# Comparison of techniques for solving the Laser Intensity Modulation Method (LIMM) equation

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**Abstract** The Laser Intensity Modulation Method (LIMM) is a technique for the determination of polarization and/or space charge profiles in the thickness direction of ceramic and polymeric samples. The experimental data are analyzed by means of a Fredholm integral equation of the first kind. This equation admits to multiple and very different solutions. A number of techniques have been developed in order to obtain the most physically reasonable profile. Several techniques based on the regularization method have been proposed. A recent version, polynomial regularization, was developed by Lang. A completely different approach is the Monte Carlo method of Tuncer and Lang. Several sets of both simulated and experimental data are analyzed by the two methods in this paper. Conclusions concerning speed and accuracy are presented.

**Keywords** LIMM · Regularization · Polarization · Pyroelectricity · Monte Carlo

## 1 Introduction

The Laser Intensity Modulation Method (LIMM) experimental technique is as follows [1]. Samples are prepared as thin plates or sheets with their flat surfaces normal to the polar axis. These surfaces are coated with very thin opaque

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electrodes. The surface of the sample is exposed to a laser beam whose intensity is modulated sinusoidally by means of an acoustooptic modulator or a built-in modulator in the laser. The energy of the laser beam is absorbed at the electrode and heat diffuses into the sample as temperature waves. The depth of penetration of the waves is greater for low modulation frequencies and less for high ones. This produces a spatially non-uniform time-varying temperature distribution that interacts with the unknown polarization distribution to produce a pyroelectric current. The real and the imaginary components of the current generated at each of a large number of different frequencies are amplified and a lock-in amplifier is used to determine the amplitude and phase relative to that of the modulated intensity of the laser beam.

The relative polarization distribution, $P(z)$, is found from a solution of the LIMM equation [2–4]:

$$I(\omega) = \alpha_P \frac{A}{L} \int_0^L P(z)\, \frac{\partial T(z,\omega)}{\partial t}\, dz \tag{1}$$

where $I(\omega)$ is the experimentally measured pyroelectric current as a function of frequency; $\alpha_P$ is the temperature dependence of the spontaneous polarization; $A$ and $L$ are the sample area and thickness; and $\partial T/\partial t$ is the time dependence of the temperature as calculated from the one-dimensional heat conduction equation. Space charge has been neglected in this analysis. Eq. 1 is a Fredholm integral equation of the first kind and is an ill-conditioned problem with a large (possibly infinite) number of very different solutions. The most successful of the methods of solving it are various forms of the regularization method and the scale transformation. The regularization method utilizes the requirement that the polarization distribution must be smooth. By specifying the degree of smoothing, extreme minima and/or maxima are excluded but moderate ones are

permitted. However, its usage requires complex computer programs. The scale transformation method is very easy to implement [3]. It is less accurate than regularization and it does not allow the determination of the polarization in regions closer to the sample electrode than the thermal diffusion length at the highest measured frequency. Recently, Lang [1] proposed a simplified regularization method called the Polynomial Regularization Method (PRM). Analysis of experimental data requires the selection of an appropriate regularization parameter. A technique called the L-curve Method (LCM) is used for that purpose. An entirely different approach is the Monte Carlo Method (MC) [5]. In this paper, two sets of current versus frequency data are simulated and the PRM and MC methods are applied.

## 2 Data simulation

The sample was assumed to be a 20-μm thick plate of lead–zirconate–titanate ceramic (PZT). Two arbitrary polarization distributions were selected as shown in the left-hand (Example 1) and right-hand (Example 2) columns of Fig. 1(a). They do not represent realistic distributions but were chosen in order to demonstrate the techniques. The real and imaginary components of the pyroelectric current were simulated using Eq. 1 at 256 frequencies logarithmically spaced between 10 Hz and 100 kHz giving 512 values of current. Random errors with a Gaussian distribution and a standard deviation equal to 2% of the range of the data were added to give the simulated data shown in Fig. 1(b).



