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Analysis of pressure-wave methods for the nondestructive determination of spatial charge or field distributions in dielectrics

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A pressure wave propagating through a charged dielectric produces an electric signal at the sample electrodes. This signal reveals information on the spatial charge or field distribution in the sample. In the present study, the mathematical relations between the signal response and the desired distribution are derived from first principles. For the derivation, the pressure dependence of the relative permittivity must be known; therefore this dependence was calculated for nonpolar as well as for polar dielectrics with the use of the Clausius-Mossotti equation. The resulting response equations are applicable to pressure-pulse, pressure-step, and arbitrary pressure-profile experiments on one- and two-sided metallized samples in open-circuit and in short-circuit configuration. The common features and the differences between the present analysis and existing theoretical descriptions are discussed in detail.

I. INTRODUCTION

Detailed knowledge of the electric charge or field distribution in the bulk of dielectrics is necessary for the understanding of charge-storage and charge-transport phenomena. The most successful techniques for the experimental determination of spatial charge or field distributions in dielectrics are based on the transit of a compression wave through the sample. This possibility was first mentioned by Collins.[1] The differences between the suggested approaches lie mainly in the methods used for the generation of the pressure wave.

A shock tube was used[2–4] to generate a pressure step of unspecified rise time. Pressure pulses of about 0.25-μs duration were produced by means of a Q-switched ruby laser.[5] A high-voltage spark gap[6,7] permitted the generation of a pressure wave with a rise time of about 0.7 μs. The ruby-laser-pulse method[5] was utilized for charge- and field-distribution measurements on electron-irradiated dielectrics[8]; the spatial resolution of the method was $50-150\,\mu$m.

Recently, a pulsed CO_2 laser was employed[9] for the production of pressure profiles which had a rise time of about 10 ns and a duration of more than 300 ns. 1.5-J acoustic pulses from a ruby laser were utilized[10] for electric field measurements inside polymethylmethacrylate disks. The full width at half maximum of the ruby-laser pulses used was 15 ns. The direct experimental resolution of all these methods is not high enough for samples thinner than a few dozen μm.

This limitation could be overcome by using an actively mode-locked and Q-switched Nd:YAG (YAG denotes yttrium aluminum garnet) laser system for the generation of about 1-ns-long pressure pulses[11] and by using a quartz crystal to produce step waves of about 1-ns rise time.[12] With these two experimental techniques field distributions in polyvinylidenefluoride foils[12] and charge distributions in Teflon fluorethylenepropylene and Mylar polyethyleneterephthalate films[13] could be measured directly.

Since all methods described[2–13] depend on the same physical process, namely a pressure wave propagating through the sample, their theoretical descriptions should be identical. This, however, appears not to be the case because different approaches have been used for the derivation of the equations which govern the response of a charged sample to a pressure wave.[3,5,7,9,12]

A clarification is attempted in the present study: The basic equations for the pressure-pulse and the pressure-step experiments are derived from first principles. The pressure dependence of the relative permittivity is investigated, and the influence of this dependence on the signal amplitude is calculated for nonpolar and for polar dielectrics. The response equations for pressure-step, pressure-pulse, and arbitrary pressure-profile experiments are derived from the basic equations. Finally, the connections between the present approach and existing theories[3,5,7,9,12] are presented explicitly.

II. BASIC EQUATIONS

All pressure-wave methods[2–13] for the determination of charge or field distributions in dielectrics depend on the temporal change of the induced electrode charges caused by two effects: (1) The compression of parts of the sample alters the distance between charges and induction charges, and (2) the compressed regions of the dielectric have a different relative permittivity. The theoretical description is based on the following assumptions:

(1) The dielectric has the shape of a sheet (area A, thickness s) with all lateral dimensions being much greater than the thickness. All quantities only change in the thickness direction, and all surfaces, interfaces, and wave fronts are exactly perpendicular to the thickness direction. Thus, a one-dimensional model can be employed.

(2) The x axis denotes the thickness direction of the dielectric, $x=0$ being the front surface through which the pressure wave enters the sample and $x=s$ its rear surface. The front electrode always contacts the front surface of the dielectric.

(3) The charge $\rho(x)$ in the sample is composed of real charges $\rho_r(x)$ and the gradient $\rho_p(x)=-dP(x)/dx$ of the polarization $P(x)$. The charge density does not change during transit of the pressure wave through the dielectric. The electric field $E(x)$ in the sample is related to the charge distribution $\rho(x)$ by use of Poisson's equation: $\rho(x)=\epsilon_0\epsilon\, dE(x)/dx$.

(4) The pressure wave travels through the sample with the velocity of sound c. Therefore, the space coordinate in the sample and the time axis are connected by the relation $x=ct$.

(5) The "mean" amplitude p of the pressure wave is given either by the average pressure during the duration τ of the pressure pulse or by the "height" of the pressure step. Attenuation and dispersion of the pressure wave in the dielectric during one transit are disregarded. The latter assumption has to be justified from the experimental evidence.

(6) The dielectric has a relative permittivity ϵ and a coefficient of one-sided compression $\chi=(1/x)(dx/dp)_T$, where p and T are the pressure and the temperature, respectively. Adiabatic compression is assumed. All quantities in the compressed region of the sample are primed.

