Microscopic Model of Nonreciprocal Optical Effects in Cr_2O_3

V. N. Muthukumar, Roser Valentí, and Claudius Gros
Institut für Physik, Universität Dortmund, 44221 Dortmund, Germany

(Received 8 June 1995)

We develop a microscopic model that explains nonreciprocal optical effects in centrosymmetric Cr_2O_3. It is shown that light can couple *directly* to the antiferromagnetic order parameter. This coupling is mediated by the spin-orbit interaction and involves an interplay between the breaking of inversion symmetry due to the antiferromagnetic order parameter and the trigonal field contribution to the ligand field at the Cr^{3+} ion.

PACS numbers: 78.20.Ls, 42.65.–k

The study of the interaction of light with magnetic substances has a long history. A classic example is the Faraday effect in ferromagnets where light couples directly to the ferromagnetic order parameter. Since the pioneering work of Argyres [1], it is known that electromagnetic radiation couples to the internal molecular field in a ferromagnet (which in turn is proportional to the order parameter) through the spin-orbit interaction. Such a coupling would, of course, be absent in antiferromagnets, where the internal molecular field is zero. In the absence of such a direct coupling between light and the antiferromagnetic order parameter, antiferromagnetic ordering could so far be probed only *indirectly*, for instance, by Raman scattering of magnetic excitations [2].

The discovery [3,4] of nonreciprocal optical effects (i.e., not invariant under time reversal) [5] below the Néel temperature T_N in optical experiments on Cr_2O_3 has therefore been considered a breakthrough in the study of antiferromagnetic ordering by light, for it is only this class of experiments that can distinguish between two magnetic states that are related to each other by the time-reversal operation. Fiebig *et al.* [4] have found that antiferromagnetic domains could be observed *directly* by nonreciprocal second harmonic generation (SHG), leading to the first photographs ever of antiferromagnetic domains [6,7]. These experiments show that light can indeed couple directly to the antiferromagnetic order parameter. Though such a coupling was anticipated earlier from symmetry considerations [8], no microscopic mechanism has been presented so far.

In this Letter, we present a microscopic mechanism that explains all nonreciprocal optical effects in Cr_2O_3. While the spin-orbit interaction is, of course, essential in coupling the charge with the spin degrees of freedom, it does not suffice for the generation of nonreciprocal effects. We find that nonreciprocal effects arise from an interplay between the breaking of crystal inversion symmetry by the antiferromagnetic order parameter and the trigonal distortion of the ligand field at the Cr^{3+} ion. This effect, in addition to the spin-orbit interaction, leads to a coupling of the antiferromagnetic order parameter with light.

We present furthermore a simple cluster model, containing the full crystal symmetry of Cr_2O_3, which allows, for the first time, the orders of magnitude of all matrix elements contributing to the nonreciprocal phenomena in Cr_2O_3 to be predicted. We apply the microscopic model to the observed phenomenon of SHG [4] and explain how antiferromagnetic domains can be distinguished experimentally. We also apply our model to another nonreciprocal effect seen experimentally in Cr_2O_3, viz., gyrotropic birefringence [3] and solve the long-standing question regarding its order of magnitude.

As an introduction to SHG in Cr_2O_3, we discuss the macroscopic theory [4] in brief. Above T_N ($\simeq 307$ K), Cr_2O_3 crystallizes in the centrosymmetric point group D_{3d}. The four Cr^{3+} ions in the unit cell occupy equivalent c positions along the C_3 (optic) axis. Since this structure has a center of inversion, parity considerations allow only magnetic dipole transitions related to the existence of an axial tensor of odd rank. Below T_N, time-reversal symmetry (R) is broken and in this case, since Neumann's principle [9] cannot be applied to nonstatic phenomena, only symmetry operations of the crystal that do not include R may be used to classify the allowed tensors for the susceptibilities. For Cr_2O_3, the remaining invariant subgroup is D_3. New tensors are allowed in this point group, for instance, a polar tensor of odd rank, that allow electric dipole transitions in SHG.

