Resonance damping in ferromagnets and ferroelectrics

A. Widom

Physics Department, Northeastern University, Boston, Massachusetts 02115, USA

S. Sivasubramanian

NSF Nanoscale Science & Engineering Center for High-Rate Nanomanufacturing, Northeastern University, Boston, Massachusetts 02115, USA

C. Vittoria and S. Yoon

Department of Electrical and Computer Engineering, Northeastern University, Boston, Massachusetts 02115, USA

Y. N. Srivastava

Physics Department and INFN, University of Perugia, 06123 Perugia, Italy (Received 22 January 2010; revised manuscript received 11 May 2010; published 7 June 2010)

The phenomenological equations of motion for the relaxation of ordered phases of magnetized and polarized crystal phases can be developed in close analogy with one another. For the case of magnetized systems, the driving magnetic field intensity toward relaxation was developed by Gilbert. For the case of polarized systems, the driving electric field intensity toward relaxation was developed by Khalatnikov. The transport times for relaxation into thermal equilibrium can be attributed to viscous sound wave damping via magnetostriction for the magnetic case and electrostriction for the polarization case.

DOI: 10.1103/PhysRevB.81.212402

I. INTRODUCTION

It has long been of interest to understand the close analogies between ordered electric polarized systems, e.g., *ferroelectricity*, and ordered magnetic systems, e.g., *ferromagnetism*. At the microscopic level, the source of such ordering must depend on the nature of the electronic energy spectra. The relaxation mechanism into thermal equilibrium state must be described by local electric field fluctuations for the electric polarization case and by magnetic intensity fluctuations for the magnetization case; Specifically, the field fluctuations for each case

$$\mathcal{G}_{ij}^{pol}(\mathbf{r},\mathbf{r}',t) = \frac{1}{\hbar} \int_0^\beta \langle \Delta E_j(\mathbf{r}',-i\lambda) \Delta E_i(\mathbf{r},t) \rangle d\lambda,$$

$$\mathcal{G}_{ij}^{mag}(\mathbf{r},\mathbf{r}',t) = \frac{1}{\hbar} \int_0^\beta \langle \Delta H_j(\mathbf{r}',-i\lambda) \Delta H_i(\mathbf{r},t) \rangle d\lambda,$$

ein

wherein

$$\beta = \frac{\hbar}{k_B T} \tag{1}$$

determine the relaxation time tensor for both cases via the fluctuation-dissipation formula $^{\rm 1-4}$

$$\tau_{ij} = \int_0^\infty \lim_{V \to \infty} \left[\frac{1}{V} \int_V \int_V \mathcal{G}_{ij}(\mathbf{r}, \mathbf{r}', t) d^3 \mathbf{r} d^3 \mathbf{r}' \right] dt.$$
(2)

We have *unified the theories of relaxation* in ordered polarized systems and ordered magnetized systems via the Kubo transport time tensor in Eqs. (1) and (2).

The transport describing the relaxation of ordered magnetization is the Landau-Lifshitz-Gilbert equation.^{5–7} This equation has been of considerable recent interest^{8–10} in describing ordered magnetic resonance phenomena.^{11–14} The equation describing the electric relaxation of an ordered po-

PACS number(s): 76.50.+g, 75.30.Sg

larization is the Landau-Khalatinikov-Tani equation.^{15–17} This equation can be simply modeled^{18–21} with effective electrical circuits.^{22–25} Information memory applications^{26–29} of such polarized system are of considerable recent interest.^{30–32}

The unification of the magnetic Gilbert-Landau-Lifshitz equations and the electric Landau-Khalatnikov-Tani equations via the relaxation time tensor depends on the notion of a *nonequilibrium driving field*. For the magnetic case, the driving magnetic intensity \mathbf{H}_d determines the relaxation of the magnetization via the torque equation

$$\mathbf{M} = \gamma \mathbf{M} \times \mathbf{H}_d,\tag{3}$$

wherein γ is the gyromagnetic ratio. For the electric case, the driving electric field \mathbf{E}_d determines the relaxation of the polarization via the equation of motion for an ion of charge *ze*

$$m\ddot{\mathbf{r}} = ze\mathbf{E}_d.$$
 (4)

The unification of both forms of relaxation lies in the close analogy between the magnetic driving intensity \mathbf{H}_d and the electric driving field \mathbf{E}_d .

