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Chiral domain walls in frustrated magnetoelastic materials

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We analyze domain walls on a two-dimensional triangular lattice with magnetoelastic coupling and find three phases: high symmetry (H) triangular, low symmetry rectangular, and spiral. The H-phase domains correspond to spin configurations of different chirality with local lattice rotation. We discuss the consequences of the interplay between chirality and magnetoelastic frustration, specifically how the latter is relieved.

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An antiferromagnetic spin configuration on a rigid two-dimensional (2D) triangular lattice is a frustrated system. If the lattice is deformable then this frustration can be relieved by the lattice adopting a (generally) lower symmetry via the magnetoelastic coupling. In addition, it is known that frustrated magnetic systems with a continuous spin symmetry may possess (an Isinglike) discrete degeneracy, namely, the right- and left-hand or chiral degeneracy. Here we illustrate these features by considering two-component XY spins on a triangular lattice and analyze various domain walls connecting domains with different chirality arising from the magnetoelastic interaction. An emerging theme here is that the magnetic frustration can be relieved by coupling to other degrees of freedom in a material, including elastic deformation as in the present case. Magnetoelastic interaction on a triangular lattice exists in materials such as solid oxygen,[1,2] $AM(SO_4)_2$ with $M=$Ti, Fe belonging to the Yavapaiite[3,4] family, and magnetic materials with face-centered cubic structure (with the [111] planes corresponding to the triangular lattice).

Chirality for the elementary triangle, with spins $\vec{S}_1$, $\vec{S}_2$, and $\vec{S}_3$ on the three vertices, is determined by the quantity[5]

$$\vec{K}_{123}=\vec{S}_1\times\vec{S}_2+\vec{S}_2\times\vec{S}_3+\vec{S}_3\times\vec{S}_1. \tag{1}$$

Two different phases that correspond to right- and left-handed spin structure are characterized by different signs of the parameter (1). In crystals with a center of inversion, the two ground states are degenerate. The chiral degeneracy is removed by applying either an electric[6] or a magnetic[7] field. Quite recently the possibility to control the sense of chirality by applying an elastic torsion was demonstrated.[8] The Hamiltonian of the spin subsystem on a triangular lattice, interacting with the displacements of the magnetic atoms, is

$$H=\frac{1}{2}\sum_{\vec{n},\vec{\delta}} J_{\vec{n},\vec{n}+\vec{\delta}}\vec{S}_{\vec{n}}\vec{S}_{\vec{n}+\vec{\delta}}. \tag{2}$$

Here $\vec{S}_{\vec{n}}$ is the spin of the atom located in the $\vec{n}$th site of the lattice: $\vec{n}=n_1\vec{c}_1+n_2\vec{c}_2$ $(n_1,n_2=0,\pm1,\pm2,\dots)$, where $\vec{c}_1=(1,0)$, $\vec{c}_2=(1/2,\sqrt{3}/2)$ are the basic vectors of the triangular lattice, $J_{\vec{n},\vec{n}+\vec{\delta}}\equiv J(|\vec{\delta}+\vec{u}_{\vec{n}+\vec{\delta}}-\vec{u}_{\vec{n}}|)$ is the exchange integral with $\vec{u}_{\vec{n}}$ being the atom displacements from their equilibrium positions (i.e., without magnetic interaction). $\vec{\delta}$ denotes the vector that connects a site with nearest neighbors.

We will consider a small spatially smooth deformation and assume that $(\vec{u}_{\vec{n}+\vec{\delta}}-\vec{u}_{\vec{n}})_i=u_{i,j}\,\delta_j$, $i,j=x,y$, where the notation $u_{i,j}=\partial u_i/\partial j$ is used. In this way the exchange integral $J_{\vec{n},\vec{n}+\vec{\delta}}$ can be written as $J_{\vec{n},\vec{n}+\vec{\delta}}=J(1-\eta\delta_i u_{i,j}\delta_j)$, where $J\equiv J(|\vec{\delta}|)$ $(J>0)$ is the exchange constant, the parameter $\eta=-d\ln J(|\vec{r}|)/d|\vec{r}|$ is a measure of the inverse radius of the exchange interaction and summation over repeated indices is assumed. The elastic energy density of the two-dimensional triangular lattice in the continuum approximation coincides with the elastic energy of the isotropic plane,

