Decomposition Of Permittivity Contributions From Reflectance Using Mechanism Models

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Abstract

In this paper we investigate the properties of a complex nonmagnetic material through the reflectance, where the permittivity is described by a mechanism model in which an unknown probability measure is placed on the model parameters. Specifically, we consider whether or not this unknown probability measure can be determined from the reflectance, the complex reflection coefficient, or both the reflectance and the derivatives of the reflectance. We also investigate the effect of measurement noise on the estimation. The numerical results demonstrate that if only the reflectance can be observed, then the distribution form cannot be recovered even in the case where the measurement noise level is small. Similar conclusions were also reached for the case where only the complex reflection coefficient can be observed. However, if both the reflectance and the derivative of the reflectance can be observed, then the estimated distribution is reasonably close to the true one even in the case where the measurement noise level is relatively high.

Key Words: Inverse problems, coefficients of reflectance, Debye and Lorentz polarization models
1 Introduction

The decomposition of a material’s electromagnetic response into the elementary component mechanisms responsible for observed phenomena is a fundamental problem of spectroscopy. In the setting of nonmagnetic materials, this involves determining the components of the permittivity using the measured spectral responses. Typically one would assume a particular combination of polarization models (Debye, Lorentz, Gaussian, etc.) with a predetermined number of dielectric parameters. However, in practice, the type of polarization model and the number of constituent dielectric mechanisms are usually unknown. In addition, the resulting decomposition may be non-unique or even nonphysical by using the reflectance (the ready observable in the primary experiment) alone.

Our approach in this work involves imposing a probability measure on model parameters as there are now incontrovertible experimental arguments for distributions of relaxation times for complex materials (e.g., see [6] and the references therein). We then develop a computational framework to nonparametrically estimate the probability measure, and investigate what type of measurement information is needed to accurately estimate the probability measure. We remark that some interferometers have the capability to obtain the derivative of the reflectance. Specifically, the Bruker 80V two beam interferometer [12, pp. 134-135] is capable of calculating derivatives up to the fifth order using the Savitzky-Golay algorithm. This algorithm smoothes the data through fitting successive sub-sets of adjacent data points with a low-degree polynomial (e.g., see Wikipedia, [18] and the references therein for details). In addition, numerical results in [15] show that the knowledge of the full complex reflection coefficient is necessary to determine uniquely a spacewise continuous index of reflection. Hence, in this effort we will consider three types of measurements, one corresponding to the reflectance, another corresponding to the reflection coefficient, and the other corresponding to both the reflectance and the derivative of the reflectance.

2 Problem Setup

For simplicity, we assume that a monochromatic uniform wave of frequency $\omega$ is incident at an angle $\theta$ on a plane interface between free space and a nonmagnetic dielectric medium as depicted schematically in Figure 1. This medium is assumed to be linear, homogeneous and isotropic. We first describe the reflection coefficient for the case where the wave is incident on a plane interface between two lossy media and then simplify to the particular situation of interest here.
2.1 General Reflection Coefficient, Interface Between Two Lossy Materials

In this section, we give details for the reflection coefficient in the case of a uniform plane wave that is incident at an arbitrary angle $\theta$ on a plane interface between two lossy media (which are assumed to be linear, homogeneous and isotropic), where the permittivity, permeability and conductivity of medium $l$ are denoted by $\varepsilon_l$, $\mu_l$, and $\sigma_l$, respectively. We remark that the arguments to derive the reflection coefficient for this case are similar to those in [14, Section 9.3]. Hence, we only sketch the ideas.

We assume that the electric field is polarized perpendicular to the plane of incidence (TE polarization). Let $d_i = (\sin(\theta), 0, \cos(\theta))^T$ denote the unit vector for the direction of the incident wave. Then the incident electric field and incident magnetic field phasors are respectively given by

$$\mathbf{E}^i = \mathbf{E}_0^i \exp(-\gamma_l d_i \cdot r),$$
$$\mathbf{H}^i = \frac{d_i \times \mathbf{E}_0^i}{\eta_{l1}} \exp(-\gamma_l d_i \cdot r).$$

In the above equation, $r = (x, y, z)^T$, and $\mathbf{E}_0^i = (0, \mathbf{E}_0^i, 0)^T$ with $\mathbf{E}_0^i$ being the incident intensity. In addition, $\gamma_l$ represents the propagation constant in medium $l$, and $\eta_{ld}$ denotes the complex intrinsic impedance in medium $l$. They are respectively given by

$$\gamma_l = i \omega \sqrt{\varepsilon_{cl}(\omega) \mu_l(\omega)}, \quad \eta_{ld} = \sqrt{\frac{\mu_l(\omega)}{\varepsilon_{cl}(\omega)}}, \quad l = 1, 2,$$

where $i$ denotes the imaginary unit, and $\varepsilon_{cl}$ represents the complex permittivity of medium $l$, and is given by

$$\varepsilon_{cl}(\omega) = \varepsilon_l(\omega) - \frac{i \sigma_l(\omega)}{\omega}. \quad (2.1)$$
The reflected electric field and the reflected magnetic field are then given by