In Sec. II the thermodynamics of ordered magnetized and polarized systems is reviewed. The notions of magnetostriction and electrostriction are given a precise thermodynamic definition. In Sec. III, the phenomenology of the relaxation equations is presented. The magnetic driving intensity \mathbf{H}_d and the electric driving field \mathbf{E}_d are defined in terms of the relaxation time tensor Eq. (2). In Sec. IV, we introduce the crystal viscosity tensor. From a Kubo formula viewpoint, the stress fluctuation correlation

$$\mathcal{F}_{ijkl}(\mathbf{r},\mathbf{r}',t) = \frac{1}{\hbar} \int_0^\beta \langle \Delta \sigma_{kl}(\mathbf{r}',-i\lambda) \Delta \sigma_{ij}(\mathbf{r},t) \rangle d\lambda \qquad (5)$$

determines the crystal viscosity

$$\eta_{ijkl} = \int_0^\infty \lim_{V \to \infty} \left[\frac{1}{V} \int_V \int_V \mathcal{F}_{ijkl}(\mathbf{r}, \mathbf{r}', t) d^3 \mathbf{r} d^3 \mathbf{r}' \right] dt.$$
(6)

For models of magnetic relaxation, acoustic heating can dominate the relaxation time tensor via magnetostriction.³³ A central new result of the work which follows concerns models of electric relaxation wherein acoustic heating dominates, via electrostriction, the relaxation time tensor in Eq. (2). The derivation employs the viscosity tensor in Eq. (6). For completeness of presentation an independent and novel microscopic derivation of viscosity—electrostriction or magnetostriction—induced relaxation is given in Appendix. It is presumed that the transport processes are quasistationary. In the concluding Sec. V, the sound wave absorption physics of the viscous damping mechanism will be noted.

II. THERMODYNAMICS

Our purpose is to review the thermodynamic properties of both magnetically ordered crystals and polarization-ordered crystals. The former is characterized by a remnant magnetization **M** for vanishing applied magnetic intensity $\mathbf{H} \rightarrow 0$ while the latter is characterized by a remnant polarization **P** for vanishing applied electric field $\mathbf{E} \rightarrow 0$.

A. Magnetically ordered crystals

Let *w* be the enthalpy per unit volume. The fundamental thermodynamic law determining the equations of state for magnetically ordered crystals is given by

$$dw = Tds + \mathbf{H} \cdot d\mathbf{M} - \boldsymbol{e}: d\boldsymbol{\sigma},\tag{7}$$

wherein s is the entropy per unit volume, T is the temperature, e is the crystal strain and σ is the crystal stress. The magnetic adiabatic susceptibility is defined by

$$\boldsymbol{\chi} = \left(\frac{\partial \mathbf{M}}{\partial \mathbf{H}}\right)_{s,\boldsymbol{\sigma}}.$$
 (8)

If

$$\mathbf{N} = \frac{\mathbf{M}}{M} \Longrightarrow \mathbf{N} \cdot \mathbf{N} = 1 \tag{9}$$

denotes a unit vector in the direction of the magnetization then the tensor Λ_{ijkl} describing adiabatic magnetostriction coefficients may be defined as³⁴

$$2\Lambda_{ijkl}N_l = M\left(\frac{\partial e_{ij}}{\partial M_k}\right)_{s,\sigma} = -M\left(\frac{\partial H_k}{\partial \sigma_{ij}}\right)_{s,\mathbf{M}}.$$
 (10)

When the system is out of thermal equilibrium, the driving magnetic intensity is

$$\mathbf{H}_{d} = \mathbf{H} - \left(\frac{\partial w}{\partial \mathbf{M}}\right)_{s,\boldsymbol{\sigma}} - \boldsymbol{\tau} \cdot \left(\frac{\partial \mathbf{M}}{\partial t}\right), \tag{11}$$

wherein τ are the relaxation time tensor transport coefficients which determine the relaxation of the ordered magnetic system into a state of thermal equilibrium.