From Maxwell's equations, one can derive the expression for SHG by considering the contributions to (nonlinear) magnetization $\mathbf{M}^{(2\omega)} = \boldsymbol{\gamma}^{(2\omega)} : \mathbf{E}^{(\omega)}\mathbf{E}^{(\omega)}$ and polarization $\mathbf{P}^{(2\omega)} = \boldsymbol{\chi}^{(2\omega)} : \mathbf{E}^{(\omega)}\mathbf{E}^{(\omega)}$. The source term $\mathbf{S}(\mathbf{r},t)$ in the wave equation $[\nabla \times (\nabla\times) + (1/c^2) \times \partial^2/\partial t^2]\mathbf{E}(\mathbf{r},t) = -\mathbf{S}(\mathbf{r},t)$ can be written in a dipole expansion as [10]

$$\mathbf{S}(\mathbf{r},t) = \mu_0\left(\frac{\partial^2\mathbf{P}(\mathbf{r},t)}{\partial t^2} + \nabla \times \frac{\partial\mathbf{M}(\mathbf{r},t)}{\partial t} + \cdots\right). \quad (1)$$

Then, by assuming that $\mathbf{E}$, $\mathbf{P}$, and $\mathbf{M}$ can be decomposed into a set of plane waves and considering a circular basis with $\mathbf{E} = E_+\mathbf{e}_+ + E_-\mathbf{e}_- + E_z\mathbf{e}_z$, where $\mathbf{e}_\pm = \mp(1/\sqrt{2})(\mathbf{e}_x \pm i\mathbf{e}_y)$ and the direction of laser light to be

0031-9007/95/75(14)/2766(4)$06.00

along the optic axis, one obtains [4]

$$\mathbf{S} = \begin{pmatrix} S_+ \\ S_- \\ S_z \end{pmatrix} = \frac{4\sqrt{2}\,\omega^2}{c^2} \begin{pmatrix} (-\gamma_m + i\chi_e)E_-^2 \\ (\gamma_m + i\chi_e)E_+^2 \\ 0 \end{pmatrix}, \quad (2)$$

where γ_m and χ_e are nonzero components of the magnetic (γ) and electric (χ) susceptibilities that are allowed by D_3 symmetry. Incoming right circularly polarized light (E_+) leads to left circularly polarized light (E_-) and vice versa in SHG. Above T_N, as $\chi_e \equiv 0$, the SHG intensities $I_\pm$ are identical while below T_N, the intensities $I_\pm \propto |\pm\gamma_m + i\chi_e|^2 E_\mp^4$ are different for right and left circularly polarized light, as observed in experiment [4].

The macroscopic theory dicussed so far is based on symmetry considerations only and does not provide estimates of the magnitude of either γ_m or χ_e, which can be obtained only from a microscopic approach. A *two ion* mechanism for a nonzero electric dipole matrix element, χ_e, in noncentrosymmetric antiferromagnets was proposed by Tanabe, Moriya, and Sugano [11]. This mechanism is unlikely to be responsible for the coherent interference effect suggested by macroscopic theory (2), since it involves magnetic excitations. Here, we propose instead, a *single ion* mechanism. In our theory the coupling of light to the antiferromagnetic order parameter is through the interference between the coherent contributions from the distinct Cr^{3+} ions in the crystal unit cell. No magnetic excitations are involved in our coupling mechanism.