**Fig. 1** Two examples in left- and right-hand columns (Examples 1 and 2, respectively). (a) True polarizations, (b) Simulated real and imaginary pyroelectric currents as functions of frequency, (c) L-curve plot for determination of optimal regularization parameter, (d) Calculated polarization distributions compared with true values for PRM
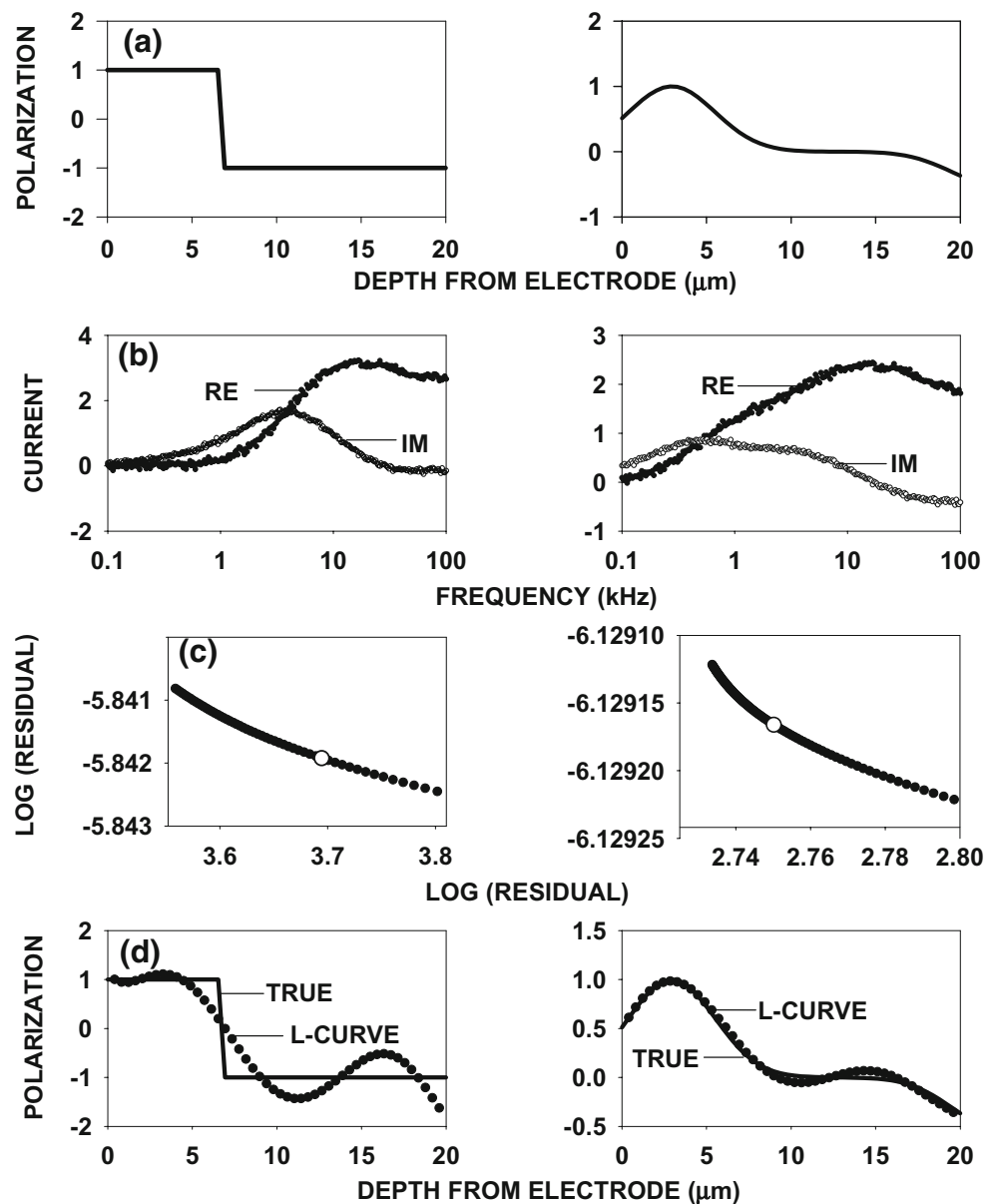
## 3 Polynomial regularization method

The PRM is used as follows:

1.  $P(z)$ is represented by an eighth-degree polynomial in the variable, $z/L$. The coefficients of the polynomial must be determined.
2.  The polynomial is substituted into Eq. 1 and evaluated at each of the 256 frequencies. This results in 512 functions of the coefficients of the polynomial.
3.  The coefficients could be found by a conventional least-squares analysis in which the sum of the squares of the differences between the simulated currents and

those calculated in step 2 is minimized. However, because of the ill-conditioned nature of the problem, this solution will invariably be the wrong one.

4.  Instead, a regularization method is used with a term to enforce smoothness on $P(z)$. The following function is minimized:

$$\sum_i \left[ I(\omega_i)_{exp} - I(\omega_i)_{calc} \right]^2 + r^2 \int_0^L \left( \frac{d^2 P(z)}{dz^2} \right)^2 dz \qquad (2)$$

If the regularization parameter $r$ is zero, a conventional least-squares solution is obtained. If $r$ is too large, all of the detail of the polarization distribution will be lost.