(7) The lateral sample dimensions do not change during transit of the compression zone. Therefore, the coefficient χ is given by $\chi=-(1+\mu)(1-2\mu)/(1-\mu)Y$, where μ and Y are Poisson's number and Young's modulus of the dielectric, respectively. Since the sound velocity c is identical to the velocity of longitudinal waves,

$$c=\left[\frac{(1-\mu)Y}{\rho_0(1+\mu)(1-2\mu)}\right]^{1/2},$$

where ρ_0 is the sample density, χ can be replaced by[10]

$$\chi=-\frac{1}{\rho_0 c^2}\;. \tag{1}$$

For short-pressure pulses or for sharp-pressure steps (ideally delta or theta functions) it is possible to derive the charge or field distribution directly from the electric signal without a deconvolution procedure. These two cases are therefore treated first; other pressure-wave profiles can be regarded as a superposition of either pulses or steps.

A. Pressure pulse

If the duration τ of the pressure pulse is much shorter than its transit time through the sample and if short-circuit conditions prevail during the duration τ, the induction charge densities $\sigma_{\mathrm{I}-}$ and $\sigma_{\mathrm{I}+}$ on the front electrode (I) before and after the pulse passes the charge layer of thickness Δx at the location x can be determined[14] [see Figs. 1(a) and 1(b)]:

$$\begin{aligned}\sigma_{\mathrm{I}-}&=-\frac{(s-x)/\epsilon+g}{(s-c\tau)/\epsilon+(c\tau)'/\epsilon'+g}\times\rho(x)\Delta x\;,\\ \sigma_{\mathrm{I}+}&=-\frac{(s-x-c\tau)/\epsilon+(c\tau)'/\epsilon'+g}{(s-c\tau)/\epsilon+(c\tau)'/\epsilon'+g}\times\rho(x)\Delta x\;.\end{aligned}\tag{2}$$

Here g is the thickness of the air gap ($\epsilon_g=1$) between rear sample surface and rear electrode (II) which equals 0 for two-sided metallized samples, and $(c\tau)'$ and ϵ' are the thickness and the relative permittivity of the compressed layer, respectively. In Fig. 1 only the coordinates x (attached to the solid dielectric) and t are shown; therefore, the small shift of the charge layer has been omitted.

When the pressure pulse of duration τ passes the charge layer of thickness Δx at the location x the induction charge density σ_{I} changes by $\Delta\sigma_{\mathrm{I}}=\sigma_{\mathrm{I}+}-\sigma_{\mathrm{I}-}$ as follows:

$$\Delta\sigma_{\mathrm{I}}=\frac{c\tau/\epsilon-(c\tau)'/\epsilon'}{s/\epsilon+g-[c\tau/\epsilon-(c\tau)'/\epsilon']}\rho(x)\Delta x\;. \tag{3}$$

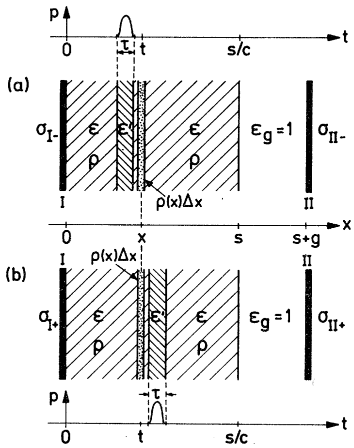

FIG. 1. Sample geometry for the pressure-pulse signal analysis: (a) before the pulse passes the charge layer of thickness Δx at the location x and (b) after the pulse has passed the location x.

In a first-order approximation the term $[c\tau/\epsilon-(c\tau)'/\epsilon']$ in the denominator can be omitted, since it is very small compared to $s/\epsilon+g$:

$$\Delta\sigma_{\mathrm{I}}=\frac{c\tau/\epsilon-(c\tau)'/\epsilon'}{s/\epsilon+g}\rho(x)\Delta x\ . \qquad (4)$$

The thicknesses of the compression layer without and with adiabatic compression are $c\tau$ and $(c\tau)'=c\tau(1+\chi p)$, respectively:

$$\Delta\sigma_{\mathrm{I}}=\frac{1/\epsilon-(1+\chi p)/\epsilon'}{s/\epsilon+g}c\tau\rho(x)\Delta x$$

$$=\frac{c\tau\delta_\epsilon}{s/\epsilon+g}\rho(x)\Delta x\ , \qquad (5)$$

where $\delta_\epsilon=1/\epsilon-(1+\chi p)/\epsilon'$ denotes the relative change of the "dielectric thickness" of the sample.

The induction charge-density change $\Delta\sigma'_{\mathrm{I}}$ resulting from the entering of the pressure pulse into the sample can be determined in a similar manner: Under short-circuit conditions, the induction charge densities $\sigma'_{\mathrm{I}-}$ and $\sigma'_{\mathrm{I}+}$ before and after the pressure pulse has entered the bulk of the dielectric are given by[14]

$$\sigma'_{\mathrm{I}-}=-\frac{(s-r)/\epsilon+g}{s/\epsilon+g}\sigma\ ,$$

$$\sigma'_{\mathrm{I}+}=-\frac{(s-r)/\epsilon+g}{(s-c\tau)/\epsilon+(c\tau)'/\epsilon'+g}\sigma\ . \qquad (6)$$

In Eq. (6), $\sigma=\int\rho(x)dx$ is the total charge per unit area in the sample volume and r is the centroid of this eventually distributed charge. The assumption that the total sample charge $\rho(x)$ is located at its centroid location can be made without loss of generality, since the influence of the distribution of this charge is covered by Eq. (5) and linear superposition is applicable.

