

# Relativistic nature of a magnetoelectric modulus of $\text{Cr}_2\text{O}_3$ crystals: A four-dimensional pseudoscalar and its measurement

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The magnetoelectric effect of chromium sesquioxide  $\text{Cr}_2\text{O}_3$  has been determined experimentally as a function of temperature. One measures the electric field-induced magnetization on  $\text{Cr}_2\text{O}_3$  crystals or the magnetic field-induced polarization. From the magnetoelectric moduli of  $\text{Cr}_2\text{O}_3$  we extract a four-dimensional relativistic invariant pseudoscalar  $\tilde{\alpha}$ . It is temperature dependent and of the order of  $\sim 10^{-4}/Z_0$ , with  $Z_0$  as vacuum impedance. We show that the new pseudoscalar is odd under parity transformation and odd under time inversion. Moreover,  $\tilde{\alpha}$  is for  $\text{Cr}_2\text{O}_3$  what Tellegen's *gyrator* is for two port theory, the *axion* field for axion electrodynamics, and the PEMC (perfect electromagnetic conductor) for electrical engineering.

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## I. INTRODUCTION

Usually, in vacuum, the constitutive relations of classical electrodynamics are  $\mathbf{D}=\varepsilon_0\mathbf{E}$  and  $\mathbf{H}=\mathbf{B}/\mu_0$ . The electric constant  $\varepsilon_0$  (permittivity of free space) alone has no direct meaning in four-dimensional spacetime; the analogous is true for the magnetic constant  $\mu_0$  (permeability of free space). However, if we combine both constants, the situation changes. As shown by Post [1], for example, it is rather the square root of the quotient of both constants, namely  $Y_0:=\sqrt{\varepsilon_0/\mu_0}$ , the *vacuum admittance* of  $Y_0\approx 1/377\ \Omega$  that represents a *scalar* in 4 dimensional spacetime in *arbitrary* coordinates; the same is true for its reciprocal, the vacuum impedance (resistance)  $Z_0:=1/Y_0$ . Thus, it is possible to extract four-dimensional information from both three-dimensional constants, provided they are taken together.

Moreover,  $c:=1/\sqrt{\varepsilon_0\mu_0}$ , the vacuum speed of light, has also a four-dimensional meaning, even though  $c$  is only a scalar under Poincaré (inhomogeneous Lorentz) transformations. This is obvious since in noninertial, that is, accelerated frames  $c$  is no longer a constant. In this sense, the vacuum admittance has a more fundamental significance than the speed of light. The vacuum admittance can be measured by a Weber-Kohlrausch type of experiment, e.g., see Raith [2] and Brown [3], the speed of light by the well-known methods of Foucault or Fizeau [4], respectively (even though, strictly speaking, the speed of light is put to a certain constant value in SI since 1983, see [5]).

Sommerfeld's fine structure constant, the dimensionless coupling constant of the electromagnetic interaction, can be written as [6,7]

$$\alpha_f = \frac{Z_0}{2R_K}, \quad (1)$$

where  $R_K$  is the quantum Hall resistance (von Klitzing constant) associated with the quantum Hall effect. It has been shown [8] that  $R_K$ , like the vacuum resistance  $Z_0$ , is *not* influenced by the gravitational field. This underlines the fundamental importance of  $Z_0$  as well as that of  $R_K$ .

Let us now turn to *media*, namely to dielectric and magnetic media that can be described by a *local and linear* constitutive law. Suppose we consider a spatially isotropic medium. In a frame where the medium is at rest, we find  $\mathbf{D}=\varepsilon\varepsilon_0\mathbf{E}$  and  $\mathbf{H}=\mathbf{B}/(\mu\mu_0)$ , with the permittivity  $\varepsilon$  and the permeability  $\mu$  of the medium; both,  $\varepsilon$  and  $\mu$ , are dimensionless and depend in general on the frequency of the wave studied ("dispersion"). By the analogous arguments as above,  $\sqrt{\varepsilon\varepsilon_0/(\mu\mu_0)}$  is a four-scalar in arbitrary coordinates and  $1/\sqrt{\varepsilon\mu\varepsilon_0\mu_0}$  a speed in inertial coordinates. The absolute refractive index  $n$  of a medium, see [9], derives from the latter expression as relation of the vacuum speed of light to the speed in the medium. Accordingly,  $n=\sqrt{\varepsilon\mu}$ .

Usually media, in particular crystalline media, behave anisotropically. We could generalize the laws above by introducing anisotropic permittivity and permeability tensors  $\varepsilon_{ab}$  and  $\mu_{ab}$ , respectively, with  $a, b, \dots = 1, 2, 3$ . But it is better to start right away with a general *local and linear* constitutive law in the context of a four-dimensional representation of electrodynamics (Sec. II). This guarantees automatically

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relativistic covariance, and the anisotropic laws mentioned will emerge as special cases.

In this context, it turns out, see Eq. (15), that the corresponding four-dimensional constitutive tensor  $\chi^{\lambda\nu\sigma\kappa} = -\chi^{\nu\lambda\sigma\kappa} = -\chi^{\lambda\nu\kappa\sigma}$  has 36 independent components in general, with  $\lambda, \nu, \dots = 0, 1, 2, 3$ . In the center of our present paper is only one component of  $\chi^{\lambda\nu\sigma\kappa}$ , namely its totally antisymmetric piece. It is the pseudoscalar  $\tilde{\alpha} := \tilde{\epsilon}_{\lambda\nu\sigma\kappa} \chi^{\lambda\nu\sigma\kappa} / 4!$  (we sum over all repeated indices), which can be formed from the constitutive tensor  $\chi^{\lambda\nu\sigma\kappa}$  with the help of the totally antisymmetric Levi-Civita symbol  $\tilde{\epsilon}_{\lambda\nu\sigma\kappa}$ . We remind ourselves that  $\tilde{\epsilon}_{\lambda\nu\sigma\kappa} = +1$  or  $-1$  depending whether  $\lambda\nu\sigma\kappa$  denotes an even or an odd permutation of the numbers 0123, respectively; it is zero otherwise, see [10]. Then,

$$\tilde{\alpha} = \frac{1}{24} (\chi^{0123} - \chi^{0213} + \chi^{0312} - \chi^{0132} + \dots). \quad (2)$$

Observing the antisymmetries of  $\chi^{\lambda\nu\sigma\kappa}$ , we find

$$\tilde{\alpha} = \frac{1}{6} (\chi^{0123} + \chi^{0231} + \chi^{0312} + \chi^{2301} + \chi^{3102} + \chi^{1203}). \quad (3)$$

The first three terms in the parentheses are related to the magnetoelectric (ME) effect in an external *magnetic* field,  $\text{ME}_B$ , the last three terms to the magnetoelectric effect in an external *electric* field,  $\text{ME}_E$ . That  $\tilde{\alpha}$  is a pseudoscalar, indeed, can be seen from the transformation properties of the quantities involved, see [11]. The dimension of  $\tilde{\alpha}$  is *1/resistance*.

The constitutive tensor  $\chi^{\lambda\nu\sigma\kappa}$  was seemingly first introduced by Tamm [12], see also Post [1], and later discussed by O'Dell [13] in the context of magnetoelectric media—that is, media in which an *electric* field  $\mathbf{E}$  induces a *magnetic* excitation  $\mathbf{H}$  and a magnetic field  $\mathbf{B}$  an electric excitation  $\mathbf{D}$ , see Fiebig [14] for a recent review. All four cited authors assumed a further symmetry, namely  $\chi^{\lambda\nu\sigma\kappa} = \chi^{\sigma\kappa\lambda\nu}$  (“vanishing of the skewon part,” see [11]). This symmetry emerges, as soon as one stipulates that the constitutive relation can be derived from a Lagrangian (thereby excluding irreversible processes). Then, in particular, i.e., for  $\chi^{\lambda\nu\sigma\kappa} = \chi^{\sigma\kappa\lambda\nu}$ ,

$$\tilde{\alpha} = \frac{1}{3} (\chi^{0123} + \chi^{0231} + \chi^{0312}). \quad (4)$$

Thus we need only three components of the constitutive tensor for the determination of  $\tilde{\alpha}$  in the case of the vanishing skewon part. Post argued [1] (not very convincingly, we should say) that the pseudoscalar (4) ought to vanish:  $\tilde{\alpha} = 0$ . This condition was dubbed “Post constraint” by Lakhtakia [15]. It was *not* assumed by O'Dell so that he was left with  $20+1$  independent components of the constitutive tensor, see [13], p.44.

Later a fierce dispute arose about the Post constraint. The situation was reviewed by Lakhtakia [15,16], de Lange and Raab [17], Raab and Sihvola [18], Raab and de Lange [19], Sihvola [20], Sihvola and Tretyakov [21], and in [22], see also [23,24], where more references to the relevant literature can be found. The evidence was mounting that there is no reason to assume the validity of the Post constraint in general. This point of view will be shown to be correct in this paper.

According to a theory of Dzyaloshinskii [25], which was based on an analysis of neutron scattering data, susceptibility measurements, and symmetry (as derived in [26]), a crystal of antiferromagnetic chromium oxide  $\text{Cr}_2\text{O}_3$  is the substance par excellence for discovering the magnetoelectric effect. In our paper, we consider only crystals being in an antiferromagnetic single domain state. In Sec. III, we will describe Dzyaloshinskii theory of  $\text{Cr}_2\text{O}_3$  and we will determine the four-dimensional pseudoscalar  $\tilde{\alpha}$  of  $\text{Cr}_2\text{O}_3$  in this framework. In Sec. IV, after a short introduction on dimensions and units, an overview will be given over magnetoelectric experiments with  $\text{Cr}_2\text{O}_3$ . Corresponding unpublished measurements by one of us (J.-P.R.) will be presented in some detail. In Sec. V, we study the experimentally determined magnetoelectric moduli of  $\text{Cr}_2\text{O}_3$ . Then, we extract from the data the relativistic pseudoscalar  $\tilde{\alpha}$  for  $\text{Cr}_2\text{O}_3$ . It turns out to be temperature dependent and is of the order of  $\tilde{\alpha} \approx 10^{-4}/Z_0$ . This is a typical magnitude for magnetoelectric moduli (Borovik-Romanov and Grimmer [27]). Thus, it is small but definitely *nonvanishing*. This proves experimentally that the Post constraint is ruled out as a generally valid law. In Sec. VI, we show that the pseudoscalar (or axion) piece of the magnetoelectric susceptibility of  $\text{Cr}_2\text{O}_3$  violates parity and time inversion invariance and doesn't contribute to the electromagnetic energy. In Sec. VII, we mention other substances that, besides  $\text{Cr}_2\text{O}_3$ , carry an axion piece, and in Sec. VIII, we will discuss the implications of our result to other disciplines within physics and electrical engineering.

In this article, we will base our considerations on the four-dimensional tensor analytical formalism as described by Post [1], which can be understood by experimentalists and theoreticians alike; moreover, Post's book, as a Dover edition, is easily available. Accordingly, the time coordinate  $x^0 = t$  has the dimension of ‘time’ whereas the spatial coordinates  $x^1, x^2, x^3$  are related to the dimension of ‘length.’ Only intermittently we will mention the formalism of exterior differential forms, as it is used in the book [11]. The equations in our paper are quantity equations that are valid for an arbitrary system of units. However, if in Secs. IV and V we turn to experiments, then we use SI and sometimes Gaussian units, which are widespread in the literature.