$$\Phi=\frac{1}{2}\int\{A_1e_1^2+A_2(e_2^2+e_3^2)\}d\vec{r}, \tag{3}$$

where $e_1=u_{x,x}+u_{y,y}$ is the dilatation and $e_2=u_{x,x}-u_{y,y}$, $e_3=u_{x,y}+u_{y,x}$ are shear strains. Here A_1 is the bulk modulus and A_2 is the shear modulus of the triangular material.[9] By using the spin variables in the form $\vec{S}_n=(\cos\phi_n,\sin\phi_n,0)$, we obtain the (spin) energy for Eq. (2) as

$$E_s=JS^2\sum_{\vec{n},\vec{\delta}}(1-\eta\delta_i u_{i,j}\delta_j)\cos(\phi_{\vec{n}-\vec{\delta}}-\phi_{\vec{n}}), \tag{4}$$

which contains both the magnetic (spin only) and magnetoelastic (spin coupled to strain) contributions to the energy.

There is an *additional* magnetoelastic contribution to the energy due to the interaction between spin chirality and lattice deformation, and satisfies both time-reversal as well as space-group symmetry operations. In the case of a triangular lattice, the simplest form of this energy is (see also Ref. 8)

$$F_\chi=\sum_{\vec{n},\vec{\delta}}C_{\vec{\delta}}(\vec{S}_{\vec{n}}\times\vec{S}_{\vec{n}-\vec{\delta}})_z\delta_j\partial_j\omega$$
$$=S^2\sum_{\vec{n},\vec{\delta}}C_{\vec{\delta}}\sin(\phi_{\vec{n}-\vec{\delta}}-\phi_{\vec{n}})\delta_j\partial_j\omega. \tag{5}$$

Here the parameter $C_{\vec{\delta}}$ determines the strength of the interaction, and $\omega=u_{x,y}-u_{y,x}$ is the z component of rotation.[9] Note that the chirality in Eq. (5) does not couple to strain but couples only to the gradient of local rotation (ω).

The spin distribution and lattice structure of the system are determined by the minima of the energy (*without* the chiral part F_χ) $E=\Phi+E_s$ with respect to the variational parameters, namely, the components of the deformation tensor $u_{i,j}$ and the angle $\phi_{\vec{n}}$. The case of the spatially uniform states of the system, when the strain tensors e_α are spatially uniform and $\phi_{\vec{n}}=\vec{q}\cdot\vec{n}$ without regard for dilatation deformations, was considered in Ref. 10. The case of a triangular lattice subject to unidirectional strain was studied in Ref. 11. Here we consider the general case when the energy of the system can be represented in the form $E=NJS^2(\mathcal{F}_m+\mathcal{F}_{el})$. The first term $\mathcal{F}_m=(1-\eta_1 e_1)W_1-\eta_2(e_2 W_2+e_3 W_3)$ is the magnetic (and magnetoelastic) energy per atom without the chiral part, with the notations $W_1=\cos q_x+2\cos(q_x/2)\times\cos[(\sqrt{3}/2)q_y]$, $W_2=\cos q_x-\cos(q_x/2)\cos[(\sqrt{3}/2)q_y]$, and $W_3=\sqrt{3}\sin(q_x/2)\sin[(\sqrt{3}/2)q_y]$.

The second term $\mathcal{F}_{el}=(a_1/2)e_1^2+(a_2/2)(e_2^2+e_3^2)$ is the elastic energy per atom. N is the number of sites in the lattice. The constants $a_j=A_j\,\sigma/(JS^2)(j=1,2)$ are the relative strengths of elastic interactions in terms of exchange energy (σ is the area of the unit cell in the lattice). We also introduced phenomenological parameters η_1 and η_2. The first one (η_1) characterizes magnetoelastic interaction caused by the dilatation of the lattice, while the second one (η_2) describes the coupling with shear deformations. Note that in the nearest-neighbor approximation, $\eta_1=\eta_2$. The wave vector $\vec{q}$ and the strain tensor components e_α ($\alpha=1,2,3$) are the variational parameters. They are determined by the equations $\partial\mathcal{F}_m/\partial\vec{q}=0$ and $\partial(\mathcal{F}_m+\mathcal{F}_{el})/\partial e_\alpha=0$, where the latter equation represents the absence of internal stresses in the body. These equations have three types of solutions.