B. Ordered polarized crystals

The fundamental thermodynamic law determining the equations of state for ordered polarized crystals is given by

$$dw = Tds + \mathbf{E} \cdot d\mathbf{P} - \boldsymbol{e}: d\boldsymbol{\sigma}, \tag{12}$$

wherein w is the enthalpy per unit volume, s is the entropy per unit volume, T is the temperature, e is the crystal strain, and σ is the crystal stress. The electric adiabatic susceptibility is defined by

$$\boldsymbol{\chi} = \left(\frac{\partial \mathbf{P}}{\partial \mathbf{E}}\right)_{s,\boldsymbol{\sigma}}.$$
 (13)

The tensor β_{ijk} describing adiabatic electrostriction coefficients may be defined as³⁴

$$\boldsymbol{\beta}_{ijk} = \left(\frac{\partial \boldsymbol{e}_{ij}}{\partial \boldsymbol{P}_k}\right)_{s,\boldsymbol{\sigma}} = -\left(\frac{\partial \boldsymbol{E}_k}{\partial \sigma_{ij}}\right)_{s,\mathbf{P}}.$$
 (14)

The piezoelectric tensor is closely related to the electrostriction tensor via

$$\gamma_{ijk} = \left(\frac{\partial e_{ij}}{\partial E_k}\right)_{s,\sigma} = \left(\frac{\partial P_k}{\partial \sigma_{ij}}\right)_{s,\mathbf{E}} = \beta_{ijm}\chi_{mk}.$$
 (15)

When the system is out of thermal equilibrium, the driving electric field is

$$\mathbf{E}_{d} = \mathbf{E} - \left(\frac{\partial w}{\partial \mathbf{P}}\right)_{s,\boldsymbol{\sigma}} - \boldsymbol{\tau} \cdot \left(\frac{\partial \mathbf{P}}{\partial t}\right), \tag{16}$$

wherein τ is the relaxation time tensor transport coefficients which determine the relaxation of the ordered polarized system into a state of thermal equilibrium.

III. RESONANCE DYNAMICS

Here we shall show how the magnetic intensity \mathbf{H}_d drives the magnetic resonance equations of motion in magnetically ordered systems. Similarly, we shall show how the electric field \mathbf{E}_d drives the polarization resonance equations of motion for polarized ordered systems.

A. Gilbert-Landau-Lifshitz equations

The driving magnetic intensity determines the torque on the magnetic moments according to

$$\frac{\partial \mathbf{M}}{\partial t} = \gamma \mathbf{M} \times \mathbf{H}_d. \tag{17}$$

Employing Eqs. (11) and (17), one finds the equations for magnetic resonance in the Gilbert form

$$\frac{\partial \mathbf{M}}{\partial t} = \gamma \mathbf{M} \times \left[\mathbf{H} - \left(\frac{\partial w}{\partial \mathbf{M}} \right)_{s, \boldsymbol{\sigma}} - \left(\frac{\boldsymbol{\alpha}}{\gamma M} \right) \cdot \frac{\partial \mathbf{M}}{\partial t} \right], \quad (18)$$

wherein the Gilbert dimensionless damping tensor α is defined as

$$\boldsymbol{\alpha} = (\gamma M)\boldsymbol{\tau}.\tag{19}$$

One may directly solve the Gilbert equations for the driving magnetic intensity according to

$$\mathbf{H}_{d} + \boldsymbol{\alpha} \cdot (\mathbf{N} \times \mathbf{H}_{d}) = \mathbf{H} - \left(\frac{\partial w}{\partial \mathbf{M}}\right)_{s,\boldsymbol{\sigma}}.$$
 (20)

Equations (17) and (20) express the magnetic resonance motion in the Landau-Lifshitz form.

B. Landau-Khalatnikov-Tani equations

The driving electric field gives rise to a polarization response according to

$$\frac{\partial^2 \mathbf{P}}{\partial t^2} = \left(\frac{\omega_p^2}{4\pi}\right) \mathbf{E}_d,\tag{21}$$

wherein ω_p is the plasma frequency. A simple derivation of Eq. (21) may be formulated as follows. In a large volume *V*, the polarization due to charges $\{z_i e\}$ is given by

$$\mathbf{P} = \left(\frac{\sum_{j} z_{j} e \mathbf{r}_{j}}{V}\right). \tag{22}$$

If the driving electric field accelerates the charges according to

$$m_j \ddot{\mathbf{r}}_j = z_j e \mathbf{E}_d \tag{23}$$

then Eq. (21) holds true with the plasma frequency

$$\omega_p^2 = 4\pi e^2 \lim_{V \to \infty} \left[\frac{\sum_j (z_j^2/m_j)}{V} \right] = 4\pi e^2 \sum_a \frac{n_a z_a^2}{m_a}, \quad (24)$$

wherein n_a is the density of charged particles of type *a*.

