Dielectric response due to stochastic motion of pinned domain walls

A. A. Fedorenko, V. Mueller, and S. Stepanow

Martin-Luther-Universität Halle, Fachbereich Physik, D-06099 Halle, Germany

(Dated: November 19, 2018)

We study the contribution of stochastic motion of a domain wall (DW) to the dielectric AC susceptibility for low frequencies. Using the concept of waiting time distributions, which is related to the energy landscape of the DW in a disordered medium, we derive the power-law behavior of the complex susceptibility observed recently in some ferroelectrics below Curie temperature.

PACS numbers: 05.20.-y, 74.25.Qt, 64.60.Ak, 75.60.Ch

During the last decade considerable interest has been attracted to the anomalously high dielectric susceptibility observed in several ferroelectrics below the paraelectric-ferroelectric phase transition temperature $T_c$. The theory of critical phenomena predicts in the vicinity of $T_c$ the Curie-Weiss law for the temperature dependent static susceptibility. However, for some ferroelectrics, such as potassium dihydrogen phosphate (KH$_2$PO$_4$) or rubidium dihydrogen phosphate (RbH$_2$PO$_4$), the complex susceptibility $\chi(\omega) = \chi'(\omega) - i\chi''(\omega)$ within the so-called plateau range $T_f < T < T_c$ remains unusually high. This anomaly is usually ascribed to DW motion, an activity which freezes out below $T_f$. According to Ref. 1, the low-frequency dielectric spectrum in the plateau range of KH$_2$PO$_4$ consists of non-Debye contribution, which has a power-law behavior with a small exponent, and several Debye-like constituents. Recent experiments performed on RbFe$_{1/2}$Nb$_{1/2}$O$_3$ and Sr$_{0.6}$Ce$_{0.4}$Ba$_{0.39}$Nb$_2$O$_6$ (SBN:Ce) revealed in the range of low frequency $\omega < \omega_c$ a power-law behavior of the DW response

$$\chi(\omega) = \chi_\infty (1 + 1/(i\omega\tau_0)^n)$$

with $0 < n < 1$. Here $\omega_c$ is the threshold frequency separating the low- and high- frequency regimes, $\tau_0$ the characteristic relaxation time and $\chi_\infty$ the high-frequency limit of the complex susceptibility. The real and imaginary parts of the complex susceptibility are related by equation

$$\chi''(\omega) = (\chi'(\omega) - \chi_\infty) \tan(n\pi/2),$$

which follows from the Kramers-Kronig relation. In a range of frequencies above $\omega_c$, the imaginary part of susceptibility $\chi''(\omega)$ was found to increase linearly on a log-log scale, while the corresponding real part $\chi'(\omega)$ decreases linearly on a linear-log scale.

An attempt to explain phenomenologically the existence of two regimes of the dynamic response was made in Ref. 2, where the low-frequency behavior was attributed to the irreversible viscous motion of DWs in a disordered medium. However, this interpretation, based on the picture of the dynamic hysteresis developed in Ref. 2, is restricted to the adiabatic regime only.

Eqs. 1 and 2 are in accordance with Jonscher’s universal dielectric response law, which describes the response of a wide class of dielectrics in a broad range of frequencies. Generally, this behavior is related to hopping of charge carriers, such as electrons, polarons or various ions, between different localized states. The exponent $n$ is determined by the spatial and energetic distribution of the corresponding states. The ferroelectric non-Debye dielectric response is expected to be due to jumps of DWs (or DW segments) between different metastable states.

In the present paper, we study the contribution of stochastic motion of a DW to the dielectric susceptibility at low frequencies, using the concept of waiting time distribution, which is related to the energy landscape of the DW in a disordered medium. We restrict our consideration to the regime in which the field driven motion of adjacent DWs is independent of each other. According to Ref. 3, the exponent $n$ depends on the poling state of dielectric samples. Poling is expected to decrease the density of DWs, which would improve the single DW approach. However, among others it may also change the distribution of impurities.