Triangular H phase.

$$\vec{q}_H=\frac{4\pi}{3}\left(\pm\frac{1}{2},\frac{\sqrt{3}}{2}\right),\ e_2=e_3=0,\quad e_1=-\frac{3\eta_1}{2a_1}. \tag{6}$$

The spin structure corresponding to each $\vec{q}_H$ is a three-sublattice antiferromagnet. The antiferromagnetic domains that correspond to two wave vectors $\vec{q}_H$ are distinguished by their chirality, i.e., they are characterized by different signs of the parameter (1). The lattice structure is a homogeneously compressed triangular lattice (with no shear) due to magnetoelastic interaction and has the space-group symmetry D_{6h} (p6mm).

Rectangular L-phase.

$$\vec{q}_L=\frac{2\pi}{\sqrt{3}}(0,1),\quad e_1=-\frac{\eta_1}{a_1},\quad e_2=\frac{2\eta_2}{a_2},\quad e_3=0. \tag{7}$$

The spin structure is a two-sublattice antiferromagnet. The triangular lattice is compressed as well as sheared to give a centered rectangular lattice. It is characterized by the space-group symmetry D_{2h} ($c2mm$). Note that the spin frustration (in the parent triangular phase) is relieved by magnetoelasticity via transition to a phase of lower symmetry.

Spiral S phase. The wave vector is given by $\vec{q}_S=(q_s,2\pi/\sqrt{3})$, with dilatation $e_1=(\eta_1/a_1)[\cos q_s-2\cos(q_s/2)]$, and shear $e_2=(\eta_2/a_2)[\cos q_s+\cos(q_s/2)]$, $e_3=0$, where $\cos(q_s/2)\equiv z$ is the root of the equation $4(\epsilon_1+\epsilon_2)z^2-(4\epsilon_1-5\epsilon_2)z+2\epsilon_1+\epsilon_2-2=0$, with $\epsilon_j=\eta_j^2/a_j$ being the dimensionless magnetoelastic energy shift due to dilatation ($j=1$) and shear ($j=2$). The spin structure is a spiral antiferromagnet.

Two more vectors $\vec{q}_L$ and $\vec{q}_s$ and deformation tensors e_2 and e_3 can be obtained from the above equations by rotations of $\pm\pi/3$ since there are three orientation states for both the centered rectangular and spiral phases.

We consider the stability of the obtained solutions for the case when $e_2\geqslant 0$, $e_3=0$, $0\leqslant q_x\leqslant 2\pi/3$, $q_y=2\pi/\sqrt{3}$. In this subspace we obtain the following results.

(1) the L phase is stable and the H phase is unstable when $\epsilon_2>(4+6\epsilon_1)/9$.

(2) Both H and L phases are stable when $(1+\epsilon_1/5)<\epsilon_2<(4+6\epsilon_1/9)$. For $\epsilon_2=(4+\epsilon_1/16)$ the H and L phases have the same energy while for $\epsilon_2>(4+\epsilon_1/16)$ the L phase becomes energetically more favorable.

(3) The H phase is stable when $\epsilon_2<(1+\epsilon_1/5)$. The spiral phase does not correspond to a minimum of the ground-state energy. In the above subspace the spiral phase in the interval $(1+\epsilon_1/5)<\epsilon_2<(4+6\epsilon_1/9)$ corresponds to a saddle point that separates the two stable phases H and L.

Domain walls. To study spatially *nonuniform* states, in the quasicontinuum approach, regarding $\vec{n}$ as a continuum variable: $\vec{n}\to\vec{r}$, $\phi_{\vec{n}}\to\phi(\vec{r})$, we assume that the wave vector $\vec{q}(\vec{r})\equiv\nabla\phi(\vec{r})$ is a smooth function of the spatial coordinate $\vec{r}$ and the difference $\phi_{\vec{n}+\vec{\delta}}-\phi_{\vec{n}}$ in Eq. (4) can be approximated as $\phi_{\vec{n}+\vec{\delta}}-\phi_{\vec{n}}\approx q_i\delta_i+\frac{1}{2}\delta_i q_{i,j}\delta_j$ and $\cos(\phi_{\vec{n}+\vec{\delta}}-\phi_{\vec{n}})\approx[1-\frac{1}{8}(\delta_i q_{i,j}\delta_j)^2]\cos(\vec{q}\cdot\vec{\delta})$, where we have taken into account that the gradient terms are small. Inserting the latter equation into Eq. (4) and neglecting small terms $(\delta_i u_{i,j}\delta_j)$, $(\delta_i q_{i,j}\delta_j)^2$ we find that the magnetic and magnetoelastic energy of the system (without chiral interaction) has the form $F_m=\int(\mathcal{F}_m+\mathcal{F}_g)d\vec{r}$, where $\mathcal{F}_m$ is given above and the second term (gradient energy) $\mathcal{F}_g=(b_1/2)(\nabla\cdot\vec{q})^2+(b_2/2)[(q_{x,x}-q_{y,y})^2+(q_{x,y}+q_{y,x})^2]$ penalizes spatial inhomogeneities in the high-symmetry H phase. Here the positive inhomogeneous exchange interaction (or gradient) constants b_1 and b_2 are

