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Master in Photonics

MASTER THESIS WORK

Evaluation of the LIPP method for determining the space charge profile in poled dielectrics

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Presented on date 11th december 2013

Registered at

ETSEP Escola Tècnica Superior
d'Enginyeria de Telecomunicació de Barcelona

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Abstract: The determination of the charge profile in corona-poled or electrode-poled samples is a key step in many dielectric studies. The purpose of this work is to evaluate the feasibility of developing a laser induced pressure pulse (LIPP) setup and to compare its performance with commercial techniques, like the electro-acoustical pulse technique (PEA). Samples of several materials (PTFE, PI, PEI and PET), with thicknesses ranging from 25 μm to 320 μm , have been poled using corona poling and electrode poling. The charge profile has been measured with LIPP and PEA, in two sample configurations: single and paired. LIPP has given in some cases more detailed results as PEA. Both methods yield coherent results for samples from 50 μm onwards. LIPP is able to determine the charge profile not only of space charge but also of induced charge by dipolar polarization.

Keywords: Laser Induced Pressure Pulse (LIPP), Electro-Acoustical Pulse (PEA), PTFE, PET, PEI, PI.

Introduction

Poled polymers (electrets) have many applications [1] Usually, in sensors and in photonics [2], the poled polymers belong to the oriented molecular dipole class of electrets even though space charge electrets can also be used.

For the application of poled polymers it is interesting to determine the charge profile [3]. For the detection of space charge or induced charge there are several techniques and methods that are available [4]. Among them, we can mention Electro-Acoustical Pulse (PEA) [5], Thermal Step Method (TSM) [6] and Laser Induced Pressure Pulse (LIPP) [7] [8] [9].

The LIPP is a pressure-wave method [10]. In this method, one side of the sample is covered with a target and irradiated with a short laser pulse. The target absorbs the energy of the laser pulse and the rapid expansion that results produces a brief but intense pressure pulse in the dielectric. When the compressed region pulse travels through the sample a current is induced in the external circuit, due to non-uniform charge distribution and permittivity change, reflecting the charge distribution. Connecting this setup with an oscilloscope the profile can be obtained. One of the advantages of the LIPP method with regards to other methods is that it produces pressure pulses with steeper rise fronts, which increases the spatial resolution of the system.

There are many ways in which the samples can be poled. Corona poling [11] is controllable and thus it can be easily used as a source of thermalized ions in order to pole dielectrics. This work has been performed mainly with corona-poled samples [11]. These samples have the problem that poling tends to decay [12, 13]. The LIPP method is interesting because it can be used to determine space charge dynamics in charged samples [14]. For example, the dynamics can be used to assess the degradation of a dielectric [15].

The LIPP method has been used over the years extensively by their creators, Sessler and Gerhard, in their investigations of charge profile in thin-film polymer materials [16]. Now it should be possible to

reproduce it on modern equipment with even better results, because of the improvement in technology.

The aim of this work is developing a LIPP setup using modern equipment and to evaluate its performance. If we combine this with modern data acquisition and treatment techniques it could be possible to build an in-house LIPP system whose performance is on par with commercial systems such as PEA.

Experimental

In this work we have used several kind of materials with different thicknesses and electrode configuration: PTFE (Teflon): 25 μm and 50 μm , aluminum electrode on one side, PET: 320 μm , aluminum electrode on one side and on both sides, PEI (Ultem), 175 μm , aluminum electrode on one side and PI (Kapton), 75 μm , aluminum electrode on one side. All samples were cut in 2.5 cm x 2.5 cm squares from as received sheets.

The aluminum electrodes were vacuum deposited on the samples. They had 2cm diameter and were deposited at the center of each sample. Samples for space charge profile have an electrode on one side and samples for polarization profile have electrodes on both sides.

Because we wanted to shine a laser on our samples, we placed a thin layer of a light absorbing material as a target on the metalized side of the samples. The target is made to absorb the energy. As a consequence it gets depleted and produces a pressure pulse on the sample, of a duration that is approximately the same as the one of the laser pulse. Two kinds of targets have been proved: A semiconducting blend of EVA with carbon black and a conductive graphite layer. The semiconducting target was discarded because it did not give sharp pressure pulses. Graphite was received as a spray tube and the weight of the layer on top of the samples was approximately 7 mg, with thickness of the order of $10^1 \mu\text{m}$.