From Eq. (6), the difference $\Delta\sigma'_{\mathrm{I}}=\sigma'_{\mathrm{I}+}-\sigma'_{\mathrm{I}-}$ becomes

$$\Delta\sigma'_{\mathrm{I}}=\sigma'_{\mathrm{I}-}\left[\frac{s/\epsilon+g}{(s-c\tau)/\epsilon+(c\tau)'/\epsilon'+g}-1\right]\ ,$$

or in linear approximation, with the use of the fact that the difference $c\tau/\epsilon-(c\tau)'/\epsilon'$ is a small quantity,

$$\Delta\sigma'_{\mathrm{I}}=\frac{c\tau/\epsilon-(c\tau)'/\epsilon'}{s/\epsilon+g}\sigma'_{\mathrm{I}-}=\frac{c\tau\delta_\epsilon}{s/\epsilon+g}\sigma'_{\mathrm{I}-}\ . \qquad (7)$$

Comparison of Eqs. (5) and (7) shows that the charge distribution $\rho(x)$ can be assumed to include the induction charge $\sigma'_{\mathrm{I}-}$ on the front electrode (I).

B. Pressure step

For a step-function-like pressure wave and open-circuit conditions during the pressure-step experiment, the potential differences V_- and V_+ between the two sample electrodes before and after the step has passed a layer Δx at the location x can be determined [see Figs. 2(a) and 2(b)]:

$$\begin{aligned}V_-&=-\int_0^{x'-}E'(\xi')d\xi'-E(x)\Delta x\\&\quad-\int_{x+}^{s}E(\xi)d\xi-E_g g\ ,\\V_+&=-\int_0^{x'-}E'(\xi')d\xi'-E'(x')\Delta x'\\&\quad-\int_{x+}^{s}E(\xi)d\xi-E_g g\ .\end{aligned} \qquad (8)$$

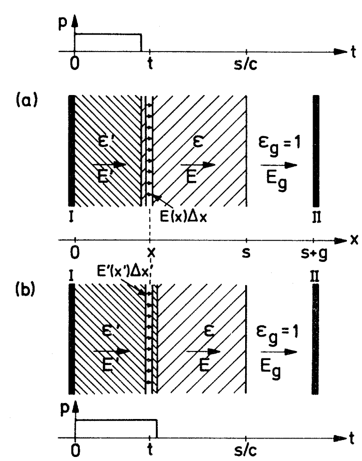

FIG. 2. Geometry of the sample during a pressure-step experiment: (a) before the step passes the location x and (b) after the step has passed the location x.

E_g is the electric field in the air gap between the rear surface and the rear electrode (II) and the integration is carried out from electrode I to just before the layer Δx $(x'-)$ and from just after this layer $(x+)$ to the other sample surface. The thickness g of this gap (which vanishes for two-sided metallized samples) stays constant during the first transit of the pressure step. Only the coordinates x (attached to the solid dielectric) and t are used in Fig. 2; thus the small thickness reduction of the sample caused by the compression has been omitted.

The voltage change $\Delta V = V_+ - V_-$ caused by the pressure step passing the layer Δx is given by

$$\Delta V = E(x)\Delta x - E'(x')\Delta x' \ . \tag{9}$$

Using $\Delta x' = \Delta x(1+\chi p)$ and Gauss's law for the dielectric displacements before and after transit of the pressure step at the location x $[\epsilon E(x) = \epsilon' E'(x')]$, one obtains

$$\Delta V = [1/\epsilon - (1+\chi p)/\epsilon']\epsilon E(x)\Delta x$$
$$= \epsilon \delta_\epsilon E(x)\Delta x \ . \tag{10}$$

C. Pressure dependence of the relative permittivity

As shown, the pressure dependence of the signal for the pressure pulse as well as for the pressure step is given by the relative change of the "dielectric thickness" of the sample

$$\delta_\epsilon = 1/\epsilon - (1+\chi p)/\epsilon' \ ,$$

where $(1+\chi p)$ accounts for the change in sample thickness caused by the adiabatic compression, and ϵ' is the relative permittivity of the compressed sample region. In order to calculate δ_ϵ the pressure dependence of the relative permittivity must be known.

1. Pressure-independent relative permittivity

It has been suggested[5,9] to assume for the sake of simplicity that the relative permittivity is pressure-independent,

$$\epsilon' = \epsilon \ . \tag{11}$$

This assumption is, however, not justified since the contribution of the pressure dependence of ϵ to the signal is of the same order as the geometric contribution (see below). The assumption results in the following expression for δ_ϵ:

$$\delta_\epsilon = -\chi p/\epsilon \ . \tag{12}$$

2. Directly pressure-dependent relative permittivity

Whereas the assumption $\epsilon = \epsilon'$ leads to a lower limit for the pressure dependence of the signal (no dielectric contribution at all), a simple relation can be found by assuming that the relative permittivity ϵ is proportional to the number of particles N per volume v: $\epsilon \propto N \propto 1/v$. This leads to the relation $\epsilon v = \epsilon' v'$ between the relative permittivities of the compressed and the noncompressed sample regions, respectively. With $v' = (1+\chi p)v$, one obtains

$$\epsilon' = \frac{\epsilon}{1+\chi p} \ , \tag{13}$$

which is, in first-order approximation, the same as the suggested[9] relation $\epsilon' = (1-\chi p)\epsilon$ [in Ref. 9 the compressibility is defined as[3] $\chi = -(1/v)(\Delta v/\Delta p)$]. From relation (13) δ_ϵ becomes

$$\delta_\epsilon = [1-(1+\chi p)^2]/\epsilon \ ,$$

or, omitting the quadratic term in χp,

$$\delta_\epsilon = -2\chi p/\epsilon \ . \tag{14}$$

The simple relation (13) is not realistic either, because it implies a pressure dependence of the vacuum part of the relative permittivity.

