On Static Dielectric Response of Microcomposites of the Type: Ferroplastics - Dielectrics

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Abstract

We describe the static dielectric response of ferroelastic-dielectric microcomposites. Its dependence on temperature, pressure and concentration is considered for smaller concentrations and for temperatures above the critical temperature. We have found a qualitative agreement with existing experimental data for $Al_2(WO_4)_3$. 
1 Introduction

The dielectric response of ferroelectric-dielectric type microcomposites was studied recently [1] - [4]. When a coupling of the elastic strain to the electric polarisation is present, then the dielectric response of such a ferroelastic material may be studied, see in [5] - [6]. Also the dielectric response of ferroelastic-dielectric type microcomposites may be studied. Response of minerals to changing hydrostatic pressure \( p \) and temperature \( T \) is also interesting property of these materials. For example materials of the perovskite type (\( \text{LaAlO}_3, \text{CaAlO}_3, \text{SrAlO}_3, \text{BaTiO}_3, \text{PbNiO}_3, \text{Pb(Zr,Ti)}_3\text{O}_3, ... \)) undergo a phase transition from a cubic phase to a phase with lower symmetry at some critical temperature. While a study of their elastic properties is usually done in literature, we will consider here the dielectric response of such materials which is due to coupling between the elastic strain tensor and the polarisation. Mechanical analysis is usually done at low frequencies (0.1Hz - 10 Hz) but also measurements at higher frequencies are done. Ferroelastic domain wall structure, twinning and other similar phenomena are studied, see in [7] and [8]. Elastic response function (compliance) shows in Cole-Cole diagrams circular and non-circular behaviour of these materials in their crystalline and ceramic form [9]. In the second case multirelaxation phenomena exist in these materials under some conditions. Under higher electric and mechanical loading nonlinear behaviour is exhibited by ferroelectric and ferroelastic ceramics [10]. While at higher temperatures there is present no ferroelastic phase, coupling of the elastic strain and electric polarisation does exist at these temperatures. So dielectric response depends then on mechanical forces acting on the microcomposite of the ferroelastic-dielectric type. This response enables us to study properties of these microcomposites using dielectric measurements for microcomposites under mechanical forces. It is known that constraint due to neighbouring material lead below the critical temperature for transition from paraelastic to ferroelastic phase and due to shape-change to several forms of the low-temperature phase [11]. As it is noted by these authors perhaps only in very small grains there exists a single variant of this form. A strain of \( 10^{-4} \) in a grain of size 10 \( \mu \text{m} \) (typical values) is an order of magnitude too large to be accommodated in a small displacements of atoms nearby and at surface. An external mechanical field may move walls between different forms of the low-temperature phase. We assume in our paper that there are small mechanical fields of the order \( 10^{-3} \) and that particles are with heir diameter of the order of 1\( \mu \text{m} \). They are microcomposites. Moreover in large concentrations of the dielectric hard material the clusters of the ferroelastic phase are still small. Thus we will consider this case in this paper only, at the percolation transition concentrations there are very large clusters of ferroelectric and dielectric material, it is not known today how they are responding to the external mechanical fields. InI ferroelastic phase long-range anisotropic forces may appear [12]. In our paper we discuss properties of the microcomposite above the critical temperature, thus these long-range forces can be neglected, they may be present
The aim of this paper is to study the static dielectric response of microcomposites of the type: ferrolastics - dielectrics. In the second chapter a model of ferroelastic-dielectric microcomposites is presented. We study the dielectric response of ferroelastic particles. Ferroelastic particles and their dielectric response to static electric field in the high temperature limit and as a function of hydrostatic pressure is studied in the next chapter. Effective Medium Approximation (EMA) is generally formulated for dielectric response of microcomposites of the type: ferroelastics - dielectrics, and then studied in low concentration limit of the ferroelastic material and in low concentration limit of the dielectric material. Microcomposites of the type: ferroelastics - dielectrics in the percolation region, i.e. for higher concentrations of the particles but not close to the limiting case $x = 1$ should be finally studied however they are from the point of view of mechanical forces more complicated than the limiting cases and will not be studied here. In the last chapter we summarize our results as concerning temperature, hydrostatic pressure and concentration dependence of the dielectric response of microcomposites of the type: ferroelastics-dielectrics. A discussion of these results and their comparison with experiments.

