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SOLID STATE

Flexomagnetic and Flexoantiferromagnetic Effects in Centrosymmetric Antiferromagnetic Materials

A. F. Kabychenkov[a] and F. V. Lisovskii[a,*]

[a]*Kotel'nikov Institute of Radio Engineering and Electronics (Fryazino Branch), Russian Academy of Sciences, Fryazino, Moscow oblast, 141190 Russia*

**e-mail: lisf@df.ru*

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Abstract—A concept of magnetic symmetry is used to show that centrosymmetric antiferromagnetic materials may exhibit linear flexomagnetic and flexoantiferromagnetic effects (magnetization induced by gradient of elastic stress or gradient of antiferromagnetic moment, respectively).

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INTRODUCTION

Antiferromagnetic (AFM) materials (about two thousand chemical elements, compounds, and alloys) form the largest group of magnetically ordered materials [1]. However, such materials have been sparsely used in technology. Note skepticism of L. Néel who was awarded the Nobel Prize in 1970 "for fundamental work and discoveries concerning antiferromagnetism and ferrimagnetism which have led to important applications in solid state physics" [2] and mentioned in the Nobel Lecture that AFM materials "are extremely interesting from the theoretical viewpoint but do not seem to have any applications" [3]. We may only mention AFM materials with weak ferromagnetic properties (e.g., orthoferrites) that are used at present in magnetooptical communication devices and computers as controlled SLMs, deflectors, isolators, and switches for fiber-optical transmission lines (see, for example, [4]).

The aforementioned commonly accepted approach to AFM materials may seem to be justified, since fabrication of real (e.g., storage) devices based on a certain material is possible if such a material is sensitive to external effects and the corresponding response can be externally detected. Conventional methods employed in magnetic elements and devices based on ferro- and ferrimagnetic materials that are sensitive to various external magnetic fields can hardly be used, since the external field strengths must be comparable with high-power effective exchange magnetic fields that provide antiferromagnetic ordering. In addition, the response of the medium is almost undetectable using magnetic fields, since the scattering does not generate AFM fields.

Interaction with antiferromagnetic medium can be implemented using nonmagnetic effects (e.g., electric fields or elastic stress) with the aid of linear magnetoelectric or piezomagnetic effects the existence of which has been predicted from the viewpoint of magnetic symmetry in crystals the symmetry group of which does not contain time reversal operation. Time reversal in such crystals enters the symmetry group only in combinations with the rotation and (spatial) inversion. In particular, the magnetoelectric effect is possible in AFM materials with a center of antisymmetry (antiinversion) and the piezomagnetism is possible in the AFM materials without the antiinversion center [5–10]. The magnetoelectric effect was detected for the first time in Cr_2O_3 [11–13], and the piezomagnetic effect was detected in MnF_2, CoF_2, and FeF_2 [14].

Unfortunately, extremely strong electric fields or elastic stresses are needed for significant response owing to relatively small constants of linear coupling of the electric or elastic subsystem and the magnetic subsystem. The problem is simplified in the presence of nonuniform effects, since significant local gradients of electric field or elastic stress can easily be generated.

In this work, we employ a concept of magnetic symmetry in the analysis of nonuniform flexomagnetic and flexoantiferromagnetic effects in centrosymmetric AFM materials. Such AFM materials do not exhibit weak ferromagnetic and piezomagnetic properties that may prevent observation of the effects under study.

FLEXOMAGNETIC EFFECT

Flexomagnetic (FM) effect lies in the magnetization in the presence of gradient of elastic stress. In the specific thermodynamic potential, the FM effect is described using a linear term with respect to components of magnetic field **H** and gradient of elastic stress σ_{kl}:

$$\Phi^{(fm)} = -\gamma_{ijkl} H_i \frac{\partial \sigma_{kl}}{\partial x_j}, \quad (1)$$

where $\Phi^{(fm)}$ is the term of the thermodynamic potential that is responsible for the FM effects and γ_{ijkl} is the tensor that is symmetric with respect to indices *kl*.

