HIGH RESOLUTION TECHNIQUE FOR MEASURING CHARGE AND POLARIZATION DISTRIBUTIONS IN DIELECTRICS BY PIEZOELECTRICALLY INDUCED PRESSURE STEP WAVES (PPS)

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Experimental technique

In the past, several experimental methods have been developed [1] for the analysis of charge and polarization distributions within dielectrics. The acoustic pressure step method originally proposed by Laurençeau et al. [2] recently could be improved significantly achieving resolutions in the order of a few µm [3, 4]. Instead of using pressure steps generated by a shock tube [2] or pressure pulses originating from short and intense laser excitation [3] our technique for pressure step generation [4] makes use of the transient response of a piezoelectric crystal plate. In the experimental arrangement, c.f. Fig. 1, a 100nsec duration, 400 volt pulse from a 50Ω-coaxial cable discharge generator is applied to the
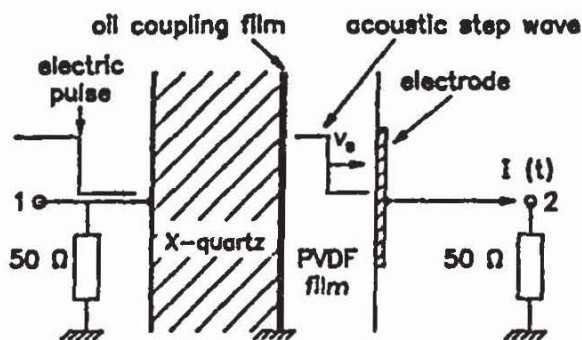

Fig. 1
Schematic representation of the experimental technique. (1) Connection to voltage pulse generator (150psec risetime), (2) Connection to wideband preamplifier and oscilloscope.

X-quartz plate of 3mm thickness and 25mm diameter. The sharp voltage step (risetime < 1nsec) at t = 0 causes an initial state of homogeneous stress in the piezoelectric crystal, with zero stress boundary condition for the acoustically unloaded crystal. This initial condition gives rise to the propagation of zero stress step waves from both surfaces into the crystal. Corresponding to the electric field switch-off after 100nsec rectangular
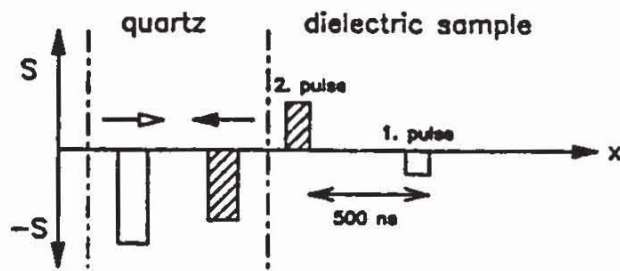
Fig. 2
Acoustic pulse sequence radiated into the sample by the X-quartz plate transient response. The second pulse leading -edge is used as step wave.

velocity (pressure) pulses are generated by the voltage pulse, Fig. 2. By mechanical contact of a dielectric film to one crystal surface a sequence of corresponding velocity (pressure) pulses with alternating sign is radiated into the sample. The first pulse is directly radiated into the dielectric film by the motion of the crystal surface in contact with the sample with no time delay. The second signal with inverted sign is caused by the pulse starting at the acoustically free crystal rear side. Since the amplitude of this signal transmitted into the sample amounts to twice the amplitude of the first pulse, all measurements are performed with the leading edge of this pulse as acoustic step wave. In addition, electrical disturbances from the cable discharge and the acoustical reflections in the sample have decayed within the acoustical delay time of 500nsec. The sequence of further multiple reflected and decaying pulses can be ignored in the measurement. Risetime limitations resulting from the finite quartz disc capacitance, the inertia loading of the X-quartz crystal and the sample by evaporated metal electrodes can be kept below 1nsec with Al-layers not exceeding 1μm thickness on the quartz crystal and 0.1μm thickness on the dielectric film sample. Mostly the dielectric film sample has not been metallized since it appeared sufficient to use the quartz crystal grounding electrode also as the sample grounding electrode, whereas the sample detecting electrode consisted of a conducting rubber disc with 3 to 5mm diameter. As coupling liquid between quartz and sample silicone oil has been used. Applying moderate static pressure to the rubber electrode the acoustic contact to the quartz plate is sufficiently good, i.e. the oil coupling film is squeezed to a thickness of 100 to 200nm, avoiding the risetime deteriorating influence of dust particles by the softness of the dielectric film and the conducting rubber electrode. The influence of acoustic attenuation leading to a risetime increase of the step wave during propagation, in samples of 100μm thickness can be reduced by cooling to liquid nitrogen temperatures.