2 Model of Ferroelastic-Dielectric Microcomposites

Microcomposites are composed of small particles. They have interesting properties especially when they consist of two or more materials with different properties. One of such examples are ferroelectric-dielectric microcomposites [1] - [4]. Ferroelectric properties of particles may appear in them due to presence of the polarisation as a primary order parameter. An interesting possibility is to consider materials in which ferroelectricity is induced as a secondary order parameter. Primary order parameter may be the corresponding component of the elastic strain tensor (or a combination of components of the elastic strain tensor). The ferroelectric state is present in such a material due to a coupling between the elastic strain tensor and the polarisation. Then we have two types of particles in the microcomposite: ferroelastic and dielectric. Changing the concentration of these two types of particles in the microcomposite the response to external fields changes. This holds for dielectric response and for other type of responses. We are calculating in this paper the dielectric response of such apredictions microcomposite. For simplicity we consider all particles in microcomposite of the same diameter $d$. When changing the concentration of the ferroelastic particles (increasing it) an infinite ferroelastic cluster of ferroelastic particles may form. The concentration at which this cluster forms is the percolation transition concentration. Then increasing further the concentration...
the ferroelastic material becomes the matrix material, and at the concentration $x = 1$ one the ferroelastic material only is formed.

When the concentration of particles changes, the dielectric response of the microcomposite changes. Thus the interplay between the percolation transition and the ferroelastic phase transition appears. In real materials there exists distribution of diameters of particles and shapes of particles. In our model we neglect this distribution for simplicity, and consider only spheres.

3 Ferroelastic Particles: Dielectric Response

The dielectric response of a ferroelastic particle will be studied using a Landau free energy expansion. We will assume cubic symmetry of the ferroelastic material for simplicity. This symmetry corresponds to high-temperature symmetry of materials mentioned in the Introduction. For materials of other symmetry the approach is similar. In ferroelastic materials the primary order parameter is an elastic strain tensor. Secondary order parameter is polarisation. While the elastic strain tensor is coupled to the external mechanical fields (hydrostatic pressure, uniaxial stress, shear stress), the secondary order parameter is coupled to an external electric field. We will use in our calculation time and space dependent external fields in general. However for microcomposites the quasi-static approximation for dielectric response is a convenient approximation because the wavelength of the electric field is usually much larger than the diameter of the particle. The space dependence of the external electric field may be neglected.

To find the free energy expansion we have to find first invariants of the primary order parameter and of the secondary order parameter, and of primary and secondary order parameter coupled. In cubic materials there are the following invariants for the primary order parameter $\epsilon_{i,j}$, where $i, j = 1, 2, 3$ denotes axis of the cubic material (Einstein sum rule is used):

$$\epsilon_{i,j} \epsilon_{j,i} = 3\epsilon^2 + 6\phi^2$$

which is of the second order in the elastic strain tensor and:

$$\epsilon_{i,j} \epsilon_{j,k} \epsilon_{k,i} = 3\epsilon(\epsilon^2 + 2\phi^2) + 6(2\epsilon\phi + \phi^2)\phi.$$  

Here we denoted on-diagonal terms as $\epsilon$ and off-diagonal terms of the elastic strain tensor as $\phi$. This term is of the third order in the elastic strain tensor. Because the coupling between the primary order parameter (elastic strain tensor) and the secondary order parameter - electric polarisation is of the first order in the elastic strain tensor and of the second order in the polarisation, and because of the fourth order in the electric polarisation order parameter (or of the sixth order for the first order phase transitions near the second order one for the ferroelectric materials), we will consider in the free energy expansion only these two invariants for the elastic strain tensor described above, (1) and (2). Thus
in the corresponding to polarisation part of the free energy expansion there will be second order term, fourth order terms and sixth order terms in general.