3. Pressure-dependent susceptibility $\epsilon - 1$ (Clausius-Mossotti equation without dipole interaction)

If only the material-related part of the relative permittivity ϵ, namely the susceptibility $\epsilon - 1$, is considered to be pressure-dependent, then Eq. (13) can be used for $\epsilon - 1$ instead of ϵ,

$$\epsilon' - 1 = \frac{\epsilon - 1}{1+\chi p} \ . \tag{15}$$

Equation (15) can also be derived from the Clausius-Mossotti equation for nonpolar materials without dipole interaction,

$$\epsilon - 1 = \frac{4\pi}{3} N\alpha \ , \tag{16}$$

where N again is the number of particles per volume v and α is the polarizability of a particle. Since the particle polarizability α and the total number of particles are constant under different pressures, relation (16) leads to

$$(\epsilon - 1)/N = (\epsilon' - 1)/N'$$

or $(\epsilon - 1)v = (\epsilon' - 1)v'$ and directly to Eq. (15).

Rewriting relation (15) and neglecting higher-order terms in χp yields

$$\epsilon' = (1-\chi p)\epsilon + \chi p \ . \tag{17}$$

This is Eq. (4) of Ref. 7 (there the relative permittivity is called K) which was derived under the assumption that the bulk polarizability $N\alpha$ is inversely proportional to the volume v. Using Eq. (17) and first-order approximation, δ_ϵ becomes

$$\delta_\epsilon = \frac{1/\epsilon - 2}{\epsilon + \chi p}\chi p \ ,$$

and finally

$$\delta_\epsilon = -(2-1/\epsilon)\chi p/\epsilon \ . \tag{18}$$

4. *Nonpolar dielectrics (Clausius-Mossotti equation with dipole interaction)*

Including dipole interaction, the Clausius-Mossotti equation is given by[15]

$$\frac{\epsilon-1}{\epsilon+2} = \frac{4\pi}{3}N\alpha \ , \tag{19}$$

which, in analogy to the derivation of Eq. (17), leads to

$$\frac{\epsilon-1}{\epsilon+2} = (1+\chi p)\frac{\epsilon'-1}{\epsilon'+2} \ ,$$

and finally to

$$\epsilon' = \frac{3\epsilon + (\epsilon+2)\chi p}{3 + (\epsilon+2)\chi p} \ . \tag{20}$$

Using Eq. (20) for the relative permittivity ϵ' of the compressed region of a nonpolar dielectric and neglecting higher-order terms in χp, one obtains

$$\delta_\epsilon = \frac{1}{\epsilon} - \frac{3+(\epsilon+5)\chi p}{3\epsilon + (\epsilon+2)\chi p}$$

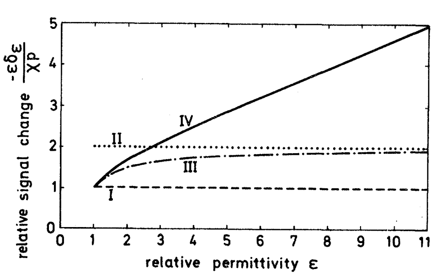

FIG. 3. Relative-permittivity dependence of the expression $-\epsilon\delta_\epsilon/\chi p$ for four different pressure dependencies of the relative permittivity ϵ: I $\epsilon'=\epsilon$ [Eq. (11)], II $\epsilon'=(1-\chi p)\epsilon$ [Eq. (13)], III $\epsilon'=(1-\chi p)\epsilon+\chi p$ [Eq. (17)], IV $\epsilon'=[3\epsilon+(\epsilon+2)\chi p]/[3+(\epsilon+2)\chi p]$ [Eq. (20)].

and

$$\delta_\epsilon = -[4/3 + \epsilon/3 - 2/(3\epsilon)]\chi p/\epsilon \ . \tag{21}$$

This relation is appropriate for dielectrics without permanent dipoles and has been used very recently in Ref. 10. Figure 3 shows how the signal amplitude would depend on the relative permittivity ϵ according to Eqs. (12), (14), (18), and (21), respectively.