Based on the definition of magnetic induction $B_j^{(fm)} = -4\pi \frac{\partial \Phi^{(fm)}}{\partial H_j}$, we obtain that the linear (with respect to the stress gradient) magnetization at $\mathbf{H} = 0$ is represented as

$$M_i^{(fm)} = \gamma_{ijkl} \frac{\partial \sigma_{kl}}{\partial x_j}. \quad (2)$$

The inverse effect lies in the generation of elastic strain $u_{kl}^{(fm)}$ in the presence of the magnetic-field gradient. Using the definition of the strain tensor, we find that

$$u_{kl}^{(fm)} = -\frac{\delta \Phi^{(fm)}}{\delta \sigma_{kl}} = -\left(\frac{\partial \Phi^{(fm)}}{\partial \sigma_{kl}} - \frac{\partial}{\partial x_n} \frac{\partial \Phi^{(fm)}}{(\partial \Phi^{(fm)}/\partial x_n)} \right).$$

Consequently, we have

$$u_{kl}^{(fm)} = \frac{\partial (\gamma_{inkl} H_i)}{\partial x_n}. \quad (3)$$

Tensor γ_{inkl} is uneven with respect to time reversal and spatial inversion. Thus, the FM effect is not observed in crystals for which the magnetic symmetry groups separately contain such operations but may exist in the crystals the point magnetic groups of which contain the operation of center-antiinversion (product of operations of time reversal and spatial inversion). For cubic AFM materials, such an operation is allowed for magnetic groups *m*'3, *m*'3*m*, and *m*'3*m*'; for tetragonal materials, such an operation is allowed for magnetic groups 4/*m*', 4'/*m*, and 4/*m*'*m*'*m*' (e.g., Fe_2TeO_6); for hexagonal materials, such an operation is allowed for magnetic groups $\bar{3}$', $\bar{3}$'*m*' (e.g., Cr_2O_3) $\bar{3}$'*m*, 6'/*m*, 6/*m*', 6'/*mmm*', 6/*m*'*m*'*m*', 6/*m*'*mm*; for rhombic materials, such an operation is allowed for magnetic groups *m*'*m*'*m*' and *mmm*' (e.g., Cr_2TeO_6, Cr_2WO_6, and V_2WO_6); and for monoclinic materials, such an operation is allowed for magnetic groups 2/*m*' and 2/*m* [15, 16].

Formula (1) shows that the term of the thermodynamic potential that is responsible for the FM effect in the Cr_2O_3 crystal is written as

$$\begin{aligned}
\Phi^{(fm)} &= -\gamma_{33} H_z \frac{\partial \sigma_{zz}}{\partial z} - \gamma_{11}\left[H_x \left(\frac{\partial \sigma_{xx}}{\partial x} + \frac{\partial \sigma_{xy}}{\partial y} \right) \right. \\
&\left. + H_y \left(\frac{\partial \sigma_{yy}}{\partial y} + \frac{\partial \sigma_{xy}}{\partial x} \right) \right] - \gamma_{12}\left[H_x \left(\frac{\partial \sigma_{yy}}{\partial x} - \frac{\partial \sigma_{xy}}{\partial y} \right) \right. \\
&\left. + H_y \left(\frac{\partial \sigma_{xx}}{\partial y} + \frac{\partial \sigma_{xy}}{\partial x} \right) \right] - \gamma_{13}\left(H_x \frac{\partial \sigma_{zz}}{\partial x} + H_y \frac{\partial \sigma_{zz}}{\partial y} \right) \\
&- 2\gamma_{88}\left(H_x \frac{\partial \sigma_{xz}}{\partial z} + H_y \frac{\partial \sigma_{yz}}{\partial z} \right) - \gamma_{31} H_z \left(\frac{\partial \sigma_{xx}}{\partial z} + \frac{\partial \sigma_{yy}}{\partial z} \right) \quad (4) \\
&- 2\gamma_{55} H_z \left(\frac{\partial \sigma_{xz}}{\partial x} + \frac{\partial \sigma_{yz}}{\partial y} \right) - 2\gamma_{24}\left[H_y \left(\frac{\partial \sigma_{yz}}{\partial y} - \frac{\partial \sigma_{xz}}{\partial x} \right) \right. \\
&\left. - H_x \left(\frac{\partial \sigma_{xz}}{\partial y} + \frac{\partial \sigma_{yz}}{\partial x} \right) \right] - \gamma_{24}\left[H_y \left(\frac{\partial \sigma_{yy}}{\partial z} - \frac{\partial \sigma_{xx}}{\partial z} \right) \right. \\
&\left. - 2H_x \frac{\partial \sigma_{xy}}{\partial z} \right] - \gamma_{72} H_z \left[\left(\frac{\partial \sigma_{yy}}{\partial y} - \frac{\partial \sigma_{xx}}{\partial y} \right) - 2\frac{\partial \sigma_{xz}}{\partial x} \right].
\end{aligned}$$