## II. LOCAL AND LINEAR MAGNETOELECTRIC MEDIA, THE PSEUDOSCALAR $\tilde{\alpha}$

### A. Excitation and field strength

Post [1] represents the four-dimensional electromagnetic field tensors according to

$$\mathfrak{G}^{\mu\nu} = -\mathfrak{G}^{\nu\mu} = \begin{pmatrix} 0 & D_1 & D_2 & D_3 \\ -D_1 & 0 & H_3 & -H_2 \\ -D_2 & -H_3 & 0 & H_1 \\ -D_3 & H_2 & -H_1 & 0 \end{pmatrix}, \quad (5)$$

$$F_{\mu\nu} = -F_{\nu\mu} = \begin{pmatrix} 0 & -E_1 & -E_2 & -E_3 \\ E_1 & 0 & B_3 & -B_2 \\ E_2 & -B_3 & 0 & B_1 \\ E_3 & B_2 & -B_1 & 0 \end{pmatrix}. \quad (6)$$

The coordinate indices  $\mu, \nu, \dots$  run from 0 to 3. We have to distinguish carefully between the upper and lower indices for reasons of general four-dimensional invariance. We use Post's conventions throughout, unless indicated otherwise.

The field  $\mathfrak{G}^{\mu\nu}$ , the electromagnetic *excitation*, represents a tensor density of weight +1.  $F_{\mu\nu}$  is a tensor. In order to transform  $\mathfrak{G}^{\mu\nu}$  to a pseudo tensor with lower indices, we introduce (Einstein's summation convention assumed)

$$\tilde{G}_{\mu\nu} := \frac{1}{2} \tilde{\epsilon}_{\mu\nu\kappa\lambda} \mathfrak{G}^{\kappa\lambda} = -\tilde{G}_{\nu\mu}, \quad (7)$$

with the totally antisymmetric Levi-Civita symbol  $\tilde{\epsilon}_{\mu\nu\kappa\lambda} = 0, \pm 1$ . Here  $\tilde{\epsilon}_{\mu\nu\kappa\lambda}$  is a pseudo tensor density of weight  $-1$ . We use  $\tilde{\epsilon}_{\mu\nu\kappa\lambda}$  for the dualization in Eq. (7), since this relation is invariant under proper as well as under improper transformations (i.e., those that include coordinate reflections). We will denote pseudo tensors by a tilde throughout our article. By simple algebra we can construct the corresponding matrix for  $\tilde{G}_{\mu\nu}$ :

$$\tilde{G}_{\mu\nu} = -\tilde{G}_{\nu\mu} = \begin{pmatrix} 0 & H_1 & H_2 & H_3 \\ -H_1 & 0 & D_3 & -D_2 \\ -H_2 & -D_3 & 0 & D_1 \\ -H_3 & D_2 & -D_1 & 0 \end{pmatrix}. \quad (8)$$

Whereas  $F_{\mu\nu}$ , the electromagnetic *field strength*, is a tensor,  $\tilde{G}_{\mu\nu}$  is a *pseudo* tensor, i.e., they behave differently under a reflection  $x^0 \rightarrow -x^0$  or  $x^1 \rightarrow -x^1$  etc. In the language of differential forms, the electromagnetic field can be represented by two two-forms, the excitation  $\tilde{G} = \tilde{G}_{\mu\nu} dx^\mu \wedge dx^\nu / 2$  and the field strength  $F = F_{\mu\nu} dx^\mu \wedge dx^\nu / 2$ , respectively. The  $\tilde{G}$  is a pseudo two-form, also called *twisted* (or odd) two-form, the  $F$  is an untwisted (or even) two-form.

### B. Maxwell equations

The Maxwell equations in premetric form are [1]

$$\partial_\nu \mathfrak{G}^{\mu\nu} = \mathfrak{J}^\mu, \quad \partial_\mu F_{\nu\lambda} + \partial_\nu F_{\lambda\mu} + \partial_\lambda F_{\mu\nu} = 0. \quad (9)$$

If we introduce the dual of the electric current density,  $\tilde{J}_{\mu\nu\lambda} := \tilde{\epsilon}_{\mu\nu\lambda\kappa} \mathfrak{J}^\kappa$ , then the inhomogeneous equation can be transformed into the equivalent equation

$$\partial_\mu \tilde{G}_{\nu\lambda} + \partial_\nu \tilde{G}_{\lambda\mu} + \partial_\lambda \tilde{G}_{\mu\nu} = \tilde{J}_{\mu\nu\lambda}. \quad (10)$$

In a more condensed way, we may also write the Maxwell equations as

$$\partial_{[\mu} \tilde{G}_{\nu\lambda]} = \frac{1}{3} \tilde{J}_{\mu\nu\lambda}, \quad \partial_{[\mu} F_{\nu\lambda]} = 0, \quad (11)$$

where  $[\mu\nu\lambda] = \frac{1}{6}(\mu\nu\lambda - \nu\mu\lambda + \lambda\mu\nu \mp \dots)$ , i.e., a plus (minus) sign occurs before even (odd) permutations. In the differen-

tial form language, the Maxwell equations (11) read

$$d\tilde{G} = \tilde{J}, \quad dF = 0, \quad (12)$$

with the three-form of the electric current  $\tilde{J} = \frac{1}{6} \tilde{J}_{\mu\nu\lambda} dx^\mu \wedge dx^\nu \wedge dx^\lambda$ .

### C. Constitutive relation

The system of the 4+4 Maxwell equations (9) for the 6 + 6 independent components of the electromagnetic field  $\mathfrak{G}^{\mu\nu}$  and  $F_{\lambda\rho}$  is evidently underdetermined. To complete this system, a constitutive relation of the form

$$\mathfrak{G}^{\mu\nu} = \mathfrak{G}^{\mu\nu}(F_{\lambda\rho}) \quad (13)$$

has to be assumed. The constitutive relation (13) is independent of the Maxwell equations and its form can be determined by using experimental results. For *vacuum*, the constitutive relation ("spacetime relation") is simple,

$$\mathfrak{G}^{\lambda\nu} = Y_0 \sqrt{-g} F^{\lambda\nu}, \quad (14)$$

with  $g := \det g_{\rho\sigma} \neq 0$  and  $F^{\mu\nu} := g^{\mu\alpha} g^{\nu\beta} F_{\alpha\beta}$ . Here  $g_{\mu\nu}$  are the covariant components of the metric of spacetime with signature  $(- + + +)$ , its contravariant components  $g^{\lambda\nu}$  can be determined via  $g_{\mu\lambda} g^{\lambda\nu} = \delta_{\mu}^{\nu}$ . For the excitation pseudotensor, we have  $\tilde{G}_{\mu\nu} = Y_0 \tilde{\epsilon}_{\mu\nu\sigma\tau} \sqrt{-g} F^{\sigma\tau} / 2$  and, in exterior calculus, with the Hodge  $*$  operator,  $\tilde{G} = Y_0 * F$ .

Turning to general *magnetolectric* media, we assume with Tamm [12], see also Post [1], the most general local [28] and linear homogeneous relation

$$\mathfrak{G}^{\lambda\nu} = \frac{1}{2} \chi^{\lambda\nu\sigma\kappa} F_{\sigma\kappa}, \quad (15)$$

where  $\chi^{\lambda\nu\sigma\kappa}$  is a *constitutive tensor density* of rank 4 and weight +1, with the dimension  $[\chi] = 1/\text{resistance}$ . Since both  $\mathfrak{G}^{\lambda\nu}$  and  $F_{\sigma\kappa}$  are antisymmetric in their indices, we have  $\chi^{\lambda\nu\sigma\kappa} = -\chi^{\lambda\nu\kappa\sigma} = -\chi^{\nu\lambda\sigma\kappa}$ . An antisymmetric pair of indices corresponds, in four dimensions, to six independent components. Thus the constitutive tensor can be considered as a  $6 \times 6$  matrix with 36 independent components.

A  $6 \times 6$  matrix can be decomposed in its tracefree symmetric part (20 independent components), its antisymmetric part (15 components), and its trace (1 component). On the level of  $\chi^{\lambda\nu\sigma\kappa}$ , this *decomposition* is reflected in

$$\chi^{\lambda\nu\sigma\kappa} = {}^{(1)}\chi^{\lambda\nu\sigma\kappa} + {}^{(2)}\chi^{\lambda\nu\sigma\kappa} + {}^{(3)}\chi^{\lambda\nu\sigma\kappa}, \quad (16)$$

$$36 = 20 \oplus 15 \oplus 1.$$

The third part, the *axion* part, is totally antisymmetric and as such proportional to the Levi-Civita symbol,  ${}^{(3)}\chi^{\lambda\nu\sigma\kappa} := \chi^{[\lambda\nu\sigma\kappa]} = \tilde{\alpha} \tilde{\epsilon}^{\lambda\nu\sigma\kappa}$ . Note that  $\tilde{\alpha}$  is a pseudoscalar since  $\tilde{\epsilon}^{\lambda\nu\sigma\kappa}$  has weight  $-1$ . Therefore, the weight of  $\tilde{\epsilon}^{\lambda\nu\sigma\kappa}$  is essential information. The second part, the *skewon* part, is defined according to  ${}^{(2)}\chi^{\mu\nu\lambda\rho} := \frac{1}{2}(\chi^{\mu\nu\lambda\rho} - \chi^{\lambda\rho\mu\nu})$ . If the constitutive equation can be derived from a Lagrangian, which is the case as long as only reversible processes are considered, then  ${}^{(2)}\chi^{\lambda\nu\sigma\kappa} = 0$ . The *principal* part  ${}^{(1)}\chi^{\lambda\nu\sigma\kappa}$  fulfills the symme-

tries  $(1)\chi^{\lambda\nu\sigma\kappa} = (1)\chi^{\sigma\kappa\lambda\nu}$  and  $(1)\chi^{[\lambda\nu\sigma\kappa]} = 0$ . The constitutive relation now reads

$$\mathfrak{G}^{\lambda\nu} = \frac{1}{2} \left( (1)\chi^{\lambda\nu\sigma\kappa} + (2)\chi^{\lambda\nu\sigma\kappa} + \tilde{\alpha} \tilde{\epsilon}^{\lambda\nu\sigma\kappa} \right) F_{\sigma\kappa}. \quad (17)$$

In order to compare this with experiments, we have to split Eq. (17) into time and space parts. As shown in [22] in detail, we can parametrize the *principal* part by the six permittivities  $\epsilon^{ab} = \epsilon^{ba}$ , the six permeabilities  $\mu_{ab} = \mu_{ba}$ , and the eight magnetoelectric pieces  $\gamma^a_b$  (its trace vanishes,  $\gamma^c_c = 0$ ) and the *skewon* part by the three permittivities  $n_a$ , the three permeabilities  $m^a$ , and the nine magnetoelectric pieces  $s_a^b$ . Then, the constitutive relation (17) can be rewritten as

$$D^a = (\epsilon^{ab} - \epsilon^{abc} n_c) E_b + (\gamma^a_b + s_b^a - \delta_b^a s_c^c) B^b + \tilde{\alpha} B^a, \quad (18)$$

$$H_a = (\mu_{ab}^{-1} - \hat{\epsilon}_{abc} m^c) B^b + (-\gamma^b_a + s_a^b - \delta_a^b s_c^c) E_b - \tilde{\alpha} E_a. \quad (19)$$

Here  $\epsilon^{abc} = \hat{\epsilon}_{abc} = \pm 1, 0$  are the three-dimensional Levi-Civita symbols. As can be seen from our derivation,  $\tilde{\alpha}$  is a four-dimensional pseudo (or axial) scalar, whereas  $s_c^c$  is only a three-dimensional scalar. The cross-term  $\gamma^a_b$  is related to the Fresnel-Fizeau effects. The skewon contributions  $m^c, n_c$  are responsible for electric and magnetic Faraday effects, respectively, whereas the skewon terms  $s_a^b$  describe optical activity. Equivalent constitutive relations were formulated by Serdyukov *et al.* [29], p. 86, and studied in quite some detail.