5. *Very short pressure pulses in polar dielectrics (Clausius-Mossotti equation for the induced polarization)*

In polar materials, only the induced polarization obeys the Clausius-Mossotti equation. For high-frequency alternating fields the permanent dipoles can no longer follow the field changes so that only the relative permittivity ϵ_∞ characteristic for the induced polarization must be used. For the very short pressure pulses of less than about 1-ns duration which are generated in the laser-induced pressure-pulse (LIPP) method[11,13] it can be assumed that only the induced polarization contributes to the pressure dependence of the signal if the relative permittivity of the sample does not change within the frequency range of the main part of the pulse spectrum (above 100 MHz). Therefore, the Clausius-Mossotti equation for the induced polarization[15]

$$\frac{\epsilon_\infty-1}{\epsilon_\infty+2} = \frac{4\pi}{3}N\alpha \tag{22}$$

can be used to calculate approximately the pressure dependence of δ_ϵ in analogy to the derivation of Eqs. (17) and (20):

$$\frac{\epsilon_\infty-1}{\epsilon_\infty+2} = (1+\chi p)\frac{\epsilon'_\infty-1}{\epsilon'_\infty+2} \ . \tag{23}$$

Furthermore, the contribution of the permanent dipoles to the relative permittivity ϵ of the material, $\epsilon-\epsilon_\infty$, can be assumed to remain constant during the transit of these very short pulses through a sample layer,

$$\epsilon - \epsilon_\infty = \epsilon' - \epsilon'_\infty \ . \tag{24}$$

Elimination of ϵ'_∞ in Eq. (23) by means of relation (24) leads to

$$\epsilon' = \epsilon - \frac{(\epsilon_\infty+2)(\epsilon_\infty-1)}{3/(\chi p)+\epsilon_\infty+2} \ ,$$

or, in first-order approximation,

$$\epsilon' = \epsilon - \frac{\chi p}{3}(\epsilon_\infty+2)(\epsilon_\infty-1) \ . \tag{25}$$

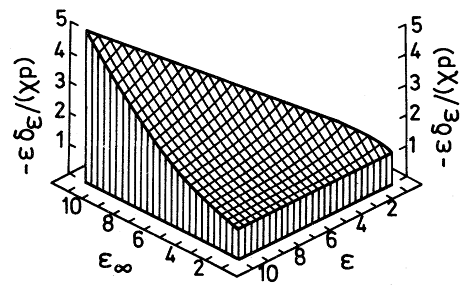

FIG. 4. Expression $-\epsilon\delta_\epsilon/\chi p$ as a function of the relative permittivities ϵ at zero frequency and ϵ_∞ at the frequency of the pressure-pulse experiment [Eq. (26)].

If higher-order terms in χp are again omitted, one obtains for very short pressure pulses in polar dielectrics,

$$\delta_\epsilon = -\left[1+\frac{(\epsilon_\infty+2)(\epsilon_\infty-1)}{3\epsilon}\right]\chi p/\epsilon \ . \tag{26}$$

For nonpolar materials, $\epsilon_\infty=\epsilon$, and Eqs. (26) and (21) become identical. The dependence of the signal amplitude on the relative permittivities ϵ at zero frequency, and ϵ_∞ at the frequency corresponding to the duration τ of the pressure pulse, is shown in Fig. 4.

III. SHORT-CIRCUIT AND OPEN-CIRCUIT RESPONSE

A. Pressure-step experiments

1. Open-circuit voltage

Dividing Eq. (10) by Δt, changing the differences Δ into differentials d, and replacing dx/dt by c yields, for a pressure step of amplitude p, a relation between the temporal change of the voltage and the electric field in the sample,

$$\frac{dV(t)}{dt} = [1-(\epsilon/\epsilon')(1+\chi p)]cE(x)$$

$$=\epsilon\delta_\epsilon cE(x) \ , \tag{27}$$

which is identical to Eq. (4) of Ref. 3 [$V(d)-V(0)$, v, z_f, $-\chi$, and Δp are used in this reference in place of $V(t)$, c, x, χ, and p, respectively]. This equation was obtained[3] by differentiation of the potential difference across a two-sided metallized dielectric during propagation of a step-function compressional wave.

Usually not the time derivative of the voltage, but the open-circuit voltage itself is determined experimentally. Integration of Eq. (27) with $V(0)=0$ and use of the relation $dx=c\,dt$ lead to

$$V(t) = [1-(\epsilon/\epsilon')(1+\chi p)]\int_0^{x=ct} E(\xi)d\xi \ . \tag{28}$$

The field integral $\int^x E(\xi)d\xi$ determines the potential $\Phi(x=ct)$ at the location $x=ct$ in the sample

$$V(t) = [1-(\epsilon/\epsilon')(1+\chi p)]\Phi(x)$$

$$=\epsilon\delta_\epsilon\Phi(x) \ . \tag{29}$$

Equation (29) corresponds to Eq. (1) of Ref. 2, to Eq. (6) of Ref. 3, and to Eq. (1) of Ref. 4, if χ, p, and $\Phi(x=ct)$ are replaced by $-\chi$, Δp, and $V(z_f)$ or $V(z_f,0)$, respectively. For the simplifying assumption (11), Eq. (29) reduces to $V(t)=-\chi p\Phi(ct)$ which is Eq. (2) of Ref. 5 with A and a replaced by $-\chi p$ and c, respectively. In Ref. 5, displacement waves are used for the derivation of response equations which contain only the geometric contribution to the signal, but not the influence of the changing relative permittivity.

2. Charge response

In Eq. (28) the open-circuit voltage $V(t)$ can be replaced by $Q(t)/C_s(t)$ where $Q(t)$ is the charge induced on the electrodes under short-circuit conditions, and $C_s(t)$ is the time-dependent capacitance between the two electrodes (all stray capacitances in the experimental setup have to be included for exact evaluations). This transition from open-circuit to short-circuit conditions is possible, since the open-circuit voltage exactly corresponds to a lack of compensation charges on the electrodes which would be provided under short-circuit conditions.

