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Nonharmonic polarization study of ferroelectric ceramics in alternating field

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The aim of this note is to study the problem of polarization in ferroelectric ceramics in the external alternating electric field at intermediate damping is considered. For the model of constant dipoles with fixed masses and electronic polarization equaled to zero, is presented the analytical expression for polarization by using a perturbation approach.

1. Introduction

Materials made of ferroelectric ceramics are widely used in different devices such as transducers, resonators, actuators, acoustic radars, motors and so on. This reason justifies significant efforts given to investigation of properties for those materials. A nonlinear regime is usually realized in a process of exploitation of the devices mentioned above. That is why the problem of research of nonlinear behaviour of ferroelectrics is currently very important. The study of this problem will allow to understand deeper the mechanisms of different processes in ferroelectrics and describe to more precisely the principles of functioning of devices and experimental data [1, 2]. Nonlinear behaviour of parameters of ferroelectrics under the influence of external factors such as temperature, electromagnetic fields, mechanical stresses and their combinations is of great importance [3-7].

To describe a repolarization of ferroelectrics we use the model of constant dipoles with fixed masses [8]. In the case of linear polarization and damping approximations this model was applied to the investigation of the repolarization of ferroelectric ceramics in alternating electric field [9]. It was shown that when damping is small, the process of repolarization includes the oscillations of two different types. The first of them describes an influence of external field. The second one is determined by the properties of material. At large damping the action of external field is practically negligible. To generalize this model the Landau thermodynamical approach was applied [10]. It allowed to obtain the equation for repolarization of ferroelectrics in nonlinear polarization approximation. This new model has been used for the study of nonharmonic polarization of ferroelectric ceramics in alternating field when damping is large [11]. In this model not only linear but a square dependence of polarization on amplitude of external field takes a place. Besides, in the first on coefficient of nonharmonicity approximation, there are two type of oscillations of ions in the crystal lattice. One with a frequency of external field and the other with a half of this frequency.

An influence of nonharmonic effects on polarization of ferroelectric ceramics in an external alternating electric field at intermediate damping is considered below. The nonlinear inhomogeneous equation for polarization P has the form

$$\frac{d^2\Delta P}{dt^2}+\gamma\frac{d\Delta P}{dt}+g\Delta P+\alpha\left(\Delta P\right)^2=aE\exp\left(i\omega t\right), \tag{1.1}$$

where $\Delta P = P - P_s$, P_s is the spontaneous polarization, γ is the damping coefficient, $a = Nq^2/m$, q is the electric charge of the oscillator, m is the mass of oscillator, N is the number of dipoles, the coefficient g characterizes harmonic oscillations, α is the nonharmonicity coefficient.

At large damping the main influence on repolarization is caused by the properties of material. The influence of external field is not essential. At small damping the main influence on repolarization is caused by external field. But there is a situation when the influence of both these factors is comparable. One can say that for such a case the damping coefficient takes an intermediate value. For Eq. (1.1) this condition has the form $\gamma^2 = 4g^2$.

If $\alpha = 0$, the general solution of Eq. (1.1) at intermediate damping may be written as follows

$$P=P_s+C_1t\exp\exp\left(-\gamma t/2\right)+C_2\exp\left(-\gamma t/2\right)+\frac{4aE\exp\left(i\omega t\right)}{\left(\gamma+2i\omega\right)^2}, \tag{1.2}$$

where C_1 and C_2 are constants of integration determined by the initial conditions [12]. Let us compare this solution with the appropriate solution at large damping [11]. If we consider a part of the solution that is determined by the properties of material (general solution of homogeneous equation), it is clear that at intermediate damping it decreases lower than for large damping due to a factor t. At large damping a term that describes the influence of external field vanishes. Now there is a non-vanishing contribution to polarization. It describes the continuous action of external field.

2. Calculation of polarization

It is known that due to various factors such as defects of structure, domain walls, mechanical stresses, variation of temperature and so on the oscillations of ions in crystal lattice have an asymmetric character. The influence of these factors are accounted for in Eq. (1.1) by the term $\alpha\,(\Delta P)^2$. This term describes the effect of nonharmonicity. Let us construct the expression for P at intermediate damping by using the perturbation technique [13].

A solution of Eq. (1.1) is represented in the form of a series

$$\Delta P = \Delta P_0 + \alpha u_1 + \alpha^2 u_2 + \ldots, \tag{2.1}$$

where u_i $(i = 1, 2, \ldots)$ are unknown functions of t and ΔP_0 is a general solution of Eq. (1.1) for $\alpha = 0$. It is determined by Eq. (1.2). Let us write it down with its real and imaginary parts shown explicitly

$$\begin{aligned}\Delta P_0 &= C_1 t \exp(-\gamma t/2) + C_2 \exp(-\gamma t/2)\\ &+ \frac{4aE\left(\gamma^2 - 4\omega^2\right)\cos(\omega t) + 8aE\gamma\omega\sin(\omega t)}{\gamma^4 - 4\gamma^2\omega^2 + 16\omega^4}\\ &+ \frac{4aE\left(\gamma^2 - 4\omega^2\right)\sin(\omega t) - 8aE\gamma\omega\cos(\omega t)}{\gamma^4 - 4\gamma^2\omega^2 + 16\omega^4}.\end{aligned} \tag{2.2}$$

