Thermal wave probing of pyroelectric distributions in the surface region of ferroelectric materials: A new method for the analysis

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For the investigation of polarization distributions in pyroelectric materials, the laser intensity modulation method (LIMM), which is based on thermal waves, is widely used. With this method, the sample under investigation is heated by the absorption of intensity modulated light at one surface, while the pyroelectric current is measured. The thermal excitation generates a thermal wave penetrating into the sample. The penetration depth is varied with the modulation frequency. A new procedure for the reconstruction of the polarization distribution from a measured pyroelectric current spectrum is introduced. This procedure is especially well suited for polarization probing near the sample surface. An approximation for the polarization distribution is calculated from a measured pyroelectric spectrum in a very simple and direct way, avoiding mathematical instabilities. The calculation can be performed during the measurement of a pyroelectric current spectrum. This makes LIMM an on-line procedure. The new technique of analysis is applied to the measurement of thin depolarized layers near the surface of homogeneously poled ferroelectric polymer films.

I. INTRODUCTION

Methods for the measurement of the spatial distribution of the polarization in dielectric materials are important for many applications of these materials. Polarization distributions influence the response of pyroelectric infrared sensors,[1] and the performance of measurement devices using the pyroelectric effect. Examples are pyroelectric microcalorimeters for specific heat[2] and thermal diffusivity measurements[3] as well as photopyroelectric spectroscopy techniques.[4] For the measurement of charge and polarization distributions, various experimental techniques are available, which are based either on the piezoelectric or on the pyroelectric effect. Among the piezoelectric techniques, the pressure pulse[5,6] and the pressure step method[7] may be noted. The methods, which are based on the pyroelectric effect, are implemented in the time or in the frequency domain. Realized in the time domain is the thermal pulse method,[8] while the laser intensity modulation method (LIMM)[9] uses thermal waves in the frequency domain. For analysis of the measured data from the thermal methods, elaborate numerical procedures are required to reconstruct the polarization profile.[9–12] The aim of this article is to present a method for the analysis of pyroelectric spectra, which is easy to implement, and which gives a good approximation for the polarization distribution in the surface region of pyroelectric films.

II. THERMAL WAVE METHOD

In this section the experimental technique for the LIMM method is described in a short form. The theoretical basis of the method and the mathematical procedures are given, which are commonly used for the evaluation of the measured pyroelectric spectra. For the measurements a film or a disk-shaped plate of the material is used, in which the spatial distribution of the polarization is to be examined. Opaque metal electrodes are deposited on both surfaces of the sample. A laser is used to heat the sample by the absorption of radiation within the opaque electrode at one surface. The intensity of the laser radiation is sinusoidally modulated. By the absorption of sinusoidally modulated intensity at the sample surface, temperature waves are generated, which penetrate into the sample. Due to this excitation, the temperature at each location within the sample varies sinusoidally. The amplitude and the phase of the temperature amplitude depend on the location within the sample and can be altered by a variation of the penetration depth of the temperature wave via the modulation frequency. Measured are the amplitude and the phase, or equivalently the in-phase and the 90° out-of-phase component of the pyroelectric current.

The experimental arrangement we use for the LIMM measurements is schematically shown in Fig. 1. The thickness of the metal electrodes is typically in the order of 30 nm, so that it is justified to view them as infinitely thin. The diameter of the heated area should be large compared with the sample thickness, so that a one-dimensional treatment of the heat conductivity equation is adequate. In this consideration, heat loss from the sample surfaces to the surroundings is neglected, an assumption which is valid for most of the common sample sizes. The pyroelectric current is measured with a current-to-voltage converter. To obtain absolute values of the pyroelectric coefficient the temperature amplitude at the surface of the sample must be known additionally. For this purpose the electrical resistance of the heated electrodes is measured simultaneously via the van der Pauw method.[13] The incoming light intensity j

$$j=j_0+j_\sim \exp(i\omega t) \tag{1}$$

is modulated with the circular frequency ω and the amplitude $j_\sim$. Solving the heat conductivity equation according