PTFE, PEI and PI samples have been poled by the corona triode method [11]. The needle of the corona triode was kept at 9 kV during the charging process (3s) while the grid was grounded and the rear side of the sample was kept at -2 kV. The distance between the sample and the grid was 8 mm. The gap between the grid and the needle was 25 mm. Voltage measurements after corona charging were carried out with a Trek model 347 electrostatic voltmeter. All samples were charged up to -1.6 kV. After poling the corona poled samples acquired a unipolar charge distribution of negative charge. PET samples were poled by electrode poling with a 1 kV voltage. The sample metallized on both sides was poled at 78 C while the sample metallized on one side was poled at 85 C. In both cases the sample was poled for 20 min and cooled 25 C at a rate of 1 C/min before removing the electric field. One of the problems with corona poling is that most of the charge is close to the surface of the sample. This condition makes the charge of the sample very sensitive and it may be discharged easily, so we have to be careful when manipulating the samples.

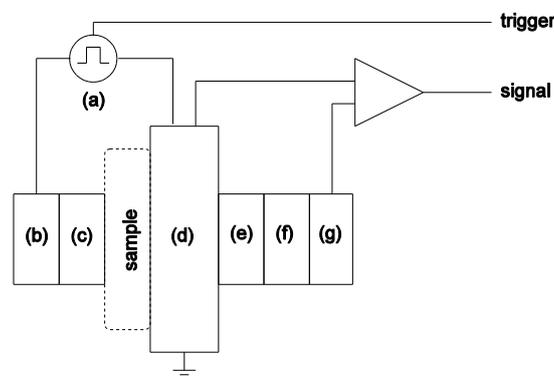


Figure 1: PEA setup: (a) pulsed source, (b) anode (Cu), (c) backing material, (d) ground electrode / cathode (Al), (e) piezo device, (f) absorber and (g) back electrode.

After charging each sample we pretend to measure its charge profile. In fact we have determined the charge profile using two methods: LIPP and PEA.

The PEA experimental set-up used in this work was a commercial PEA equipment "Techimp PEA system". Data were recorded by means of a "Tektronix TDS 5032" digital oscilloscope. The pulse amplitude was 300 V and the pulse length was 40 ns. Charge distribution measurements were performed in a 1 cm circular section at the middle of the sample.

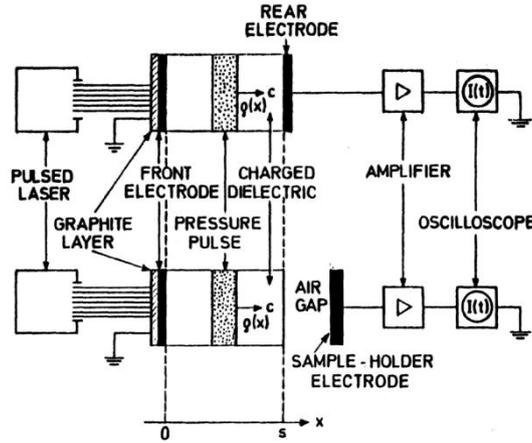


Figure 2: Scheme of LIPP setup (from reference [8]).

Instead, in the LIPP method we place the sample in a sample holder and we shine the laser on the sample for detecting the charge distribution. The laser that is used is Nd:YAG with a pulse energy up to 50 mJ, even though we have not reached this value in our experiments. Unless otherwise stated, 15 mJ were used in the experiments. The duration of the laser pulse is 8 ns and the frequency of the shots has been set so it is easy to manually interrupt the beam to control the number of shots received by the target. The laser beam is focused with several lenses before it reaches the sample and gets absorbed by the target. The target then delivers a pressure pulse that where ever it finds the charge gives an electric signal that we can see in the oscilloscope. For increasing the magnitude of this signal we connect to a FEMTO DHPCA high speed variable gain amplifier to our sample holder that enlarges the signal coming from the charge profile and also acts as an impedance adaptor. The amplifier has a capacitance that grows as a larger gain is selected. Rise time is 2 ns, 4.4 ns and 25 ns for 10^2 , 10^3 and 10^4 gain respectively. The setup has been shielded to avoid interference from the Pockels cell. A fast light detector has been used to trigger the oscilloscope.