The DW, which separates domains of different polarization, can be viewed as a two-dimensional ($d = 2$) elastic interface moving in a disordered environment. The configuration of a $d$-dimensional elastic interface is described by the profile function $z(x, t)$, which obeys the Langevin-type equation

$$\mu^{-1} \frac{\partial z(x, t)}{\partial t} = \gamma \nabla^2 z + h_0 \cos \omega t + g(x, z) + \eta(x, t),$$

where $\mu$ is the mobility, $\gamma$ the stiffness constant and $h(t) = h_0 \cos \omega t$ the AC driving force density. Note that $x$ is the $d$-dimensional vector. We assume Gaussian distribution of thermal noise, with zero mean and the correlator

$$\langle \eta(x, t) \eta(x', t') \rangle = 2 \mu^{-1} T \delta^{(d)}(x - x') \delta(t - t').$$

Note that in real ferroelectrics the dependence of $\mu$ and $\gamma$ on temperature $T$ may complicate the comparison of the computed temperature dependence of the susceptibility with experiment. Pinning to impurities, which are
described by the quenched random force $g(x, z)$, strongly suppresses the DW mobility. The quenched forces are assumed to be Gaussian distributed with zero mean and the variance

$$\langle g(x, z)g(x', z') \rangle = \delta^{(d)}(x - x') \Delta(z - z').$$  \hfill (5)

To make this model well-defined, we introduce the cutoff $\Lambda_0^{-1}$ in the Dirac’s delta function $\delta^{(d)}(x)$ at scales in the order of the impurity separation. In what follows we consider random field (RF) disorder, where for $z > 0$ the correlation $\Delta(z) = \Delta(-z)$ is a monotonically decreasing function of $z$, and decays rapidly to zero over the distance $a$.

To describe different regimes of the DW motion, we recall the fundamental scales of the problem. On scales smaller than the Larkin scale $L_c \approx (a^2 \gamma^2/\Delta(0))^{1/\varepsilon}$, the elasticity wins over the disorder for $d < 4$ and the DW remains flat, while on larger scales the DW becomes rough and self-affine with a nontrivial roughness exponent $\zeta$. At $T = 0$ and $\omega \to 0$, the system undergoes a second order depinning transition at $h_0 = h_c$. The depinning threshold $h_0 \approx a^2 \gamma L_c^{-2}$ can be estimated from balancing the pinning forces and the driving forces on the scale $L_c$. At finite temperature, the depinning transition smears out, and the DW moves at any field strength. In the creep regime, which sets in for low fields, the motion is very slow and controlled by thermal activation. The energy barrier $E_B(L_c) \approx U_c(L_c/L_c)^\varepsilon$ to be overcome during one period of the field. Thus the threshold frequency

$$\omega^* = 1/\tau(L_{\text{opt}}) = \tau_0^{-1} \exp(-U_c/T(h_0/h_c)^\mu)$$  \hfill (7)

separates the sliding regime from the pinned regime. The critical value $\omega^*$, which corresponds to the experimental setup used in Refs. 4 and 8, can be estimated as follows. Since the activity of DWs for $T < T_f$ is almost frozen, we conclude that temperatures in the range $T_f < T < T_c$ are of order $U_c$. The creep exponent is $\mu(d = 2) = 1$ for RF disorder. The amplitude of the electric field in the dielectric experiments of Ref. 4 $h_0 \approx 300V/m$ is far below the coercive field $150kV/m$. Assuming that the threshold $h_c$ is of order of the coercive field we find that the experimental frequencies ($\omega > 0.01Hz$) are likely to be much higher than the threshold frequency estimated from Eq.(7). To explain the observed low-frequency response we suppose that DWs can also move for $\omega^* < \omega < \omega_c$ ($\omega_c$ is defined below) via hopping between different minima of the energy landscape. However, this motion is extremely slow and can be characterized by the displacement $z \propto t^n$ with $n < 1$ so that in the adiabatic limit $t \to \infty$ the average velocity will vanish. In analogy with the stochastic transport phenomena in disordered solids, we refer to the DW-hopping in the frequency range $\omega^* < \omega < \omega_c$ as the stochastic regime.