According to Post [1], the pseudoscalar  $\tilde{\alpha}$  should vanish for the vacuum and for all media. We will show in the next section that, in general, this is not the case. For Cr<sub>2</sub>O<sub>3</sub> the pseudoscalar  $\tilde{\alpha}$  turns out to be finite.

### III. ANTIFERROMAGNET Cr<sub>2</sub>O<sub>3</sub> AND THE THEORY OF DZYALOSHINSKII

On the basis of neutron scattering data [26] and susceptibility measurements [30] of the antiferromagnetic chromium sesquioxide [31] Cr<sub>2</sub>O<sub>3</sub>, Dzyaloshinskii [25] was able to establish the magnetic symmetry class  $\bar{3}'m'$  of the Cr<sub>2</sub>O<sub>3</sub> crystals. In accordance with these results, Dzyaloshinskii developed, by starting from a thermodynamic potential quadratic and bilinear in  $\mathbf{E}$  and  $\mathbf{H}$ , as foreseen by Landau & Lifshitz [32], a theory for the electromagnetic constitutive relations for Cr<sub>2</sub>O<sub>3</sub>. We write them here as *quantity equations* that are valid in an arbitrary system of units [33]:

$$D_x = \epsilon_{\perp} \epsilon_0 E_x + \alpha_{\perp} \sqrt{\epsilon_0 \mu_0} H_x, \quad (20)$$

$$D_y = \epsilon_{\perp} \epsilon_0 E_y + \alpha_{\perp} \sqrt{\epsilon_0 \mu_0} H_y, \quad (21)$$

$$D_z = \epsilon_{\parallel} \epsilon_0 E_z + \alpha_{\parallel} \sqrt{\epsilon_0 \mu_0} H_z, \quad (22)$$

and

$$B_x = \mu_{\perp} \mu_0 H_x + \alpha_{\perp} \sqrt{\epsilon_0 \mu_0} E_x, \quad (23)$$

$$B_y = \mu_{\perp} \mu_0 H_y + \alpha_{\perp} \sqrt{\epsilon_0 \mu_0} E_y, \quad (24)$$

$$B_z = \mu_{\parallel} \mu_0 H_z + \alpha_{\parallel} \sqrt{\epsilon_0 \mu_0} E_z. \quad (25)$$

The  $z$  axis is parallel to the optical axis of Cr<sub>2</sub>O<sub>3</sub>. Remember that  $\sqrt{\epsilon_0 \mu_0} = 1/c$  and  $\sqrt{\epsilon_0/\mu_0} = 1/Z_0$ , with  $c$  as velocity of light and  $Z_0$  as vacuum impedance. Here we have permittivities parallel and perpendicular to the  $z$  axis of the crystal, namely  $\epsilon_{\parallel}, \epsilon_{\perp}$ , analogous permeabilities  $\mu_{\parallel}, \mu_{\perp}$  and magnetoelectric moduli  $\alpha_{\parallel}, \alpha_{\perp}$ . Note that all these moduli are dimensionless, and this is true for all systems of units. Our dimensionless  $\alpha$ 's are different from the ones used by experimentalists and theoreticians up to now. We will discuss the transition to different systems of units in Sec. IV A below.

As we can see from Eqs. (5) and (15), we have to get  $(\mathbf{D}, \mathbf{H})$  on the left-hand side and  $(\mathbf{E}, \mathbf{B})$  on the right-hand side in order to end up with a constitutive law that is written in a relativistically covariant form. For this purpose, we resolve the last three equations with respect to  $H$ :

$$H_x = \frac{1}{\mu_{\perp} \mu_0} B_x - \frac{\alpha_{\perp}}{\mu_{\perp}} \sqrt{\frac{\epsilon_0}{\mu_0}} E_x, \quad (26)$$

$$H_y = \frac{1}{\mu_{\perp} \mu_0} B_y - \frac{\alpha_{\perp}}{\mu_{\perp}} \sqrt{\frac{\epsilon_0}{\mu_0}} E_y, \quad (27)$$

$$H_z = \frac{1}{\mu_{\parallel} \mu_0} B_z - \frac{\alpha_{\parallel}}{\mu_{\parallel}} \sqrt{\frac{\epsilon_0}{\mu_0}} E_z. \quad (28)$$

On substitution into Eqs. (20)–(22), we find

$$D_x = \left( \epsilon_{\perp} - \frac{\alpha_{\perp}^2}{\mu_{\perp}} \right) \epsilon_0 E_x + \frac{\alpha_{\perp}}{\mu_{\perp}} \sqrt{\frac{\epsilon_0}{\mu_0}} B_x, \quad (29)$$

$$D_y = \left( \epsilon_{\perp} - \frac{\alpha_{\perp}^2}{\mu_{\perp}} \right) \epsilon_0 E_y + \frac{\alpha_{\perp}}{\mu_{\perp}} \sqrt{\frac{\epsilon_0}{\mu_0}} B_y, \quad (30)$$

$$D_z = \left( \epsilon_{\parallel} - \frac{\alpha_{\parallel}^2}{\mu_{\parallel}} \right) \epsilon_0 E_z + \frac{\alpha_{\parallel}}{\mu_{\parallel}} \sqrt{\frac{\epsilon_0}{\mu_0}} B_z. \quad (31)$$

Now we have to compare with the local and linear constitutive relation (18) and (19). Since Dzyaloshinskii assumed that his constitutive relations can be derived from a Hamiltonian, it is clear that the *skewon piece* with its 15 independent components *vanishes* identically, see [11], Eq. (D.1.44). But this can be also read off from comparing Eqs. (18) and (19) with (26)–(31). The skewon pieces  $n_c$  and  $m^c$  must be zero, since the  $D_a$  in Eqs. (29)–(31) are proportional to  $E_a$ , and the  $H_a$  in Eqs. (26)–(28) are proportional to  $B_a$ . A similar consideration shows that  $s_a^b = 0$ , since  $s_a^b$  only provides off-diagonal pieces. Consequently, Eqs. (18) and (19) reduce to

$$D^a = \epsilon^{ab} E_b + \gamma^a_b B^b + \tilde{\alpha} B^a, \quad (32)$$

$$H_a = \mu_{ab}^{-1} B^b - \gamma^b_a E_b - \tilde{\alpha} E_a, \quad (33)$$

with 21 independent moduli. The permittivity matrix  $\epsilon^{ab}$  and the impermeability matrix  $\mu_{ab}^{-1}$  are both symmetric and possess 6 independent components each, the magnetoelectric cross term with  $\gamma^a_b$ , which is tracefree,  $\gamma^c_c = 0$ , has 8 inde-



pendent components. The four-dimensional pseudoscalar (we call it also the axion parameter) represents 1 component.

By comparing Eqs. (32) and (33) with the above equations (29)–(31) and (26)–(28), we can read off the permittivity

$$\varepsilon^{ab} = \varepsilon_0 \begin{pmatrix} \varepsilon_{\perp} - \frac{\alpha_{\perp}^2}{\mu_{\perp}} & 0 & 0 \\ 0 & \varepsilon_{\perp} - \frac{\alpha_{\perp}^2}{\mu_{\perp}} & 0 \\ 0 & 0 & \varepsilon_{\parallel} - \frac{\alpha_{\parallel}^2}{\mu_{\parallel}} \end{pmatrix} \quad (34)$$

and the impermeability

$$\mu_{ab}^{-1} = \mu_0^{-1} \begin{pmatrix} \mu_{\perp}^{-1} & 0 & 0 \\ 0 & \mu_{\perp}^{-1} & 0 \\ 0 & 0 & \mu_{\parallel}^{-1} \end{pmatrix}. \quad (35)$$

For the magnetoelectric cross terms, we have

$$\gamma_x^x B^x + \tilde{\alpha} B^x = \frac{\alpha_{\perp}}{\mu_{\perp}} \sqrt{\frac{\varepsilon_0}{\mu_0}} B_x, \quad (36)$$

$$\gamma_y^y B^y + \tilde{\alpha} B^y = \frac{\alpha_{\perp}}{\mu_{\perp}} \sqrt{\frac{\varepsilon_0}{\mu_0}} B_y, \quad (37)$$

$$\gamma_z^z B^z + \tilde{\alpha} B^z = \frac{\alpha_{\parallel}}{\mu_{\parallel}} \sqrt{\frac{\varepsilon_0}{\mu_0}} B_z, \quad (38)$$

and

$$-\gamma_x^x E_x - \tilde{\alpha} E_x = -\frac{\alpha_{\perp}}{\mu_{\perp}} \sqrt{\frac{\varepsilon_0}{\mu_0}} E_x, \quad (39)$$

$$-\gamma_y^y E_y - \tilde{\alpha} E_y = -\frac{\alpha_{\perp}}{\mu_{\perp}} \sqrt{\frac{\varepsilon_0}{\mu_0}} E_y, \quad (40)$$

$$-\gamma_z^z E_z - \tilde{\alpha} E_z = -\frac{\alpha_{\parallel}}{\mu_{\parallel}} \sqrt{\frac{\varepsilon_0}{\mu_0}} E_z. \quad (41)$$

Note that in the Cartesian coordinates used by Dzyaloshinskii we have  $B^x = B_x$ , etc., since the spatial metric is Euclidean with signature (+ + +). Thus we are left with

$$\gamma_x^x + \tilde{\alpha} = \frac{\alpha_{\perp}}{\mu_{\perp}} \sqrt{\frac{\varepsilon_0}{\mu_0}}, \quad (42)$$

$$\gamma_y^y + \tilde{\alpha} = \frac{\alpha_{\perp}}{\mu_{\perp}} \sqrt{\frac{\varepsilon_0}{\mu_0}}, \quad (43)$$

$$\gamma_z^z + \tilde{\alpha} = \frac{\alpha_{\parallel}}{\mu_{\parallel}} \sqrt{\frac{\varepsilon_0}{\mu_0}}. \quad (44)$$

One of the triplets of Eqs. (36)–(41) is redundant because of the vanishing of the skewon piece of  $\chi^{\lambda\nu\sigma\kappa}$ .