$$b_1=-\frac{1}{32}\sum_{\vec{\delta}}\vec{\delta}^4\cos(\vec{q}_H\cdot\vec{\delta})=\frac{3}{64},\quad b_2=\frac{1}{2}b_1.$$

Thus the *total* magnetic and magnetoelastic energy density of the triangular lattice (with chiral interaction), which we consider here to study domain walls, has the form

$$\mathcal{F}=(1-\eta_1 e_1)W_1-\eta_2(e_2W_2+e_3W_3)+\tfrac{1}{2}[a_1e_1^2+a_2(e_2^2+e_3^2)]+\mathcal{F}_g+\mathcal{F}_\chi, \tag{8}$$

where the functions W_α ($\alpha=1,2,3$) are given above with a space dependent vector $\vec{q}(\vec{r})$. The spatial structure of the system is determined by the Euler-Lagrange equations for the wave vector $\vec{q}$ and the displacements $\vec{u}$:

$$(b_1+b_2)\nabla^2(\boldsymbol{\nabla}\cdot\vec{q})-\boldsymbol{\nabla}\cdot\frac{\partial}{\partial\vec{q}}\{(1-\eta_1 e_1)W_1-\eta_2[e_2W_2+e_3W_3]+\mathcal{F}_\chi\}=0,\quad a_2\nabla^2\vec{u}+a_1\boldsymbol{\nabla}(\boldsymbol{\nabla}\cdot\vec{u})-\vec{f}=0, \tag{9}$$

with the body force $\vec{f}$ given by

$$f_x=\partial_x(\eta_1W_1+\eta_2W_2)+\eta_2\partial_yW_3-\chi(\partial_{xy}^2W_4+\partial_y^2W_5),$$

$$f_y=\partial_y(\eta_1W_1-\eta_2W_2)+\eta_2\partial_xW_3+\chi(\partial_x^2W_4+\partial_{xy}^2W_5),$$

where $W_4=\sin(q_x)+\sin(q_x/2)\cos(\sqrt{3}q_y/2)$ and $W_5=\cos(q_x/2)\sin(\sqrt{3}q_y/2)$. The energy density of the spin chirality-lattice deformation interaction $\mathcal{F}_\chi$ is

$$\mathcal{F}_\chi=\chi\left\{\sin(q_x)\partial_x\omega+\sin\left(\frac{q_x}{2}\right)\cos\left(\frac{\sqrt{3}q_y}{2}\right)\partial_x\omega+\sqrt{3}\cos\left(\frac{q_x}{2}\right)\sin\left(\frac{\sqrt{3}q_y}{2}\right)\partial_y\omega\right\}, \tag{10}$$

where the parameter $\chi=C/J$ represents the strength of the interaction in terms of the spin-exchange integral. We now analyze the domain walls linking two variant phases. In what follows we neglect, for simplicity, the spin-dilatation interaction and assume $\eta_1=0$. This interaction is responsible for the unit-cell area change and does not influence qualitative behavior of the system.

We assume that the domain wall is oriented along the y axis and in this way we are interested in solutions to Eqs. (9), which depend on one spatial variable x. With the domain walls present in the system there exist spontaneous nonuniform elastic stresses which are determined by the expressions $\sigma_{xx}=(a_1+a_2)\partial_xu_x-\eta_2W_2$, $\sigma_{yy}=(a_1-a_2)\partial_xu_x+\eta_2W_2$, and $\sigma_{xy}=a_2-\eta_2W_3$.

Let us consider a domain wall that links the variants $H_\pm$ with $\vec{q}_H=4\pi/3(\pm1/2,\sqrt{3}/2)$. This means that we look for solutions under boundary conditions

$$q_x\to\pm\frac{2\pi}{3},\ q_y\to\frac{2\pi}{\sqrt{3}},\ \partial_x\vec{u}\to0,\ x\to\pm\infty. \tag{11}$$

We consider separately the cases without and with spin chirality-lattice deformation interaction.