The crystal field due to the oxygen ions in Cr_2O_3 splits the fivefold degenerate $3d$ orbitals of the Cr^{3+} ions into two levels, the lower one (t_2 level) being triply degenerate (d_{xy}, d_{yz}, d_{zx}) and the upper (e level) doubly degenerate ($d_{x^2-y^2}$, $d_{3z^2-r^2}$). The three t_2 orbitals are occupied in the ground state and the two e orbitals are empty. In SHG, the Cr^{3+} ion absorbs two photons and is excited to a $(t_2)^2e$ configuration via two consecutive electric dipole (ED) processes, corresponding to an $\mathbf{r}\cdot\mathbf{E}^{(\omega)}$ term in the Hamiltonian. A contribution to γ_m is then obtained for an emission via a magnetic dipole (MD) process, corresponding to an $\mathbf{L}\cdot\mathbf{B}^{(2\omega)}$ term in the Hamiltonian. This contribution to γ_m is allowed at all temperatures. The key point of a microscopic model is then to find the mechanism that allows a contribution to χ_e via an electric dipole matrix element, $\mathbf{r}\cdot\mathbf{E}^{(2\omega)}$, in emission.

We present our theory in two steps. As a first step, we explain the coupling mechanism between light and the order parameter, and later we proceed to discuss the role of the D_{3d} crystal symmetry. In order to understand the origin of the ED transition below T_N, let us consider, for the moment, just two d orbitals per Cr^{3+} ion: the d_{xy} orbital (ground state) and the $d_{x^2-y^2}$ orbital (excited state). The MD contribution to SHG, i.e., to $\mathbf{M}^{(2\omega)}$ is

$$\langle d_{xy}, m_s|\mathbf{L}|d_{x^2-y^2}, m_s\rangle\langle d_{x^2-y^2}, m_s|(\mathbf{r}\cdot\mathbf{E}^{(\omega)})^2|d_{xy}, m_s\rangle,$$

where the label m_s denotes the spin quantum number of the relevant orbital. (Energy denominators have been omitted for clarity.) Here $(\mathbf{r}\cdot\mathbf{E}^{(\omega)})^2$ denotes $\mathbf{r}\cdot\mathbf{E}^{(\omega)}|\alpha\rangle\langle\alpha|\mathbf{r}\cdot\mathbf{E}^{(\omega)}$ together with a sum over the intermediate states $|\alpha\rangle$ and corresponding energy denominators.

In order to develop the theory for the ED contribution, we treat the spin-orbit interaction and the trigonal distortion of the ligand field at the Cr^{3+} ion as perturbations to the $3d$ Cr states. It is easy to see that the diagonal part of the spin-orbit interaction mixes the t_2 and e states, viz.,

$$\langle d_{xy}, m_s|\mathbf{L}\cdot\mathbf{S}|d_{x^2-y^2}, m_s\rangle = 2i\langle S_z\rangle, \quad (3)$$

where $\langle S_z\rangle \equiv \langle m_s|S_z|m_s\rangle$.

Next, we note that the $3d$ and the $4p$ Cr orbitals are mixed by the trigonal field [12], which breaks local parity. The local eigenstates of the Cr^{3+} ion can therefore be written in the lowest order of perturbation theory as

$$|\tilde{d}_{xy}\rangle = |d_{xy}, m_s\rangle + \lambda\langle S_z\rangle|d_{x^2-y^2}, m_s\rangle + \eta'|p', m_s\rangle,$$

$$|\tilde{d}_{x^2-y^2}\rangle = |d_{x^2-y^2}, m_s\rangle - \lambda\langle S_z\rangle|d_{xy}, m_s\rangle + \eta|p, m_s\rangle,$$

where λ is proportional to the spin-orbit coupling; η, η' are proportional to the trigonal field and $|p, m_s\rangle, |p', m_s\rangle$ are Cr $4p$ orbitals that will be specified later. The ED contribution to the SHG, i.e., to $\mathbf{P}^{(2\omega)}$, can now be expanded in powers of λ and η as

$$\langle\tilde{d}_{xy}|\mathbf{r}|\tilde{d}_{x^2-y^2}\rangle\langle\tilde{d}_{x^2-y^2}|(\mathbf{r}\cdot\mathbf{E}^{(\omega)})^2|\tilde{d}_{xy}\rangle = \eta\lambda\langle d_{xy}, m_s|\mathbf{L}\cdot\mathbf{S}|d_{x^2-y^2}, m_s\rangle\langle d_{x^2-y^2}, m_s|\mathbf{r}|p, m_s\rangle \times \langle d_{x^2-y^2}, m_s|(\mathbf{r}\cdot\mathbf{E}^{(\omega)})^2|d_{xy}, m_s\rangle + \cdots, \quad (4)$$

where the contribution $\sim\lambda\eta$ is shown for illustration. All nonzero contributions are $\sim\lambda\eta\langle S_z\rangle$. To see this, consider all possible contributions order by order.