To calculate the dielectric response in the stochastic regime, we consider a 180° domain structure of quasiperiodicity 2l, where the DW separates homogeneously polarized regions with spontaneous polarization $P_l$ and $-P_l$, respectively. The time dependence of the macroscopic polarization reads

$$dP(t)/dt = (2P_0/l)\langle v(t) \rangle.$$  \hfill (8)

The linear susceptibility $\chi(\omega)$ defined through the Fourier transform $P(\omega) = \chi(\omega)h(\omega)$ of $P(t)$ can be expressed as

$$\chi(\omega) = \frac{2P_0 \mu(\omega)}{l \omega}. $$ \hfill (9)

where $\mu(\omega)$ is the renormalized DW mobility given by $\langle v(t) \rangle = \mu(\omega)h(\omega)$. The center of mass of the field driven DW probes different local minima of the rugged energy landscape, corresponding to different metastable DW-configurations (see Fig. 1). This motion is similar to the stochastic motion of a particle in the RF environment. The Langevin equation for the particle is given by

$$\frac{dz}{dt} = f(z) + h + \eta(t),$$ \hfill (10)

where $f(z)$ is the Gaussian distributed RF with $\langle f(z) \rangle = 0$, $\langle f(z)f(z') \rangle = \Delta_0 \delta(z - z')$, and $\eta(t)$ the thermal noise. At low temperatures $T$ and small driving forces $h$, the motion of the particle can be described by the thermally activated dynamics, which is controlled by the probability distribution density $W(E)$ of energy barriers $E$ separating different metastable states. Using the standard methods of the field theory, $W(E)$ can be written as a path integral, the estimate of which by the steepest descent method gives

$$W(E) = E_0^{-1} \exp(-E/E_0)$$ \hfill (11)
with \( E_0 = \Delta_0/(2\hbar) \). Due to the thermally activated dynamics, the waiting time \( \tau(E) = \tau_0 \exp(-E/T) \) needed to overcome the barrier \( E \) is characterized by the distribution \( \Psi(\tau) d\tau = W(E) dE \), which decays as a power law for large \( \tau \)

\[
\Psi(\tau) = \frac{n\pi^n}{T^{1+n}}, \quad n = T/E_0.
\]

As follows from Eq. (12) the average waiting time \( \int d\tau \Psi(\tau) \) diverges for low temperatures \( T < E_0 \). This indicates that the particle motion is dominated by very rare, but extremely deep potential wells, which leads to sublinear drift \( z \propto t^n \), \( n < 1 \).

The distribution of waiting times which controls the motion of the center of mass of DW can also be related to the distribution of the energy barriers \( W(E) \). As shown in Ref. 13, the energy barriers scale as the energy minima, and therefore, have identical distributions. Generalizing the loop expansion of the energy distribution developed in Ref. 13 for driven elastic interface we have obtained the energy distribution of the DW in the presence of nonzero external force \( h \) as

\[
W(E) = \int_{-\infty}^{\infty} ds 2\pi \exp(isE) \exp \left[ -\frac{1}{2} L^2 \sum_k \left( 1 + \frac{is}{\gamma^2 k^2} \int q \frac{\Delta_g}{1 + q^2 h^2/(\gamma^2 k^2)} \right) \right],
\]

where \( \Delta_g \) is the Fourier transform of \( \Delta(z) \). Assuming that the width \( a \) of the disorder correlator \( \Delta_g \) is small, we approximate the integral over \( q \) in Eq. (13) by \( \Delta(0)/(1 + h^2 a^2/\gamma^2 k^4) \). The inverse Fourier transform of the discrete version of Eq. (13) for \( d = 1 \) gives \( f(x;\kappa) = \sum_j C_j(\kappa) \exp(-(j^2 + \kappa^2)/2) x \), where \( x = 4\pi^2 \gamma E/\Delta(0) L^2 \), \( \kappa = (h a L^2)/(4\pi^2 \gamma) \), and the coefficients \( C_j(\kappa) \) are given by

\[
C_j(\kappa) = \frac{2^{j^2 - 1}}{\pi^2 \kappa} \Gamma(2j) \Gamma\left( j - \frac{\kappa}{2} \right) \Gamma\left( j +\frac{\kappa}{2} \right) \prod_{i=1}^{j} \left( j^2 + i^2 \right)^{\cos^2(\pi \sqrt{2i}) - \cos^2(\pi \sqrt{2j})/2} \left( j^2 + i^2 \right).
\]