The matrix  $\gamma$  is traceless:  $\gamma_x^x + \gamma_y^y + \gamma_z^z = 0$ . If we add up all three equations, we find for the pseudoscalar (or axion) piece

$$\tilde{\alpha} = \frac{1}{3} \left( 2 \frac{\alpha_{\perp}}{\mu_{\perp}} + \frac{\alpha_{\parallel}}{\mu_{\parallel}} \right) \sqrt{\frac{\varepsilon_0}{\mu_0}}. \quad (45)$$

Resubstituted into Eqs. (42)–(44), the magnetoelectric  $\gamma$  matrix becomes

$$\gamma^a_b = \frac{1}{3} \left( \frac{\alpha_{\perp}}{\mu_{\perp}} - \frac{\alpha_{\parallel}}{\mu_{\parallel}} \right) \sqrt{\frac{\varepsilon_0}{\mu_0}} \begin{pmatrix} 1 & 0 & 0 \\ 0 & 1 & 0 \\ 0 & 0 & -2 \end{pmatrix}, \quad (46)$$

that is, it has only nonvanishing diagonal components! The magnetoelectric matrix  $\gamma^a_b$  as well as the pseudoscalar  $\tilde{\alpha}$  carry the dimension of 1/resistance. Conventionally, in the “magnetoelectric literature” the  $\gamma$ -matrix and  $\tilde{\alpha}$  are collected in the “relativistic” matrix

$$\text{rel} \alpha^a_b := \gamma^a_b + \tilde{\alpha} \delta^a_b = \sqrt{\frac{\varepsilon_0}{\mu_0}} \begin{pmatrix} \frac{\alpha_{\perp}}{\mu_{\perp}} & 0 & 0 \\ \frac{\alpha_{\perp}}{\mu_{\perp}} & 0 & 0 \\ 0 & 0 & \frac{\alpha_{\parallel}}{\mu_{\parallel}} \end{pmatrix}. \quad (47)$$

It is called relativistic, since it occurs in the context of the relativistic ( $\mathbf{E}, \mathbf{B}$ ) system, see Eqs. (32) and (33).

Since there are doubts in the literature about the correctness of Dzyaloshinskii’s theory, see Lakhtakia [15], it is important to note that O’Dell [13], pp. 115/116, and Janner [34], p. 205, (see also Rado and Folen [35] and O’Dell [36]) analyzed the crystal structure of  $\text{Cr}_2\text{O}_3$  and determined the form the matrices  $\varepsilon^{ab}$ ,  $\mu_{ab}^{-1}$ , and  $\text{rel} \alpha^a_b$  ought to possess. They found

$$\varepsilon^{ab} \sim \mu_{ab}^{-1} \sim \text{rel} \alpha^a_b \sim \begin{pmatrix} \bullet & \cdot & \cdot \\ \cdot & \bullet & \cdot \\ \cdot & \cdot & * \end{pmatrix}, \quad (48)$$

where nonvanishing entries are denoted by  $\bullet$  and  $*$ , respectively. A comparison with Eqs. (34), (35), and (46) confirms Dzyaloshinskii’s theory.

Summing up: The nonvanishing magnetoelectric moduli for  $\text{Cr}_2\text{O}_3$  can be determined with the help of Eqs. (45) and (46). Let us stress that  $\varepsilon_{\parallel}$ ,  $\varepsilon_{\perp}$ ,  $\mu_{\parallel}$ ,  $\mu_{\perp}$ ,  $\alpha_{\parallel}$ , and  $\alpha_{\perp}$ , according to their definitions (20)–(25), are measured in an external  $\mathbf{E}$  and/or an external  $\mathbf{H}$  field.

## IV. Magnetoelectric experiments with $\text{Cr}_2\text{O}_3$

### A. Dimensions and units

Since in the literature, which we need for extracting data, different systems of units are used, we want to underline again that usually all our equations are *quantity equations*, which are valid in all systems of units; only in the “experimental” Secs. IV and V, we will turn to specific systems of units and some equations may be unit-dependent; see also

Rivera [37] in this context. We will go into some detail here, since these questions often lead to misunderstandings between theoreticians and experimentalists.

A *physical quantity*  $Q$  is given by

$$Q = \{Q\}[Q]. \quad (49)$$

Here  $\{Q\}$  is a numerical value and  $[Q]$  the physical dimension of the quantity  $Q$ . For instance, we have for a (one-dimensional) *displacement*  $s$ ,

$$s = \{s\}[s] = \{s\} \text{ length}, \quad (50)$$

where length is the dimension of  $s$ . So far, all of the equations in Secs. I–III are quantity equations. They interrelate “physical quantities” that consist of numbers and dimensions. Like in Eqs. (49) and (50), they are totally independent of any system of units. In many papers and books, the equations are only valid in one system of units, they are *numerical* equations, like in Jackson’s book on electrodynamics [38], for example. In our paper, as in Post [1] or in [11], the equations are quantity equations and are *always* valid, independent of the units chosen.

In a second step, if we relate our equations to measured values, we need a *system of units*. Then, for the example above,

$$\begin{aligned} s &= 15 \text{ m} = 1500 \text{ cm} \approx 45 \text{ ft} = \dots = \{s\}' \text{ m} = \{s\}''' \text{ cm} \\ &= \{s\}'' \text{ ft} = \dots \end{aligned} \quad (51)$$

The numerical value  $\{s\}$  depends on the unit chosen. In fact,

$$\frac{\{s\}'}{\{s\}''} = \frac{[s]''}{[s]'}, \quad \frac{\{s\}'}{\{s\}'''} = \frac{[s]'''}{[s]'}, \quad (52)$$

etc., that is, we have *reciprocal proportionality*. This is one of the fundamental laws of dimensional theory. The physical quantity  $s$  is *invariant*, i.e., it does not change, but its numerical value  $\{s\}$  may change according to the choice of the unit.

In the center of our interest is the pseudoscalar  $\tilde{\alpha}$ . According to Eq. (33), it has the dimension

$$[\tilde{\alpha}] = \frac{[H_a]}{[E_a]} = \frac{\text{current}}{\text{length}} \times \frac{\text{length}}{\text{voltage}} = \frac{1}{\text{resistance}}. \quad (53)$$

Moreover, the permittivity matrix  $\varepsilon^{ab}$ , according to Eq. (32), has the dimension

$$[\varepsilon^{ab}] = \frac{[D^a]}{[E_a]} = \frac{\text{charge}}{\text{area}} \times \frac{\text{length}}{\text{voltage}} = \frac{1}{\text{velocity}} \times \frac{1}{\text{resistance}} \quad (54)$$

and the impermeability matrix  $\mu_{ab}^{-1}$ , according to Eq. (33),

$$\begin{aligned} [\mu_{ab}^{-1}] &= \frac{[H_a]}{[B^a]} = \frac{\text{current}}{\text{length}} \times \frac{\text{area}}{\text{voltage} \times \text{time}} \\ &= \text{velocity} \times \frac{1}{\text{resistance}}. \end{aligned} \quad (55)$$

In particular, we have

$$[\varepsilon_0] = \frac{1}{\text{vel.}} \times \frac{1}{\text{resist.}}, \quad [\mu_0^{-1}] = \text{vel.} \times \frac{1}{\text{resist.}},$$

$$\left[ \sqrt{\frac{\varepsilon_0}{\mu_0}} \right] = \frac{1}{\text{resist.}}. \quad (56)$$

Accordingly, we can summarize these considerations in

$$\frac{[\varepsilon^{ab}]}{[\varepsilon_0]} = \frac{[\mu_{ab}^{-1}]}{[\mu_0^{-1}]} = \frac{[\gamma^a_b]}{[\sqrt{\varepsilon_0/\mu_0}]} = \frac{[\tilde{\alpha}]}{[\sqrt{\varepsilon_0/\mu_0}]} = 1. \quad (57)$$

These quotients are dimensionless in *all* systems of units. Here  $\varepsilon_0$  and  $\mu_0$  are universal constants. They have in different systems of units different *numerical values*. We have  $\sqrt{\varepsilon_0\mu_0} = 1/c$  and, in particular,

$$\sqrt{\frac{\varepsilon_0}{\mu_0}} = \frac{1}{Z_0} \approx \frac{1}{376.73\Omega} \text{ in SI,}$$

$$\sqrt{\frac{\varepsilon_0}{\mu_0}} = \frac{c}{4\pi} \text{ in Gaussian units,}$$

$$\sqrt{\frac{\varepsilon_0}{\mu_0}} = c \text{ in Heaviside-Lorentz}$$

(“rationalized Gaussian”) units. (58)

Therefore, if one spots an  $\varepsilon_0$  and/or a  $\mu_0$  in our equations, like in Eqs. (20)–(25), it does *not* mean that we are in SI, but rather that we use quantity equations with correct dimensions.

The thermodynamic potential (an energy density) relevant for the magnetoelectrical effect contains cross terms between electric and magnetic fields. As a quantity equation, it reads, restricting ourselves to the linear regime,

$$-g(\mathbf{E}, \mathbf{H}; T) = \dots + \underbrace{\alpha^{ab}}_{\neq \ell} E_a H_b = \dots + \sqrt{\varepsilon_0\mu_0} \underbrace{\alpha_*^{ab}}_{\text{dim.-less}} E_a H_b. \quad (59)$$

This linear part of a power series development is valid for “small” values of the  $E_a$  and  $H_b$  fields, at least relatively to the internal crystal fields. The  $\alpha_{\perp}$  and  $\alpha_{\parallel}$  of Sec. III are components of the matrix  $\alpha_*^{ab}$ . Hence, strictly speaking, we should have put a star to all of them. However, for convenience we dropped these stars.

We will concentrate here on SI and on the Gaussian system. In the Gaussian system of units, a mixed system consisting of electrical (electrostatic) and magnetic cgs units, see, e.g., Sommerfeld [39], Panofsky and Phillips [40], or Jackson [38], we have the following field redefinitions:

$${}^G E_a = E_a, \quad {}^G D_a = 4\pi D_a, \quad {}^G H_a = \frac{4\pi}{c} H_a, \quad {}^G B_a = c B_a. \quad (60)$$

The speed of light  $c$  is instrumental for making the dimensions of  ${}^G E_a$  and  ${}^G B_a$  equal to each other,  $[{}^G E_a] = [{}^G B_a]$ , and, similarly,  $[{}^G D_a] = [{}^G H_a]$ . The  $4\pi$  removes this factor from the Coulomb law. With the field redefinitions (60) and with the

convention in the Gaussian system  ${}^G\alpha^{ab} := c\alpha^{ab}$ , Eq. (59) can be rewritten as (see Landau-Lifshitz [32] and Dzyaloshinskii [25] for the Gaussian system) [41]

$$\begin{aligned} -g(\mathbf{E}, \mathbf{H}; T) &= \dots + \underbrace{{}^{\text{SI}}\alpha^{ab}}_{\text{dim.-less}} {}^{\text{SI}}E_a {}^{\text{SI}}H_b \\ &= \dots + \sqrt{\varepsilon_0\mu_0} \underbrace{{}^{\text{SI}}\alpha^{ab}}_{\text{dim.-less}} {}^{\text{SI}}E_a {}^{\text{SI}}H_b \\ &= \dots + \frac{1}{4\pi} \underbrace{{}^G\alpha^{ab}}_{\text{dim.-less}} {}^GE_a {}^GH_b. \end{aligned} \quad (61)$$

In SI,  $[{}^{\text{SI}}\alpha^{ab}] = \text{s/m}$ ,  $[{}^{\text{SI}}E_a] = \text{V/m}$ , and  $[{}^{\text{SI}}H_b] = \text{A/m}$ . Then,

$$[{}^{\text{SI}}g] = \frac{\text{VAs}}{\text{m}^3} = \frac{\text{J}}{\text{m}^3} = \frac{\text{kg}}{\text{m s}^2} = 10 \frac{\text{g}}{\text{cm s}^2} = 10[{}^Gg]. \quad (62)$$

On the other hand, in the Gaussian system, we have for the electric field

$$[{}^GE_a] = \frac{\text{statvolt}}{\text{cm}} = 3 \times 10^4 \frac{\text{V}}{\text{m}} = 3 \times 10^4 [{}^{\text{SI}}E_a] \quad (63)$$

and for the magnetic excitation

$$[{}^GH_a] = \text{Oe} = \frac{1000 \text{ A}}{4\pi \text{ m}} = \frac{1000}{4\pi} [{}^{\text{SI}}H_a]. \quad (64)$$

Thus we have for the magnetoelectric moduli

$$[{}^G\alpha^{ab}] = [{}^{\text{SI}}\alpha^{ab}] = 1 = c [{}^{\text{SI}}\alpha^{ab}] \approx 3 \times 10^8 \frac{\text{m}}{\text{s}} [{}^{\text{SI}}\alpha^{ab}]. \quad (65)$$

Accordingly, we have the rule that multiplying  ${}^{\text{SI}}\alpha^{ab}$ , given in s/m, by  $c = 3 \times 10^8 \text{ m/s}$  yields the dimensionless Gaussian value  ${}^G\alpha^{ab}$ . Incidentally, in some papers Heaviside-Lorentz (“rationalized Gaussian”) units are still in use, see, e.g., Borovik-Romanov and Grimmer [27], p. 139.

