

## Magnetocapacitance in Nonmagnetic Composite Media

Meera M. Parish<sup>1,2,\*</sup> and Peter B. Littlewood<sup>3</sup>

<sup>1</sup>*Department of Physics, Princeton University, Princeton, New Jersey 08544, USA*

<sup>2</sup>*Princeton Center for Theoretical Science, Princeton University, Princeton, New Jersey 08544, USA*

<sup>3</sup>*Cavendish Laboratory, Madingley Road, Cambridge CB3 0HE, United Kingdom*

(Received 23 July 2008; published 17 October 2008)

The dielectric response in a magnetic field is routinely used to probe the existence of coupled magnetic and elastic order in the multiferroics. However, here we demonstrate that magnetism is not necessary to produce a magnetocapacitance when the material is inhomogeneous. By considering a two-dimensional, two-component composite medium, we find a characteristic dielectric resonance that depends on magnetic field. We propose this as a possible signature of inhomogeneities and we argue that this behavior has already been observed in nanoporous silicon and some manganites.

DOI: [10.1103/PhysRevLett.101.166602](https://doi.org/10.1103/PhysRevLett.101.166602)

PACS numbers: 72.20.My, 72.80.Ng, 75.47.Pq

The behavior of a material's dielectric constant under an applied, alternating electric field provides a powerful probe of its microscopic properties. For example, dielectric relaxation (characterized by a delay in the electrical polarization with respect to the changing electric field) can signify the presence of noninteracting, freely rotating dipoles [1]. More recently, the dielectric response as a function of an applied static magnetic field has been used to determine whether or not a material is a *multiferroic* [2–5]. In multiferroic systems, magnetic order and electrical polarization are coupled, thus giving rise to a magnetocapacitance. This magnetoelectric coupling makes multiferroics a topic of interest owing to its potential application [6].

It is well known that Maxwell-Wagner extrinsic effects, such as inhomogeneities and contact effects, can enhance the dielectric constant and yield dielectric relaxation in the absence of intrinsic dipolar relaxation [7]. However, it was only recently appreciated that the Maxwell-Wagner effect can also yield a magnetocapacitance *without* multiferroicity and its associated magnetoelectric coupling, provided the material exhibits an intrinsic magnetoresistance [8]. Such effects clearly demonstrate that a magnetocapacitance is not sufficient to establish multiferroicity. On the other hand, having a magnetocapacitance without magnetoelectric coupling may be more practical for technological applications.

In this Letter, we show that in an inhomogeneous medium one does not require even an intrinsic magnetoresistance to produce a magnetic-field-dependent dielectric constant. Thus, there is the possibility of a sizable magnetocapacitance without any magnetic order.

As far as we are aware, our work is the first to address the dielectric response of composite media in the presence of a magnetic field, in a regime relevant to semiconductors. While the dc magnetotransport in classical, disordered media has been studied extensively [9–14], there are very few works on the ac dielectric response. Moreover, the latter are restricted to metal-insulator composites which are either in the absence of a magnetic field [15] or they

have an electric-field frequency  $\omega$  that is close to the plasma resonance frequency [16]. Such a regime is only applicable to highly metallic materials rather than the semiconducting compounds we are interested in here.

We find that the dielectric function of a composite medium exhibits unexpected behavior in a magnetic field and, as such, could be used as a probe for inhomogeneities. Specifically, we find a strong dielectric resonance as a function of frequency, where the position of the dissipation peak sensitively depends on magnetic field. Furthermore, the real part of the dielectric response even switches sign in the vicinity of the resonance at finite field. Using these results, we argue that inhomogeneities are a probable cause of the magnetic-field-dependent “dielectric relaxation” observed in nanoporous silicon [17] and in some manganese compounds [18,19].

To focus our investigation, we restrict ourselves to a two-component, two-dimensional (2D) composite medium. This should be sufficient to describe the salient features of a disordered material, but simple enough for us to derive analytical expressions. We assume that the length scale of the inhomogeneities is much larger than all the microscopic length scales of the system, e.g., the mean free path. Thus, the local current density  $\mathbf{j}(\omega, \mathbf{r})$  is related to the local electric field  $\mathbf{E}(\omega, \mathbf{r})$  via Ohm's law:  $\mathbf{j}(\omega, \mathbf{r}) = \hat{\sigma}(\omega, \mathbf{r})\mathbf{E}(\omega, \mathbf{r}) \equiv i\omega\hat{\epsilon}(\omega, \mathbf{r})\mathbf{E}(\omega, \mathbf{r})$ , where  $\hat{\sigma}(\omega, \mathbf{r})$  is the local conductivity tensor and  $\hat{\epsilon}(\omega, \mathbf{r})$  is the local dielectric function. Furthermore, we take the regime where (i)  $1/\omega$  is much larger than the scattering time, and (ii) the intrinsic conductivity and dielectric constant of each component are frequency independent. Then, the conductivity tensor in a transverse magnetic field  $\mathbf{H} = H\hat{z}$  is given by