**Fig. 2** Results of Monte Carlo Method for Examples 1 and 2 of Fig. 1(**a**). *Solid lines* are the calculated values and *dotted lines* are the true ones
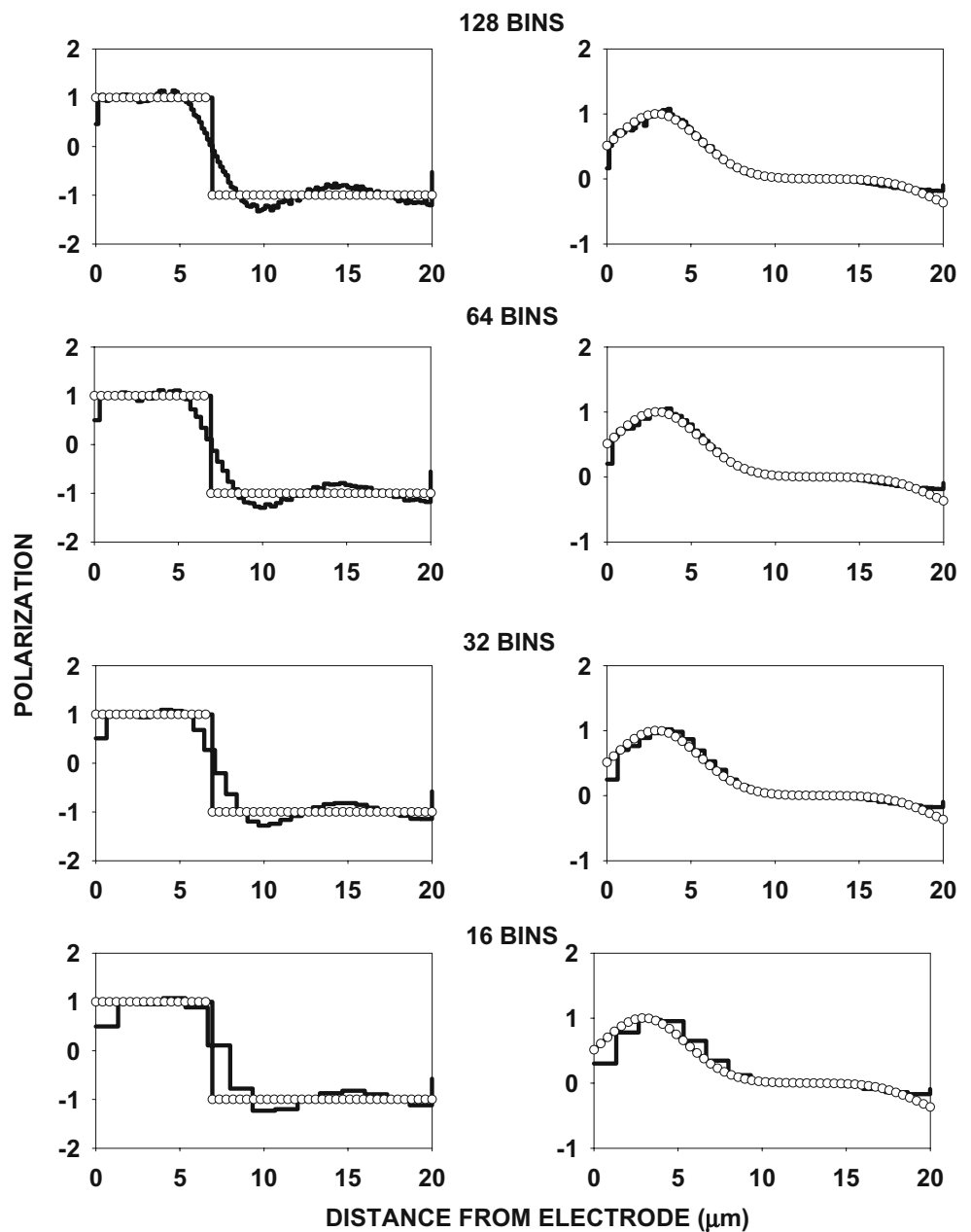
5.  The optimal value of $r$ is found using the L-curve method [6]. The logarithm of the summation on the left-hand side of Eq. 2 (the residual) is plotted against the logarithm of the integral on the right-hand side (roughness) for a range of values of $r$ as shown in Fig. 1(c). The $r$ corresponding to the point of maximum curvature is the optimal value (denoted by a larger circle in the figure).
6.  The resulting polarization distributions are shown on graphs with the true values in Fig. 1(d).

## 4 Monte Carlo method

The Monte Carlo method is implemented as follows:

1.  A random number generator is used to choose a large number of $z$ values. The number should be about 90% of the number of frequency data points.
2.  The real and imaginary parts of $\partial T(z,\omega)/\partial t$ are evaluated for each of the $z$ and $\omega$ values.
3.  A matrix $\mathbf{T}$ is formed in which the rows are the simulated frequencies and the columns are the real and imaginary parts of $\partial T(z,\omega)/\partial t$.
4.  The frequency values are entered into a vector $\mathbf{j}$.
5.  The least-squares problem $T \bullet g = j$ is solved for $\mathbf{g}$.

6.  The $z$ axis is divided into 128, 64, 32 or 16 bins and the $\mathbf{g}$ values contained in each bin are averaged.
7.  These histograms give the polarization distribution as shown for the two examples in Fig. 2.

## 5 Discussion and conclusions

Both methods accurately reproduced the true function of Example 2. Example 1 was satisfactory but less accurate due to the discontinuous nature of the true function. The computations took less than 2 min on a computer with a 1.8 MHz Pentium M processor for the PRM method and a 2.4 MHz Pentium 4 processor for the Monte Carlo Method. The PRM program was written with Mathematica and the Monte Carlo with Matlab.

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