The polarization resonance equation of motion follows from Eqs. (16) and (21) as¹⁷

$$\left(\frac{4\pi}{\omega_p^2}\right)\frac{\partial^2 \mathbf{P}}{\partial t^2} + \boldsymbol{\tau} \cdot \frac{\partial \mathbf{P}}{\partial t} + \frac{\partial w(\mathbf{P}, s, \boldsymbol{\sigma})}{\partial \mathbf{P}} = \mathbf{E}.$$
 (25)

The electric field **E** induces the polarization **P** at resonant frequencies which are eigenvalues of the tensor Ω for which

$$\mathbf{\Omega}^2 = \frac{\omega_p^2 \boldsymbol{\chi}^{-1}}{4\pi} \equiv \omega_p^2 (\boldsymbol{\epsilon} - \mathbf{1})^{-1}.$$
 (26)

The decay rates for the polarization oscillations are eigenvalues of the tensor Γ for which

$$\Gamma = \frac{\omega_p^2 \tau}{4\pi}.$$
(27)

If the decay rates are large on the scale of the resonant frequencies, then the equation of motion is over damped so that

$$\min_{j} \Gamma_{j} \gg \max_{i} \Omega_{i}$$

implies

$$\boldsymbol{\tau} \cdot \frac{\partial \mathbf{P}}{\partial t} + \frac{\partial w(\mathbf{P}, s, \boldsymbol{\sigma})}{\partial \mathbf{P}} = \mathbf{E}.$$
 (28)

Equation (28) represents the Landau-Khalatnikov equation for polarized systems.

IV. HEATING RATE PER UNIT VOLUME

Let us here consider the heating rate implicit in relaxation processes. Independently of the details of the microscopic mechanism for generating such heat, the rates of energy dissipation are *entirely determined* by τ . Explicitly, the heating rates per unit volume for magnetization and polarization are given, respectively, by

$$\dot{q}_{\mathbf{M}} = \frac{\partial \mathbf{M}}{\partial t} \cdot \boldsymbol{\tau} \cdot \frac{\partial \mathbf{M}}{\partial t}$$
(29)

and

$$\dot{q}_{\mathbf{P}} = \frac{\partial \mathbf{P}}{\partial t} \cdot \boldsymbol{\tau} \cdot \frac{\partial \mathbf{P}}{\partial t}.$$
(30)

Finally, the notion of crystal viscosity η_{ijkl} is introduced into elasticity theory³⁵ via the heating rate per unit volume from rates of change in the strain $\partial e / \partial t$; it is

$$\dot{q}_{e} = \frac{\partial e_{ij}}{\partial t} \eta_{ijkl} \frac{\partial e_{kl}}{\partial t}.$$
(31)

Crystal viscosity is employed to describe, among other things, sound wave attenuation. Our purpose is to describe how heating rates in Eqs. (29) and (30) can be related to the heating rate in Eq. (31). This allows us to express the transport coefficients τ in terms of the crystal viscosity.

A. Relaxation via magnetostriction

From the magnetostriction Eq. (10), it follows that magnetic relaxation gives rise to a strain

$$\frac{\partial e_{ij}}{\partial t} = \frac{2}{M} \Lambda_{ijkl} N_k \frac{\partial M_l}{\partial t}$$
(32)

and thereby to the heating rate

$$\dot{q} = \frac{4}{M^2} \frac{\partial M_i}{\partial t} (\Lambda_{mnqi} N_q) \eta_{mnrs} (\Lambda_{rskj} N_k) \frac{\partial M_j}{\partial t}$$
(33)

in virtue of Eq. (31). Employing Eqs. (29) and (33), we find that the magnetic relaxation transport coefficient in the magnetostriction model

$$\tau_{ij} = \frac{4}{M^2} (\Lambda_{mnqi} N_q) \,\eta_{mnrs}(\Lambda_{rskj} N_k) \,. \tag{34}$$