$$\hat{\sigma}(\omega) = \frac{\sigma}{1 + \beta^2} \begin{pmatrix} 1 & \beta \\ -\beta & 1 \end{pmatrix} + i\omega\epsilon\mathbf{1}. \quad (1)$$

Here,  $\sigma$  is the dc scalar conductivity,  $\epsilon$  is the bare dielectric constant and  $\beta = \mu H$ , where  $\mu$  is the carrier mobility. In general, the quantities  $\sigma$ ,  $\epsilon$  and  $\mu$  will be random functions

of space, but here we will only allow them to take on two different values.

By taking volume averages over the composite medium, we can define an effective conductivity such that  $\langle \mathbf{j}(\omega) \rangle = \hat{\sigma}_e(\omega) \langle \mathbf{E}(\omega) \rangle$ . However, for the typical measuring setup depicted in Fig. 1(a), we clearly have the boundary condition  $j_y = 0$ , and so it is more natural to use the effective resistivity  $\hat{\rho}_e(\omega) \equiv \hat{\sigma}_e(\omega)^{-1}$ . Thus, the components of the dielectric function that are actually probed in experiment are given by  $\varepsilon_{xx}(\omega) = (i\omega\rho_{e,xx}(\omega))^{-1}$  and  $\varepsilon_{xy}(\omega) = (i\omega\rho_{e,xy}(\omega))^{-1}$ . The longitudinal response  $\varepsilon_{xx}(\omega)$  is simply the capacitance of the configuration in Fig. 1(a), while the transverse response  $\varepsilon_{xy}(\omega)$  can be extracted from a measurement of the transverse electric field  $E_y(\omega) = (\varepsilon_{xx}(\omega)/\varepsilon_{xy}(\omega))E_x(\omega)$ . Note that in the limit  $\beta \rightarrow 0$ , we must have  $E_y(\omega) \rightarrow 0$  and thus  $\varepsilon_{xy}(\omega) \rightarrow \infty$ .

First, it is instructive to consider a homogeneous medium described by the conductivity tensor in Eq. (1). One can represent this as a capacitor in parallel with a resistor [see Fig. 1(b)], where both circuit elements are frequency independent. For this simple case, we find the real part of the longitudinal dielectric response to be

$$\Re[\varepsilon_{xx}(\omega)] = \frac{\varepsilon(1 - \beta^2 + (\omega\tau)^2(1 + \beta^2)^2)}{1 + (\omega\tau)^2(1 + \beta^2)^2} \quad (2)$$

where  $\tau = \varepsilon/\sigma \equiv \rho\varepsilon$ . Thus, we see that  $\Re[\varepsilon_{xx}(\omega)]$  can be *negative* at sufficiently low  $\omega\tau$  when  $\beta > 1$ . Usually, at zero magnetic field,  $\Re[\varepsilon_{xx}(\omega)]$  only goes negative at a dielectric resonance, and this signifies the presence of an inductive element in the system. However, for our simple resistor-capacitor circuit in a strong magnetic field, the boundary condition  $j_y = 0$  causes the Hall component of the resistive element to be mixed into the longitudinal dielectric response, resulting in a negative real part.

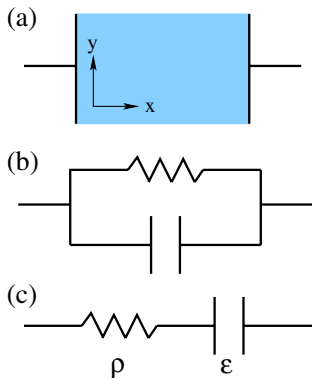


FIG. 1 (color online). Simple circuits of resistors  $\hat{\rho} \equiv \hat{\sigma}^{-1}$  and capacitors  $\varepsilon$  that illustrate the basics of dielectric response. Diagram (a) depicts the measuring setup, where a rectangular sample is confined between metallic plates and subjected to an applied electric field  $E_x\hat{x}$ , (b) represents a simple homogeneous medium, and (c) represents the simplest metal-insulator composite that exhibits the Maxwell-Wagner effect.