The contribution $\sim\lambda^0\eta^0$ vanishes because all d orbitals have even parity, viz., $\langle d_{xy}, m_s|\mathbf{r}|d_{x^2-y^2}, m_s\rangle \equiv 0$. The contribution $\sim\lambda^0\eta^1$, viz.,

$$\eta\langle d_{xy}, m_s|\mathbf{r}|p, m_s\rangle\langle d_{x^2-y^2}, m_s|(\mathbf{r}\cdot\mathbf{E}^{(\omega)})^2|d_{xy}, m_s\rangle, \quad (5)$$

is finite for every Cr site and is proportional to η. However, note that the trigonal field at Cr sites that are related by inversion symmetry have opposite signs, viz., if $A1/B2$ and $B1/A2$ are pairs of Cr^{3+} ions in the unit cell that are related by inversion symmetry, $\eta_{A_1} = -\eta_{B_2}$ and $\eta_{A_2} = -\eta_{B_1}$ [12]. On summing the contributions to the matrix element $\sim\lambda^0\eta^1$ from each Cr ion in the unit cell, we see that this matrix element has to vanish identically.

The term $\sim\lambda^1\eta^0$ vanishes, again because of the parity of the d orbitals, but the contribution $\sim\lambda^1\eta^1$ shown in (4) does not vanish generally. Using Eq. (3), we can sum up the total contribution $\sim\lambda^1\eta^1$ from the four (equivalent) Cr ions in the unit cell of Cr_2O_3 as

$$\lambda\eta\mathbf{X}^{(2\omega)}(\langle S_z\rangle_{A1} - \langle S_z\rangle_{B1} + \langle S_z\rangle_{A2} - \langle S_z\rangle_{B2}) = \lambda\eta\mathbf{X}^{(2\omega)}\Delta(T),$$

where $\mathbf{X}^{(2\omega)} = \langle d_{x^2-y^2}, m_s|\mathbf{r}|p, m_s\rangle\langle d_{x^2-y^2}, m_s|(\mathbf{r}\cdot\mathbf{E}^{(\omega)})^2|d_{xy}, m_s\rangle$ at site $A1$. Thus, we have shown that the ED contribution to the SHG, $\mathbf{P}^{(2\omega)}$, couples directly to the spin part of the antiferromagnetic order parameter $\Delta(T) = \langle S_z\rangle_{A1} - \langle S_z\rangle_{B1} + \langle S_z\rangle_{A2} - \langle S_z\rangle_{B2}$. Note that $\mathbf{P}^{(2\omega)}$ is nonreciprocal, as it changes sign under time reversal and allows us to differentiate between the different antiferromagnetic domains in Cr_2O_3.

We now complete the microscopic description by incorporating the full symmetry of Cr_2O_3, i.e., D_{3d}. This is done by considering a $(CrO_6)_2$ cluster model shown in Fig. 1. In order to reproduce the full D_{3d} symmetry, one has to choose the locations of the oxygen ions in this cluster in such a way that they do not coincide exactly with those in the actual crystal structure. This is because the cluster model contains only two Cr sites, while there are four in the unit cell of Cr_2O_3.