### B. Astrov, Rado and Folen, and Wiegelmann *et al.*

In our task to determine the pseudoscalar  $\tilde{\alpha}$ , we can take recourse to already published experimental data. Our main sources are Astrov [42] (Fig. 1) for the *electrically* induced magnetoelectric effect (called  $\text{ME}_E$  in future) and Rado and Folen [44] for the *magnetically* induced magnetoelectric effect ( $\text{ME}_H$ ), see also O’Dell [13] and Refs. [45–49]. In both investigations single crystals of  $\text{Cr}_2\text{O}_3$  were used. In the  $\text{ME}_E$  experiments [42,44], Eqs. (23)–(25) were verified ( $H$  switched off) and in the  $\text{ME}_H$  experiments [44] Eqs. (20)–(22) ( $E$  switched off). In particular, Rado and Folen made both type of experiments and found that the magnetoelectric moduli  $\alpha_{\perp}$  and  $\alpha_{\parallel}$  for  $\text{ME}_E$  experiments coincide with those of the  $\text{ME}_H$  experiments. This proves the vanishing of the skewon part of the constitutive tensor  $\chi^{\lambda\nu\sigma\kappa}$  for  $\text{Cr}_2\text{O}_3$ .

Accordingly, these experiments confirmed Dzyaloshinskii’s theory for  $\text{Cr}_2\text{O}_3$  below the spin-flop phase. Further experiments were then done mainly for the  $\text{ME}_H$  case since (i) it is easier to conduct an experiment with very high mag-

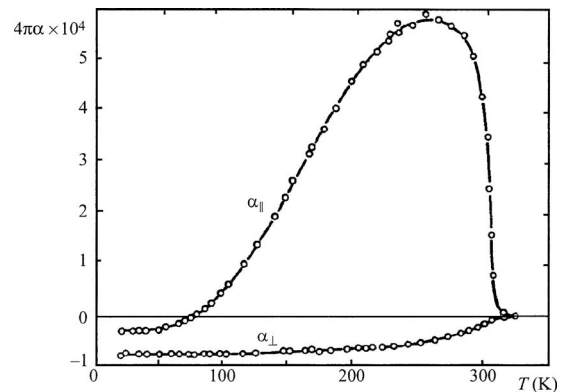


FIG. 1. The  $\text{ME}_E$  effect (linear magnetoelectric effect with electric field-induced magnetization) of  $\text{Cr}_2\text{O}_3$ : Temperature dependence of the magnetoelectric components  $\alpha_{\parallel}$  and  $\alpha_{\perp}$  according to Astrov [42]; see also the tables [43].

netic fields rather than with high electric fields. (ii) Even at low  $\mu_0 H$  fields, say below 1 T, the  $\text{ME}_H$  effect needs no calibration of the measuring system, contrary to the  $\text{ME}_E$  case with the quasistatic method, as  $D_a = -\delta g / \delta E_a = \alpha^{ab} H_b$  and  $D_a = Q / S_a$ ,  $\alpha^{ab} = Q / (S_a H_b)$ . The charges  $Q$ , usually in the pC range, are measured with a high input impedance electrometer, the magnetic field  $\mu_0 H_b$  with a Hall probe and the area  $S_a$  of one of the electrode by taking a picture of it. (iii) Quasistatic as well as AC measurements can be done easily. For  $\text{ME}_E$  quasistatic experiments, a SQUID must be used, see Kita [50,51].

With either methods,  $\text{ME}_H$  or  $\text{ME}_E$ , (i) best results are obtained with gold electrodes rather than with silver paste ones. (ii) The resistivity of the sample must be high enough, especially when the  $\text{ME}_H$  quasistatic method of measurements is used. (iii) One has to make sure that the antiferromagnetic magnetoelectric crystal forms a single domain by cooling it in appropriate simultaneous electric and magnetic fields through the Néel temperature; this is the so-called magnetoelectric annealing, see [54,55], and [13], p.124. Magnetoelectric crystals may also be (weakly) ferromagnetic or ferrimagnetic.

Our third main source of information are the measurements of Wiegelmann *et al.* [53], see also [52,56]. He took magnetic fields  $B$  as high as 20 T and measured from liquid helium up to room temperature. Wiegelmann *et al.* took a quasistatic magnetic field and thereby disproved explicitly claims by Lakhtakia [15] that measurements with magnetic fields of some kilo hertz cannot be extrapolated to static measurements. The  $\alpha_{zz}$  values of Wiegelmann *et al.* [53] were in very good agreement with independent  $\alpha_{zz}$  measurements presented below (Sec. IV C). However, the sign of  $\alpha_{\perp}(T)$  relative to  $\alpha_{\parallel}(T)$  was left open. Hence we took that from Astrov [42].

The values of  $\alpha_{\perp}(T)$  and  $\alpha_{\parallel}(T)$  of Fig. 2 are thus taken from Wiegelmann [52], Fig. 2.8, p.41 [see also Wiegelmann *et al.* [53] (Fig. 2, p.143)] after digitalization, interpolation, and correction for the relative sign. These values are given here in SI units in ps/m (picosecond/meter).

Independently, see Sec. IV, Rivera measured quasistatically at 133 Hz  $\alpha_{\parallel}$  of  $\text{Cr}_2\text{O}_3$  between 1.6 K and 305 K. He

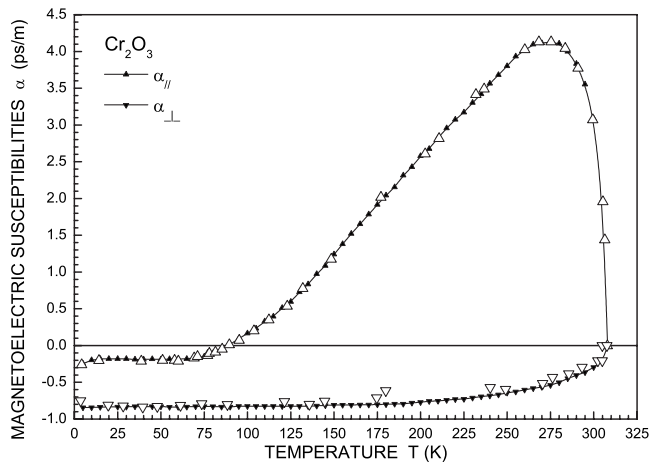


FIG. 2. The  $ME_H$  effect (linear magnetoelectric effect with magnetic field-induced polarization) of  $Cr_2O_3$ : Plot of  $\alpha_{\parallel}(T)$  and  $\alpha_{\perp}(T)$  after digitalization and interpolation (small full triangles) of Fig. 2.8, p. 41, of Wiegmann [52], (see also Fig. 2, p. 143, Wiegmann *et al.* [53]). The sign of  $\alpha_{\perp}(T)$  was set negative according to Astrov [42]. The curves ( $B$ -splines) are only guides for the eyes.

normalized that at  $T=275$  K, the temperature of the maximum value of  $\alpha_{\parallel}$ . He found the maximum value of  $\alpha_{\parallel}(\text{at } 275 \text{ K})=4.13 \text{ ps/m}$ . As we saw in Eq. (65),  $^{SI}\alpha^{ab}$ , given in s/m must be multiplied by the speed of light [57] in order to yield the dimensionless  $^{SI}\alpha^{ab}$ . Dropping again the star, we have

$$\begin{aligned} \alpha_{\parallel}(\text{at } 275 \text{ K}) &= 4.13 \times 10^{-12} \frac{\text{S}}{\text{m}} \times 2.99792 \times 10^8 \frac{\text{m}}{\text{s}} \\ &\approx 1.238 \times 10^{-3}. \end{aligned} \quad (66)$$

### C. Quasistatic magnetoelectric measurement technique and complementary results on the symmetry of $Cr_2O_3$

This section is dedicated to a detailed description of the quasistatic magnetoelectric measurement technique, applied to  $Cr_2O_3$ , which has proved to be the most accurate method for measuring magnetoelectric coefficients, but is not well known worldwide. It has not been detailed in Tolédano [33] and was described incompletely in Rado and Folen [35]. In addition, so far unpublished results are presented [37], supporting the questioned magnetic point group  $\bar{3}'m'$  of  $Cr_2O_3$  [62].

After grinding and polishing a crystal, polarized light microscopy was used, see, e.g., Wahlstrom [58] or Hartshorne [59], to check the orientation of a perpendicular cut to the optical axis. Actually, conoscopy allowed such tests (Fig. 3) in the near infrared, 260  $\mu\text{m}$  thick  $Cr_2O_3$  crystals being absorbing in visible light. The symmetry was found to be uniaxial to the accuracy of the conoscopy method, consistent with the symmetry  $\bar{3}'m'$  [26]. Recently the lower symmetry point group  $\bar{3}'$  has been suggested [60], however, it is being questioned by other authors [61,62]. The optical axis was computed to be inclined less than  $3^\circ$  away from the normal

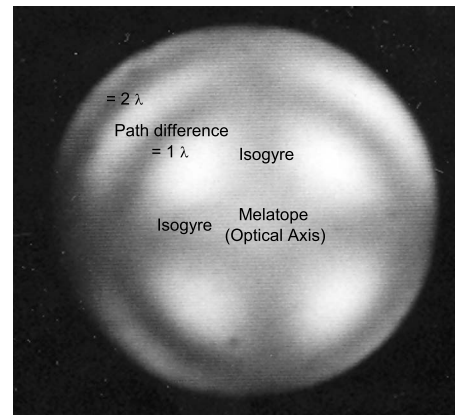


FIG. 3. Conoscopic pictures, in the near infrared, of a  $Cr_2O_3$  platelet at room temperature and between linear crossed polarizers, prepared for measuring the  $\alpha_{\parallel}(T)$  coefficient. We can see one or two isochromatic curve(s), the circle(s), the cut being slightly inclined, less than  $3^\circ$  relatively to the optical axis, the  $z$  axis. At the center of the cross, the melatope emerges as the optical axis with a possible rotation of the  $\mathbf{E}$  vector of the light along this axis, evidenced by a lighter center (Zeiss objective  $125 \times 1.30$  oil  $P$ , and condenser with head and oil, numerical aperture=1.30).

to the cut. Incidentally, this platelet was then sent to Wiegmann in Grenoble; see [53].