Now let's switch off the magnetic field and consider the simplest realization of an inhomogeneous medium: a resistor and capacitor in series [Fig. 1(c)]. Here, we have a dielectric function

$$\varepsilon_{xx}(\omega) = \frac{\varepsilon(1 - i\omega\tau)}{1 + (\omega\tau)^2}. \quad (3)$$

For low frequencies  $\omega\tau < 1$ , the capacitor dominates the response, while at high frequencies  $\omega\tau > 1$ , the voltage drop falls primarily over the resistor instead since there is insufficient time for the capacitor to build up charge. Thus, at the characteristic frequency  $\omega\tau = 1$ , we have a rapid change in  $\Re[\varepsilon_{xx}(\omega)]$  and an associated peak in  $\Im[\varepsilon_{xx}(\omega)]$ . This is a basic illustration of the Maxwell-Wagner effect, where the inhomogeneity mixes the real and imaginary modes of the response to create an apparent dielectric relaxation.

From these simple circuits, we see that geometric effects and macroscopic inhomogeneities can mix the different “modes” (longitudinal, transverse, real and imaginary) of the system, and thus generate unexpectedly rich behavior, such as the dielectric resonance we will describe below. Inhomogeneities play a similar role in the dc magnetotransport of heavily disordered media, since they can mix the Hall resistivity into the longitudinal component to produce a linear magnetoresistance [14].

We now address the full problem of 2D isotropic inhomogeneous media. We suppose that the material is composed of two phases  $\hat{\sigma}_1(\omega)$  and  $\hat{\sigma}_2(\omega)$  in proportions  $p_1 = p$  and  $p_2 = 1 - p$ , respectively. To model the dielectric response of a strongly inhomogeneous medium, we take the extreme case  $\sigma_1 = 0$ ,  $\varepsilon_1 = \varepsilon$  and  $\sigma_2 = \sigma$ ,  $\varepsilon_2 = 0$ ; i.e., we consider purely capacitive and purely resistive regions. When the proportions of each phase are equal ( $p = 1/2$ ), we can use a symmetry transformation for the current density and electric field [20–22] to derive the *exact* result for the components of the effective dielectric function:

$$\varepsilon_{xx}(\omega) = \varepsilon \frac{(1 + i\omega\tau)}{\sqrt{i\omega\tau}\sqrt{(1 + i\omega\tau)^2 - (\omega\tau\beta)^2}}, \quad (4)$$

$$\varepsilon_{xy}(\omega) = \frac{\varepsilon}{\beta} \left(1 - \frac{i}{\omega\tau}\right). \quad (5)$$

We immediately see that the Hall component  $\varepsilon_{xy}(\omega)$  is equivalent to the longitudinal dielectric response of a homogeneous material [Fig. 1(b)] in the absence of a magnetic field, with bare dielectric constant  $\varepsilon/\beta$  and resistivity  $\beta\rho$  (which is just the Hall resistivity for dc magnetotransport). As we will see below, this is unique to the case where the proportions are equal.

The longitudinal dielectric response  $\varepsilon_{xx}(\omega)$ , on the other hand, involves a nontrivial mixture of dielectric and resistive components in both the real and imaginary parts. The behavior is plotted in Fig. 2 and represents the key result of

this paper. For large magnetic fields  $\beta > 1$ , a dielectric resonance occurs at  $\beta\omega\tau = 1$ , where  $\Re[\epsilon_{xx}(\omega)]$  has a pronounced peak and  $\Im[\epsilon_{xx}(\omega)]$  varies rapidly. In particular, we see that the resonant frequency is determined by the Hall resistivity  $\beta\rho$ , instead of  $\rho$  like in the usual Maxwell-Wagner effect. Thus, the position of the peak in frequency space is inversely proportional to  $\beta$ . By substituting  $\beta\omega\tau = 1$  into Eq. (4), we see that the peak height  $\Re[\epsilon_{xx}^p] \approx \epsilon\beta/\sqrt{2}$ . However, the actual dissipation at the peak remains constant because here we always have  $\Re[\rho_{e,xx}] \approx \sqrt{2}\rho$ . In addition, the real part  $\Re[\epsilon_{xx}(\omega)]$  becomes *negative* for  $\beta\omega\tau \geq 1$  and  $\beta > 1$ , but it exhibits ordinary Maxwell-Wagner dielectric relaxation at zero magnetic field. Surprisingly, there is no corresponding peak in  $\Re[\epsilon_{xx}(\omega)]$  when  $\beta = 0$ , and this holds for arbitrary insulator fraction  $p$  according to results obtained from the effective medium approximation and numerical resistor network studies [15]. This is particularly unexpected given that we obtain a dielectric relaxation peak from the simplest metal-insulator composite in Fig. 1(c). However, we speculate that one may recover the peak when the disorder is anisotropic since Fig. 1(c) is obviously highly anisotropic.