The intensity of the short circuited sample should be [17]

$$I(t) = -(2 - \epsilon_r^{-1})x \frac{A}{B} c \int_0^s P(ct - z) \rho(z) dz$$

where ϵ_r is the relative permittivity of the sample, x is the compressibility A is the surface of the electrode, S is the thickness of the sample, c is the speed of sound, $P(x)$ is the profile of the pressure pulse and $\rho(x)$ is the charge profile. When the length of the pressure pulse is short with regards to the charge profile, we can approximate this expression by

$$\sim -(2 - \epsilon_r^{-1})x \frac{A}{B} c^2 \tau P \rho(ct)$$

where τ is the duration of the pressure pulse.

As it is mentioned before, corona poled samples may get discharged very fast. One of our solutions for this is to make a "sandwich" of samples (paired configuration). In this configuration we put the non-metallized side of the two samples together so the samples do not get discharged with manipulation. Samples are measured in paired configuration and alone (single configuration). Paired configuration is particularly useful to calibrate the speed of sound in the material, identifying the place where the boundary between the surfaces is placed.

LIPP measurements that have produced damage on the metallized electrode have been discarded since there is an optoelectric effect when the laser light enters into the material. The number of shots before the target is depleted is around 20 with the graphite target.

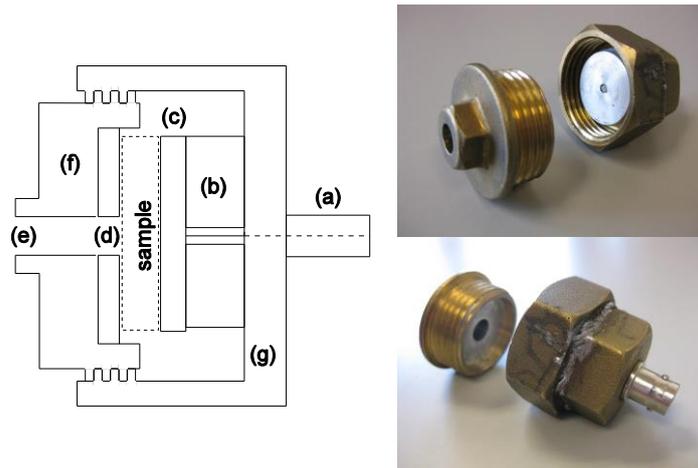


Figure 3: LIPP sample holder: (a) BNC connector, (b) Teflon padding, (c) signal electrodes, (d) ground electrode, (e) pupil, (f) inner body and (g) outer body.

As it is shown in figure 3, the sample holder that we made, is formed by two parts, one part has a circular shape with 2.5 cm radius. This part is connected to the oscilloscope. After putting the sample it will be closed with a head that has small pupil for the laser beam. The side of sample that is covered by graphite has to be heading to the pupil. The capacity of the sample holder has been measured with an impedance analyzer and it has been found to be 0.15 nF.

Results and discussion

Two different configurations were used for corona-poled sample measurement: single and paired configuration. In all figures the ground electrode is placed at the left. Different samples are used for LIPP and PEA even though they have been treated in the same way.

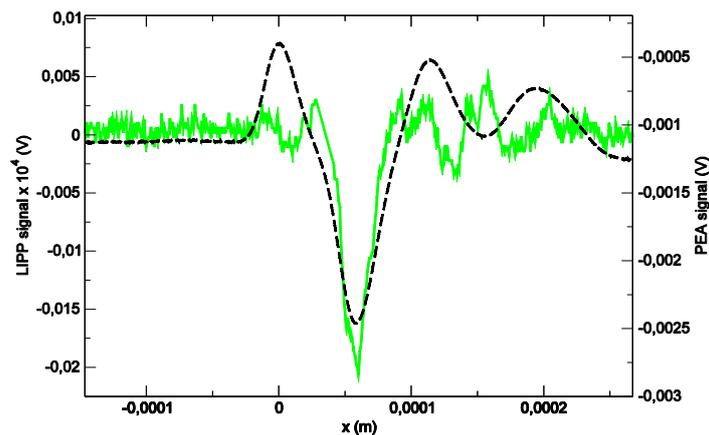


Figure 4: Comparison of LIPP (continuous line) and PEA (dashed line) results for PTFE 2x50 μm samples.

In figure 4 we present a pair of 50 μm PTFE samples measured in paired configuration by LIPP and PEA. The signal coming from the LIPP method is noisier due to internal noise from the oscilloscope and also electromagnetic noise from the Pockels cell.