We define the Cr sites to be located at $z_0(0,0,\pm 1)$ in the cluster model (z_0 is an arbitrary constant). Neglecting the trigonal distortion for the moment, let us choose the oxygens around the Cr sites to be located at

$$\begin{aligned} &z_0(0,0,\epsilon) + \vartheta\epsilon(\sqrt{2/3}, 0, \sqrt{1/3}), \\ &z_0(0,0,\epsilon) + \vartheta\epsilon(-\sqrt{1/6}, \sqrt{1/2}, \sqrt{1/3}), \\ &z_0(0,0,\epsilon) + \vartheta\epsilon(-\sqrt{1/6}, -\sqrt{1/2}, \sqrt{1/3}), \end{aligned} \tag{6}$$

where $\vartheta = \pm 1$ in order to obtain the sites of all the 12 oxygen ions (see Fig. 1). For any of the four combinations ($\epsilon = \pm 1, \vartheta = \pm 1$), the three oxygen sites given in (6) form a plane. It is obvious that this cluster model has a center of inversion at $(0,0,0)$. In addition, it is easy to verify that this model has all the symmetry elements belonging to the group D_{3d} [9], in particular, the C_3 and the 2_y symmetries that correspond to threefold and twofold rotations about the z and y axes, respectively.

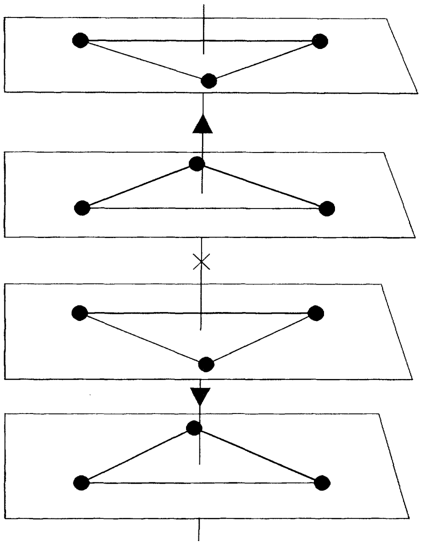

FIG. 1. An illustration of the $(CrO_6)_2$ cluster model. The circles and the triangles indicate the positions of the oxygen and chromium ions, respectively. The direction of the triangles indicates the ordering of the Cr^{3+} moments in the antiferromagnetic state at a given domain. The cross is the location of the center of inversion.

The crystal field at the Cr site in the $(CrO_6)_2$ cluster is, in first approximation, cubic. The trigonal distortion of the ligand field can be treated as a perturbation. On doing this and rotating the crystal field axes with respect to the crystallographic axes (to facilitate calculations), we find that a convenient representation for the Cr one-particle orbitals is given by

$$\begin{aligned} t_2^{(1)} &= -|d_{3z^2-r^2}\rangle + \eta_1|p_z\rangle, \\ t_2^{(2)} &= 3^{-1/2}[\sqrt{2}\,|d_{x^2-y^2}\rangle + |d_{zx}\rangle] + \eta_2|p_x\rangle, \\ t_2^{(3)} &= 3^{-1/2}[\sqrt{2}\,|d_{xy}\rangle + |d_{yz}\rangle] + \eta_2|p_y\rangle, \\ e^{(1)} &= 3^{-1/2}[|d_{xy}\rangle - \sqrt{2}\,|d_{yz}\rangle] + \eta_3|p_y\rangle, \\ e^{(2)} &= 3^{-1/2}[|d_{x^2-y^2}\rangle - \sqrt{2}\,|d_{zx}\rangle] + \eta_3|p_x\rangle, \end{aligned} \tag{7}$$

where the spin quantum numbers have been suppressed. For the first (second) Cr ion ($\epsilon = \pm 1$), one replaces z in the right-hand side of (7) by ($z \mp z_0$), respectively. The above expression also includes the effect of the hemihedral part of the trigonal distortion, which is the most dominant interaction. This interaction is of the form ηz, and it leads to a mixing of Cr $3d$ states and Cr $4p$ states with coefficients η_1, η_2, and η_3 being proportional to η. Using (7), together with the spin-orbit interaction, one obtains after a lengthy but straightforward calculation the dynamical current operator $\mathbf{J}(\mathbf{r},t) = \langle\Psi(t)|\hat{\mathbf{J}}|\Psi(t)\rangle$. Here we report the results of this calculation; we will discuss the details elsewhere [13]. The source term entering (2) is then obtained to be

$$\mathbf{S}(\mathbf{r},t) = \frac{4\pi}{c^2}\frac{\partial}{\partial t}\mathbf{J}(\mathbf{r},t).$$