At the time t the step wave has reached the location $x=ct$ in the dielectric, and the sample part between $x=0$ and $x=ct$ is compressed, thus having the thickness $x'=(1+\chi p)x$ and the relative permittivity ϵ'. Therefore, the capacitance C_1' of the compressed part of the sample is given by

$$C_1' = \frac{\epsilon_0\epsilon' A}{x'} = \frac{\epsilon_0\epsilon A}{(\epsilon/\epsilon')x'} \ ,$$

where A is the sample area. The capacitance C_2 of the noncompressed sample part is $C_2=\epsilon_0\epsilon A/(s-x)$. The capacitance $C_g=\epsilon_0 A/g$ of the rear gap stays constant during the first transit of the pressure step. If all stray capacitances are negligible the capacitance $C_s(t)$ between the electrodes can be calculated, using the formula $1/C_s=1/C_1'+1/C_2+1/C_g$ for a series of capacitors:

$$C_s(t)=\frac{\epsilon_0\epsilon A}{(\epsilon/\epsilon')x'+(s-x)+\epsilon g}\ . \tag{30}$$

The charge $Q(t)$ which builds up on the short-circuited electrodes during transit of the pressure step can thus be expressed by

$$Q(t)=\frac{\epsilon_0\epsilon A[1-(\epsilon/\epsilon')(1+\chi p)]}{(\epsilon/\epsilon')x'+(s-x)+\epsilon g}\times\int_0^{x=ct}E(\xi)d\xi\ . \tag{31}$$

On the other hand, the charge $Q(t)$ is given by the intergral $\int I(\tau)d\tau$ of the current $I(t)$ between the electrodes; the field integral $\int E(\xi)d\xi$ is the potential $\Phi(x)$ at the location x in the dielectric. For a two-sided metallized sample ($g=0$), the integral of the short-circuit current is thus given by

$$Q(t)=\int_0^t I(\tau)d\tau=\frac{\epsilon_0\epsilon A}{(\epsilon/\epsilon')x'+(s-x)}\times[1-(\epsilon/\epsilon')(1+\chi p)]\Phi(x)\ , \tag{32}$$

which is identical to Eq. (3) of Ref. 2 if the correct $Q(t)/A$ is used instead of $Q(t)$ only, to Eq. (8) of Ref. 3, and to Eq. (2) of Ref. 4 [S, $J(t)$, 1, $-\chi$, Δp, $V(z_f,0)$, z_f, and $d-z_f$ are utilized in these references in place of A, $I(t)$, ϵ_0, χ, p, $\Phi(x=ct)$, $s-x$, and $x'=(1+\chi p)x$, respectively].

3. Short-circuit current

Differentiation of Eq. (31) with use of the relation $dx=c\,dt$ and Poisson's equation results in an expression for the short-circuit current $I(t)=dQ(t)/dt$ of pressure-step experiments:

$$I(t)=\frac{Ac[1-(\epsilon/\epsilon')(1+\chi p)]}{(\epsilon/\epsilon')x'+(s-x)+\epsilon g}\int_0^{x=ct}\rho(\xi)d\xi=\frac{Ac\delta_\epsilon}{s/\epsilon-\delta_\epsilon x+g}\int_0^{x=ct}\rho(\xi)d\xi\ , \tag{33}$$

where the charge distribution $\rho(x)$ has to include the compensation charges at $x=0$ and at $x=s$ because the field in the sample is caused not only by the volume charge distribution but also by the electrode charges.

In Eq. (33), the charge-density integral $\int\rho(\xi)d\xi$ can be divided into three parts, namely (1) the compensation-charge density $\sigma_{\rm I}$ on the front electrode (I) at $x=0$, (2) the integral of the real-charge density

$$\int_{0+}^x\rho_r(\xi)d\xi\ ,$$

and (3) the polarization $P(x)=-\int_{0+}^x\rho_p(\xi)d\xi$:

$$I(t)=\frac{A}{s}c\epsilon\delta_\epsilon\left[\sigma_{\rm I}+\int_{0+}^x\rho(\xi)d\xi-P(x)\right]\ ,$$

where $s-\epsilon\delta_\epsilon x+\epsilon g$ has been replaced by s (no air gap and $\epsilon\delta_\epsilon x\ll s$) and ρ_r by ρ. The relative permittivity ϵ' of the compressed sample part can be written as sum of ϵ and an expression in χp, and first-order approximation can be employed as follows:

$$\epsilon\delta_\epsilon=1-(\epsilon/\epsilon')(1+\chi p)\approx\epsilon'/\epsilon-1-\chi p\ .$$

Insertion of this expression and separation of the polarization term yield

$$I(t)=\frac{A}{s}c\left[\left[\frac{\epsilon'}{\epsilon}-1-\chi p\right]\left[\sigma_{\rm I}+\int_{0+}^x\rho(\xi)d\xi\right]-\left[\frac{\epsilon'}{\epsilon}-1\right]P(x)+\chi pP(x)\right]\ .$$