The Gilbert damping tensor follows from Eqs. (19) and (34) as

$$\alpha_{ij} = \frac{4\gamma}{M} (\Lambda_{mnqi} N_q) \,\eta_{mnrs}(\Lambda_{rskj} N_k). \tag{35}$$

The central relaxation tensor Eq. (35) describes the magnetic relaxation in terms of the magnetostriction coefficients and the crystal viscosity.

B. Relaxation via electrostriction

From the electrostriction Eq. (14), it follows that a timevarying polarization gives rise to a time varying strain

$$\frac{\partial e_{ij}}{\partial t} = \beta_{ijk} \frac{\partial P_k}{\partial t}$$
(36)

and thereby to the heating rate

$$\dot{q} = \frac{\partial P_i}{\partial t} \beta_{kli} \eta_{klmn} \beta_{mnj} \frac{\partial P_j}{\partial t}$$
(37)

in virtue of Eq. (31). Employing Eqs. (30) and (37), we find that the electric relaxation transport coefficient in the electrostriction model

$$\tau_{ij} = \beta_{kli} \eta_{klmn} \beta_{mnj}. \tag{38}$$

The central relaxation tensor Eq. (38) describes the polarization relaxation time tensor coefficients in terms of the electrostriction coefficients and the crystal viscosity. The implications of the electrostriction model for the Landau-Khalatnikov equation is to the authors' knowledge a new result.

V. CONCLUSIONS

For ordered polarized and magnetized systems, we have developed phenomenological equations of motion in close analogy with one another. For the magnetized case, the relaxation is driven by the magnetic intensity \mathbf{H}_d yielding the Gilbert equation of motion.⁷ For the polarized case, the relaxation is driven by the electric field \mathbf{E}_d yielding the Tani equation of motion.¹⁷ In both cases, the relaxation time tensor $\boldsymbol{\tau}$ is determined by the crystal viscosity as derived in the Appendix; i.e., in Eqs. (A3) and (A6). The viscosity can be

- ¹L. Onsager and S. Machlup, Phys. Rev. **91**, 1505 (1953).
- ²S. Machlup and L. Onsager, Phys. Rev. **91**, 1512 (1953).
- ³H. B. Callen, in *Fluctuation, Relaxation, and Resonance in Magnetic Systems*, edited by D. ter Haar (Oliver & Boyd, London, 1962), p. 176.
- ⁴R. Kubo et al., Statistical Physics II: Nonequilibrium Statistical Mechanics (Springer, Berlin, 1998).
- ⁵L. Landau and L. Lifshitz, Phys. Z. Sowjetunion 8, 153 (1935).
- ⁶T. L. Gilbert, Armor Research Foundation Report No. 11, 1955 (unpublished).
- ⁷T. L. Gilbert, IEEE Trans. Magn. **40**, 3443 (2004).
- ⁸M. Fähnle *et al.*, J. Phys. D **41**, 164014 (2008).
- ⁹E. Rossi et al., Phys. Rev. B 72, 174412 (2005).
- ¹⁰V. Kamberský, Phys. Rev. B **76**, 134416 (2007).
- ¹¹I. Garate and A. H. MacDonald, Phys. Rev. B **79**, 064403 (2009).
- ¹²A. Brataas et al., Phys. Rev. Lett. **101**, 037207 (2008).
- ¹³K. Gilmore et al., Phys. Rev. Lett. 99, 027204 (2007).
- ¹⁴R. D. McMichael and A. Kunz, J. Appl. Phys. **91**, 8650 (2002).
- ¹⁵L. D. Landau and I. M. Khalatnikov, Dokl. Akad. Nauk SSSR 96, 469 (1954).
- ¹⁶Y. Makita et al., J. Phys. Soc. Jpn. 28, 1221 (1970).
- ¹⁷K. Tani, J. Phys. Soc. Jpn. **26**, 93 (1969).
- ¹⁸S. Sivasubramanian *et al.*, Ferroelectrics **300**, 43 (2004).
- ¹⁹H. Li et al., Integr. Ferroelectr. 97, 69 (2008).