For both signals, time has been converted to length taking the speed of sound in PTFE as 1400m/s [18]. In the LIPP method the signal has been amplified by a factor 10^4 . There is reasonable correspondence between the two signals, PEA and LIPP. They seem to measure the same charge profile because the shape of the diagrams is similar. We can see the electrodes at the positions 0 and 0.0001 m and the boundary between the samples at 0.00005 m. Electrodes are not visible on LIPP.

This is one advantage of the method but it makes more difficult to interpret the position of the electrodes without additional information.

There is a large symmetrical negative peak that represents the charge injected during corona poling. This peak is centered at the boundary between the samples. This indicates that most of the charge is at the surface of the samples.

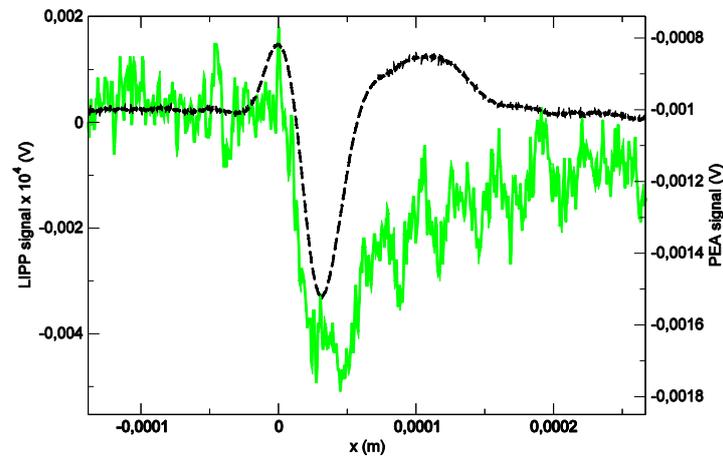


Figure 5: Comparison of LIPP (continuous line) and PEA (dashed line) results for PTFE 25 μm samples.

In figure 5 we illustrate the limits of both techniques measuring a very thin PTFE sample of just 25 μm . Both techniques give a charge profile that is wider than the sample size. This happens because the width of the pressure pulse is comparable to the time it takes to travel through the width of the sample. In the case of LIPP the width of the pressure pulse is about 8 ns while the PEA method uses a pulse of 40 ns.

The figure also shows a limitation due to the current amplifier. Since amplification is 10^4 , the capacitance of the amplifier can be noticed as slow signal decay.

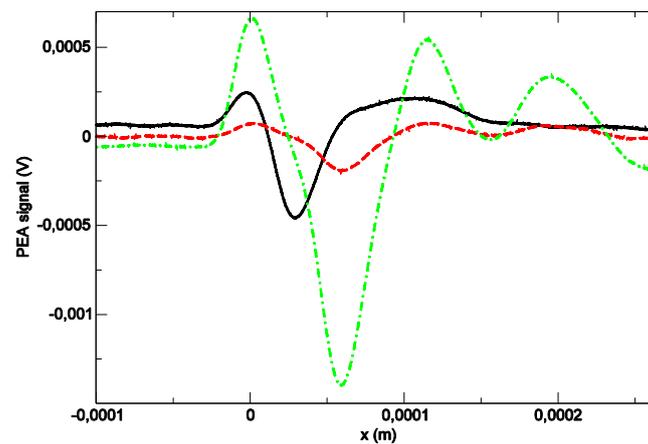


Figure 6: Comparison of PEA results for PTFE samples of three different thicknesses 25 μm (continuous), 50 μm (dashed), 2x50 μm (dot-dashed).

The previous figures show paired samples and a single sample respectively. An important question is how results depend on the configuration of the sample. Figure 6 tries to address this question. It can be seen in this figure that the sharpness of the paired samples is more pronounced than the one of the other samples. This happens because the paired sample has kept more charge inside and we did not have depletion of the charge. The length of the signal is coherent with the thickness of the samples. PEA has a problem with single samples. Charge is so close to the surface that the signal is overlapped with the one coming from the electrodes. From this figure and the previous one, it seems clear that we

can work with paired samples configuration, which is easier to manipulate, taking advantage of a stronger signal.