Step wave response signal and evaluation

As measurements with a thin quartz disc as detector have verified, it is easy to calibrate the generator in terms of the calculated acoustic step wave velocity or pressure amplitude [4] radiated into the sample. Thus, the acoustic step wave velocity amplitude in the sample is known from the measured generator pulse voltage. The observed step wave short circuit current response $I_S(t)$ [4] can be expressed by charge, polarization and "true" piezoelectricity distributions in the dielectric sample:

$$I_s(t) = Av/L \ [(1+\gamma)(\sigma_1 + \int_0^{x_s - v_s t} \rho(x)dx) - e(x_s) - \gamma P_0(x_s)] \tag{1}$$

with $I_S(t)$ signal current under short circuit conditions, c.f. Fig. 1; $x_S = v_S t$ step front position after propagation time t; A = sample area; L = sample thickness;

$$v = 2Z_S \ e_{xx} \ V_p[\ (Z_S + Z_q) \ Z_q L_q]^{-1} \tag{2}$$

velocity amplitude in the step wave with Z_S, Z_Q = acoustic impedance of the sample and the quartz plate respectively, e_{xx} = piezoelectric constant of quartz, L_Q = thickness of the quartz plate, V_p = applied pulse voltage; v_S = step propagation velocity; $\gamma = -1/\epsilon \ \partial\epsilon/\partial s$ relative electro-striction coefficient;

$$\sigma_1 = 1/L \ \int_0^L P_0(x)dx - \int_0^L (1 - x/L)\rho(x)dx \tag{3}$$

static counter charge per area on the grounding electrode of the sample; $\rho(x)$ = density of true excess charges in the sample volume coupled to the particle velocity of the step wave; $e_{33}(x_S)$ = intrinsic piezoelectric constant, i.e. changes of elementary dipolemoment by strain; $P_0(x_S)$ persistent polarization in the unstrained film, note the elementary dipolemoments of this contribution do not change under strain. In deriving Eq. 1 Maxwells equations and the strain relation in the step wave $s = -v/v_S$ have been used. For the homogeneous strained sample, i.e. complete step transit, we obtain from Eq. 1 the total charge transport:

$$Q = \int_0^{L/v_s} I_s(t)dt = As/L \ \int_0^L [e(x) - P_0(x)]dx \tag{4}$$

in agreement with the expectation that the charge distribution $\rho(x_S)$ and electrostriction do not contribute to the homogeneous piezoelectricity. In contrast, the transient response $I_S(t)$ with $e(x_S) = 0$ and $P_0(x_S) = 0$ allows for the determination of the charge distribution by

$$\rho(x) = dI_s/dt \ L[Avv_s(1+\gamma)]^{-1} \tag{5}$$

as follows from Eq. 1. Alternatively, a pure $P_0(x)$ distribution, i.e. $e(x_S) = 0$ and $\rho(x_S) = 0$ results in

$$I_s(t) = Av/L \ [(1+\gamma)\sigma_1 - \gamma P_o(x_s)] \tag{6}$$

This indicates that $P_o(x_s)$ can be resolved only by finite electrostriction $\gamma \neq 0$; the σ_1 contribution results in a positive constant current during the step transit, thus the $P_o(x_s)$ signal is negative with a constant positive background signal resulting from σ_1. With $\rho(x_s) = 0$ and $e(x_s) = 0$, $P_o(x_s)$ follows from the short circuit current response according to:

$$P_o(x_s) = (Av\gamma)^{-1} \ [(1+\gamma)v_s \int_0^t I_s(t)dt - LI_s(t)] \tag{7}$$

t_L = acoustic transit time for the sample with thickness L and $x_s = v_s t$. We finally discuss the situation of complete field neutralization by the condition $\rho(x) = \partial P/\partial x$, i.e. the charge distribution is in electrostatic equilibrium with the persistent polarization. In this case, Eq.1 with finite γ reduces to

$$I_s(t) = Av/L \ [P_o(x_s) - e(x_s)] \tag{8}$$

where $P_o(x_s)$ can be replaced by

$$\int_0^x \rho(x)dx$$

which corresponds to the physical interpretation that the origin of $I_s(t)$ lies in the movement of charges and not in the movement of elementary dipoles with constant dipole-moment. Electrostriction, i.e. $\gamma \neq 0$ does not enter in this case since the internal macroscopic field is zero by field neutralization, i.e. finite conductance of the dielectric material. With respect to the relative electro-striction coefficient entering the evaluation for pure charge or polarization distributions, a simple experiment-al determination is possible by applying a static DC-voltage V_B to the sample still under dynamic short circuit conditions. The resulting rectangular contribution to the short circuit signal response

$$I_s(t) = Av/L \ (1+\gamma) \ V_B/L \ \varepsilon\varepsilon_o \tag{9}$$

allows for the evaluation of γ with ε known from dielectric measurements. Thus, the determination of $I_s(t)$ allows quantitative evaluation in terms of Eqs. 1, 5, 7, 8 if it is already known that either "pure" piezoelectricity, pure charge, pure polarization, or zero field conditions are valid.