$\chi=0$. Under the boundary conditions (11) we find $q_y=2\pi/\sqrt{3}$, $\partial_xu_x=(\eta_2/a)W_2$, $\partial_xu_y=0$, and

$$b\partial_x^2q_x-\frac{\partial}{\partial q_x}\left\{\cos(q_x)-2\cos\left(\frac{q_x}{2}\right)-\frac{\epsilon}{2}\left[\cos(q_x)+\cos\left(\frac{q_x}{2}\right)\right]^2\right\}=0, \tag{12}$$

where the parameter $\epsilon=\eta_2^2/a$ characterizes the intensity of (non-chiral) magnetoelastic coupling for the case of one-dimensional longitudinal deformations and $a=a_1+a_2$, $b=b_1+b_2$. For $\epsilon<1/4$, the solution of Eq. (12) under the boundary condition (11) is given by the expression

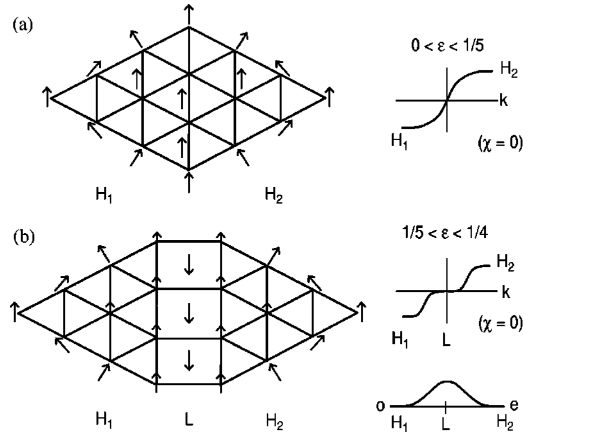

FIG. 1. Schematic representation of domain walls for no interaction between spin chirality and lattice deformation ($\chi=0$)). (a) Spin configuration for a domain connecting two triangular variants (H_1, H_2). (b) Spin structure [chirality variation k, see Eq. (1)] and strain e for a domain with the rectangular (L) phase sandwiched between two triangular (H_1, H_2) variants.

$$\frac{x(8-7m)}{\sqrt{8(2-m)b}}=\{mF(\psi|m)+8m_1\Pi(n,\psi|m)\}, \tag{13}$$

where $F(\psi|m)$ $[\Pi(n,\psi|m)]$ is the incomplete elliptic integral of the first (third) kind[12] with the argument ψ and modulus m given by

$$\tan\frac{q_x}{4}=\sqrt{\frac{2(1-m)}{2-m}}\tan\psi,\quad n=\frac{8-7m}{2-m},\quad m=\frac{4\sqrt{\epsilon}}{1+2\sqrt{\epsilon}},$$

$m_1=1-m$. For $\epsilon<1/5$ the function (13) gives a single domain wall that links two variants of the H phase [Fig. 1(a)]. For $1/5<\epsilon<1/4$ it represents a sandwichlike structure: H_1-L-H_2 [see Fig. 1(b)]. Note that when $\epsilon\in(1/5,\ 1/4)$ the low symmetry L phase is metastable: its energy is higher than the energy of the H phase. The spatial extension of the intermediate L phase depends on ϵ: it increases when $\epsilon\to1/4$. The corresponding lattice structure is determined by

$$u_x=\frac{2\eta_2\sqrt{b}}{\epsilon a}\left\{F\left(\frac{\pi}{2}+\frac{q_x}{4}\middle|4\epsilon\right)-E\left(\frac{\pi}{2}+\frac{q_x}{4}\middle|4\epsilon\right)-F\left(\frac{\pi}{3}\middle|4\epsilon\right)-E\left(\frac{\pi}{3}\middle|4\epsilon\right)\right\}$$

and $u_y=0$. For $\epsilon\in(1/4,\ 4/9)$ when both H and L phases exist but the L phase has lower energy than the H phase, the solution to Eq. (12) under the boundary conditions (11) represents a pulse solution [Fig. 2(a)] with an amplitude which depends on the strength of the magnetoelastic coupling ϵ.