Hereafter, abbreviated indices are used for the fourth-rank tensors: 11 → 1, 22 → 2, 33 → 3, 23 → 4, 31 → 5, 12 → 6, 32 → 7, 13 → 8, and 21 → 9.

Expression (4) shows that the components of magnetization $\mathbf{M}^{(fm)}$ induced by the gradient of elastic stress are represented as

$$\begin{aligned}
M_x^{(fm)} &= \gamma_{11}\left(\frac{\partial \sigma_{xx}}{\partial x} + \frac{\partial \sigma_{xy}}{\partial y} \right) + \gamma_{12}\left(\frac{\partial \sigma_{yy}}{\partial x} - \frac{\partial \sigma_{xy}}{\partial y} \right) \\
&+ \gamma_{13}\frac{\partial \sigma_{zz}}{\partial x} + 2\gamma_{88}\frac{\partial \sigma_{xz}}{\partial z} - 2\gamma_{24}\left(\frac{\partial \sigma_{xz}}{\partial y} + \frac{\partial \sigma_{yz}}{\partial x} \right) - 2\gamma_{42}\frac{\partial \sigma_{xy}}{\partial z}, \\
M_y^{(fm)} &= \gamma_{11}\left(\frac{\partial \sigma_{yy}}{\partial y} + \frac{\partial \sigma_{xy}}{\partial x} \right) + \gamma_{12}\left(\frac{\partial \sigma_{xx}}{\partial y} + \frac{\partial \sigma_{xy}}{\partial y} \right) \\
&+ \gamma_{13}\frac{\partial \sigma_{zz}}{\partial y} + 2\gamma_{88}\frac{\partial \sigma_{yz}}{\partial z} + 2\gamma_{24}\left(\frac{\partial \sigma_{yz}}{\partial y} - \frac{\partial \sigma_{xz}}{\partial x} \right) \quad (5) \\
&+ \gamma_{42}\left(\frac{\partial \sigma_{yy}}{\partial z} - \frac{\partial \sigma_{xx}}{\partial z} \right), \\
M_z^{(fm)} &= \gamma_{33}\frac{\partial \sigma_{zz}}{\partial z} + \gamma_{31}\left(\frac{\partial \sigma_{xx}}{\partial z} + \frac{\partial \sigma_{yy}}{\partial z} \right) \\
&+ 2\gamma_{55}\left(\frac{\partial \sigma_{xz}}{\partial x} + \frac{\partial \sigma_{yz}}{\partial y} \right) + \gamma_{72}\left[\left(\frac{\partial \sigma_{yy}}{\partial y} - \frac{\partial \sigma_{xx}}{\partial y} \right) - 2\frac{\partial \sigma_{xz}}{\partial x} \right],
\end{aligned}$$

and the components of the strain tensor are represented as

$$\begin{aligned}
u_{xx}^{(jm)} &= -\frac{\partial(\gamma_{11}H_x)}{\partial x} - \frac{\partial(\gamma_{12}H_y)}{\partial y} - \frac{\partial(\gamma_{31}H_z)}{\partial z} \\
&+ \frac{\partial(\gamma_{42}H_y)}{\partial z} + \frac{\partial(\gamma_{72}H_z)}{\partial y}, \\
u_{yy}^{(jm)} &= -\frac{\partial(\gamma_{11}H_y)}{\partial y} - \frac{\partial(\gamma_{12}H_x)}{\partial x} - \frac{\partial(\gamma_{31}H_z)}{\partial z} \\
&- \frac{\partial(\gamma_{42}H_z)}{\partial y} - \frac{\partial(\gamma_{72}H_z)}{\partial y},
\end{aligned}$$