Comparison with experiment

Especially with finite electrical conductance as in PVDF the condition of vanishing field by charge compensation
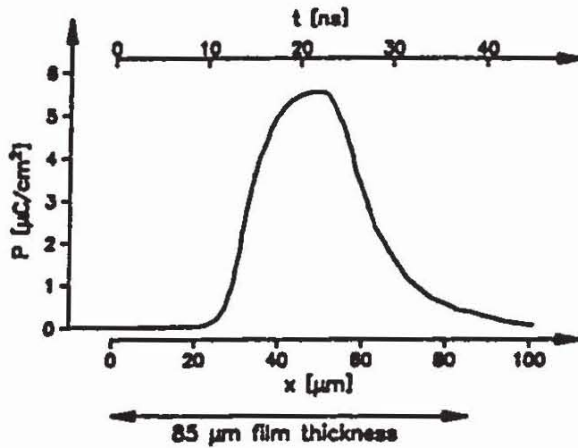
Fig. 3
Acoustic step response
signal for a corona
poled PVDF film.
Polarization evaluated
using Eq. 8 with
$e(x_s) = 0$.

leads to the simple result of Eq. 8 by which $\rho(x_s)$ and
$P_o(x_s)$ can be calculated from the experimental $I_s(t)$
dependence if $e(x_s)$ is either known [5] or comparatively
small, c.f. Fig. 3. For charge compensated internal polar-
ization zones, as in Fig. 3, the absence of signal con-
tributions at the film boundary is characteristic. In the
experimental result, Fig. 3, the equilibrium or zero
field condition in principle could also arise from local
neutralization of the dipole field of polarized crystal-
lites. This "counter charge" situation corresponds to a
vanishing $\rho(x_s)$ and leads to a reduced signal amplitude
[5]. Experimentally, for the sample of Fig. 3 the pres-
ence of space charges according to $\rho(x) = \partial P/\partial x$ has been
verified by microtome slicing combined with charge
measurement. Also the measured total charge flow during
the poling process corresponds to the polarization
measured by the acoustic step wave response. The build-up
of counter charges in contrast would imply significantly
higher total charge flow than observed. An experimental
example for a pure charge distribution [6] is shown in
Fig. 4. A PTFE sample of 25μm thickness has been
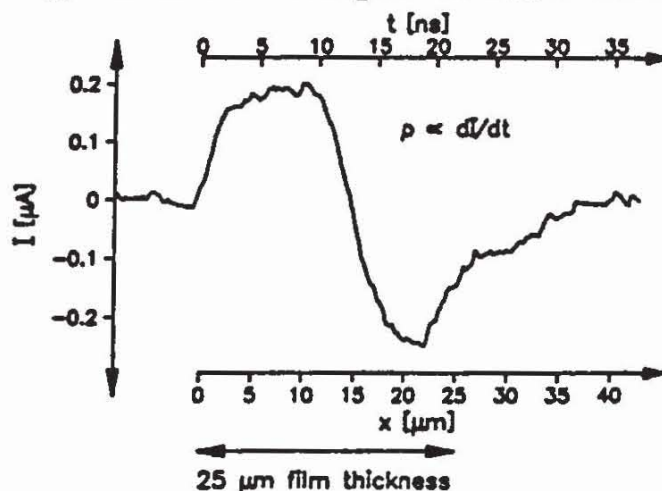

$\rho \propto dI/dt$

Fig. 4
Acoustic step re-
sponse signal for
a 30kV electron
beam charged [6]
PTFE sample.
Discussion see text.

irradiated with electrons, producing a negative charge
layer in the film. As expected the step wave signal
begins with positive constant current caused by the
positive counter charge on the grounding electrode and
changes abruptly to the negative as the step wave passes
the real charge layer. Quantitative evaluation according
to Eq. 5 requires the knowledge of γ which can be obtain-
ed by the additional measurement discussed before.

Time evolution of charge and polarization
Our step wave technique, so far, has been discussed with
respect to the investigation of already prepared samples.
In some situations it is of interest to study the build-
up of charge and polarization distributions during the
poling procedure. This can be accomplished by inserting
an additional insulating film, e.g. mylar between the
conducting rubber electrode and the sample film. The
insulating film acts as a series capacitor reducing the
signal amplitude with no influence on the signal shape.
Poling of the sample is possible by DC-current(voltage)
supply with a thin metal electrode at the insulating film
evaporated on the face in sample contact. The same
technique is used for measuring the electrostriction
coefficient. In both cases quantitative evaluation
additionally requires the measurement of the insulating
film capacitance.

References
1. For a general review see Electrets (Edited by
   G.M. Sessler) Topics in Applied Physics, Vol. 33.
   Springer Verlag, Berlin (1980).
2. P. Laurençeau et al., Phys. Rev. Lett. 38, 46 (1977)
3. G.M. Sessler, J.E. West, R. Gerhard
   Phys. Rev. Lett. 48, 563 (1982)
4. W. Eisenmenger, M. Haardt,
   Solid State Communications, 41, 917 (1982)
5. M.G. Broadhurst et al., J. Appl. Phys. 49, 4992 (1978)
6. To be published in co-operation with G.M. Sessler and
   R. Gerhard having also prepared the PTFE sample.