To determine whether or not this dielectric resonance is a generic feature of 2D conductor-insulator composites, we use the effective medium approximation [23,24] to exam-

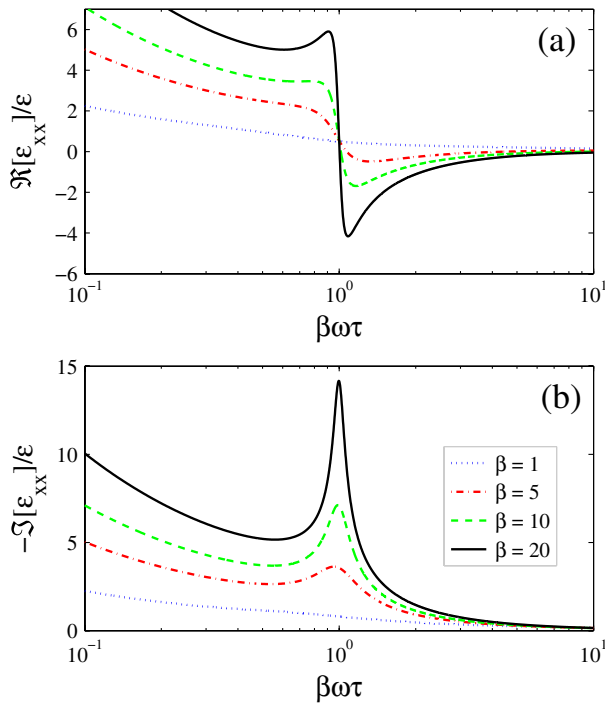


FIG. 2 (color online). Dielectric response versus frequency over a range of magnetic fields  $\beta$  for a 2D two-component medium with equal proportions ( $p = 1/2$ ). When  $\beta > 1$ , there is a resonance at normalized frequency  $\beta\omega\tau = 1$ , where the real part (a) varies rapidly, eventually changing sign, while the imaginary part (b) exhibits a peak.

ine the case where  $p \neq 1/2$ . This amounts to numerically solving the coupled equations

$$\sum_i p_i (\hat{\sigma}_i - \hat{\sigma}_e) \left( \mathbf{1} - \frac{\hat{\sigma}_i - \hat{\sigma}_e}{2\sigma_{e,xx}} \right)^{-1} = 0. \quad (6)$$

When  $\beta > 1$ , we find that there is always a resonance at  $\beta\omega\tau = 1$  for arbitrary  $p$ , but that the height of the dissipation peak  $\Re[\epsilon_{xx}^p(\omega)]$  decreases with increasing  $p$  (decreasing conductor fraction) as shown in Fig. 3. Also, the behavior of  $\Re[\epsilon_{xx}(\omega)]$  is qualitatively unchanged at the resonance, apart from the fact that the point at which it switches sign occurs at a higher field for increasing  $p$ . This makes sense given that it is the Hall component of the conducting region that is responsible for the negative part of  $\Re[\epsilon_{xx}(\omega)]$ .

The largest variation with respect to  $p$  occurs in the Hall component  $\Re[\epsilon_{xy}(\omega)]$ . As shown in Fig. 4, while  $\Re[\rho/\rho_{e,xy}]$  is relatively insensitive to frequency for  $p \leq 1/2$ , it goes negative beyond a critical value of  $\beta\omega\tau > 1$  when  $p > 1/2$ . Indeed, in the limit  $p \rightarrow 1$ , we have  $\Re[\rho/\rho_{e,xy}] \rightarrow \infty$  for  $\beta\omega\tau < 1$  and  $\Re[\rho/\rho_{e,xy}] \rightarrow -\infty$  for  $\beta\omega\tau > 1$ , with  $\Re[\rho/\rho_{e,xy}] = 1/\beta$  always at  $\beta\omega\tau = 1$ . Thus, we see there is some symmetry to the “mode-mixing”: the dielectric region reverses the sign of the Hall component at high frequencies in the same way that the Hall resistivity forces the dielectric constant to become negative at low frequencies. In principle, one could exploit this property to estimate the insulator fraction  $p$  of a composite medium.