$$u_{zz}^{(jm)} = -\frac{\partial(\gamma_{33}H_z)}{\partial z} - \frac{\partial(\gamma_{13}H_x)}{\partial x} - \frac{\partial(\gamma_{13}H_y)}{\partial y},$$

$$u_{xy}^{(jm)} = u_{yx}^{(jm)} = -\frac{\partial(\gamma_{11}H_x)}{\partial y} - \frac{\partial(\gamma_{11}H_y)}{\partial x} + \frac{\partial(\gamma_{12}H_x)}{\partial y} + \frac{\partial(\gamma_{12}H_y)}{\partial x} + 2\frac{\partial(\gamma_{42}H_x)}{\partial z}, \quad (6)$$

$$u_{xz}^{(jm)} = u_{zx}^{(jm)} = -2\frac{\partial(\gamma_{88}H_x)}{\partial z} - 2\frac{\partial(\gamma_{55}H_z)}{\partial x} + 2\frac{\partial(\gamma_{24}H_y)}{\partial x} + 2\frac{\partial(\gamma_{24}H_x)}{\partial y} + 2\frac{\partial(\gamma_{72}H_z)}{\partial x},$$

$$u_{yz}^{(jm)} = u_{zy}^{(jm)} = -2\frac{\partial(\gamma_{88}H_y)}{\partial z} - 2\frac{\partial(\gamma_{55}H_z)}{\partial y} - 2\frac{\partial(\gamma_{24}H_y)}{\partial y} + 2\frac{\partial(\gamma_{24}H_x)}{\partial x}.$$

At constant coefficients, the strains are linearly related to the gradient of the components of magnetic field. For example, consider a cylinder made of a uniaxial ferromagnetic material that is placed vertically (the vertical direction is collinear to the antiferromagnetism vector) on a solid base. Uniform gravity with gravity constant g causes nonuniform deformation in such a cylinder, so that the induced magnetization can be represented as

$$M_z^{(fm)} = \gamma_{33}\frac{\partial\sigma_{zz}}{\partial z} = \gamma_{33}\rho g,$$

where ρ is the density of the AFM material.

A similar effect is observed when the upper end surface of the cylinder is rigidly fixed on a horizontal surface. An AFM cylinder that is suspended on an elastic thread will be rotated owing to an angular momentum of $\mathbf{M}^{(fm)}/\gamma^{(g)}$ ($\gamma^{(g)}$ is the gyromagnetic coefficient) that is acquired due to magnetization. Such an effect is an analog of the Einstein–de Haas effect for ferromagnetic materials.

Similar effects are possible for different configurations. Bending of a thin AFM plate with the base plane that is parallel to the neutral plane causes nonuniform elastic stress that induces the following magnetization components:

$$M_x^{(fm)} = 2\gamma_{42}\frac{E}{1+\sigma_P}\frac{\partial^2\varsigma}{\partial x\partial y},$$

$$M_y^{(fm)} = -\gamma_{42}\frac{E}{1+\sigma_P}\left(\frac{\partial^2\varsigma}{\partial y^2} - \frac{\partial^2\varsigma}{\partial x^2}\right), \quad (7)$$

$$M_z^{(fm)} = -\gamma_{31}\frac{E}{1-\sigma_P}\left(\frac{\partial^2\varsigma}{\partial x^2} + \frac{\partial^2\varsigma}{\partial y^2}\right),$$

where $\varsigma(x, y)$ are the coordinates of the neutral plane [17], E is the tensile (Young) modulus, and σ_P is the ratio of the transverse compression to longitudinal tension (Poisson coefficient). Note that the magnetization can easily be detected using excitation of elastic waves.

FLEXOANTIFERROMAGNETIC EFFECT

Flexoantiferromagnetic (FAM) effect lies in magnetization in the presence of the gradient of antiferromagnetic moment. The effect is described in the thermodynamic potential using a term that is linear with respect to components of magnetic field **H** and gradient of antiferromagnetic moment:

$$\Phi^{(fam)} = -\eta_{ijk}H_i\frac{\partial L_j}{\partial x_k}. \quad (8)$$

Potential (8) causes magnetic induction or magnetization (in the absence of magnetic field):

