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High-Resolution Laser-Pulse Method for Measuring Charge Distributions in Dielectrics

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A laser-induced pressure-pulse method for measuring the charge distribution in (10 to 100 μm thick) dielectric samples with a resolution of about 4 μm is described. A short (< 1 ns) energetic light pulse from a neodymium-doped yttrium aluminum garnet laser is applied to a specially coated surface of the sample. The recoil due to ablation of material from the coating generates a pressure pulse (~ 2 ns) which propagates through the sample. The charge distribution is evaluated by measuring the electrode currents from 25- and 75-μm polymers charged with corona and electron-beam methods.

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During the past few years a number of methods to investigate charge distributions in dielectrics have been developed.[1] While the centroid can be conveniently measured with the thermal-pulse method,[2] it is more difficult to obtain detailed information about the actual distribution of the charge. Recently, however, progress has been made with two measuring techniques, namely the acoustic method[3-6] and the electron-beam method.[7-9]

Existing acoustic methods use either a shock tube,[3] a spark gap,[4] or a ruby laser[5] to excite pressure pulses or steps of about 0.1 μs duration, or a quartz crystal[6] to generate a pressure step of about 1-ns risetime. The present acoustic method utilizes a very short laser-induced pressure pulse (LIPP) and differs in three important respects from the existing techniques: (1) The duration of LIPP is only about 1 ns which leads to high resolution without further signal processing; (2) the coupling of the LIPP into the sample is achieved directly without parallel-mounting or bonding problems; and (3) the LIPP technique is used not only for two-sided, but also for one-sided metalized samples.

The light pulses were generated by a mode-locked and Q-switched 1.064-μm neodymium-doped yttrium aluminum garnet (Nd:YAlG) laser[10] system which allows a single-pulse operation. The length of the light pulses can be varied within the range of 70 to 1200 ps and the energy of a single pulse can be chosen as high as 100 mJ, corresponding to a peak power of 1.4 GW. For most of the measurements, pulse energy of 50 mJ was used. The cross-sectional area of the beam at the sample surface was about 0.2 cm^2.

The surface of the sample was mechanically excited by the laser pulse causing ablation of a graphite layer deposited on the sample surface prior to the experiment. The emission of the material generates a recoil which launches the desired pressure pulse. For a laser pulse of 70-ps duration, the beam intensity of 3.5 GW/cm^2 is sufficient to cause ablation.[11] The effect of the heat diffusion into the sample is beyond the time scale of the present experiment.

The LIPP of duration τ propagates with the velocity of sound c through the sample, as illustrated in Fig. 1. It is assumed that the length $c\tau$ of the LIPP is smaller than any characteristic length describing changes in the charge distribution of the sample. In an open-circuit arrangement of the sample electrodes, the pulse yields, for pressures not exceeding the elastic limit of the dielectric, a voltage response $V(t)$ given by[4]

$$V(t) = -(2-\epsilon^{-1})\chi P c\tau E(ct), \quad (1)$$

where χ is the compressibility of the sample, P is the pressure amplitude of the pulse, and $E(ct) = E(x)$ is the local electric field in the sample.

Differentiation of Eq. (1) with respect to time and use of Poisson's equation $\epsilon\epsilon_0 dE/dx = \rho(x)$ with

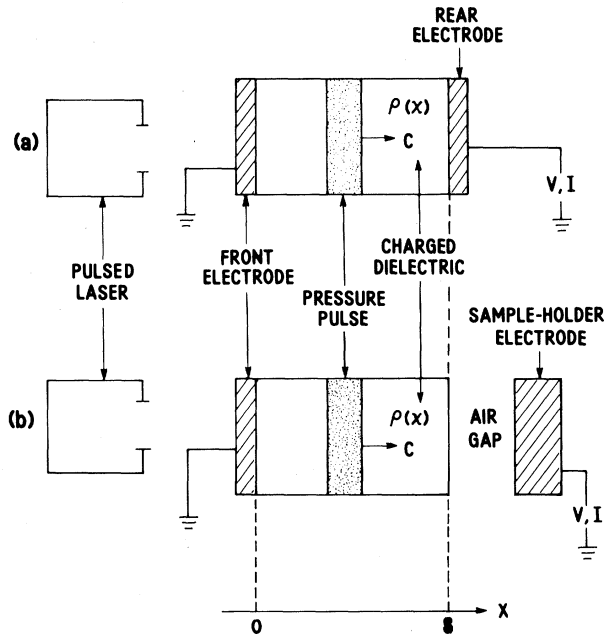

FIG. 1. Schematic setup and geometry of the experiment for (a) two-side-metalized samples and (b) one-side-metalized samples.