To calculate the first order term u_1, with α in the power of 1, let us substitute Eq. (2.1) into Eq. (1.1). For u_1 the next differential equation is obtained

$$\frac{d^2 u_1}{dt^2} + \gamma\frac{du_1}{dt} + g u_1 = -\Delta P_0^2. \tag{2.3}$$

The general solution $u_{1,a}$ of homogeneous equation corresponding to Eq. (2.3) may be found by analogy with solution (1.2). It has the form

$$u_{1,a} = C_3 t \exp(-\gamma t/2) + C_4 \exp(-\gamma t/2), \tag{2.4}$$

where C_3 and C_4 are constants of integration determined by the initial conditions.

Equation (2.4) determines the fundamental system of functions for homogeneous equation corresponding to Eq. (2.3). By using this system the partial solution $u_{1,b}$ of Eq. (2.3) may be written as follows [12]

$$u_{1,b} = \exp(-\gamma t/2) \int \frac{aEt \exp[(i\omega - \gamma/2)\,t]}{W} dt - t \exp(-\gamma t/2) \int \frac{aE \exp[(i\omega - \gamma/2)\,t]}{W} dt, \qquad (2.5)$$

where $W = -\exp(-\gamma t)$ is the Wronskian of fundamental system of functions for homogeneous equation. After very cumbersome calculations, Eq. (2.5) can written in the form

$$\begin{aligned}
u_{1,b} = &-\left(4\gamma^2 t^2 + 42\gamma t + 96\right) \frac{C_1^2 \exp(-\gamma t)}{\gamma^4} \\
&- \frac{4C_2^2 \exp(-\gamma t)}{\gamma^2} - (4\gamma t + 6)\frac{2C_1 C_2 \exp(-\gamma t)}{\gamma^3} \\
&- \frac{16a^2E^2\gamma^4 - 192a^2E^2\gamma^2\omega^2 + 256a^2E^2\omega^4}{\left(\gamma^4 - 4\gamma^2\omega^2 + 16\omega^4\right)^2} \\
&\cdot \left[\frac{32\gamma\omega}{\left(\gamma^2 + 16\omega^2\right)^2}\sin(2\omega t) + \frac{4\left(\gamma^2 - 16\omega^2\right)}{\left(\gamma^2 + 16\omega^2\right)^2}\cos(2\omega t)\right] \\
&+ \frac{128a^2E^2\gamma\omega\left(\gamma^2 - 4\omega^2\right)}{\gamma^4 - 4\gamma^2\omega^2 + 16\omega^4} \\
&\cdot \left[\frac{16\gamma\omega}{\left(\gamma^2 + 16\omega^2\right)^2}\cos(2\omega t) - \frac{2\left(\gamma^2 - 16\omega^2\right)}{\left(\gamma^2 + 16\omega^2\right)^2}\sin(2\omega t)\right] \\
&- \frac{8aE\left(\gamma^2 - 4\omega^2\right)C_1}{\gamma^4 - 4\gamma^2\omega^2 + 16\omega^4}\left[\frac{3t}{\omega^2}\cos(\omega t) + \frac{2}{\omega^3}\sin(\omega t)\right]\exp(-\gamma t/2) \\
&+ \frac{16aE\gamma\omega C_1}{\gamma^4 - 4\gamma^2\omega^2 + 16\omega^4}\left[\frac{2}{\omega^3}\cos(\omega t) + \frac{t}{\omega^2}\sin(\omega t)\right]\exp(-\gamma t/2) \\
&+ \frac{8aE\left(\gamma^2 - 4\omega^2\right)C_2}{\left(\gamma^4 - 4\gamma^2\omega^2 + 16\omega^4\right)\omega^2}\cos(\omega t)\exp(-\gamma t/2) \\
&+ \frac{16aE\gamma C_2}{\left(\gamma^4 - 4\gamma^2\omega^2 + 16\omega^4\right)\omega}\sin(\omega t)\exp(-\gamma t/2) \\
&+ i\frac{8aE\left(\gamma^2 - 4\omega^2\right)C_1}{\gamma^4 - 4\gamma^2\omega^2 + 16\omega^4}\left[\frac{2}{\omega^3}\cos(\omega t) + \frac{t}{\omega^2}\sin(\omega t)\right]\exp(-\gamma t/2) \\
&+ i\frac{16aE\gamma\omega C_1}{\gamma^4 - 4\gamma^2\omega^2 + 16\omega^4}\left[\frac{2}{\omega^3}\sin(\omega t) + \frac{3t}{\omega^2}\cos(\omega t)\right]\exp(-\gamma t/2) + \cdots
\end{aligned} \qquad (2.6)$$