For the  $ME_H$  experiments of  $\alpha_{\parallel}(T)$  presented below, semi-transparent gold electrodes were evaporated on both sides of a platelet with area (one side)  $S_z=4.70 \text{ mm}^2$  (thickness  $th_z=260 \mu\text{m}$ ). On Fig. 4, this  $Cr_2O_3$  platelet is shown mounted with its thin gold wires ( $\varnothing=40 \mu\text{m}$ ) on Cu wires and then on two coaxial 50  $\Omega$  low noise cables 1 and 2, on a stainless steel sample holder. On the top left (forefront) of the picture, we see the temperature sensor, a calibrated (1.5 to 300 K) Carbon Glass Resistor (CGR-2000, “Lake Shore,”  $\varnothing$  about 3 mm). This sample holder was then inserted in a copper can with He exchange gas, in an Helium bath cryostat.

As already mentioned, before the measurements, the crystal was always cooled using the so-called magnetoelectric annealing with dc magnetic and electric fields, see Martin, Anderson, and Schmid [54,55,63] in this general context. It was easy to apply an electric field because the electrical resistivity of the crystal was very high. We measured the charges  $Q$  with a low noise electrometer (Keithley, 642-LNFPA) and  $H$  with a Hall probe. The  $H$  was produced by an old 12-inch Varian “V 4012-3B” electromagnet with Varian “Mark I” magnetic field regulator. We measured at more than 130 temperature values. Typically, the record of  $Q$  was as follows (quasistatic  $ME_H$  method): for about half a minute at zero field, then increasing linearly  $H$  with time, from 0 to 10 kOe in about 2.5 min, maintaining  $H$  for half a minute at maximum, decreasing  $H$  linearly with time to 0 and then maintaining  $H$  at 0 for half a minute. This procedure was used to cancel any shift of the base line, if any, see also Rivera [37,64]. The final curve looks like a volcano with a flat top and straight sides, proving the linear character of the  $ME_H$  effect for  $\alpha_{\parallel}$  of  $Cr_2O_3$ , see Fig. 5 for  $T=291.7$  K. Note the very good signal over signal+noise ratio. Remember that  $1 \text{ Oe}=1000/(4\pi) \text{ A/m} \approx 79.6 \text{ A/m}$  and



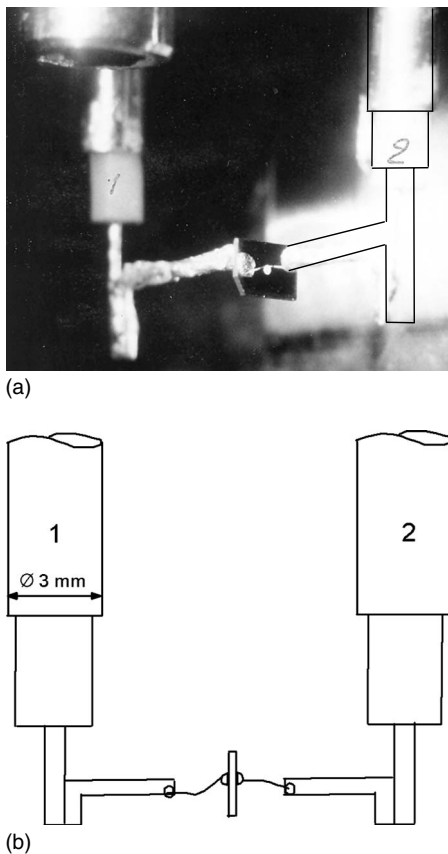


FIG. 4. (a) A  $\text{Cr}_2\text{O}_3$  platelet ( $S_z=4.70 \text{ mm}^2$ ,  $th_z=260 \mu\text{m}$ ) for  $\alpha_{\parallel}(T)$  measurements, connected to two low noise coaxial cables 1 and 2. One can see, by reflection on the platelet, the gold wire and the Cu wire 2 to the right. Black lines were added on the right of this unique Polaroid picture for clarity. (b) A schematic drawing of the setup depicted under (a).

$H=10 \text{ kOe}$  yields  $B=\mu_0 H=1 \text{ T}$ . As  $D_z=Q/S_z=\alpha_{\parallel} H_z$ , we compute directly  $\alpha_{\parallel}=Q/(S_z H_z)$ .

In Fig. 6, we show the curve of  $\alpha_{\parallel}(T)$ , in SI units, i.e., ps/m. The curve, around  $T=10 \text{ K}$ , has an elbow which could come from a possible crystallographic and/or magnetic phase transition. Further experiments should be done below  $10 \text{ K}$  to clarify this point. Closely below  $T_N$ , from  $293$  to  $304 \text{ K}$ , we plotted (not shown)  $\ln[\alpha_{\parallel}(T)/\alpha_{\parallel}^{\text{max}}]$  vs  $\ln[(T_N-T)/T_N]$ . It is a straight line, the slope gives the exponent  $\approx 0.34$  (thus about  $1/3$ ). Astrov [42], from the cut for measuring  $\alpha_{\perp}(T)$  by a  $\text{ME}_E$  experiment, found an exponent  $\approx 1/2$ , as expected according to the Landau theory.

In Table 1, Wiegelmann *et al.* [53] compared the values of the temperature  $T$  at  $\alpha_{\parallel}=0$  (where the sign changes) and  $T$  at  $\alpha_{\parallel}^{\text{max}}$  that were obtained by different authors. This is better than giving an error on the results of  $\alpha$  because with the  $\text{ME}_H$  effect, the larger error, apart from the one on the area of the crystal (about  $3-5 \%$ ), comes from the uncertainty about the success in the magnetoelectric annealing, which could also be influenced by the quality of the crystal.

In Fig. 7, we display  $Q(\theta)$  for  $H=5 \text{ kOe}$  and  $10 \text{ kOe}$ , also at  $T=292 \text{ K}$ , showing very good parts of cosine curves. The angle  $\theta$  is measured between the optical axis (the  $z$  axis) and the  $H$  field direction. This demonstrates that  $\alpha_{zx}$  and  $\alpha_{zy}$  are

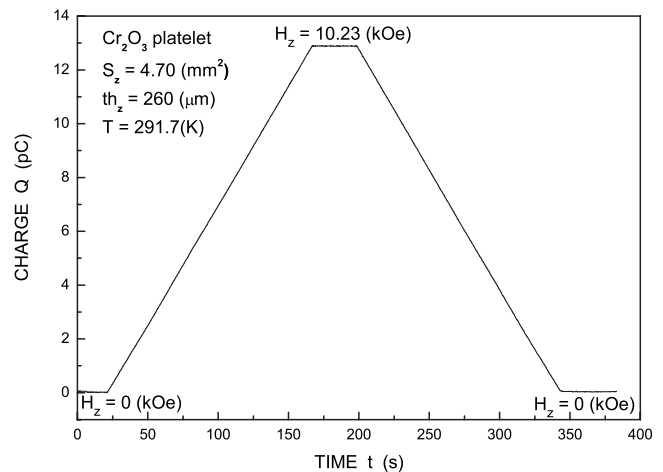


FIG. 5. Quasistatic  $\text{ME}_H$  experiment for  $\text{Cr}_2\text{O}_3$ , charge vs time. The field  $H$  varies linearly in time, from 0 to  $10 \text{ kOe}$  and from  $10 \text{ kOe}$  to 0, proving the linearity of the magnetoelectric effect in  $\text{Cr}_2\text{O}_3$ , here for  $\alpha_{\parallel}$  (at  $292 \text{ K}$ ).

very small or even null, at least, at that temperature. This again supports Dzyaloshinskii's theory; see Eq. (47). After transformation from rectangular to polar coordinates, we could obtain a continuous curve  $\cos^2 \theta$ , similar to the one of Fig. 3 given by Astrov [42,65] for the case  $\alpha_{\parallel}$  at  $T=103 \text{ K}$ .

Figure 8 concerns the  $\text{ME}_H$  effect at a low frequency of  $133 \text{ Hz}$ . It represents a dynamic measure of  $\alpha_{\parallel}(T)$ , with an ac field  $h_z \approx 18 \text{ Oe}_{\text{rms}}$  (root mean square) superimposed to a low dc field  $H_z \approx 580 \text{ Oe}$ , just to maintain the domains close to  $T_N$ . The rate of heating from  $4.2 \text{ K}$  to  $330 \text{ K}$  was  $+2 \text{ K/min}$ . As shown in Fig. 4, the crystal was mounted "floating" between the coaxial cables 1 and 2. As  $Q=CU$ , by measuring with a "lock-in" amplifier (SR 530) the voltage induced by the ac magnetic field  $h_z$  we have a measure of the  $Q(T)$  which is proportional to  $\alpha_{\parallel}(T)$ . To a first approximation, the

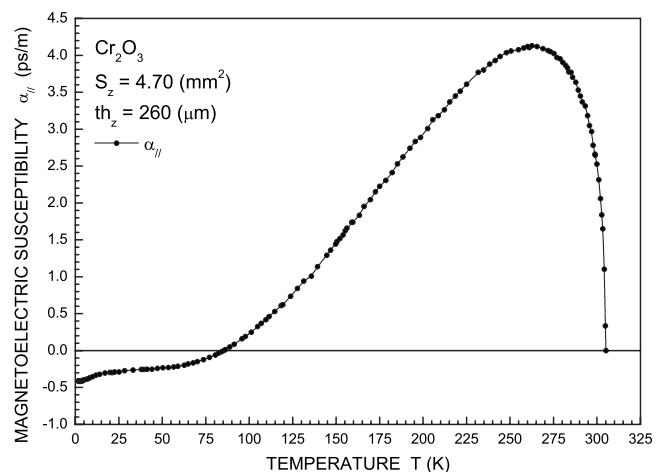


FIG. 6. Magnetoelectric susceptibility  $\alpha_{\parallel}(T)$  for  $\text{Cr}_2\text{O}_3$ , obtained by quasistatic  $\text{ME}_H$  experiments,  $H$  varying at each temperature from 0 to  $10 \text{ kOe}$  and back to 0, as described in the text. The interpolation between the points was made by means of  $B$ -splines and is only a guide for the eyes.

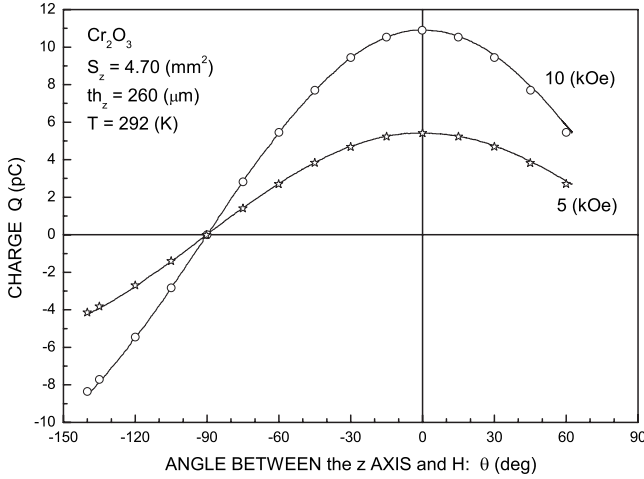


FIG. 7.  $ME_H$  experiments for  $Cr_2O_3$  at  $T=292$  K and constant  $H$  fields. By rotating the electromagnet, the measured charges (continuous lines) follow cosine curves. Superimposed are normalized computed points (open circles at  $H=10$  kOe; open stars at  $H=5$  kOe). This proves that  $\alpha_{zx}$  and  $\alpha_{zy}$  are very small or null [62]. At  $\theta=0^\circ$ , the field  $H$  is parallel to the optical axis ( $z$  axis).

capacitance  $C$  of the crystal was supposed to be independent of temperature. If we superimposed the normalized curves at  $T_{\max}$  of  $\alpha_{\parallel}(T)$ , measured quasistatically and dynamically, from 70 K to 270 K, the superposition is very good. The Néel temperature found dynamically (308 K) is 3 K above the one found quasistatically (305 K). Probably the heating rate (+2 K/min) was too high.