$dx = cdt$ yields

$$\frac{dV(t)}{dt} = -\frac{1}{\epsilon\epsilon_0}\left(2-\frac{1}{\epsilon}\right)\chi P c^2 \tau \rho(x). \quad (2)$$

This equation relates the change of the open-circuit voltage to the charge density.

Under short-circuit conditions the voltage change dV/dt is replaced by $I(t)/C$, where $I(t)$ is the short-circuit current and C is the capacitance of the sample. With $C = \epsilon\epsilon_0 A/s$, where A and s are the sample area and thickness, respectively, one obtains

$$I(t) = -(2-\epsilon^{-1})\chi P c^2 \tau (A/s)\rho(x). \quad (3)$$

According to this equation, the charge density in the dielectric can be determined directly from the current observed in the external circuit upon propagation of the LIPP through the sample. Since the pressure amplitude is presently not known, Eq. (3) will not be evaluated. Quantitative measurements of this quantity with a piezoelectric crystal are, however, planned.

The electret samples used in the present study are 25- and 75-μm-thick films of polyethyleneterephthalate (PETP), available under the trade name Mylar, and films of the same thicknesses of polyfluoroethylenepropylene (FEP), available under the trade name Teflon. The films were

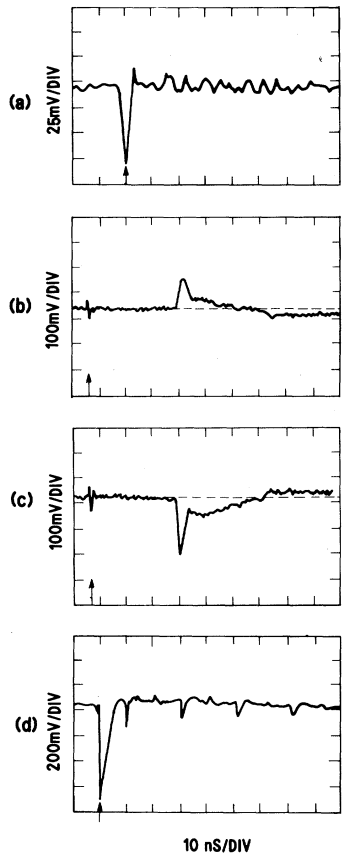

FIG. 2. Current response of one-side-metalized PETP films; arrows indicate initial spikes. (a) Uncharged 75-μm sample, (b) positive-corona 75-μm sample, (c) negative-corona 75-μm sample, and (d) negative-corona 25-μm sample.

first metalized with a 0.7-cm-diam, 250-nm-thick layer of aluminum by evaporation on one or both surfaces. Thereafter, the samples were charged by corona or electron-beam methods[1] to projected charge densities of about 10^{-8} C/cm^2. The samples were then mounted in a sample holder which also provides a rear electrode for the one-side-metalized films as shown in Fig. 1(b). A small air gap (~ 10 μm) exists between the rear surface of the samples and the electrode of the sample holder. For better absorption of the laser light, the front electrode of the samples was coated with about a 2-μm layer of aerosolized graphite.

Initial experimental results are shown in Figs. 2 through 4. Measurements on the same sample could be repeated many times without signal degradation; this indicates that heating by the laser pulse does not discharge the sample. The time constants of the electronics and sample capaci-

tance are such that the observed voltages are proportional to $I(t)$ introduced above except that the display is inverted. Thus, according to Eq. (3), the observed temporal current responses directly correspond to the spatial charge distributions.

Results obtained on a series of 25- and 75-μm-thick PETP films, metalized on the front surface only, are shown in Fig. 2. While the sample whose response is depicted in Fig. 2(a) was uncharged, the other samples were charged with positive or negative corona, as indicated. As expected, the uncharged film shows no signal except for an initial spike probably due to photoionization by the laser pulse. The charged samples exhibit not only the light-induced spikes but also additional spikes which are attributed to the charge layers in these electrets. Since these films were charged by a corona process, the charges are located at the free (rear) surfaces. The separation of the first and second spikes is thus the transit time of the pulse. Transit times of 11 and 32 ns for the 25- and 75-μm samples, respectively, correspond to a sound velocity of 2.3 km/s in PETP, in agreement with literature values.[12] After transit, the LIPP is reflected from the rear surface of the film. Since phase and direction of propagation are both reversed, the signals recorded after reflection are expected to be of the same sign as the signals seen before reflection. When the pulse reaches the front electrode, which carries a small induction charge, a signal of opposite polarity is generated. While some degradation of the signals after the first reflection on the rear surface is seen for the 75-μm films [Figs. 2(b) and 2(c)], the 25-μm film clearly shows signals due to a sequence of reflections [Fig. 2(d)]. Most of the induction charge is located on the electrode of the sample holder for the 75-μm films which is not affected by the pressure pulse and therefore does not give a signal.