Using the steepest descent method to estimate the sum in \( f(x;\kappa) \), we find that for large energy \( E \) the distribution is exponential \( W(E) \propto \exp(-2ah E/\Delta(0)) \). However, this expression of the energy distribution is incorrect for the same reason as the perturbation result for the roughness. The appropriate method to extract the correct behavior of elastic interfaces in random environments is the functional renormalization group. To find the effect of renormalization on the energy distribution for driven interface, we follow Ref. 13 and replace all bare quantities in Eq. (13) by the renormalized ones. We obtain the renormalized energy distribution for \( d = 1 \) as \( W(E) \propto \exp(-2(\gamma ah)^1/2 E/\Delta(0) L_c) \). The evaluation of Eq. (13) for \( d > 1 \) is more complicated. However, for \( d = 2 \) the tail of the energy distribution for large energies is exponential, and is given by Eq. (11) with \( E_0 \) of order \( U_c \). Consequently, the distribution of waiting times has the same form.

To calculate the renormalized mobility \( \mu(\omega) \), we now employ the Montroll-Weiss formalism developed in Refs. 10 and 22 for continuous time random walks. According to Ref. 10, the mobility \( \mu(\omega) \) can be written as

\[
\mu(\omega) = \frac{\sigma^2}{2T} \int_0^\infty dt e^{-i\omega t} \langle (z(t) - z(0))^2 \rangle
\]

where \( \sigma \) is the average distance between adjacent potential wells and \( \Psi(\omega) \) is the Laplace transform of \( \Psi(\tau) \). For \( \Psi(\tau) \) given by Eq. (12) we obtain

\[
\Psi(\omega) = \int_0^\infty dt e^{-\omega t} \Psi(\tau) \approx 1 - A(\omega \tau_0)^n,
\]

where \( A = \pi/(\Gamma(n) \sin(n\pi)) \). Combining Eqs. (9), (15), and (16), we obtain the complex susceptibility as

\[
\chi(\omega) = \frac{2P_0 \sigma^2}{A L_c^{1/2} (\omega \tau_0)^n}.
\]

The above consideration applies for low frequencies, where the center of mass of the DW explores many valleys during one period \( 2\pi/\omega \). For \( \omega \) larger than some frequency \( \omega_c \), the center of mass of the DW stays in one valley. The characteristic frequency \( \omega_c \) can be estimated from the condition that the center of mass of the DW remains within one well during period \( 2\pi/\omega \) with the probability 1. This yields \( \omega_c \approx \tau_0^{-1} \).

For frequencies \( \omega > \omega_c \), the DW is captured in one valley, the potential of which we approximate by

\[
U(z) = \frac{1}{2} \tau_1 z^2 + \int_0^z dz' g(z'),
\]

where the 1st term describes the average shape of the well, and the 2nd term describes the fluctuations due to the residual part of the random force \( g \). Neglecting the second term in Eq. (18) yields Debye-response

\[
\chi''(\omega) \propto \omega \tau_1/(1 + (\omega \tau_1)^2).
\]

The 2nd term in Eq. (18) results in distribution of relaxation times \( \Psi(\tau) \) instead of the single relaxation time \( \tau_1 \). Expecting the exponentially distributed barriers on all scales, the distribution of relaxation times is likely to obey a power-law \( \Psi(\tau) = n_1 \tau_1^{n_2} \tau^{1+n_1} \) with some exponent \( n_1 \) which is not necessarily equal to \( n \). As a result the Debye response changes to

\[
\chi''(\omega) \propto (\omega \tau_1)^{n_1}.
\]
To describe $\chi''(\omega)$ for $\omega \leq \omega_c$ we propose the following scaling ansatz

$$\chi''(\omega) = (\omega \tau_0)^{n_2} F(L_\omega/\sigma_0). \quad (21)$$

$F(x)$ is a scaling function and $L_\omega$ is the distance in $z$ direction explored by the DW during one period of the AC field. The distance $L(\tau_m)$ explored by the DW during the time $\tau_m$ can be estimated from the condition

$$\frac{L(\tau_m)}{\sigma_0} \int_{\tau_m}^{\infty} \Psi(\tau)d\tau = 1, \quad (22)$$

which means that the DW at most once is captured by the deepest well with the waiting time larger than $\tau_m$. For $\tau_m$ equal to the period of AC field, Eq. (22) gives $L_\omega \equiv L(\tau_m \simeq 1/\omega) = \sigma_0/(\omega \tau_0)^{n_2}$. Substituting the latter into Eq. (24) we obtain

$$\chi''(\omega) = (\omega \tau_1)^{n_2} F(1/(\omega \tau_0)^{n_2}). \quad (23)$$