$$M_i^{(fam)} = \eta_{ijk}\frac{\partial L_j}{\partial x_k}. \quad (9)$$

As distinct from weak ferromagnetic effect [5], such magnetization is proportional to the gradient of the antiferromagnetic moment that can be related to nonuniform composition, presence of domain walls or crystal interfaces, temperature gradients, etc. For Cr_2O_3, the terms that are responsible for the FAM effect are written as

$$\Phi^{(fam)} = -\eta_{111}\left(H_x\left(\frac{\partial L_x}{\partial x} - \frac{\partial L_y}{\partial y}\right) - H_y\left(\frac{\partial L_x}{\partial y} + \frac{\partial L_y}{\partial x}\right)\right) - \eta_{123}\left(H_x\frac{\partial L_y}{\partial z} - H_y\frac{\partial L_x}{\partial z}\right) - \eta_{231}\left(H_y\frac{\partial L_z}{\partial x} - H_x\frac{\partial L_z}{\partial y}\right) - \eta_{312}H_z\left(\frac{\partial L_x}{\partial y} - \frac{\partial L_y}{\partial x}\right), \quad (10)$$

and the magnetization is given by

$$M_x^{(fam)} = \eta_{111}\left(\frac{\partial L_x}{\partial x} - \frac{\partial L_y}{\partial y}\right) + \eta_{123}\frac{\partial L_y}{\partial z} - \eta_{231}\frac{\partial L_z}{\partial y},$$

$$M_y^{(fam)} = \eta_{111}\left(\frac{\partial L_x}{\partial y} - \frac{\partial L_y}{\partial x}\right) - \eta_{123}\frac{\partial L_x}{\partial z} - \eta_{231}\frac{\partial L_z}{\partial x}, \quad (11)$$

$$M_z^{(fam)} = -\eta_{312}\left(\frac{\partial L_x}{\partial y} - \frac{\partial L_y}{\partial x}\right).$$

Temperature nonuniformity in the (x, y) plane leads to magnetization with the following components: $M_x^{(fam)} = -\eta_{231}\frac{\partial L_z}{\partial y}$ and $M_y^{(fam)} = -\eta_{231}\frac{\partial L_z}{\partial x}$.

In the presence of external magnetic field $\mathbf{H}_0$, the total thermodynamic potential must be represented as

$$\Phi = \Phi_0 - \mathbf{M}\mathbf{H}_0 + (\mathbf{H}_m^2/8\pi), \quad (12)$$

where $\mathbf{H}_m$ is the field generated by the magnetization [1]. In the quadratic approximation with respect to **L** and **M** and derivatives of **L** (small derivatives of **M** are disregarded), the thermodynamic potential of Cr_2O_3 is given by the following expression taking into account the terms that are responsible for the FAM effect:

$$\Phi_0 = \frac{1}{2}\left[a_1^{(l)}\mathbf{L}^2 + a_1^{(m)}\mathbf{M}^2 + a_1^{(ln)}\left(\frac{\partial \mathbf{L}}{\partial x_i}\right)^2 + a_2^{(l)}L_z^2 + a_2^{(m)}M_z^2 + a_2^{(ln)}\left(\frac{\partial L_z}{\partial x_i}\right)^2\right] - \eta_1\left[M_x\left(\frac{\partial L_x}{\partial x} - \frac{\partial L_y}{\partial y}\right) - M_y\left(\frac{\partial L_x}{\partial y} + \frac{\partial L_y}{\partial x}\right)\right] - \eta_2\left(M_x\frac{\partial L_y}{\partial z} - M_y\frac{\partial L_x}{\partial z}\right) - \eta_3\left(M_y\frac{\partial L_z}{\partial x} - M_x\frac{\partial L_z}{\partial y}\right) - \eta_4 M_z\left(\frac{\partial L_x}{\partial y} - \frac{\partial L_y}{\partial x}\right). \quad (13)$$