This concludes the detailed presentation of the highly accurate quasistatic  $ME_H$  measurement technique, demonstrated on  $Cr_2O_3$  and consistent with Tolédano [33] and Wiegmann *et al.* [53].

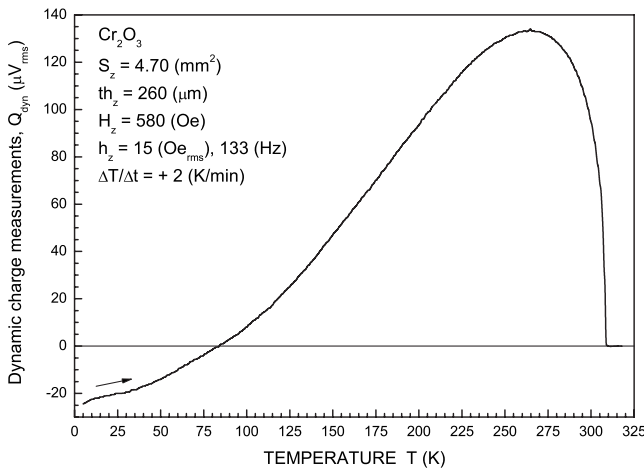


FIG. 8. Magnetoelectric susceptibility  $\alpha_{\parallel}(T)$  for  $Cr_2O_3$  obtained by low frequency (133 Hz) ac  $ME_H$  experiments ( $h_z \approx 18$  Oe<sub>rms</sub>). Knowing  $T \approx 85.0$  K at  $\alpha_{\parallel}(T)=0$  from quasistatic experiments and the base line above  $T_N$ , the curve was corrected for a small drift.

## V. EXTRACTING THE PSEUDOSCALAR $\tilde{\alpha}$

### A. Permeabilities $\mu_{\perp}$ and $\mu_{\parallel}$

The magnetic susceptibility tensor of  $Cr_2O_3$ , namely  $\chi := 1 - (1/\mu)$  or, in components,  $\chi_{ab} = \delta_{ab} - \mu_{ab}^{-1}$ , was determined by Foner [66] as a function of the temperature [67]. From the caption of his Fig. 8, we can take the static susceptibility perpendicular to the  $c$  axis  $\chi_{\perp}^s$  of  $Cr_2O_3$ . The superscript  $g$  stands for specific (or mass) magnetic susceptibility. Foner used the old unit emu/g. From the inside of the front cover of Landolt-Börnstein [68] we learn that 1 emu/g  $\equiv 1$  cm<sup>3</sup>/g. Accordingly,

$$\chi_{\perp}^s(\text{at } 4.2 \text{ K}) = 2.24 \times 10^{-5} \frac{\text{cm}^3}{\text{g}}. \quad (67)$$

Let us now determine the *volume* susceptibility  $\chi^v$ . Again from Landolt-Börnstein we take  $^{\text{SI}}\chi^v = 4\pi^G \chi^s$ . The density for the mineral eskolaite [69], containing 94%  $Cr_2O_3$ , is 5.23 g/cm<sup>3</sup> and for  $Cr_2O_3$  ceramics [70] a bit less, namely 5.21 g/cm<sup>3</sup>. Then, in SI,

$$\chi_{\perp}^v(\text{at } 4.2 \text{ K}) = 4\pi \times 2.24 \times 10^{-5} \times 5.22 \approx 1.47 \times 10^{-3}. \quad (68)$$

We can read off from Fig. 8 of Foner [66] that both, the parallel and the perpendicular susceptibilities at the Néel temperature  $T_N$  are about 13% higher than  $\chi_{\perp}^v$  (at 4.2 K). Consequently, we find

$$\begin{aligned} \chi_{\perp}^v(\text{at } T_N) &\approx \chi_{\parallel}^v(\text{at } T_N) \approx 1.13 \times \chi_{\perp}^v(\text{at } 4.2 \text{ K}) \\ &\approx 1.62 \times 10^{-3}. \end{aligned} \quad (69)$$

O'Dell [13], App.1, found the slightly higher value of  $\approx 1.64 \times 10^{-3}$ .

Now we can determine the permeabilities: Below and close to the Néel temperature, we have

$$\mu_{\max} = \frac{1}{1 - \chi^v(\text{at } T_N)} \approx 1 + \chi^v(\text{at } T_N) \approx 1.00162. \quad (70)$$

At 4.2 K, we find

$$\mu_{\perp}(\text{at } 4.2 \text{ K}) \approx 1 + \chi_{\perp}^v(\text{at } 4.2 \text{ K}) \approx 1.00147. \quad (71)$$

Since  $\chi_{\parallel}$  is even smaller, we have  $\mu \approx 1$ . Because  $Cr_2O_3$  is antiferromagnetic, this was to be expected [71].

### B. Pseudoscalar (or axion piece) $\tilde{\alpha}$

Now we can come back to Eq. (45). Since  $\mu_{\perp} \approx \mu_{\parallel} \approx 1$ , the pseudoscalar (or axion piece) of the constitutive tensor  $\chi^{\lambda\nu\sigma\kappa}$  becomes  $\tilde{\alpha}\tilde{\epsilon}^{\lambda\nu\sigma\kappa}$ , with

$$\tilde{\alpha} \approx \frac{1}{3}(2\alpha_{\perp} + \alpha_{\parallel}) \sqrt{\frac{\epsilon_0}{\mu_0}}. \quad (72)$$

It is the arithmetic mean of the trace of the magnetoelectric tensor  ${}^{\text{rel}}\alpha_b^a$  of Eq. (47). Going back to Fig. 2, we can take the values of  $\alpha_{\perp}$  and  $\alpha_{\parallel}$  and compute  $\tilde{\alpha}$ . The result is plotted in Fig. 9. As we can see, for temperatures of up to about 163 K, the pseudoscalar is negative, for higher temperatures

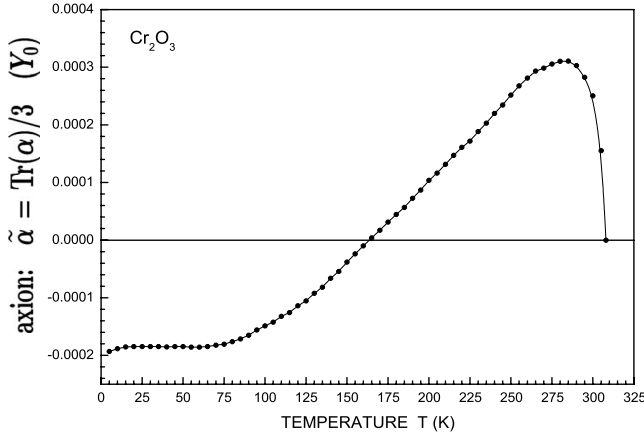


FIG. 9. The pseudoscalar or axion piece  $\tilde{\alpha}$ , see Eq. (4), of the constitutive tensor  $\chi^{\lambda\nu\sigma\kappa}$  of  $\text{Cr}_2\text{O}_3$  in units of  $Y_0=1/Z_0$  as a function of the temperature  $T$  in K; here  $Z_0$  is the vacuum impedance which, in SI, is  $\approx 377$  ohm.

positive until it vanishes at the Néel temperature of about 308 K. For example, at 285 K, we find  $\tilde{\alpha}_{\text{max}} \approx 1.035$  ps/m. Multiplied by  $c$  we get

$$\begin{aligned} \tilde{\alpha}_{\text{max}}(\text{at } 285 \text{ K}) &\approx 3.10 \times 10^{-4} \sqrt{\frac{\epsilon_0}{\mu_0}} \approx \frac{1}{3226Z_0} \\ &\stackrel{\text{SI}}{\approx} 8.22 \times 10^{-7} \frac{1}{\Omega} \approx \frac{1}{1.216M\Omega}. \end{aligned} \quad (73)$$

## VI. PSEUDOSCALAR OR AXION PIECE OF THE MAGNETOELECTRIC SUSCEPTIBILITY OF $\text{Cr}_2\text{O}_3$ VIOLATES $P$ AND $T$ ; IT CARRIES NO ELECTROMAGNETIC ENERGY DENSITY

As has been pointed out by Janner [72], amongst others, the violation of the invariances under space reflection (parity  $P$ ) and time inversion ( $T$ ) of the corresponding crystal, in our case  $\text{Cr}_2\text{O}_3$ , are necessary conditions for the emergence of the magnetoelectric effect. The same is true for the emergence of the pseudoscalar or axion piece  $\tilde{\alpha}$ . The constitutive relation for the axion piece alone can be read off from Eqs. (32) and (33) as

$$D^a = +\tilde{\alpha}B^a, \quad (74)$$

$$H_a = -\tilde{\alpha}E_a, \quad (75)$$

see [11], Eqs. (D.1.112) and (D.1.111). If we denote, as in crystallography, a space reflection by  $\bar{1}$  and a time inversion by  $1'$ , see Janner [72], then we have

$$\bar{1}D^a = -D^a, \quad \bar{1}H_a = H_a, \quad \bar{1}E_a = -E_a, \quad \bar{1}B^a = B^a, \quad (76)$$

$$1'D^a = D^a, \quad 1'H_a = -H_a, \quad 1'E_a = E_a, \quad 1'B^a = -B^a, \quad (77)$$

see also Marmo *et al.* [73]. If we now apply a space reflection to Eqs. (74) and (75), then they transform into their negatives,

$$D^a = -\tilde{\alpha}B^a, \quad (78)$$

$$H_a = +\tilde{\alpha}E_a. \quad (79)$$

The same is true for a time inversion. In other words, the constitutive relations for the axion piece *violate  $P$  and  $T$  invariance*; see also Dzyaloshinskii [74] and Tolédano [33]. They are only invariant under the combined  $PT$  transformation. Consequently, the violation of  $P$  and  $T$  invariance is an essential characteristics of the axion piece  $\tilde{\alpha}$ .

The first, to our knowledge, who tried to utilize the constitutive laws (74) and (75) for vacuum electrodynamics was Schrödinger [75], p.25. He made the ansatz, compare Eq. (17),

$$\mathfrak{G}^{\lambda\nu} = \frac{1}{2}\tilde{\epsilon}^{\lambda\nu\sigma\kappa}F_{\sigma\kappa} \text{ or } \tilde{G}_{\mu\nu} = F_{\mu\nu}. \quad (80)$$

A look at Eqs. (8) and (6) shows that this, for  $\tilde{\alpha}=1$ , yields the laws (74) and (75). But Schrödinger rejected it as being unphysical if taken for vacuum electrodynamics.