The coupling between the elastic strain tensor $\epsilon_{j,k}$ and the polarisation vector $P_i$ has the form:

$$\epsilon_{i,j} P_i P_j = \epsilon P^2.$$  \hspace{1cm} (3)

The polarisation vector $P_i$ is assumed to have the nonzero component only in the x-direction, we assume that external electric field will be applied in this direction. Then the free energy $F$ expansion has the form:

$$F = \int dV \left[ B_2 (\epsilon^2 + 2\phi^2) + C_3 (\epsilon (\epsilon^2 + 2\phi^2) + 2(2\epsilon\phi + \phi^2)\phi) + \Gamma P^2 \epsilon + \frac{\alpha}{2} P^2 + \frac{\beta}{4} P^4 + \frac{\gamma}{6} P^6 - E_P - \epsilon_{i,j} \sigma_{j,i} \right].$$ \hspace{1cm} (4)

The last term in the free energy expansion depends on the stress tensor. This tensor may be hydrostatic pressure $p$, $\sigma_{j,i} = \delta_{j,i} p$, uniaxial stress $\sigma_{x,x} = \sigma$ or shear stress $\sigma_{x,y}$. The constants in the free energy expansion (4) are positive and temperature independent, with the exception of the constant $B$, for which $B = B_0(T - T_c)$ where $B_0$ is positive, $T_c$ is a critical temperature for the transition from the paraelastic to the ferroelastic phase.

Depending on the field applied (electric, mechanical) we calculate the response of the material described by the free energy expansion (4). We will assume that the surface charge is compensated in the case of polarized particles. The expansion constants in (4) may be in fact temperature and hydrostatic pressure dependent. We will not consider this dependence as we mentioned above with the exception of the constant $B$. However it may happen that in some materials these constants are weakly temperature and hydrostatic pressure dependent. This dependence should be taken into account when there are reasons of the experimental origin.

Let us now consider the high-temperature paraelastic phase. In this case the free energy expansion from (4) takes the form in which second order terms in the polarisation are taken into account and the first order terms in the elastic strain tensor are taken into account. Note that the second order in the electric polarisation is corresponding to the first order in the elastic strain tensor. However the coupling term is of the fourth order in polarisation, so also the second order term in elastic strain tensor is taken into account. The fourth order term in polarisation, with the constant beta, is not taken into account because at high temperatures this term does not play any essential role. Then the free energy $F$ expansion has the form:

$$F = \int dV \left[ \frac{B}{2} (\epsilon^2 + 2\phi^2) + \Gamma P^2 \epsilon + \frac{\alpha}{2} P^2 - E_P - \epsilon_{i,j} \sigma_{j,i} \right].$$ \hspace{1cm} (5)

As we can see from (5) we still may consider different situations as concerning the influence of the external electric and mechanical fields. Let us first consider
the static case of the electric field and of the hydrostatic pressure.

4 Dielectric Response of Ferroelastic Particles: Static High Temperature Limit and Hydrostatic Pressure Dependence

In this case the free energy expansion has the form:

\[ F = \int dV \left[ \frac{B}{2} (\epsilon^2 + 2\phi^2) + \Gamma P^2 \epsilon + \frac{\alpha}{2} P^2 - E.P - 3\epsilon p \right]. \] (6)

Here \( p \) is the hydrostatic pressure, it is the same in all directions. The Lagrange-Euler equations for the most stable state at a given electric field and hydrostatic pressure have the form:

\[ B\epsilon - 3p + \Gamma P^2 = 0 \] (7)
\[ \alpha P + 2\Gamma \epsilon P - E = 0 \]

