Use of scanning LIMM (Laser Intensity Modulation Method) to characterise polarisation variability in dielectric materials

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Abstract. The Laser Intensity Modulation Method (LIMM) has traditionally been used to characterise the depth dependence of polarisation of piezoelectric materials. Although the technique is simple, it is difficult to extract the polarisation / depth data from the measured pyroelectric current because of the complex mathematics pertaining to the physics of the technique. However, the laser probe may still be used as a comparative or qualitative tool in mapping out the polarisation across the surface of a material. A novel scanning LIMM system has been developed to map the variation in piezoelectric activity across a range of samples. The system has been upgraded with a galvanometer mirror scanner to increase speed and reduce sensitivity to acoustic noise. The improvements are discussed and tested on a range of case studies. The technique can be used to show differences in piezoelectric activity caused by features such as inhomogeneous material composition, porosity and mechanical damage. The method has application as a quality control tool for materials and device manufacturers.

1. Introduction
The laser intensity modulation method (LIMM) was developed in the early 1980’s as a method to characterize the through thickness polarization of polymeric ferroelectric thin films. In this method an intensity modulated radiation source is used to generate a harmonic thermal wave in a sample, via an absorbing electrode, which in turn generates a pyroelectric current in the material. Because of the time constant of this thermal wave it can sample to different depths, depending on the frequency of modulation and the thermal diffusivity of the material. Most work has concentrated on quantifying the results of the experiment, and because of the complexity of this task workers have tended to use the method in a one-dimensional mode. Recently the method has been used in a more qualitative manner to get three-dimensional information from the technique. In the scanning LIMM method a focused laser beam must be moved across the test sample surface, and until now this has been achieved by moving the sample on an x-y table. This traveling stage can introduce acoustic noise into the measurements, particularly when examining piezoelectric ceramic samples with a relatively high mechanical quality factor. In this work we report for the first time the use of galvanometer mirror scanners to scan the beam across the sample.

The generation of current in the scanning LIMM technique is also more involved than in the one-dimensional case. In general, thermally generated currents are termed pyroelectric currents, and for a ferroelectric material the measured pyroelectric coefficient is given by the following.
where the first term is the pyroelectric current measured at constant strain, termed the primary pyroelectric effect, \( p_i^e \), and the second term is current produced from piezoelectric effects via thermal expansion. In the scanning LIMM technique there are further possibilities, such as where thermal expansion in an inactive part of the sample causes a thermoacoustic wave, which is then picked up piezoelectrically in the sample. All these mechanisms can lead to a signal, so quantitative evaluation is difficult, nevertheless as will be shown the technique can produce useful qualitative data.

The harmonic heat flux problem can be solved analytically for the one-dimensional case, where the laser beam diameter is much broader than the penetration depth, to give the temperature at any depth. From this it can be shown that the penetration depth of the thermal wave, \( x \), depends on the angular frequency, \( \omega \), and the thermal diffusivity, \( \alpha \), as follows

\[
x = \sqrt{\frac{2\alpha}{\omega}}
\]

The thermal diffusivity of the material studied here, Lead Zirconate Titanate, PZT, is \( 4.5 \times 10^{-7} \text{ m}^2/\text{s} \), which means the wave will penetrate 0.38mm at a frequency of 1Hz and only 3.78 \( \mu \text{m} \) at a frequency of 10kHz.

2. Experimental
A schematic of the LIMM system developed at NPL is shown in figure 1. The system is based around a 785nm, 75mW diode laser with an analogue input to control laser intensity that is capable of modulation frequencies of up to 1MHz. The analogue modulation is controlled by the oscillator output of a Stanford Research Systems SR830 Lock-in amplifier, which is also used to acquire the pyroelectric current. A FEMTO DLPCA-200 current to voltage converter is used to process the pyroelectric signal before it is sent to the lock-in amplifier. The sample stage is a stepper motor, Danaher Precision x-y stage, capable of 50mm travel with a step size of 1 \( \mu \text{m} \). In the present system the laser is mounted horizontally on the side of the case and the beam is deflected through 90º by the x-y mirror scanner before reaching the sample. The scanners are Cambridge Technology 6210 galvanometer scanners with 3mm mirrors, and controlled by a PositionPro-2 hardware controller. The

Figure 1 Schematic of scanning LIMM system.
angular range of the scanners is ±20 degrees, with a step size of 1.14 μm. The instrument enclosure is a metal box that is interlocked for the purpose of laser safety, and also acts as a Faraday cage to mitigate the influence of external electromagnetic fields on the sensitive current measurements. The system is mounted on vibration dampening rubber feet, but as the size of the box has been increased to accommodate the necessary components it has become more susceptible to airborne noise. This aspect of the performance could be improved by adding an extra acoustic isolation layer around the enclosure.