Here, $(\epsilon'/\epsilon-1)P(x)$ is the dipole contribution caused by the change of the relative permittivity, and $\chi pP(x)$ is the dipole contribution resulting from the compression. For piezoelectric materials, the latter can be expressed by $\chi pe(x)$ where $e(x)$ is the piezoelectric strain constant. Using the electrostriction constant

$$\gamma=-\frac{1}{\epsilon}\left[\frac{\Delta\epsilon}{\Delta S}\right]\approx-\frac{\epsilon'-\epsilon}{\epsilon\chi p}\ ,$$

where $S=\chi p$ is the strain in the compressed region of the dielectric, one obtains

$$I(t)=\frac{A}{s}c\chi p\left[-(\gamma+1)\left[\sigma_{\rm I}+\int_{0+}^x\rho(\xi)d\xi\right]+\gamma P(x)+e(x)\right]\ . \tag{33'}$$

This equation can be transcribed into Eq. (2) of Ref. 12 by replacing s, c, χp, x, and ξ with 1, v_s, $-v/v_s$, x_s, and x, respectively. The analysis used in Ref. 12 is based on a general description of piezoelectricity in polymers.[16] This description takes into account real charges, polarization, electrostriction, and piezoelectricity. The transformation of Eq. (33) into Eq. (33') shows that eventual piezoelectric contributions are contained in the formulas of the present study and need not be taken into account separately.

The electric field $E(x=ct)$ can be substituted for

$$\frac{1}{\epsilon_0\epsilon}\int_0^{x=ct}\rho(\xi)d\xi$$

(Poisson's equation) in Eq. (33) because the field $E(0)$ in the front electrode is zero,

$$I(t)=\frac{\epsilon_0\epsilon A c\delta_\epsilon}{s/\epsilon-\delta_\epsilon x+g}E(x)\;, \tag{34}$$

which reduces to Eq. (2) of Ref. 5, if Eqs. (12) and (30), their sound velocity a, and their relative compression $A_0=-\chi p$ are employed.

For polar materials the relative permittivity ϵ' in the compressed region behind the pressure step may not be constant, since some of the permanent dipoles may have relaxation times shorter than the transit time of the step. This eventual time- and depth-dependent change of the relative permittivity ϵ' leads to signal contributions from sample parts that are not actually passed by the step wave. Correct results from such signals can only be obtained by means of a deconvolution process.

4. *Pressure-step experiments on nonpolar dielectrics*

For nonpolar materials, combination of Eqs. (21), (1), (29), and (34) yields the open-circuit voltage

$$V(t)=\left[\frac{4}{3}+\frac{\epsilon}{3}-\frac{2}{3\epsilon}\right]\frac{p\Phi(x)}{\rho_0 c^2} \tag{35}$$

and the short-circuit current

$$I(t)=\frac{\epsilon_0\epsilon A}{s+\epsilon g}\left[\frac{4}{3}+\frac{\epsilon}{3}-\frac{2}{3\epsilon}\right]\frac{pE(x)}{\rho_0 c}\;, \tag{36}$$

respectively; in Eq. (36), the time-dependent term $[4/3+\epsilon/3-2/(3\epsilon)]\chi px/\epsilon$ in the denominator has been omitted because it is very small compared to $s/\epsilon+g$.

B. Pressure-pulse experiments

1. *Short-circuit current*

When a pressure pulse propagates through the sample, the temporal change of the induction surface-charge density [Eq. (5)] produces a current density $i=d\sigma_\mathrm{I}/dt$ and thus a current $I=A\,d\sigma_\mathrm{I}/dt$ flowing to the front electrode (I) with the active area A. Using $dx=c\,dt$, Eq. (5) and the transition from differences Δ to differentials d, the short-circuit current in a pressure-pulse experiment can be calculated as follows:

$$I(t)=\frac{A\delta_\epsilon}{s/\epsilon+g}c^2\tau\rho(x)\;. \tag{37}$$

In this case, the capacitance C_p between the two electrodes is constant because the thickness $(1+\chi p)c\tau$ and the relative permittivity ϵ' of the traveling compression layer do not change. Disregarding the very small change of the "dielectric thickness" of the sample caused by the pressure pulse, the capacitance C_p is given by

$$C_p=\frac{\epsilon_0\epsilon A}{s+\epsilon g}\;. \tag{38}$$

Combination of Eqs. (37) and (38) and use of B_0, C, and a, instead of $-c\tau\chi p$, C_p, and c, respectively, yields, under the simplification of relation (11), Eq. (5) of Ref. 5. Combination of Eqs. (18) and (37) results for $g=0$ (no rear gap) in Eq. (1) of Ref. 11 and in Eq. (3) of Ref. 13.

2. *Open-circuit voltage*

Under open-circuit conditions, the change $\Delta\sigma_\mathrm{I}$ of the induction-charge density given by Eq. (5) is not possible since no charges can flow. Instead, a voltage change $\Delta V=(A/C_p)\Delta\sigma_\mathrm{I}$ between the two electrodes is caused by the lack of compensation charges. In this case, transition to differentials and integration lead to

$$\begin{aligned}V(t)&=c\tau(\delta_\epsilon/\epsilon_0)\int_0^{x=ct}\rho(\xi)d\xi\\&=c\tau\epsilon\delta_\epsilon E(x)\;,\end{aligned} \tag{39}$$

whereby Eqs. (5) and (38), Poisson's equation, and the assumptions $V(0)=0$ and $E(0)=0$ have been used.