Neglecting higher harmonics in the hard dielectrics we obtain that the dielectric permittivity has the form:

\[ \epsilon_f = \frac{1}{\alpha^*} \equiv \frac{1}{\alpha} (1 + \frac{6\Gamma p}{\alpha B}) \] (8)

As we can see from the equation (8) the dielectric permittivity increases with increasing hydrostatic pressure for \( \Gamma \) positive, and it decreases with decreasing hydrostatic pressure for \( \Gamma \) positive. We assume for simplicity that \( \alpha \) is temperature independent, and that \( B \) is temperature dependent, \( B = B_0(T - T_0) \leq 0 \). Thus the dielectric permittivity of the ferroelastic material is temperature dependent for nonzero hydrostatic pressure. With increasing temperature decreases and the dielectric permittivity tends to the zero hydrostatic pressure value. Note that for negative \( \Gamma \) there exists a critical hydrostatic pressure \( p_c = \frac{\alpha B}{\alpha \Gamma} \) at which the permittivity becomes zero. For hydrostatic pressures higher that the critical the permittivity is negative. We will not consider the cases in which the hydrostatic pressure is higher than critical for negative \( \Gamma \).

5 Effective Medium Approximation - General Formulation for Dielectric Response of Composites of the Type: Ferroelastics - Dielectrics

The effective medium approximation is valid for the whole interval of concentrations \( 0 \leq x \leq 1 \). The effective permittivity \( \epsilon_{eff} \) may be obtained from [1]:
\[ x \frac{\epsilon_d - \epsilon_{\text{eff}}}{\epsilon_d + 2\epsilon_{\text{eff}}} + (1 - x) \frac{\epsilon_f - \epsilon_{\text{eff}}}{\epsilon_f + 2\epsilon_{\text{eff}}} = 0 \] (9)

This approximation is based on the response of a spherical particle to the whole effective microcomposite. Both components are taken into account symmetrically. The percolation transition occurs at \( x = \frac{1}{3} \) for the F component. For the permittivity of the hard material we take the constant dielectric permittivity \( \epsilon_d \) and \( \frac{1}{\alpha}(1 + \frac{6\Gamma p}{\alpha B}) \) for the permittivity of the ferroelastic material. The effective permittivity calculated from (9) will describe response of the microcomposite on the electric field and on the hydrostatic pressure. Let us now consider two limiting case: the limit of small concentration of the ferroelastic material, and the limit of small concentration of the hard dielectric material.

6 Low Concentration of the Ferroelastic Material

For low concentration of ferroelastic particles in the hard dielectric material we may calculate the dielectric response of the microcomposite from (9). In this case a small parameter is \( 1 - x \). The effective dielectric permittivity \( \epsilon_{\text{eff}} \) has the form:

\[ \epsilon_{\text{eff}} = \epsilon_d + 3(1 - x)\epsilon_d \frac{\frac{1}{\alpha} - \epsilon_d}{\frac{1}{\alpha} + 2\epsilon_d} \] (10)

Substituting for the ferroelastic permittivity from (8) we obtain hydrostatic pressure, temperature and concentration dependence of the effective dielectric permittivity in the limit \( x \) near the value 1:

\[ \epsilon_{\text{eff}} = \epsilon_d + 3(1 - x)\epsilon_d \left( \frac{\frac{1}{\alpha} + \frac{6\Gamma p}{\alpha B}}{\frac{1}{\alpha} + 2\epsilon_d} \right) \] (11)

As we can see with increasing concentration of the ferroelastic component, decreasing \( x \), the dielectric response increases assuming that the permittivity of the hard material is lower than that of the ferroelastic material. If the permittivity of the hard material is higher than that of the ferroelastic material, then with increasing concentration of the ferroelastic component the effective dielectric permittivity decreases. Increasing temperature the effective dielectric response of the microcomposite is tending to that of the dielectric response of microcomposite with zero hydrostatic pressure. Increasing hydrostatic pressure to higher values the effective dielectric response becomes less hydrostatic pressure dependent.
7 Low Concentration of the Dielectric Material