Since the full D_{3d} symmetry of Cr_2O_3 is taken into account, the calculations based on the $(CrO_6)_2$ model correctly predict all selection rules also found by the macroscopic approach. In addition, we also obtain estimates for all nonzero matrix elements of the nonlinear susceptibilities.

The interference between the MD and the ED processes in SHG can be observed experimentally when the matrix elements γ_m and χ_e occurring in (1) are roughly of the same order of magnitude. We estimate their relative order of magnitude by

$$\frac{\chi_e}{\gamma_m} \sim 4\frac{\lambda_0}{a_0}\frac{\lambda}{E_e - E_{t_2}}\frac{\eta}{E_p - E_d}\Delta(T). \tag{8}$$

Here $\lambda_0 \approx 5000$ Å is the wavelength of the emitted light, $a_0 \approx 0.69$ Å is the radius of Cr^{3+}, $\lambda \approx 100$ cm^{-1} is the spin-orbit interaction [14,15], $E_e - E_{t_2} \approx 8000$ cm^{-1} is the difference in energy between the t_2 and the e orbitals, $\eta \approx 350$ cm^{-1} is the trigonal field [15], $E_p - E_d \approx 8 \times 10^4$ cm^{-1} is the difference in energy between the d and the p orbitals that are mixed by the trigonal distortion, and $\Delta(T) \approx 1$ is the antiferromagnetic order parameter.

The additional factor of 4 occurs since there are 4 matrix elements of the same order of magnitude [see (4)]. The above expression gives the right order of magnitude, and we therefore conclude that the ED matrix element in this mechanism can indeed interfere with the MD matrix element. Clearly, the ED matrix element vanishes above T_N when time-reversal symmetry is restored.

We now consider the phenomenon of gyrotropic birefringence (GB). This is another nonreciprocal effect, the possible existence of which was first pointed out by Brown, Shtrikman, and Treves [8]. GB is a one-photon process appearing as a shift in the principal optic axes along with a change in the velocity of propagation of light. The first quantum mechanical treatment of this problem was presented by Hornreich and Shtrikman [16], who estimated that GB in Cr_2O_3 would lead to a shift in the optical axes of roughly 10^{-8} rad, viz., a very small effect. Recently, however, Krichevtsov *et al.* measured this nonreciprocal rotation and the related magnetoelectric susceptibility of Cr_2O_3 in the optical region [3]. They found that the observed values were at least 4 orders of magnitude larger than those predicted by Hornreich and Shtrikman. They also found that the temperature dependence of the nonreciprocal effects mimicked that of the order parameter. The observed intensities and temperature dependence suggest that these effects originate from the ED process we have proposed.

Now it is known that the dominant contribution to GB is from the magnetoelectric susceptibility defined by $\mathbf{M}^{(\omega)} = \alpha : \mathbf{E}^{(\omega)}$ [16]. Using (7), we have calculated the ED contribution to α in the optical region. We find that

$$\alpha_{xx} \sim 4\mu_0 ce \frac{g\mu_B}{\hbar(\omega - \omega_n)} \frac{\lambda}{E_e - E_{t_2}} \frac{\eta}{E_p - E_d} n_0 \Delta(T), \tag{9}$$

in dimensionless units. Here, n_0 is the density of Cr ions in Cr_2O_3 ($\simeq 3.3 \times 10^{28}\ \mathrm{m}^{-3}$). In the region of experimental interest, $\hbar(\omega - \omega_n) \sim 0.5$ eV. Thus, we estimate $\alpha_{xx} \sim 0.2 \times 10^{-4}$, which is of the same order of magnitude as that observed experimentally. This also means that the nonreciprocal rotation would be $\sim 10^{-4}$ rad. Since the ED process we consider couples light to the order parameter, the observed temperature dependence follows naturally from our mechanism.