Finally, we emphasize that the existence of the dielectric resonance is not conditional on there being a magnetoresistance at  $\omega = 0$ . One clearly sees this from the fact that there is always a resonance at  $\beta\omega\tau = 1$  when  $\beta > 1$ , even though the magnetoresistance is zero for  $p < 1/2$  and undefined for  $p \geq 1/2$ . One can obtain a nonzero magnetoresistance when the dielectric region has a nonzero conductivity  $\sigma_1 \neq \sigma_2$ , but this saturates with magnetic field unless  $p = 1/2$  [24,25].

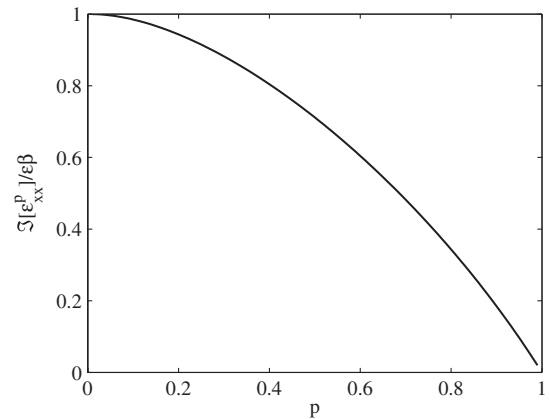


FIG. 3. Behavior of the resonance peak  $\Re[\epsilon_{xx}^p]$  at  $\beta\omega\tau = 1$  as a function of insulating fraction  $p$ . This holds for all  $\beta > 1$ .

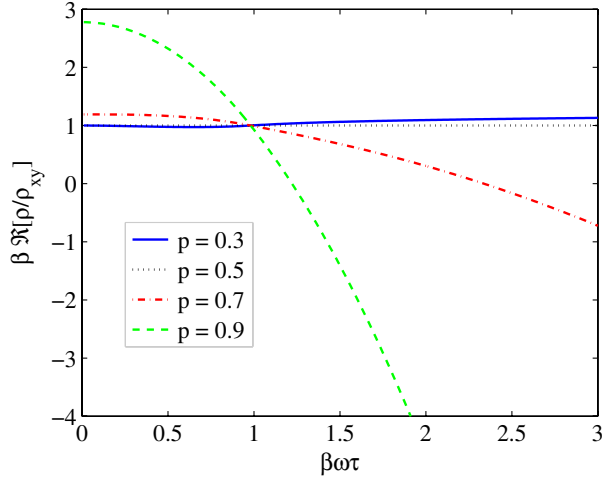


FIG. 4 (color online). Hall component  $\Re[\rho/\rho_{xy}] \equiv -\omega\tau\Im[\epsilon_{xy}]/\epsilon$  at  $\beta = 5$  as a function of frequency for different insulating fractions  $p$ . Similar curves are obtained for other  $\beta > 1$ . At  $\beta\omega\tau = 1$ ,  $\Im[\epsilon_{xy}]/\epsilon = 1$  for all  $p$  and  $\beta$ .

The magnetic-field-dependent dielectric resonance described above has already been observed experimentally in nanoporous silicon [17], a material which clearly resembles a conductor-insulator composite. Specifically, the dielectric response was measured as a function of temperature  $T = 25\text{--}40$  K at fixed frequency  $\omega = 100$  kHz and magnetic field  $H = 0\text{--}32$  T, but this is equivalent to varying  $\omega\tau$  if we assume that  $\epsilon$  is temperature-independent and that the electrical transport in the semiconductor is activated:  $\rho = \rho_0 e^{\Delta/k_B T}$ , where  $\Delta$  is the activation gap. Then the temperature at which the resonance occurs is  $k_B T = \Delta/\log(1/\beta\omega\epsilon\rho_0)$ , so that we expect the dielectric resonance to shift to higher  $T$  with increasing  $H$ , as indeed was observed. Assuming that  $H = 1$  T corresponds to  $\beta \approx 1$ , we can fit the data with  $\Delta \approx 30$  meV, consistent with activation from an impurity band, and  $\omega_0 \equiv 1/\rho_0\epsilon \approx 4 \times 10^{10}$  Hz. Our results should be contrasted with an alternative proposal [26] which interprets the data with a model of Debye relaxation, requiring that charge carriers are pinned to a distribution of sites, but which does not explicitly introduce inhomogeneous fields.