measured independently from the magnetic or electrical relaxation by employing sound absorption techniques.³⁶

APPENDIX: KUBO FORMULAS

From the thermodynamic Eq. (10), the fluctuations in the magnetic intensity are given by magnetostriction, i.e.,

$$\Delta H_k(\mathbf{r},t) = -\left(\frac{2\Lambda_{ijkl}N_l}{M}\right)\Delta\sigma_{ij}(\mathbf{r},t).$$
 (A1)

Equations (A1), (1), and (5) imply

$$\mathcal{G}_{ij}^{mag}(\mathbf{r},\mathbf{r}',t) = \frac{4}{M^2} (\Lambda_{mnqi} N_q) \mathcal{F}_{mnrs}(\mathbf{r},\mathbf{r}',t) (\Lambda_{rskj} N_k).$$
(A2)

Employing Eqs. (A2), (2), and (6), one finds the central result for the magnetic relaxation time tensor; It is

$$\pi_{ij}^{mag} = \frac{4}{M^2} (\Lambda_{mnqi} N_q) \,\eta_{mnrs} (\Lambda_{rskj} N_k) = \frac{\alpha_{ij}}{\gamma M}.$$
 (A3)

From the thermodynamic Eq. (14), the fluctuations in the electric intensity are given by electrostriction, i.e.,

$$\Delta E_k(\mathbf{r},t) = -\beta_{iik} \Delta \sigma_{ii}(\mathbf{r},t). \tag{A4}$$

Equations (A4), (1), and (5) imply

$$\mathcal{G}_{ij}^{pol}(\mathbf{r},\mathbf{r}',t) = \beta_{kli}\mathcal{F}_{klmn}(\mathbf{r},\mathbf{r}',t)\beta_{mnj}.$$
 (A5)

Employing Eqs. (A5), (2), and (6), one finds the central result for the electric relaxation time tensor; It is

$$\tau_{ij}^{pol} = \beta_{kli} \eta_{klmn} \beta_{mnj}. \tag{A6}$$

- ²⁰V. In *et al.*, Phys. Rev. E **73**, 066121 (2006).
- ²¹A. Gordon, Solid State Commun. **147**, 201 (2008).
- ²²S. Sivasubramanian *et al.*, IEEE Trans. Ultrason. Ferroelectr. Freq. Control **50**, 950 (2003).
- ²³H. Li and G. Subramanyam, IEEE Trans. Ultrason. Ferroelectr. Freq. Control 56, 1861 (2009).
- ²⁴D. Guyomar et al., J. Phys. D: Appl. Phys. **40**, 6048 (2007).
- ²⁵B. Andó et al., IEEE Trans. Instrum. Meas. **12**, 31 (2009).
- ²⁶Y. Kato et al., Jpn. J. Appl. Phys., Part 1 46, 2157 (2007).
- ²⁷H. Kohlstedt *et al.*, Microelectron. Eng. **80**, 296 (2005).
- ²⁸L. Eun Sun et al., Jpn. J. Appl. Phys., Part 1 47, 2725 (2008).
- ²⁹R. E. Jones Jr., *Custom Integrated Circuits Conference*, Proceedings of the IEEE (IEEE, New York, 1998), p. 431.
- ³⁰I. Vrejoiu et al., J. Vac. Sci. Technol. B 27, 498 (2009).
- ³¹N. Inoue and Y. Hayashi, IEEE Trans. Electron Devices **48**, 2266 (2001).
- ³²T. Mikolajick et al., Microelectron. Reliab. 41, 947 (2001).
- ³³C. Vittoria *et al.*, Phys. Rev. B **81**, 014412 (2010).
- ³⁴L. D. Landau and E. M. Lifshitz, *Electrodynamics of Continuous Media* (Pergamon Press, Oxford, 1960).
- ³⁵L. D. Landau and E. M. Lifshitz, *Theory of Elasticity* (Pergamon Press, Oxford, 1986), Secs. 10, 23, and 34.
- ³⁶A. B. Bhatia, *Ultrasonic Absorption* (Oxford University Press, Oxford, 1967).