According to Eqs. (17) and (28), the scaling function $F(x)$ must behave for small and large $x$ as

$$F(x) \simeq \begin{cases} x^{n_1}, & x \ll 1, \\ x^{1+n_1/n}, & x \gg 1. \end{cases} \quad (24)$$

The fit of the experimental data of Kleemann et al.\textsuperscript{23} using the scaling function

$$\chi''(\omega) = \omega^{-n} \left[ 1 + \frac{1}{2\kappa - 1} \left( \frac{\omega}{\omega_c} \right)^{n_2} \right]^{2n}. \quad (25)$$

is shown in Fig. 2.

In contrast to the imaginary part, the real part of the susceptibility behaves as a power of $\ln \omega$. To explain this behavior, we suggest that the intrawell dynamics of DW is a superposition of center of mass motion and relaxational motion of internal modes. Indeed, in the pinned phase $\omega > \omega_c$, the DW segments can also jump between metastable states with close energies, which gives rise to additional dissipation. The relaxation contribution of internal modes to the susceptibility reads\textsuperscript{20,21}

$$\chi' \propto (\ln(1/\omega \tau_0))^{2/\theta}, \quad (26)$$

$$\chi'' \propto (\ln(1/\omega \tau_0))^{2/\theta-1}. \quad (27)$$

We briefly remind the derivation of Eqs. (26) and (27). As suggested in Refs.\textsuperscript{21} and \textsuperscript{24}, one can treat the pinned DW as an hierarchical ensemble of noninteracting two-level systems (TLS), each is a DW segment of linear size $L$. The separation between the two configurations of the given TLS is $w(L) \propto L^\zeta$, and the energy difference is $\Delta E$. Transitions caused by thermal activation may occur only in TLSs with the energy difference $\Delta E \leq T$. The corresponding rate of transitions is given by $\tau^{-1}(L) = \tau_0^{-1} \exp(-E_B(L)/T)$, where $E_B(L)$ is the energy barrier. In the presence of AC field, these transitions lead to power dissipation\textsuperscript{25}

$$Q(L) \propto \frac{(\delta E(L))^2}{4T \cosh^2(\Delta E/2T)} \frac{\omega^2 \tau(L)}{1 + \omega^2 \tau^2(L)}. \quad (28)$$

Due to the exponential increase of $\tau$ with $L$, the main contribution to the integral comes from the length scale $L_\omega$ given by the condition $\omega \tau(L_\omega) \sim 1$. On the other hand, the power dissipation is related to the imaginary part of the complex susceptibility\textsuperscript{26}

$$\overline{Q} = \frac{1}{2} \omega \chi'' h_0^2. \quad (29)$$

Equating (28) and (29) we arrive at Eq. (20) for the imaginary part of the susceptibility. The corresponding real part can be restored from the imaginary part with the help of the Kramers-Kronig relation in the form of $\pi/2$ rule\textsuperscript{27}

$$\chi''(\omega) = -\frac{\pi}{2} \frac{d\chi'(\omega)}{d\ln \omega}. \quad (30)$$
The real part of the susceptibility, which corresponds to Eq. (21), decreases as a power-law and can be hidden by the slower decreasing logarithmic contribution (25). In contrast, the power-law contribution (20) dominates the behavior of the imaginary part of the susceptibility, so that the logarithmic contribution (26) is irrelevant.

Let us finally discuss in brief the nonlinear response associated with the DW motion. We define the nonlinear susceptibilities as $\chi_m' = \chi_m' - i\chi_m''$ with

$$\chi_m' = \frac{1}{\pi h_0} \int_0^{2\pi} P(t) \cos(m\omega t) d(\omega t), \quad (31)$$

$$\chi_m'' = \frac{1}{\pi h_0} \int_0^{2\pi} P(t) \sin(m\omega t) d(\omega t). \quad (32)$$

The linear susceptibility corresponds to $\chi_1$. To calculate the nonlinear susceptibility in the sliding regime, i.e. at very low frequencies $\omega < \omega^*$, we integrate Eq. (5) with $\langle v(t) \rangle$ given by Eq. (6) and $h(t) = h_0 \cos(\omega t)$, and then substitute $P(t)$ in Eqs. (31) and (32). As a result we obtain $\chi_m' = 0$ and $\chi_m'' \propto 1/\omega$ for odd $m$. Note that in the linear case ($m = 1$) this gives a conduction-type susceptibility $\chi(\omega) = 1/\omega \tau_0$. The computation of the nonlinear susceptibility in the stochastic regime is a more difficult task which is left for further investigations.