The response of a one-side-metalized 25-μm FEP film is shown in Fig. 3. The sample was negatively corona charged prior to the experiment. It exhibits an initial spike due to the light pulse and another spike after 19 ns, corresponding to the charge. The delay yields a sound velocity of 1.3 km/s. No reliable literature value of the sound velocity in FEP in the thickness direction has been found for comparison.

In Fig. 4, the responses of 55-keV electron-beam-charged 75-μm PETP and FEP samples, metalized on both surfaces, are depicted. After initial spikes due to the laser light, the positive induction charge on the front electrode is seen. This charge is much larger than on the previously discussed samples since the charge injected by the electron beam extends deep into the film. This signal is followed by a broad peak due to the negative charge, which is distributed over a certain thickness range of the films. A charge of positive sign follows which may be partially an electrode charge. Particularly for the FEP film, however, the positive-charge signal is so broad that injection of positive charges possibly generated by secondary emission has to be assumed (also see Ref. 9). The presence of a positive charge close to the electrode of incidence in electron-beam-charged PETP samples has also been seen in distribution measurements performed with the electron-beam method.[9] From the rear-electrode location in Fig. 4 (corresponding to the peak of the positive-current signal), the location of the maximum negative charge density is then

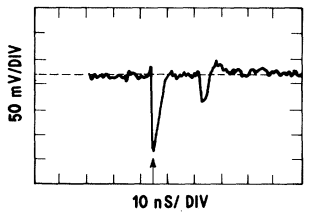

FIG. 3. Current response of negative-corona charged, one-side-metalized 25-μm FEP film; arrow indicates initial spike.

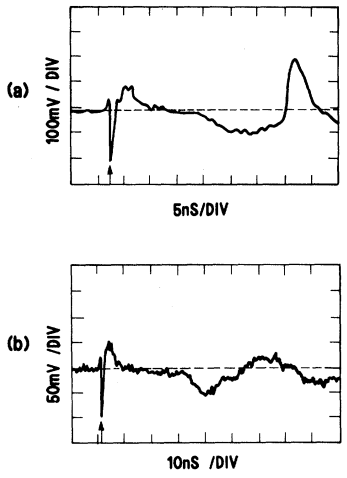

FIG. 4. Current responses of two-side-metalized 75-μm films, charged with a 55-keV electron beam through the rear electrode; arrows indicate initial spikes. (a) PETP, (b) FEP.

23 and 27 μm from the rear surface for PETP and FEP, respectively. The FEP values agree approximately with the centroid location in electron-beam–charged samples, as determined by split-Faraday-cup measurements.[13]

The resolution in the present experiment can be estimated from the data in Figs. 2 and 3. Since the charges in corona-charged samples are initially located close to the surface, the width of the corresponding current spikes yields the resolution. Inspection of Fig. 2(d), for example, indicates a resolution of 2 ns or about 4 μm in PETP. The resolution is, under the present conditions, determined by the time constants of the available amplifiers and can be improved considerably by using more appropriate electronics. A 500-ps laser pulse should, with proper electronics, yield a resolution of about 0.5 μm and 1 μm in FEP and PETP, respectively.

The LIPP method described in this paper is, therefore, capable of higher resolution than other known pressure-pulse techniques. It is also simpler than existing techniques since synchronous coupling of the pulse into the sample over its entire surface is no problem. The LIPP method should therefore find applications not only in the study of polymer electrets but also for investigations of charge storage in metal oxides and other insulting structures. In addition, the present LIPP method can be used to determine sound velocity and attenuation in the thickness direction of thin films.

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Note added.—After this manuscript was submitted, a related technique and result was reported by Alquie, Dreyfus, and Lewiner.[14] In their technique, a laser pulse is used to excite a pressure pulse in a thick aluminum plate which is coupled to the sample under investigation. Because of the long fall time of the pressure pulse in this case, a mathematical deconvolution is necessary to determine the electric field distribution. In the present paper, the laser pressure pulse impinges directly on the sample under investigation and the distribution is directly determined with resolution better than 4 μm.

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