Combination of Eqs. (11) and (39) with use of B_0 and a for $-\chi pc\tau$ and c, respectively, results in Eq. (5) of Ref. 5. Equations (18) and (39) can be combined with Eq. (7) of Ref. 6, Eq. (16) of Ref. 7 (the relative permittivity is named K in these references), and Eq. (1) of Ref. 13. The equations of Refs. 6 and 7 were derived from the open-circuit voltage change across the capacitance which is formed by the two-sided metallized dielectric.

3. *Pressure-pulse experiments on polar dielectrics*

For very short laser-induced pressure pulses (LIPP's),[13] the short-circuit current $I(t)$ and the open-circuit voltage $V(t)$ can be calculated from

Eqs. (1), (26), (37), and (39) even for polar dielectrics as follows:

$$I(t)=\frac{A}{s+\epsilon g}\left[1+\frac{(\epsilon_\infty+2)(\epsilon_\infty-1)}{3\epsilon}\right]\times\frac{p\tau\rho(x)}{\rho_0} \tag{40}$$

and

$$V(t)=\left[1+\frac{(\epsilon_\infty+2)(\epsilon_\infty-1)}{3\epsilon}\right]\frac{p\tau E(x)}{\rho_0 c}\ . \tag{41}$$

For nonpolar materials, these equations also hold true, since use of $\epsilon=\epsilon_\infty$ transforms Eq. (26) into (21).

C. Arbitrary pressure profiles $p(t-\tau')$

If the pressure wave used for the probing of the charge or field distribution is neither a step nor a pulse, the response can be calculated by means of a convolution integral over the product of the unknown distribution and the pressure profile $p(t-\tau')$ of duration τ. For this purpose the pressure dependence of the relative permittivity ϵ has to be known.

In Ref. 9 the assumption (11) is used for simplicity; thus, following Eqs. (12) and (39), the open-circuit voltage response to a pressure wave of arbitrary profile is given by

$$V(t)=-c\chi\int_{t-\tau}^{t}E(\xi=c\tau')p(t-\tau')d\tau'\ , \tag{42}$$

which is identical to Eq. (2) of Ref. 9 if $-cd\tau'$ is replaced by dz. Similarly, combination of Eqs. (18) and (39) yields Eq. (8) of Ref. 6 and Eq. (17) of Ref. 7.

Combining Eqs. (12) and (37), integrating the result, and assuming a constant sample thickness s during propagation of the pressure wave, the charge $Q(t)$ flowing in the external circuit can be determined as follows:

$$Q(t)=-\frac{\epsilon_0\epsilon A}{s}c\chi\times\int_{t-\tau}^{t}E(\xi=c\tau')p(t-\tau')d\tau'\ , \tag{43}$$

which corresponds to Eq. (4) of Ref. 9, if the notation is changed accordingly.

More accurate formulas for the current and voltage responses to arbitrary pressure-wave profiles can be obtained by using Eqs. (1), (21), (37), and (39),

$$I(t)=\frac{A}{s+\epsilon g}\left[\frac{4}{3}+\frac{\epsilon}{3}-\frac{2}{3\epsilon}\right]\frac{1}{\rho_0}\times\int_{t-\tau}^{t}\rho(\xi=c\tau')p(t-\tau')d\tau'\ , \tag{44}$$

$$V(t)=\left[\frac{4}{3}+\frac{\epsilon}{3}-\frac{2}{3\epsilon}\right]\frac{1}{\rho_0 c}\times\int_{t-\tau}^{t}E(\xi=c\tau')p(t-\tau')d\tau'\ . \tag{45}$$

Equation (45) corresponds to Eq. (5) of Ref. 10 if the necessary changes of the notation are made.

The actual charge or field distribution can only be determined by a numerical deconvolution procedure for which the exact shape of the pressure profile $p(t-\tau')$ must be known. Despite the fact that the deconvolution yields a unique solution in principle,[9] the accuracy of the deconvolution result is limited by the experimental and numerical errors. This problem is analogous to the reported[17] influence of measurement errors on the evaluation of heat-pulse[1,18,19] experiments. For laser-generated pressure waves the reproducibility is rather poor, and therefore the deconvolution procedure[9] should be avoided which is possible even for thin dielectrics if very short and energetic LIPP's are used.[13] In addition, the response to pressure profiles can become very complicated because of partial reflections at the sample surfaces.

IV. CONCLUSION

The current or voltage response of a charged dielectric during the transit of a pressure wave was analyzed starting from a simple geometrical model and including the pressure dependence of the relative permittivity. This pressure dependence was calculated for nonpolar and for polar materials using the Clausius-Mossotti equations for the total polarization and for the induced polarization, respectively. From the results the dependence of the signal amplitude on the relative permittivities at zero frequency and at the frequency of the experiment was derived. It could be shown that all existing theories[3,5,7,10,12] can be derived from the given simple formulation of the problem if the different assumptions on the pressure dependence of the relative permittivity are taken into account.

The detailed analysis revealed that all methods using pressure steps or pressure profiles involve the possibility of complex signal contributions from sample parts not actually passed by the leading edge of the pressure wave. These contributions are caused either by slow (compared to the temporal resolution of the experiment) changes of the relative permittivity or by partial reflections at the sample surfaces. To avoid these difficulties as well as the problems of a deconvolution procedure, very short pressure pulses should be used for the determination of spatial charge distributions in dielectrics.

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