$$\cdots + i\frac{8aE\left(\gamma^2-4\omega^2\right)C_2}{\left(\gamma^4-4\gamma^2\omega^2+16\omega^4\right)\omega^2}\sin(\omega t)\exp(-\gamma t/2)$$
$$- i\frac{16aE\gamma C_2}{\left(\gamma^4-4\gamma^2\omega^2+16\omega^4\right)\omega}\cos(\omega t)\exp(-\gamma t/2)$$
$$+ i\frac{64a^2E^2\gamma\omega\left(\gamma^2-4\omega^2\right)}{\left(\gamma^4-4\gamma^2\omega^2+16\omega^4\right)\omega^4}$$
$$\cdot\left[\frac{4\left(\gamma^2-16\omega^2\right)}{\left(\gamma^2+16\omega^2\right)^2}\cos(2\omega t)+\frac{32\gamma\omega}{\left(\gamma^2+16\omega^2\right)^2}\sin(2\omega t)\right] \qquad (2.6)\ \text{[cont.]}$$
$$+ i\frac{96a^2E^2\gamma^2\omega^2}{\left(\gamma^4-4\gamma^2\omega^2+16\omega^4\right)^2}$$
$$\cdot\left[\frac{2\left(\gamma^2-16\omega^2\right)}{\left(\gamma^2+16\omega^2\right)^2}\sin(2\omega t)-\frac{16\gamma\omega}{\left(\gamma^2+16\omega^2\right)^2}\cos(2\omega t)\right].$$

Finally, up to the first order terms in α, the polarization of ferroelectrics in external alternating electric field at intermediate damping is

$$P = P_s + \Delta P_0 + \alpha\left(u_{1,a}+u_{1,b}\right). \qquad (2.7)$$

In Fig. 1 the behaviour of the real component of relative polarization P/P_s determined by Eq. (2.7) is shown. Calculations have been carried out for material like barium titanate [9].

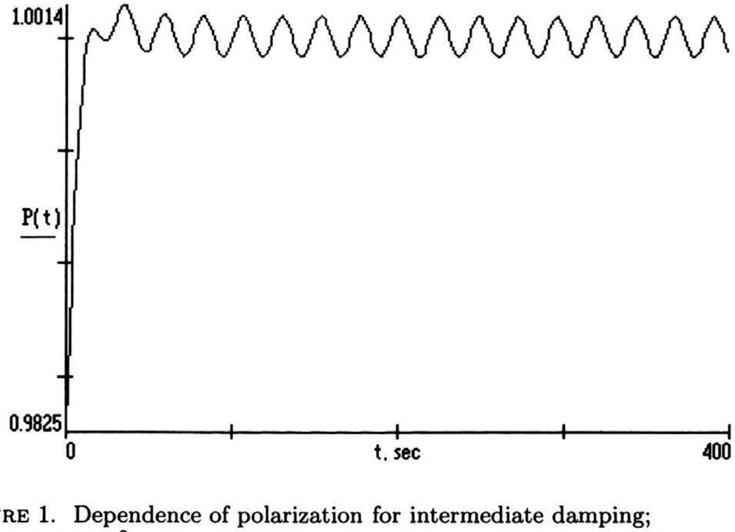

FIGURE 1. Dependence of polarization for intermediate damping; $P_s = 18\,\mu\text{C/cm}^2$, $E = 20\,\text{kV/cm}$, $a = 2$, $\omega = 50\,\text{Hz}$, $\gamma = 0.2$, $\alpha = 0.05$, $C_1 = C_3 = 0.01$, $C_2 = C_4 = 0$.

From Fig. 1 one can see that after some time the oscillations of ions in crystal will be determined by external field. As a result, the polarization will oscillate with some frequency. This frequency will be determined by the frequency of external field but not equal to it. The amplitude of oscillations is not high because damping is not small enough. In the linear theory the amplitude of oscillation increases when damping decreases [9]. Now, it is not so obvious. Not only damping but nonlinearity also influence on the repolarization. Nevertheless, it follows from calculations that the tendency mentioned above occurs. For smaller γ, the amplitude is higher if other parameters don't change. At the beginning of the repolarization the influence of damping is more essential. Some distortion of the shape of the curve in Fig. 1 indicates that when t is small, the process of repolarization is not stable. But after some time the influence of external field will become important. At large damping the process of repolarization is determined by the properties of material. In this case the interaction between two neighboring ions in crystal are so large that for any physically reasonable amplitudes of external field the oscillating component of polarization vanishes in time.

3. Conclusions

The specific feature of repolarization of ferroelectric ceramics in external alternating electric field at intermediate damping consists in the presence of non-vanishing oscillating component. For the first order terms in α approximation at large damping there are oscillations with two frequencies (with a frequency of external field and a half frequency). As damping large these oscillations vanish. At intermediate damping for the same approximation there are oscillations of polarization with a frequency of external field and a twice frequency.

To describe ferroelectric ceramics a model of constant dipoles with fixed masses is used. In this model every dipole was considered as a mechanical oscillator. If damping is large, a vanishing of oscillations indicates their decoherence. Hence, at intermediate damping some coherence appears during mechanical oscillators. One can suppose that at small damping this coherence will be higher.

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