$\chi\neq0$, $\eta_2=0$. Assuming no shear magnetoelastic coupling ($\eta_2=0$) for simplicity, under the boundary conditions (11) we obtain $\partial_xu_x=0$ and $\partial_xu_y=(\chi/a_2)\partial_xW_4$. We define a parameter $\xi=\chi^2/a_2$ which gives the strength of the coupling between the spin chirality and lattice deformation. The resulting Euler-Lagrange equation is similar to a double sine-Gordon equation[13] but with a wave-vector-dependent gradi-

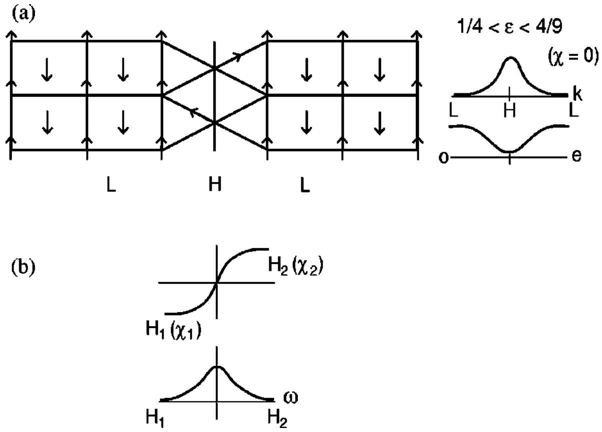

FIG. 2. (a) Spin structure and strain for a domain containing the triangular (*H*) phase sandwiched between the rectangular (*L*) phase. (b) For $\chi \neq 0$ spin structure is similar to Fig. 1(a) but with a local lattice rotation (ω) in the region of the chiral domain wall.

ent coefficient. It has the associated free energy $f = \frac{1}{2} L^2(q_x)(\partial_x q_x)^2 + \cos q_x - 2\cos(q_x/2)$, where

$$L(q_x) = \sqrt{b - \xi\left[\cos q_x - \frac{1}{2}\cos\left(\frac{q_x}{2}\right)\right]^2} \tag{14}$$

is an effective correlation length. It is seen from this expression that when the coupling between spin chirality and lattice deformation is sufficiently strong (ξ is large) the system is unstable with respect to the creation of spatially inhomogeneous excitations. To stabilize the system the higher derivatives of the wave vector q_x and/or the strain gradients $(\nabla e_i)^2 (i=1,2,3)$ should be taken into account.[14] In what follows we restrict ourselves to the case of weak spin chirality-lattice deformation interaction $\xi < b$, when the effective correlation length is real $[L^2(q_x) > 0\ ]$.

Using the boundary conditions (11) for q_x we get $(\partial_x q_x)^2 L^2(q_x) - 2[\cos q_x - 2\cos(q_x/2)] = 3$. The solution to this equation describes a domain wall that links the two variants with different chiralities [see Fig. 2(b)]. The z component of local lattice rotation given by

$$\omega = \frac{\chi}{a_2}\,\frac{(1-2\cos(q_x/2))[2\cos(q_x) - \cos(q_x/2)]}{2\,L(q_x)}, \tag{15}$$

is a pulselike distribution [Fig. 2(b)]. Thus, within the domain wall the lattice experiences two kinds of rotation: clockwise and counterclockwise. The structure of the lattice in the domain wall region is determined by the relations $u_x = 0$ and $u_y = (\chi/a_2)[\sin(q_x) - \sin(q_x/2)]$. The corresponding lattice and spin structures (not shown) are similar to Fig. 1(a). For $\eta_2 \neq 0$ the solutions must be obtained numerically.

In summary, we have investigated spatially inhomogeneous ground states of quasi-two-dimensional magnetoelastic materials such as solid oxygen. We found three different ground states—high symmetry (triangular), low symmetry (rectangular), and spiral phases—for magnetoelastically coupled XY spins on a triangular lattice. In addition to finding chiral antiferromagnetic domain wall solutions (spin orientation and lattice deformation), we found that the magnetic frustration can be relieved by coupling magnetism to elasticity. We anticipate that similar effects will be obtained more generally by coupling to other degrees of freedom such as shuffles, out-of-plane distortions and external fields (stress, magnetic field). Finally, we note that an antiferromagnetic Heisenberg model was also used to describe triangular monolayer adsorption on a surface.[15] Removal of frustration on a compressible Ising antiferromagnet on a triangular lattice has been studied previously.[16] An understanding of magnetoelastic domains in the antiferromagnetic phase is also important in many materials.[17]

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