To conclude, we have developed a microscopic model that explains all nonreciprocal optical effects observed below T_N in Cr_2O_3. We have shown that these effects can be explained by an electric dipole process that arises from an interplay between the spin-orbit coupling and the trigonal distortion of the ligand field. Such a process couples light directly to the antiferromagnetic order parameter. Although we have applied the theory to explain nonreciprocal optical effects in Cr_2O_3, it can be generalized to all materials where (i) the magnetic ion is not at a center of inversion and (ii) inversion symmetry is broken below T_N. In particular, we predict that such effects should be observed in V_2O_3 and $MnTiO_3$ as also in the cuprate Gd_2CuO_4 below the ordering temperature of the gadolinium magnetic subsystem, $T_N(\mathrm{Gd}) = 6.5$ K [17].

This work was stimulated by intensive discussions with M. Fiebig, D. Fröhlich, G. Sluyterman v. L., and H. J. Thiele. This work was supported by the Deutsche Forschungsgemeinschaft, by the Graduiertenkolleg "Festkörperspektroskopie," and by the European Community Human Capital and Mobility program.

[1] P. N. Argyres, Phys. Rev. **97**, 334 (1955).
[2] R. A. Fleury and R. Loudon, Phys. Rev. **166**, 514 (1968). For a comprehensive review, see M. G. Cottam and D. J. Lockwood, *Light Scattering in Magnetic Solids* (Wiley-Interscience, New York, 1986).
[3] B. B. Krichevtsov, V. V. Pavlov, R. V. Pisarev, and V. N. Gridnev, J. Phys. Condens. Matter **5**, 8233 (1993).
[4] M. Fiebig, D. Fröhlich, B. B. Krichevtsov, and R. V. Pisarev, Phys. Rev. Lett. **73**, 2127 (1994).
[5] For a discussion of reciprocity in optics, see A. L. Shelankov and G. E. Pinkus, Phys. Rev. B **46**, 3326 (1992).
[6] M. Fiebig, D. Fröhlich, and G. Sluyterman v. L., Appl. Phys. Lett. (to be published).
[7] Note, however, that domain walls can be observed optically in certain antiferromagnets when they are associated with crystal distortions. See W. L. Roth, J. Appl. Phys. **31**, 2000 (1960).
[8] W. F. Brown, Jr., S. Shtrikman, and D. Treves, J. Appl. Phys. **34**, 1233 (1963).
[9] R. R. Birss, *Symmetry and Magnetism* (North-Holland, Amsterdam, 1964).
[10] L. Rosenfeld, *Theory of Electrons* (Interscience Publishers, Inc., New York, 1951).
[11] Y. Tanabe, R. Moriya, and S. Sugano, Phys. Rev. Lett. **15**, 1023 (1965).
[12] Y. Tanabe, Prog. Theor. Phys. Suppl. **14**, 17 (1960); S. Sugano, *ibid.* **14**, 66 (1960); Takeo Izuyama and G. W. Pratt, Jr., J. Appl. Phys. **34**, 1226 (1963).
[13] V. N. Muthukumar, R. Valentí, and C. Gros (to be published).
[14] G. T. Rado, Phys. Rev. Lett. **6**, 609 (1961).
[15] S. Sugano and Y. Tanabe, J. Phys. Soc. Jpn. **13**, 880 (1958).
[16] R. M. Hornreich and S. Shtrikman, Phys. Rev. **171**, 1065 (1968).
[17] H. Wiegelmann, A. A. Stepanov, I. M. Vitebsky, A. G. Jansen, and P. Wyder, Phys. Rev. B **49**, 10 039 (1994).