rewritten in the form of Eqs. (A5) and (A8), respectively. Therefore the account for the modulation of the positions $\mathbf{u}^{\mu_{ij}}$ in the general expression for the superexchange, Eq. (A1), simply amounts to amending the coefficients j'_{ij} and j''_{ij} in Eqs. (A5) and (A8), in accordance with Eqs. (A14) and (A15). For example, additional first-order terms which appear in Eq. (A15) change the expressions of Eqs. (A6) and (A7) as follows:

$$j'_{ij}\rightarrow j'_{ij}+\sum_{\mu_{ij}}\left(\boldsymbol{\epsilon}_1^{\mu_{ij}}\cdot\frac{\partial}{\partial\mathbf{u}^{\mu_{ij}}}\right)J(r_{ij},\{\mathbf{u}^{\mu_{ij}}\}), \tag{A16}$$

$$j''_{ij}\rightarrow j'_{ij}-\sum_{\mu_{ij}}\left(\boldsymbol{\epsilon}_2^{\mu_{ij}}\cdot\frac{\partial}{\partial\mathbf{u}^{\mu_{ij}}}\right)J(r_{ij},\{\mathbf{u}^{\mu_{ij}}\}). \tag{A17}$$

Using Eqs. (A14) and (A15) it is easy to write out similar expressions for the coefficients of the second-order contribution of Eq. (A8).

Clearly, a number of symmetry properties of the coeffi-

cients j'_{ij} and j''_{ij} given by Eqs. (A6)–(A12) which are present for $J_{ij}=J(r_{ij})$, disappear upon account for the modulation of the positions $\mathbf{u}^{\mu_{ij}}$. In particular, for the first-order corrections to vanish, not only should the displacements of the lattice sites (magnetic ions) be perpendicular to the bonds, but all of the displacements $\boldsymbol{\epsilon}_{1,2}^{\mu_{ij}}$ should be perpendicular to the corresponding gradients of $J_{ij}=J(r_{ij},\{\mathbf{u}^{\mu_{ij}}\})$ with respect to $\mathbf{u}^{\mu_{ij}}$. However, it is clear that, because j'_{ij}, j''_{ij} and, in general, $\tilde{J}_{ij}$, are functions on the nondistorted lattice which also depend on the modulation wave vector $\mathbf{Q}_c$ and the polarizations $\boldsymbol{\epsilon}_{1,2}^{\mu_{ij}}$, they are invariant with respect to all symmetry operations of that initial lattice which do not change $\mathbf{Q}_c$ and $\boldsymbol{\epsilon}_{1,2}^{\mu_{ij}}$. Importantly, this includes the translation group of the nondistorted lattice, which means that the new couplings and the exchange modulation amplitudes which are related by that lattice translations are equal.

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