3. Results and Discussion

3.1. Thermal Modelling

To help to understand the physical processes in the LIMM experiment, a 1-D thermal finite volume model, TherMOL\textsuperscript{12}, was constructed for a 0.15mm thick sample of a PZT material illuminated with a laser. Figure 2 shows the modeled temperature rise at two depths on the sample, the front surface where the laser is incident, and the back surface. These are also shown for two different angular frequencies of the intensity modulation of the incident laser radiation. The incident laser is modeled as a sinusoidal heat flux, which is applied as a boundary condition, that is, all the laser radiation, (modified by a suitable absorption coefficient) is converted to heat in an infinitely thin layer on the top surface. In reality this may not be the case since this depends on the transparency of the absorbing electrode to the incident radiation. The temperature rise on the front surface shows two effects, a slow temperature rise which will eventually reach a steady state, and on top of this a higher frequency temperature modulation at the frequency of the incident radiation. The slow temperature rise is independent of the modulation frequency as it is based on the average laser intensity, and although this does give rise to a pyroelectric charge this component is removed by use of lock in techniques. The higher frequency component is measured using the lock-in and for a particular frequency and other experimental conditions this is constant over time. This implies that in the experiment the material under test should not have temperature dependent, or time dependent properties. The temperature rise at the back surface also shows a slow temperature rise, but for the low frequency case the amplitude is

![Figure 2](image_url)

Figure 2 Finite volume 1D thermal model of temperature rise of 150 μm thick PZT sample with a harmonic heat flux applied to the front surface.
reduced and there is a large phase shift, whilst in the high frequency case there is no visible modulation. This illustrates the depth sensitivity of the LIMM technique, at high frequencies the signal is generated from the top surface, and reducing frequency samples through more of the thickness.

3.2. Scanning System

One of the original aims of adding the mirror scanners to the system was to reduce the sensitivity to acoustic noise pickup. Figure 3 shows the pyroelectric current over time whilst moving either the x-y stage or the beam every ten seconds. Moving the x-y stage generates a sharp pulse in the signal every time the stage moves, whilst moving the beam with the mirrors does not. Secondly there is additional noise generated by the x-y stage due to the energizing of the stepper motor coils needed to maintain position. This can be seen when the motors are turned off after forty seconds in the ‘move stage’ experiment, and is entirely missing from the ‘move beam’ experiment as the motors were off.

Although it would appear that by changing to moving the beam, rather than moving a mechanical stage, the acoustic noise of the stage has been eliminated, the use of the mirror scanners has revealed a further artifact of the scanning LIMM method. The effect, which is similar to the acoustic noise problem, is a consequence of a pyroelectric signal only being generated by a change in temperature. This implies a constant beam focused in one spot, will after thermal equilibrium is reached, not generate any further current. If however the focused beam is moved to another position, as in a scanning system, then the original spot will see a sharp drop in temperature and the new position a sharp rise leading to a current. As they are opposite in sense they will tend to cancel out, but this effect has been seen in scanning LIMM measurements and is most prominent when the step size of the scan is larger than the thermal diffusion zone of the beam.

Overall the addition of the mirror scanner has led to an improvement in the speed of the system of roughly 5 fold. Typically in the previous system when moving the stage it was necessary to wait for 500ms after each step to be sure that any transient noise in the system had time to settle. In theory moving the stage at a constant velocity and matching the acquisition time accordingly could reduce this transient. However as has been shown, even energizing the stage introduces a small amount of noise which is not seen when using the mirror scanners.
3.3. Scanning Results

To illustrate the use of the system a series of scans were performed on typical piezoelectric materials and devices.

