Nonlinear capacitance dilatometry for investigating elastic and electromechanical properties of ferroelectrets

S. Bauer-Gogonea,[a] F. Camacho-Gonzalez, R. Schwödiauer, B. Ploss,[b] and S. Bauer
Soft-Matter Physics, Johannes Kepler University, Altenberger Str. 69, A-4040 Linz, Austria

(Received 19 July 2007; accepted 23 August 2007; published online 17 September 2007)

Nonlinearities in ferroelectret polymer foam capacitors arise from voltage-dependent thickness changes. Such thickness changes are caused by the converse piezoelectric and electrostrictive effects in these soft materials. The authors show that the higher harmonics of the current response during application of a sinusoidal voltage to ferroelectret capacitors provide information on the elastic and electromechanical properties of the foam. The authors demonstrate the potential of this versatile measurement technique by investigating the temperature dependence of the piezoelectric response and by monitoring the changes in the elastic and electromechanical properties during inflation of cellular polypropylene. © *2007 American Institute of Physics*. [DOI: 10.1063/1.2784960]

Charged cellular ferroelectrets are soft materials for electromechanical energy conversion,[1–3] ideally suited for acoustical applications,[4] airborne ultrasound,[5] large area piezoelectric sensors,[6] and flexible ferroelectret field-effect transistors.[7] Though ferroelectrets are based on nonpolar, low-dielectric-constant polymers, they surprisingly display hysteresis and switching, very similar to phenomena observed in ferroelectric materials.[8] Ferroelectrics are also known to be strongly nonlinear in their dielectric response,[9] a feature which is used, in nonlinear scanning dielectric microscopy[10] for imaging polarized regions in ferroelectric thin films.[11] Extending dielectric investigations to the nonlinear regime provides additional information on the micro-Brownian motion of molecular dipoles in noncrystalline segments of polar polymers[12,13] or on ferro-to-paraelectric phase transitions in ferroelectric polymers.[14–17] In poled second-order nonlinear optical polymers, dielectric nonlinearities provide direct access to molecular hyperpolarizabilities.[18] Since ferroelectrets are based on nonpolar polymers, molecular dipole contributions can be ruled out as a source of dielectric nonlinearities. While the understanding of material properties of ferroelectrets advanced recently,[3] their nonlinear dielectric properties have not been explored yet. An analysis of nonlinearities is essential in applications such as microphones and loudspeakers, where nonlinearities lead to signal distortion.[19]

Here, we show that nonlinearities in the current response of ferroelectrets arise from thickness changes in the ferroelectret capacitor, caused by the piezoelectric and electrostrictive responses of the ferroelectret. We describe a simple experimental setup for measuring these electrical nonlinearities and demonstrate that they can be used for measuring the elastic and electromechanical properties of ferroelectrets, Furthermore, the technique is interesting for reading out the piezoelectric response of cellular polymers in sensor applications.

The experimental arrangement for investigating dielectric nonlinearities in ferroelectrets is shown in Fig. 1. The signal from an ultralow-distortion function generator (Stanford Research DS 360) is fed into a bipolar voltage amplifier (Kepco BOP 1000M) and then applied to the cellular polymer. The cellular polymer sample is mounted on a heating stage (Linkam TM90), to allow for temperature-dependent measurements. The current from the ferroelectret is measured with a digital lock-in amplifier (EG&G 7260) as a voltage drop at a resistor, with an impedance much smaller than the impedance of the ferroelectret. Also shown in Fig. 1 is a simple layer model of the cellular material used for analyzing the nonlinear response of the ferroelectrets. Here, the complex morphology of the foam is represented by a serial connection of polymer layers with a thickness s_1 and an air gap with thickness s_2,[20] and the internal charge is described by a sheet charge density σ. E_1 and E_2 are the electric fields in the polymer and air gap, respectively, caused by σ and by an applied voltage V. This simple layer model was developed for explaining the piezoelectric response of ferroelectrets,[20] and extended for the analysis of the strain-dependent capacity of ferroelectrets.[21]

In general, dielectric nonlinearities are derived from thermodynamic arguments using a polynomial expansion of the electric enthalpy as a function of the electric field.[10] In cellular materials, the nonlinear response directly follows from the voltage-induced shape change of the ferroelectret. It is assumed that the compressive Maxwell stress in the polymer leads to a negligible thickness change of the polymer layers, whereas the Maxwell stress in the air gap $p=\varepsilon_0 E_2^2/2$ results in the observed thickness change of the sample $\Delta s_2(\sigma,V)=s_0 p/Y$, where s_0 and Y are the total foam thickness and Young modulus. When expressed in terms of the piezoelectric $d_{33}=\varepsilon_0 s_1 s_0 \sigma/(2\varepsilon_0 Y(s_1+s_2\varepsilon_1)^2)$ coefficient,[20] the thickness change of the air gap is:

$$\Delta s_2(d_{33}^0,V)=\frac{\varepsilon_0 A^2 Y}{2\varepsilon_0}\frac{d_{33}^{0\,2}}{C_0^2}+d_{33}^0 V+\frac{s_0 C_0^2}{2Y\varepsilon_0 A^2}V^2, \tag{1}$$

where ε_0 is the vacuum permittivity, $C_0=\varepsilon_0\varepsilon s_1 s_2 A/(\varepsilon s_1+s_2)$ is the initial capacity of the polymer foam capacitor, and ε is the dielectric constant of the polymer. Due to the voltage-dependent thickness change in the air gap, the capacity of the ferroelectret is nonlinear in the applied voltage V,

$$C(d_{33}^0,V)=C_0-\frac{C_0^2 d_{33}^0}{\varepsilon_0 A}V+\frac{s_0 C_0^4}{2Y\varepsilon_0^2 A^3}V^2. \tag{2}$$

[a]Electronic mail: sbauer@jku.at
[b]On leave from the SciTec Department, University of Applied Sciences, Jena, Germany.