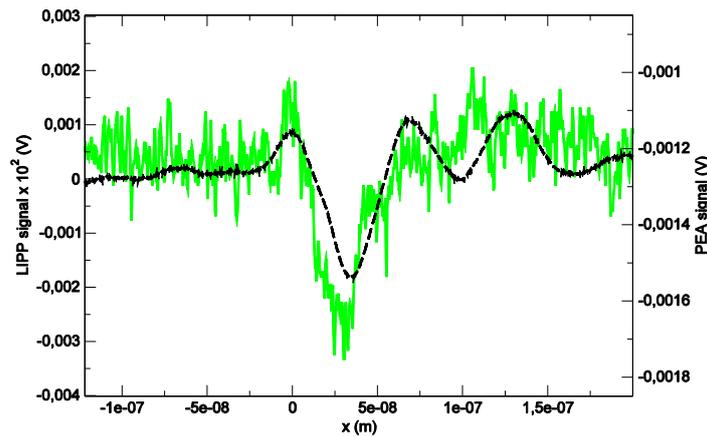


Figure 7: Comparison of LIPP (continuous line) and PEA (dashed line) results for PI 75µm samples.

Figure 7 presents results with a single PI sample. PI has turned out to be a good material to work with LIPP because it delivers a more intense signal than PTFE. In this case, an amplification of just $1e2$ can be employed giving a faster response. Also, it should be mentioned that for PI LIPP seems to yield better results than PEA, where we can only see the position of the electrodes and some reflections. We have used speed of sound 2600m/s [19] to convert time to length even though this value seems to be somewhat high.

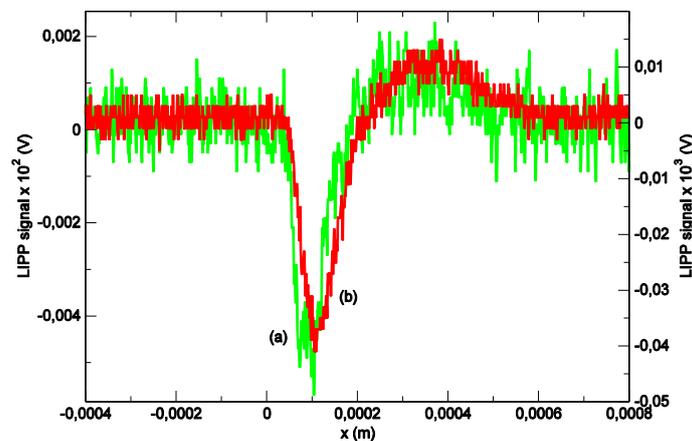


Figure 8: Comparison of LIPP results obtained with (a) 10^2 amplification and (b) 10^3 amplification. PI samples $2 \times 75 \mu\text{m}$ samples were employed.

Another interesting question is which the effect of the amplification is over the signal. PI is idoneous because it does not need too much amplification and therefore we can explore how it affects the signal. Figure 8 shows two different amplifications at a scale at which they can be compared. 10^2 Amplification shows a sharper rise and decrease of the charge profile. It also seems to show more detail, like a split in the peak at the boundary between the samples. The drawback is that the limited resolution of the digital acquisition means that there is less resolution. Also there seems to be more noise. The 10^3 amplification has less noise and more resolution but shows less detail due to integration of the signal by the capacitance from the amplifier, which is greater as the amplification is larger.

Another material suitable for charge profile tests is PEI. In figure 9 we present results from a rather thick corona poled sample. The speed of sound in PEI is not available in the literature, but from this

plot it can be estimated in about 1.5×10^3 m/s. Anyway we have preferred to leave the plot in terms of time.

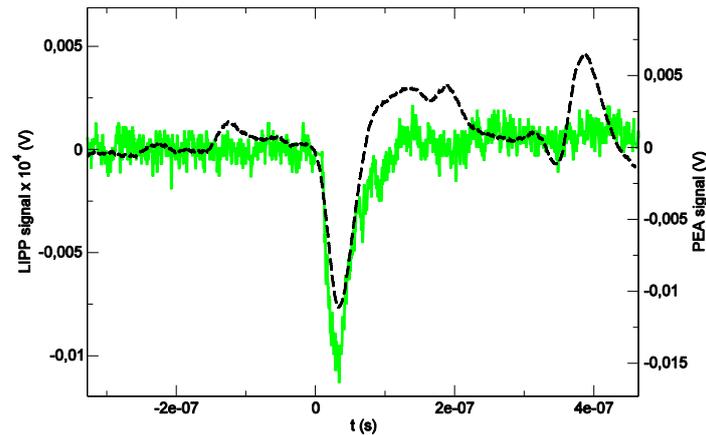


Figure 9: Comparison of LIPP (continuous line) and PEA (dashed line) results for PEI 175 μ m samples.