For low concentration of dielectric particles in the ferroelastic matrix we may calculate the dielectric response of the microcomposite again from (9) in a similar way as in the preceding section. In this case we have \( x \) near the value 0. Thus a small parameter is now \( x \). The effective dielectric permitivity \( \epsilon_{eff} \) is in this case given as:

\[
\epsilon_{eff} = \epsilon_f + 3x\epsilon_f \frac{\epsilon_d - \epsilon_f}{\epsilon_d + 2\epsilon_f}
\]

(12)

Substituting for the ferroelastic permitivity from (8) we obtain hydrostatic pressure, temperature and concentration dependence of the effective dielectric permitivity in this limit as:

\[
\epsilon_{eff} = \frac{1}{\alpha}(1 + \frac{6\Gamma p}{\alpha B}) + 3x\frac{1}{\alpha}(1 + \frac{6\Gamma p}{\alpha B}), \frac{\epsilon_d - \frac{1}{\alpha}(1 + \frac{\epsilon_f}{\alpha B})}{\epsilon_d + 2\frac{1}{\alpha}(1 + \frac{\epsilon_f}{\alpha B})}
\]

(13)

8 Summary

We studied here the static dielectric response of microcomposites: ferroelastics-dielectrics. A model for such a microcomposite was formulated. Dielectric properties of ferroelastic particles were studied. Cubic symmetry of the material was assumed. This corresponds to most of the known ferroelastic materials at high temperatures. Materials with other symmetry may be studied in a similar way. We consider a coupling of the elastic strain tensor to the electric polarization. While the primary order parameter is coupled to external mechanical fields, the secondary order parameter is coupled to external electric field. For microcomposites a quasistatic approximation may be used for the dielectric response. We have found the free energy expansion using invariants of the primary order parameter, of the secondary order parameter, and of their mixed terms. We assumed that in the free energy expansion only the coefficient \( B \) of the second order of the primary order parameter is temperature dependent, and that other parameters are temperature independent. All of these coefficients are assumed to be hydrostatic pressure independent. Face charges are assumed to compensate the dipole moment in ferroelastic particles in which an electric dipol appears. We apply hydrostatic pressure on the microcomposite and consider high-temperature properties of such a microcomposite here. The low-temperature properties may be considered in the same way easily. The most important role in the free energy expansion play terms of the second order in the elastic strain tensor and of the second order in electric polarisation. The fourth order terms of the polarisation are small and we do not consider them.
Firstly we studied the dielectric response of the ferroelastic particles for high temperatures (temperatures above the transition temperature to the ferroelastic phase) and the hydrostatic pressure dependence. We have found the dielectric constant in the static limit here, which correspond to mechanical measurements from 0.1Hz to 10 Hz. For higher frequencies the dynamic dielectric constant may be calculated directly. It turns out that the dielectric response of a particle is depending on hydrostatic pressure: increasing hydrostatic pressure the response is larger (for the constant $\Gamma$ positive, which is the most usual case), its increase is linear in hydrostatic pressure. The nearer we are to the ferroelastic transition temperature the higher is the change in the dielectric response for the same change of hydrostatic pressure. At the transition temperature the dielectric response constant diverges in this static limit for nonzero hydrostatic pressures. The dielectric response of a particle decreases increasing hydrostatic pressure for the constant $\Gamma$ negative. We consider only such hydrostatic pressures in this case which are above the critical hydrostatic pressure $p_c = \frac{aB}{6|\Gamma|}$.