In principle, one can exploit the dielectric resonance to construct a magnetic sensor that is sensitive to fields in the neighborhood of  $\beta \sim 1/\omega\tau$ . However, the challenge is to make it operational within the ideal frequency range  $\omega = 1$  kHz–1 MHz at room temperature and for  $H < 1$  T, since this requires a semiconductor that has both a large band gap  $\Delta > 0.5$  eV and a high mobility  $\mu > 1$  T $^{-1}$ .

A dielectric resonance has also been observed in the manganite  $\text{La}_{2/3}\text{Ca}_{1/3}\text{MnO}_3$  at zero magnetic field, just above the ferromagnetic transition temperature [18]. Moreover, Rairigh *et al.* [19] observed a “colossal mag-

netocapacitance” coinciding with a regime of phase separation between magnetic metal and charge-ordered insulator. Magnetic materials have, of course, an intrinsic magneto-capacitive coupling, but we suggest that the magnetodielectric response is strongly enhanced in such a phase-separated composite, where the magnetized domains combine to produce a large internal magnetic field which acts on the transport currents.

To conclude, we have shown that a unique magnetic-field-dependent dielectric resonance is produced by strongly inhomogeneous media and, as such, may be used as a probe of inhomogeneities or as a magnetic sensor.

We are grateful to Neil Mathur and Gustau Catalan for fruitful discussions. This research was supported in part by the National Science Foundation under Grant Number DMR-0645461.

\*mparish@princeton.edu

- [1] A. K. Jonscher, *Dielectric Relaxation in Solids* (Chelsea Dielectrics Press, London, 1983).
- [2] J. Hemberger *et al.*, *Nature (London)* **434**, 364 (2005).
- [3] T. Kimura *et al.*, *Phys. Rev. B* **67**, 180401(R) (2003).
- [4] N. Hur *et al.*, *Phys. Rev. Lett.* **93**, 107207 (2004).
- [5] M. P. Singh, W. Prellier, C. Simon, and B. Raveau, *Appl. Phys. Lett.* **87**, 022505 (2005).
- [6] W. Eerenstein, N. D. Mathur, and J. F. Scott, *Nature (London)* **442**, 759 (2006).
- [7] P. Lunkenheimer *et al.*, *Phys. Rev. B* **66**, 052105 (2002).
- [8] G. Catalan, *Appl. Phys. Lett.* **88**, 102902 (2006).
- [9] M. B. Isichenko, *Rev. Mod. Phys.* **64**, 961 (1992).
- [10] C. Herring, *J. Appl. Phys.* **31**, 1939 (1960).
- [11] Y. A. Dreizin and A. M. Dykhne, *Sov. Phys. JETP* **36**, 127 (1973).
- [12] D. Stroud and F. P. Pan, *Phys. Rev. B* **13**, 1434 (1976).
- [13] B. Y. Balagurov, *Sov. Phys. Solid State* **28**, 1694 (1986).
- [14] M. M. Parish and P. B. Littlewood, *Nature (London)* **426**, 162 (2003); *Phys. Rev. B* **72**, 094417 (2005).
- [15] T. B. Murtanto, S. Natori, J. Nakamura, and A. Natori, *Phys. Rev. B* **74**, 115206 (2006).
- [16] D. J. Bergman and Y. M. Strelniker, *Phys. Rev. Lett.* **80**, 857 (1998).
- [17] R. Vasic *et al.*, *Curr. Appl. Phys.* **7**, 34 (2007).
- [18] J. Rivas *et al.*, *Appl. Phys. Lett.* **88**, 242906 (2006).
- [19] R. P. Rairigh *et al.*, *Nature Phys.* **3**, 551 (2007).
- [20] A. M. Dykhne, *Sov. Phys. JETP* **32**, 348 (1971).
- [21] B. Y. Balagurov, *Sov. Phys. Solid State* **20**, 1922 (1978).
- [22] A. M. Dykhne and I. M. Ruzin, *Phys. Rev. B* **50**, 2369 (1994).
- [23] D. Stroud, *Phys. Rev. B* **12**, 3368 (1975).
- [24] V. Guttal and D. Stroud, *Phys. Rev. B* **71**, 201304(R) (2005).
- [25] R. Magier and D. J. Bergman, *Phys. Rev. B* **74**, 094423 (2006).
- [26] J. S. Brooks *et al.*, *Phys. Rev. B* **78**, 045205 (2008).