Dielectric Nonlinearity of TGS

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Abstract

Dielectric nonlinearities in triglycine sulfate (TGS) have been measured by the analysis of harmonic components of the electric current, while a sinusoidal electric field was applied to the sample. The investigations were motivated by two aims. The first one was to develop a sensitive method for a measurement of the Landau parameters in the paraelectric phase. The second aim was to determine small bias fields and their temperature dependence. A high resolution scanning system (16 bit) has been realized, which allows the resolution of harmonics with an amplitude of 10^{-5} of the fundamental current. The nonlinearities have been measured near the Curie temperature as a function of the temperature and of an overlayed dc field. The experimental results are compared with theoretical calculations from the Landau theory with literature data. A small bias field in the order of 100 V/m has been observed in the paraelectric phase. The bias field is temperature dependent, with a temperature dependency according to a constant polarization.

Theory

For a phenomenological description of the ferroelectric properties of a material by the Landau theory, the ferroelectric contribution to the free energy F is written as a polynom of the dielectric displacement D:

$$F = F_0 + \frac{1}{2}\alpha D^2 + \frac{1}{4}\gamma D^4 + \frac{1}{6}\delta D^6 \tag{1}$$

The coefficients α, γ, δ i. e., the Landau parameters, are temperature dependent in general. A measurement of the dielectric nonlinearities i.e., of the coefficients ϵ_n of the power series representation of the function $D(E)$:

$$D = P_s + \varepsilon_0\varepsilon_1\, E + \varepsilon_0\varepsilon_2\, E^2 + \varepsilon_0\varepsilon_3\, E^3 + \ldots = P_s + \sum_{n=1}^{\infty} \varepsilon_0\varepsilon_n\, E^n \tag{2}$$

gives access to the Landau parameters. In the paraelectric phase i. e., for $P_s = 0$, the first nonlinear dielectric coefficients calculated from the Landau parameters are:

$$\varepsilon_0\varepsilon_1 = 1/\alpha\,, \quad \varepsilon_0\varepsilon_3 = -\gamma/\alpha^4\,, \quad \varepsilon_0\varepsilon_5 = (3\gamma^2 - \alpha\delta)/\alpha^7 \tag{3}$$

while $\varepsilon_0\varepsilon_2 = \varepsilon_0\varepsilon_4 = 0$. The Landau parameters α and γ can be calculated from measured ε_1 and ε_3. Especially the sign of ε_3 indicates the order of the phase transition. In principle, the measurement of ε_5 gives access to δ. A practical determination of δ is only possible, if $\alpha\delta$ is not negligible small compared with γ^3, however.

As an effect of doping or radiation defects, a bias field E_i may occur, which shifts the $D(E)$ curve:

$$D = P_s^0 + \varepsilon_0\varepsilon_1^0\,(E + E_i) + \varepsilon_0\varepsilon_2^0\,(E + E_i)^2 + \ldots = P_s^0 + \sum_{n=1}^{\infty} \varepsilon_0\varepsilon_n^0\,(E + E_i)^n \tag{4}$$

Here ε_n^0 and P_s^0 denote the nonlinear dielectric coefficients and the spontaneous polarization of the defect free material. The coefficients ε_n can be calculated as a function of E_i and ε_n^0, if Eq. (4) is