In PEI space charge seems to have migrated more towards the middle of the sample than in the other materials. This is due to a larger conductivity than other polymers [20].

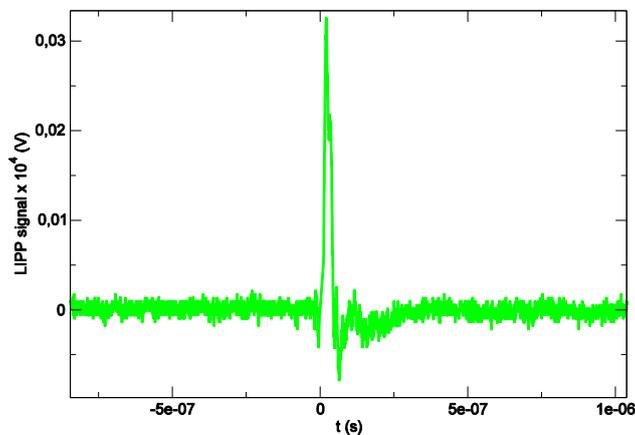


Figure 10: Optoelectric effect capture using the LIPP method in. Results obtained in PEI 175 μ m samples.

When the target gets depleted, the aluminum vacuum deposited electrode is damaged in a few shots and the laser pulse enters into the sample. There is an optoelectric effect that can be seen in figure 10. One interesting fact about this effect is that the voltage is positive. Taking this into account, the most probable cause of this effect is photoconduction due to the laser pulse releasing negative charge carriers from shallow traps that recombine with positive charge from the electrodes. Photoconductivity in PI and PEI has been reported previously [21].

All the results presented until this point refer to corona poled samples. It is desirable to know if we can also detect the charge profile of samples poled by electrode poling, usually at a certain temperature to increase and discriminate the effect of the field. For this reason we have taken PET samples with one and two aluminum vacuum deposited electrodes. In the first case we can expect space charge injection through the electrode free surface while in the second case the charge profile will correspond to induced charge arising from orientation of molecular dipoles and probably also a macroscopic space charge dipole.

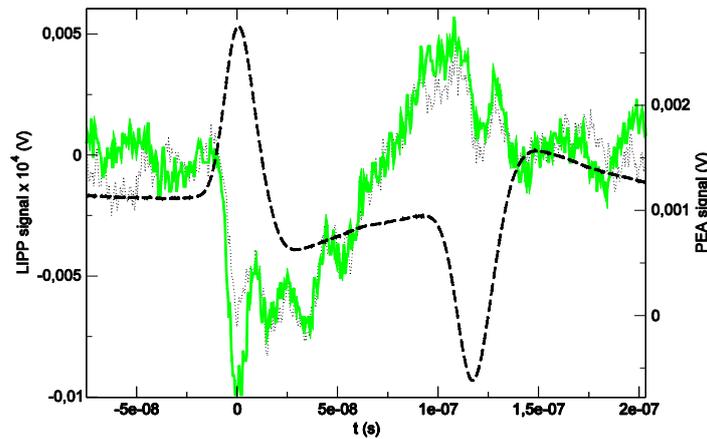


Figure 11: LIPP results (continuous and dotted line) for PET sample charged by direct application of a poling field. Results from a PEA experiment on a sample with the same thickness are also plotted (dashed line) to show the position of the electrodes.

We had no positive results with the one electrode PET sample. Instead, we were able to detect the charge profile with the LIPP method in the two electrode sample. Figure 11 presents these results. It should be noted that electrode poled samples have not been so much studied as corona poled or irradiated samples [16]. Since also the charge profile in this sample was not detected with PEA, we plot LIPP results side by side with a PEA experiment done on a similar sample polarized in the same PEA setup, so we can at least see the position of the electrodes. The plot is also shown in terms of time since there is a discrepancy between the value of the speed of sound in the literature and our own estimation.

The main visible feature should be the induced charge at the electrodes due to molecular dipole polarization. Effectively a sharp signal can be seen at the position of the electrodes, particularly at the first one. Since this is a superficial charge distribution, the signal should have the same shape as the pressure pulse. At the first electrode we can see a peak with a width of approximately 10 ns, which corresponds to the induced charge and has the same duration of the pressure pulse.