Another property is characteristic for the axion piece: It does not contribute to the electromagnetic energy density. This can be seen easily since the energy density in electrodynamics is  $\frac{1}{2}(D^aE_a + H_aB^a)$ . If Eqs. (74) and (75) are substituted, this expression vanishes. But even more so, also the energy flux density vanishes. In order to prove this, we turn to the energy-momentum tensor  $\mathfrak{T}_\lambda{}^\nu$  of the electromagnetic field that is built up from the energy density  $\mathfrak{T}_0{}^0$ , the energy flux density  $\mathfrak{T}_0{}^b$  (with  $b=1,2,3$ ), the momentum density  $\mathfrak{T}_a{}^0$  (with  $a=1,2,3$ ), and the momentum flux density  $\mathfrak{T}_a{}^b$  according to

$$\mathfrak{T}_\lambda{}^\nu = \begin{pmatrix} \text{energy } d & \text{energy flux } d \\ \text{momentum } d & \text{momentum flux } d \end{pmatrix} = \begin{pmatrix} \mathfrak{T}_0{}^0 & \mathfrak{T}_0{}^b \\ \mathfrak{T}_a{}^0 & \mathfrak{T}_a{}^b \end{pmatrix}. \quad (81)$$

$\mathfrak{T}_0{}^b$  is also called the Poynting flux and  $\mathfrak{T}_a{}^b$  the Maxwell stress. The energy-momentum tensor reads [cf. Post [1], Eq. (9.55)]

$$\mathfrak{T}_\lambda{}^\nu = \mathfrak{L}\delta_\lambda{}^\nu - \mathfrak{G}^{\nu\sigma}F_{\lambda\sigma}, \quad (82)$$

with

$$\mathfrak{L} := \frac{1}{4}\mathfrak{G}^{\sigma\tau}F_{\sigma\tau} = \frac{1}{2}(H_aB^a - D^aE_a); \quad (83)$$

the last equation can be read off directly from Eqs. (5) and (6). With some algebra, Eq. (82) can be rewritten as

$$\mathfrak{T}_\lambda{}^\nu = \frac{1}{4}\tilde{\epsilon}^{\nu\mu\rho\sigma}(\tilde{G}_{\lambda\mu}F_{\rho\sigma} - F_{\lambda\mu}\tilde{G}_{\rho\sigma}), \quad (84)$$

see [11], Eq. (B.5.40). Since the laws (74) and (75) can be put together in the manifestly covariant form

$$\tilde{G}_{\mu\nu} = \tilde{\alpha}F_{\mu\nu}, \quad (85)$$

we see immediately from Eq. (84) that

$$\mathfrak{T}_\lambda{}^\nu(\text{of axion piece } \tilde{\alpha}) = 0. \quad (86)$$

Thus, in particular, the electromagnetic energy density  $\mathfrak{T}_0{}^0$  of the axion piece vanishes.

## VII. OTHER SUBSTANCES AND SYMMETRIES PERMITTING MAGNETOELECTRICITY WITH THE AXION PIECE

In the present article the relativistic analysis is based on data of the antiferromagnet  $\text{Cr}_2\text{O}_3$ , because it represents so far probably the best studied magnetoelectric material and has diagonal components of the linear magnetoelectric effect tensor  $\alpha$ ; see Eq. (47). However, other materials and symmetries could have served the same purpose, in principle. Among the 122 Heesch-Shubnikov point groups 58 ones are permitting the linear magnetoelectric effect [76], and therefrom 32 ones possess diagonal components of the magnetoelectric tensor  $\alpha$  [27,37,77]. Strictly speaking, our magnetoelectric tensor  $\text{rel}\alpha$  in Eq. (47) belongs to the *EB* scheme, see Eqs. (32) and (33), whereas the corresponding tensor in the literature [78] is the one of the *EH* scheme. However, as we can see in Eq. (47), because of  $\mu \approx 1$  the differences are marginal and do not touch our arguments.

One can distinguish three types of diagonals (for the complete set of magnetoelectric tensors see, e.g., Refs. [37,77,27], Table 1.5.8.1, for the examples cited, see Ref. [27], Table 1.5.8.2, except for [60,78]):

- (i) 19 point groups with  $\alpha_{11} \neq \alpha_{22} \neq \alpha_{33}$ .

Examples:

Point group  $m'm'm'$ :  $\text{DyAlO}_3$ ,  $\text{GdAlO}_3$ ,  $\text{TbAlO}_3$ ,

Point group  $m'$ :  $\text{Ni}_3\text{B}_7\text{O}_{13}\text{I}$ ,

Point group  $m'm'2$ :  $\text{Cu}_3\text{B}_7\text{O}_{13}\text{Cl}$ .

- (ii) 8 point groups with  $\alpha_{11} = \alpha_{22} \neq \alpha_{33}$  ( $\alpha_{11} = \alpha_{22} = \alpha_\perp$ ,  $\alpha_{33} = \alpha_\parallel$ ).

Examples:

Point group  $\bar{3}'m'$ :  $[\text{Cr}_2\text{O}_3]$  [62],  $\text{Nb}_2\text{Mn}_4\text{O}_9$ ,  $\text{Nb}_2\text{Co}_4\text{O}_9$ ,  $\text{Ta}_2\text{Mn}_4\text{O}_9$ ,  $\text{Ta}_2\text{Co}_4\text{O}_9$ ,

Point group  $\bar{3}'$ :  $\text{Cr}_2\text{O}_3$  [60,62],

Point group  $4/m'm'm'$ :  $\text{Fe}_2\text{TeO}_6$ .

- (iii) 5 point groups with  $\alpha_{11} = \alpha_{22} = \alpha_{33}$ .

Examples:

Point group  $\bar{4}'3m'$  (expected):  $\text{Cr}_3\text{B}_7\text{O}_{13}\text{Br}$ ,  $\text{Cr}_3\text{B}_7\text{O}_{13}\text{I}$  [78],  $\text{Cu}_3\text{B}_7\text{O}_{13}\text{I}$  [79].

Thus it is clear that the pseudoscalar  $\tilde{\alpha}$  occurs in quite a number of different substances. Its existence can no longer be denied.

## VIII. DISCUSSION

The structure of the constitutive law (85) is not unprecedented, as we already discussed in [22]. In electrical engineering, in the theory linear networks, more specifically in the theory of two ports (or four poles), Tellegen [80,81] came up with the new structure of a *gyrator*, which is defined via

$$v_1 = -s i_2,$$

$$v_2 = s i_1, \quad (87)$$

where  $v$  are voltages and  $i$  currents of the ports 1 and 2, respectively. Let us quote from Tellegen [81], p. 189: “The ideal gyrator has the property of ‘gyrating’ a current into a voltage, and vice versa. The coefficient  $s$ , which has the dimension of a resistance, we call the gyration resistance;  $1/s$  we call the gyration conductance.” The gyrator is a nonreciprocal network element.

If we turn to the electromagnetic field, then because of dimensional reasons the quantities related to the *currents*  $i_1$ ,  $i_2$  are the excitations  $D^a$ ,  $H_a$  and the quantities related to the *voltages*  $v_1$ ,  $v_2$  the field strengths  $E_a$ ,  $B^a$ . Then we find without problems straightforwardly the relations

$$E_a = -s H_a,$$

$$B^a = s D^a. \quad (88)$$

If we rename the admittance  $s$  according to  $s = 1/\tilde{\alpha}$ , then Eqs. (88), (74), and (75) coincide. Without the least doubt, the gyrator is in the theory of two ports what the axion piece is in magnetoelectricity. The axion piece “rotates” the excitations, modulo an admittance, into the field strengths, as the gyrator the currents into voltages.

These analogies or rather isomorphisms carry even further. In 2005, Lindell and Sihvola [82,83], see also [84], introduced the new concept of a *perfect electromagnetic conductor* (PEMC). It also obeys the constitutive law  $\tilde{G}_{\mu\nu} = \tilde{\alpha} F_{\mu\nu}$  or Eq. (88). The PEMC is a generalization of the perfect electric and the perfect magnetic conductor. In this sense, it is the “ideal” electromagnetic conductor that can be hopefully built by means of a suitable *metamaterial*, see Sihvola [85]. The pseudoscalar  $\tilde{\alpha}$  is called Tellegen parameter by Lindell *et al.*; see [86], p.13 (for a more general view, see [87]); artificial Tellegen material has been produced and positively tested by Tretyakov *et al.* [88], amongst others.

Continuing with our search for isomorphisms, we turn to axion electrodynamics, see Ni [89], Wilczek [90], and, for more recent work, Itin [91,92]. If for vacuum electrodynamics we add to the usual Maxwell-Lorentz expression specified in Eq. (14) an axion piece patterned after the last term in Eq. (17), then we have the constitutive law for axion electrodynamics [93],

$$\mathfrak{G}^{\lambda\nu} = \frac{1}{Z_0} \sqrt{-g} F^{\lambda\nu} + \frac{1}{2} \tilde{\alpha} \tilde{\epsilon}^{\lambda\nu\sigma\kappa} F_{\sigma\kappa}. \quad (89)$$

Alternatively, with the excitation pseudotensor (7) we find

$$\tilde{G}_{\mu\nu} = \frac{1}{2Z_0} \tilde{\epsilon}_{\mu\nu\kappa\lambda} \sqrt{-g} F^{\kappa\lambda} + \tilde{\alpha} F_{\mu\nu} \quad (90)$$

and, in exterior calculus,

$$\tilde{G} = \left( \frac{1}{Z_0} * + \tilde{\alpha} \right) F. \quad (91)$$

We discussed this “spacetime relation” and also the corresponding Lagrangian in some detail in [11,22]. The Hodge



star operator  $*$  is odd; it transforms a form into a twisted form, and vice versa. In  $\text{Cr}_2\text{O}_3$  we had  $\tilde{\alpha} \approx 10^{-4}/Z_0$ . It is everybody's guess what it could be for the physical vacuum. In elementary particle theory one adds in the corresponding Lagrangian also kinetic terms of the axion  $\tilde{a}$   $\sim g^{\mu\nu} \partial_\mu \tilde{a} \partial_\nu \tilde{a}$  and possibly a massive term  $\sim m_{\tilde{a}}^2 \tilde{a}^2$ . However, this hypothetical  $P$  odd and  $T$  odd particle has not been found so far, in spite of considerable experimental efforts; see Davis *et al.* [94] and references given.

The axion shares its  $P$  odd and  $T$  odd properties with the  $\tilde{a}$  piece of  $\text{Cr}_2\text{O}_3$ , with the gyrotator, and with the PEMC. One may speculate whether an axion detector made of  $\text{Cr}_2\text{O}_3$  crystals could enhance the probability of finding axions.

We pointed out the presence of the axion parameter or the axion field in four different domains of physics: (i) In condensed matter physics in the antiferromagnet  $\text{Cr}_2\text{O}_3$ , (ii) in the gyrotator of linear networks, (iii) in the PEMC of electrical engineering, and (iv) in the axion field in axion electrodynamics. Wilczek (private communication) remarked to these isomorphisms between these four systems: "It's a nice demonstration of the unity of physics." Already in his paper of 1987 [90] he argued that "...it is...not beyond the realm of possibility that fields whose properties partially mimic those of axion fields can be realized in condensed-matter systems." This was prophetic. In our paper we proved this.

Nevertheless, these four physical systems are different. The isomorphism is similar as that between an LC oscillator

(oscillating circuit) and a mechanical pendulum. Two catchwords characterizing jointly those systems are "Lagrangian system" and "linear restoring force." In (i), e.g., it is the structural arrangement of the Cr and the O atoms, together with the electronic filling per ion [95], that in the end cause the  $P$ -odd and  $T$ -odd behavior of the electronic distribution in  $\text{Cr}_2\text{O}_3$ . In (iv), in the vacuum of electrodynamics, the axion field represents some new type of a pseudoscalar vacuum permittivity or permeability that is made propagating by adding kinetic axion terms to the Lagrangian. Since the vacuum carries no atoms, we cannot visualize how the axion field may arise. In spite of this, in the Lagrangian of axion electrodynamics we have the same term  $\tilde{a}F \wedge F$  as in the Lagrangian of chromium sesquioxide.

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