The effective medium approximation theory is formulated for dielectric response of the microcomposite. The two limiting cases are considered: the limit of small concentration of the ferroelastic particles, and the limit of small concentrations of the hard dielectric material. The effective dielectric constant is calculated in both cases. In the first case we have found that increasing the concentration of the ferroelastic component the dielectric response increases assuming that the permittivity of the hard material is lower than that of the ferroelastic. In the opposite case it decreases. Increasing temperature the effective dielectric constant of the microcomposite is tending to the dielectric response of the microcomposite which is under no hydrostatic pressure. If temperature decreases to the transition temperature from paraelastic to ferroelastic phase then the effective response is becoming independent on the hydrostatic pressure. For temperatures higher that the critical it is hydrostatic pressure dependent. For small hydrostatic pressures linearly, for larger hydrostatic pressures and positive $\Gamma$ the effective response increases, however with smaller and smaller velocity. In the second case of low concentration of the dielectric material the effective permittivity increases with hydrostatic pressure for positive constant $\Gamma$. Increasing the concentration of the dielectric hard material the effective dielectric constant decreases if the ferroelastic dielectric constant is higher than that of the dielectric constant. For high temperatures the effective dielectric constant becomes such as if no hydrostatic pressure was applied. For temperature decreasing to critical temperature the effective ferroelectric constant becomes very sensitive to hydrostatic pressure values however with larger concentrations of the dielectric hard material this sensitivity is lower.

We described a theory of microcomposites of the ferroelastic-dielectric type in the limiting cases of concentrations. Quasistatic approximation is used. The Effective Medium Approximation as described above is less usefull for description of the region of percolation transition. For the composite theory of the
elastics-elastics composite, which is of the EMA type, see in [13]. In \( \text{Sn} - \text{VO}_2 \) composites for example [14] 5 percents of inclusions displayed various mechanical instabilities, and 0.5 volume percents of inclusion shows no signs of such instabilities. According to authors this concentration dependence is in agreement with composite theory, which is of the EMA type, see in [13].

In our model above we did not consider the mechanical inclusion/matrix interactions. The ferroelastic crystals as ferroelastic particles may have domain wall structure. However we discuss temperatures in which there is no transition to the ferroelastic phase, the paraelastic phase. Effect of elastic clamping was not considered here. For its discussion for an improper and a pseudopropoer ferroelastic inclusion see [15]. For ellipsoidal shapes of the ferroelastic inclusions the order parameter and strain are uniform inside the inclusion. For improper and pseudopropoer ferroelastic inclusions and polycrystals inclusion/matrix interaction renormalizes the constant of the Landau free energy expansion of the order parameter. For proper ferroelastic materials which we consider here 3D clamping of the crystal inclusion in the matrix is not considered, we assume the mechanical equilibrium of the inclusion/matrix system is present at temperatures higher than the critical temperature. Results of measurements of low frequency dielectric constant and dielectric loss on orthorhombic \( \text{Al}_2(\text{WO}_4)_3 \) show [16] for polycrystalline material, where voids may play the role of dielectric material, linear increasing dependence with hydrostatic pressure. This is qualitatively in agreement with our theory. In epitaxial films proper ferroelastic phase transition with symmetry-conserving and symmetry-breaking misfit strains may be present [17]. Most of microcomposites are in the form of films. The authors [17] have found that if the extrinsic misfit strain, does not break the symmetry of the high-temperature phase, the transition in the film occurs at somewhat lower temperature than in the bulk. This phenomenon we neglect in our paper. In cubic-tetragonal systems like \( \text{Nb}_3\text{Sn}, \text{V}_3\text{Si}, \text{In} - \text{Tl} \) alloys, \( \text{Fe} - \text{Pd} \) alloys and \( \text{Ni}_2\text{MnGa} \) the cubic cell elongates (or contracts) in one of the main axis to form tetragonal cell [18]. While these interesting materials are in general different from those about which we wrote in the introduction above, their high-temperature phase is cubic and our approach may be used to study them. Results of our paper may be interesting for interpretation of experiments on ferroelastic-dielectric microcomposites.

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