Minimization of the potential using the conditions $\delta\Phi/\delta\mathbf{M} = 0$ and $\delta\Phi/\delta\mathbf{L} = 0$ yields the following system of equations:

$$M_x = \eta_1^{(m)}\left(\frac{\partial L_z}{\partial y} - \frac{\partial L_y}{\partial y}\right) + \eta_2^{(m)}\frac{\partial L_y}{\partial z} - \eta_3^{(m)}\frac{\partial L_y}{\partial z} + \zeta_\perp H_x,$$

$$M_y = -\eta_1^{(m)}\left(\frac{\partial L_x}{\partial y} + \frac{\partial L_y}{\partial x}\right) - \eta_2^{(m)}\frac{\partial L_x}{\partial z} + \eta_3^{(m)}\frac{\partial L_z}{\partial x}y + \zeta_\perp H_y,$$

$$M_z = \eta_4^{(m)}\left(\frac{\partial L_x}{\partial y} - \frac{\partial L_y}{\partial x}\right) + \zeta_\parallel H_z,$$

$$L_x + a_{11}^{(ln)}\frac{\partial^2 L_x}{\partial x_i^2} - \eta_1^{(l)}\left(\frac{\partial M_x}{\partial x} - \frac{\partial M_y}{\partial y}\right) + \eta_2^{(l)}\frac{\partial M_y}{\partial z} - \eta_4^{(l)}\frac{\partial M_z}{\partial y} = 0, \quad (14)$$

$$L_y + a_{11}^{(ln)}\frac{\partial^2 L_y}{\partial x_i^2} + \eta_1^{(l)}\left(\frac{\partial M_x}{\partial y} + \frac{\partial M_y}{\partial x}\right) - \eta_2^{(l)}\frac{\partial M_x}{\partial z} + \eta_4^{(l)}\frac{\partial M_z}{\partial x} = 0,$$

$$L_z + a_{12}^{(ln)}\frac{\partial^2 L_y}{\partial x_i^2} + \eta_3^{(l)}\left(\frac{\partial M_x}{\partial y} - \frac{\partial M_y}{\partial x}\right) = 0,$$

where $\eta_i^{(m)} = \eta_i/a_1^{(m)}$ for $i = 1, 2, 3$, $\eta_4^{(m)} = \eta_4/(a_1^{(m)} + a_2^{(m)})$, $\eta_j^{(m)} = -\eta_j/a_1^{(l)}$ for $j = 1, 2, 4$, $\eta_3^{(l)} = -\eta_3/(a_1^{(l)} + a_2^{(l)})$, $a_{11}^{(ln)} = -a_1^{(ln)}/a_1^{(l)}$ and $a_{12}^{(ln)} = -(a_1^{(ln)} + a_2^{(ln)})/(a_1^{(l)} + a_2^{(l)})$ are the reduced constants, $\zeta_\perp = 1/a_1^{(m)}$, $\zeta_\parallel = 1/(a_1^{(m)} + a_2^{(m)})$, and $\mathbf{H} = \mathbf{H}_0 + \mathbf{H}_m$ is the internal magnetic field.

In the absence of external magnetic field, easy-axis homogeneous state is obtained at negative constants of uniform exchange $a_1^{(l)}$ and anisotropy $a_2^{(l)}$ and positive constant of nonuniform exchange $a_1^{(ln)}$. When the antiferromagnetic moment is relatively small $((\eta_i^{(m)}/d)L \ll \zeta H$ and $a_{11}^{(ln)}/d^2 \ll 1$, where d is the nonuniformity scale), the gradient of the magnetic field induces the following components of the antiferromagnetic moment:

$$L_x = \chi_1^{(l)}\left(\frac{\partial H_x}{\partial x} - \frac{\partial H_y}{\partial y}\right) + \chi_2^{(l)}\frac{\partial H_z}{\partial y};$$
$$L_y = -2\chi_1^{(l)}\frac{\partial H_x}{\partial y} - \chi_2^{(l)}\frac{\partial H_z}{\partial x}; \quad L_z = 0, \quad (15)$$

where $\chi_1^{(l)} = -\eta_1^{(m)}/a_1^{|l|}$, and $\chi_2^{(l)} = (\eta_2^{(m)} - \eta_4^{(m)})/a_1^{|l|}$ are generalized susceptibilities.

CONCLUSIONS

The theoretical analysis supplements the modern concepts of the properties of centrosymmetric AFM materials (that are usually mentioned as materials that exhibit the magnetoelectric effect) and possible practical applications. The latter are facilitated by the fact that flexomagnetic and flexoantiferromagnetic effects emerge in the presence of relatively large gradients of nonuniform elastic stress or magnetic fields that can easily be implemented.

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