In conclusion, we have considered the dielectric response of ferroelectric DWs below the paraelectric-ferroelectric phase transition temperature. As a function of the frequency of the external electric AC-field there are three regimes in the behaviour of the complex susceptibility: the sliding, the stochastic, and the pinned regimes. The response in the sliding regime, which corresponds to very low frequencies, is due to the creep-like motion of DWs. The response in the stochastic regime is related to jumps of the DW as a whole, and can be described using the concept of waiting time distributions. The response in the pinned regime at high-frequencies is expected to be due to superposition of center of mass motion and relaxational motion of internal modes. The existence of all three regimes for the dielectric response was recently observed in SrTi$_{18}$O$_{30}$.

The support from the Deutsche Forschungsgemeinschaft (SFB 418) is gratefully acknowledged.

1. V. Mueller, Y. Shchur, H. Beige, A. Fuith, and S. Stepanow, Europhys. Lett. 57, 107 (2002).
2. V. Mueller, Y. Shchur, H. Beige, S. Mattauch, J. Glinne, and G. Heger, Phys. Rev. B 65, 134102 (2002).
3. Y. Park, Solid State Commun. 113, 379 (2000).
4. W. Kleemann, J. Dec, S. Miga, Th. Woike, and R. Pankrath, Phys. Rev. B 65, 220101(R) (2002).
5. X. Chen et al., Phys. Rev. Lett. 89, 137203 (2002).
6. T. Nattermann, A. Pokrovsky, and V.M. Vinokur, Phys. Rev. Lett. 87, 197005 (2001).
7. A.K. Jonscher, Nature, 267, 673 (1977).
8. A.K. Jonscher, Dielectric Relaxation in Solids (Chelsea Dielectric Press, London, 1983).
9. W. Kleemann, J. Dec, and R. Pankrath, Ferroelectrics 286, 21 (2003); 291, 75 (2003).
10. D. S. Fisher, Phys. Rev. Lett. 56, 1964 (1986).
11. T. Nattermann, S. Stepanow, L.-H. Tang, and H. Leschhorn, J. Phys. II 2, 1483 (1992).
12. O. Narayan and D. S. Fisher, Phys. Rev. B 48, 7030 (1993).
13. P. Chauve, P. Le Doussal, and K. J. Wiese, Phys. Rev. Lett. 86, 1785 (2001).
14. Y.N. Huang et al., Phys. Rev. B 55, 16159 (1997).
15. M. V. Feigel’man and V. M. Vinokur, J. Phys. France 49, 1731 (1988).
16. H. Scher and M. Lax, Phys. Rev. B 7, 4491 (1973).
17. V. M. Vinokur, M. C. Marchetti, and L.-W. Chen, Phys. Rev. Lett. 77, 1845 (1996).
18. B. Drossel and M. Kardar, Phys. Rev. E 52, 4841 (1995).
19. A. A. Fedorenko and S. Stepanow, Phys. Rev. E 68, 056115 (2003).
20. T. Nattermann, Y. Shapir, and I. Vilfan, Phys. Rev. B 42, 8577 (1990).
21. L.B. Ioffe and V.M. Vinokur, J. Phys. C 20, 6149 (1987).
22. E.W. Montroll and G.H. Weiss J. Math. Phys. 6, 167 (1965).
23. L.D. Landau and E.M. Lifshitz, Statistical Physics (Moscow, 1976).
24. E. Pytte and Y. Imry, Phys. Rev. B 35, 1465 (1987).
25. S. Brazovskii and T. Nattermann, e-print cond-mat/0312375.
26. J. Dec, W. Kleemann, and M. Itoh, Ferroelectrics 298, 163 (2004).