3.3.1. Piezo Composite Material.

Figure 4 shows a scanning LIMM image of a piezo composite sample. The composite consists of 250\(\mu\)m diameter piezoelectric fibres embedded in an epoxy matrix. The regions of high pyroelectric activity are piezoelectric fibres seen end-on, whilst the epoxy is inactive. The scanned images consist of 80 by 80 points at a step size of 50\(\mu\)m and took approximately 30 minutes to acquire. The depth sensitivity of the technique can be seen in the different frequency images, one at 1kHz, which samples approximately 12\(\mu\)m and one at 100Hz which samples to a depth of approximately 38\(\mu\)m, with the sample thickness being 1mm. The lower frequency image shows the piezoelectric fibres more clearly, and some are not visible at all in the high frequency image. This could be because either the fibre is not piezoelectrically active, or more likely that there is a thin layer of epoxy covering the face of the fibre that the high frequency thermal wave cannot penetrate. The sample has gold electrodes both top and bottom which are needed first to pole the sample and then later to use the device. One of the issues with piezocomposite materials is the difficulty of generating a field across the piezoelectric material in the presence of an insulating polymer. This technique offers a way of examining which areas of the sample have had a field high enough to pole the piezoelectric.

![Image of scanning LIMM image of piezo composite material](image)

3.3.2. Piezoelectric multilayer with artificial defects.

In a piezoelectric multilayer layers of piezoelectric material are alternately sandwiched with metal electrodes in order to reduce the operating voltage whilst still maintaining a high field. Figure 5 shows the top surface of a 15mm square, 3mm thick, piezoelectric multilayer which has model defects introduced in the form of 2.5mm diameter circles patterned as the number five on a dice. The piezoelectric layers are stacked in the plane of the image and the electrical contacts to the layers are taken off at the left and right of the sample. In this case the LIMM image is formed where the top absorbing layer acts only as a light to thermal converter, and does not form the collecting electrode. The material is slightly transparent in the visible spectrum and so not all of the heat is produced directly at the top surface. The two images taken at 100Hz and 1Hz clearly show the reduced output.

![Image of scanning LIMM image of piezoelectric multilayer with artificial defects](image)
from the defect areas, and it can be seen that the defect layers on alternate layers are offset left to right in the direction of the electrical contacts. This is because each metal layer must stop short of going across the full width of the sample otherwise it will short circuit. In the higher frequency image the circles are slightly more defined whereas in the low frequency image they tend to blur together. The high frequency image shows the right hand circles as slightly larger and appear more "in-focus", as this layer is nearer to the top surface. The piezoelectric induced displacement of these multilayers has already been investigated\textsuperscript{13}, and it is interesting to note that the patterns seen were elongated ellipses, as in the low frequency LIMM image, and it was not possible to distinguish two separate layers. This is because the displacement generated comes from the entire thickness of the multilayer, rather than just a few layers in the scanning LIMM results.

3.3.3. Piezoelectric multilayer stack.

Figure 6 shows a scanning LIMM image of a piezoelectric stack, where several individual 2.2mm thick chips have been stacked together, with the electrodes being taken off top and bottom of the image. The image is taken from the side of the stack, and again there is no absorbing electrode, simply a capping layer of PZT for electrical isolation purposes. The individual stacks can be identified and they would appear to show different levels of activity. At the right hand side there is an extra PZT chip without electrodes and this gives almost no signal, whereas the left hand one gives the highest output. This is probably due to the differences in the proximity of the electrodes to the surface rather than a real difference in activity of the piezoelectric material. The individual layers of the multilayer cannot be identified as by the time the thermal wave has reached this depth it has already passed through the capping layer thus degrading the resolution. The image also shows some spots of higher activity at the top and bottom between each stack, (the width of the piezoelectric part of the stack is 7mm). This is an artifact where there are regions of solder on the sample and there is a good thermal and mechanical coupling to the rest of the sample leading to a phantom signal.

4. Conclusions

The scanning LIMM system developed at NPL has been upgraded to include an x-y galvanometer mirror scanner in order to scan the beam across the sample, rather than move the sample with an x-y stage. This has led to improved scanning speeds, with a typical step speed of the scanner of less than

\begin{figure}[h]
\centering
\includegraphics[width=\textwidth]{figure5.png}
\caption{Scanning LIMM image of piezoelectric multilayer samples, left hand image at 100Hz, right hand image at 1Hz. Z scale is current in Amps.}
\end{figure}
1msec, but more importantly less acoustic noise. Overall this has given roughly a 5-fold increase in the speed of the system.

The system has been used to examine a broad range of piezoelectric devices and has been demonstrated to show excellent x-y spatial sensitivity and also the depth sensitivity has been used to highlight differences in samples. Conventionally the LIMM method needs an additional radiation absorbing electrode layer, however it has been demonstrated that as long as there are electrodes present in the device for current collection, then this extra step is not needed. This is useful in practical terms if the technique is to be used as a rapid investigative tool.

5. References

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