At the second electrode, it can be observed a different behavior of the reflection of the pressure pulse in PEA (no phase inversion) and LIPP (phase inversion), probably due to the fact that in the PEA setup the sample is placed in the sample holder over a layer of oil that does not limit the movement of the surface.

The signal between the electrodes is difficult to interpret. In fact, there seems to be structure in the sample since some peaks are repeated on different runs. The resolution of the technique is between 10 and 20 μm which is more or less the size of the features revealed by the peaks.

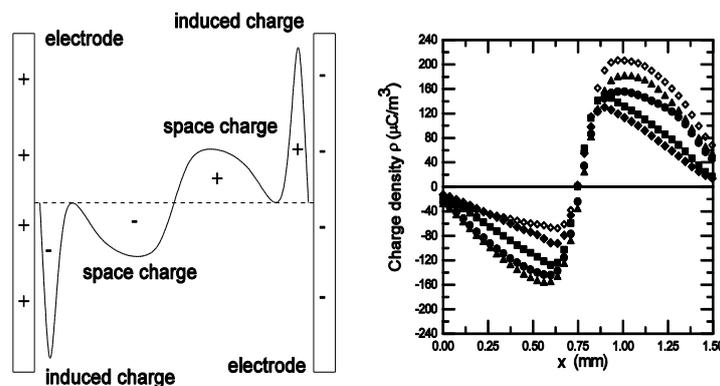


Figure 12: (a) Interpretation of PET LIPP results. (b) Charge profile measured by the thermal step method in PMMA poled in the same way as a PET sample, from reference [22].

A possible interpretation of the signal between the electrodes is given in figure 12(a). In this figure we plot a macroscopic space charge dipole formed at the same time that the molecular dipole polarization. We have found charge profile results using another method [19], the thermal step method that supports this interpretation. These results are plotted in figure 12(b) and were obtained in another material (PMMA) with a different thickness but nonetheless show that this interpretation is feasible. Alternatively these results could be explained if one assumes that the molecular polarization inside the sample is not homogeneous.

Conclusions:

We have succeeded in developing a working LIPP setup and we have evaluated its performance. It has been very useful to have a PEA setup available so that we had an alternative method to determine the charge profile. We have determined the charge profile in several materials and thicknesses. The method has yield useful results between 50 μm and up to at least 320 μm . The determination of charge profile in 25 μm is limited by the resolution of the LIPP method which is about 20 μm in most polymeric samples (speed of sound of the order of 2×10^3 m/s).

We have also worked with different types of charge profiles such as space charge and induced charge. In some cases, LIPP has provided more resolution than PEA and certainly it has the potential to give more detailed results since its pressure pulse is four times shorter than the one of PEA. Also it does not detect the charge at the electrodes giving a cleaner signal from the sample. As a drawback, data is considerably noisier due to interference from the Pockels cell and to a less capable acquisition system. When the surface is touched 80% of the charge of the sample is lost. Even manipulation the paired samples they can get them discharged. Anyway, paired samples seem to be more robust and to yield a better signal. The target is also critical. It has to deliver the appropriate pressure pulse and have to be strong enough to withhold several laser shots. If it gets depleted too fast, the aluminum electrode is damaged and the laser enters inside the sample, giving an optoelectrical effect that masks the signal we are looking for. We have tried two different kinds of targets and found that the graphite target is the most appropriate.

The use of low frequency shots together with manual interruption of the beam and the use of the "single" option in the oscilloscope allows us to control the number of shots needed in a better way than with ancient analog oscilloscopes that required a periodical signal. This gives us more flexibility in our experiments.

Another point to take into account is amplification. High amplification reduces bandwidth, and too little amplification reduces resolution. Therefore a compromise between both requirements has to be adopted.

The amplifier not only does enlarge the signal but also acts as an impedance adaptor. Several improvements can be made to the setup such as trying an acquisition system with more resolution and less noise and filtering the signal, finding a more durable target that gives more predictable results and, in general, improving reproducibility of data, which is highly dependent on the state of the target. Also there is room to understand better how results depend on the target and to improve manipulation of samples so the charge is not modified before measurement.

This work can be continued in many promising ways. LIPP results can be used to complement many research works. Charge profile of corona charged samples can be used to give deeper insight on results from voltage decay experiments. On the other hand, samples charged with direct application of a poling field could be discharged by TSDC to correlate the charge profiles with the peaks in the TSDC thermo gram.

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