\[
\begin{align*}
\vec{E}_r &= \vec{E}_0^r \exp(-\Gamma^r \cdot \mathbf{r}) = \vec{E}_0^r \exp(-(\Gamma_x^r x + \Gamma_z^r z)), \\
\vec{H}_r &= \frac{1}{\omega \mu_1(\omega)}(-i\Gamma^r \times \hat{\vec{E}}_0) \exp(-(\Gamma_x^r x + \Gamma_z^r z)),
\end{align*}
\]

where \(\vec{E}_0^r = (0, \vec{E}_0^r, 0)^T\) with \(\vec{E}_0^r\) being the reflected intensity, and \(\Gamma^r\) denotes the propagation vector of the reflected field and is given by \(\Gamma^r = (\Gamma_x^r, 0, \Gamma_z^r)^T\) with \((\Gamma_x^r)^2 + (\Gamma_z^r)^2 = \gamma_1^2\). In addition, the transmitted fields can be written as

\[
\begin{align*}
\vec{E}_t &= \vec{E}_0^t \exp(-\Gamma^t \cdot \mathbf{r}) = \vec{E}_0^t \exp(-(\Gamma_x^t x + \Gamma_z^t z)), \\
\vec{H}_t &= \frac{1}{\omega \mu_2(\omega)}(-i\Gamma^t \times \hat{\vec{E}}_0) \exp(-(\Gamma_x^t x + \Gamma_z^t z)),
\end{align*}
\]

where \(\vec{E}_0^t = (0, \vec{E}_0^t, 0)^T\) with \(\vec{E}_0^t\) being the transmitted intensity, and \(\Gamma^t\) denotes the propagation vector of transmitted field and is given by \(\Gamma^t = (\Gamma_x^t, 0, \Gamma_z^t)^T\) with \((\Gamma_x^t)^2 + (\Gamma_z^t)^2 = \gamma_2^2\).

Since the tangential components of both the electric field and the magnetic field are continuous on the interface, we can show that the angle of reflection is equal to the angle of incidence (the law of reflection), and that

\[
\begin{align*}
\vec{E}_0^r + \vec{E}_0^t &= \vec{E}_0^t, \\
r_{1s}(\vec{E}_0^r - \vec{E}_0^t) &= -r_{2s}\vec{E}_0^t,
\end{align*}
\]

where \(r_{1s}\) and \(r_{2s}\) are given by

\[
\begin{align*}
\frac{1}{r_{1s}} &= \sqrt{\frac{\varepsilon_{c1}(\omega) \cos(\theta)}{\mu_1(\omega)}}, \\
\frac{1}{r_{2s}} &= \sqrt{\frac{\varepsilon_{c2}(\omega) \mu_2(\omega) - \varepsilon_{c1}(\omega) \mu_1(\omega) \sin^2(\theta)}}.
\end{align*}
\]

Note that the reflection coefficient \(r_s\) is the ratio of reflected intensity \(\vec{E}_0^r\) to the incident intensity \(\vec{E}_0^t\). Hence, by solving (2.2) we find

\[
r_s(\omega, \theta) = \frac{r_{1s} - r_{2s}}{r_{1s} + r_{2s}}.
\]

Note that the reflection coefficient \(r_s\) is dependent on frequency, and in general is complex. Next we turn our attention to simplifying the expression for the reflection coefficient to match the formulation of the problem of interest.

### 2.2 Reflection Coefficient, Interface Between Free Space and a Lossy Material

In free space, \(\mu_1 \equiv \mu_0\) and \(\varepsilon_{cl} \equiv \varepsilon_0\), where \(\varepsilon_0, \mu_0\) are, respectively, the permittivity and permeability in free space. Also, since our material is nonmagnetic, we have that \(\mu_2 \equiv \mu_0\).
Note that the conductivity in the dielectric material is very small. Hence, we assume that it is negligible, that is, $\sigma \equiv 0$. Thus, by (2.1) we have $\hat{\varepsilon}_{r2}(\omega) = \hat{\varepsilon}_{2}(\omega)$.

Once we make the requisite substitutions in (2.3) and (2.4) we obtain

\[
\begin{align*}
    r_{1s} &= \sqrt{\frac{\varepsilon_0}{\mu_0}} \cos(\theta), \\
    r_{2s} &= \frac{1}{\mu_0} \sqrt{\hat{\varepsilon}_2(\omega)\mu_0 - \varepsilon_0\mu_0 \sin^2(\theta)} \\
    &= \frac{1}{\sqrt{\mu_0}} \sqrt{\hat{\varepsilon}_2(\omega) - \varepsilon_0 \sin^2(\theta)},
\end{align*}
\]

and so the reflection coefficient for the given scenario is

\[
\begin{align*}
    r_s(\omega, \theta) &= \frac{\sqrt{\varepsilon_0} \cos(\theta) - \sqrt{\hat{\varepsilon}_2(\omega) - \varepsilon_0 \sin^2(\theta)}}{\sqrt{\varepsilon_0} \cos(\theta) + \sqrt{\hat{\varepsilon}_2(\omega) - \varepsilon_0 \sin^2(\theta)}} \\
    &= \frac{\cos(\theta) - \sqrt{\hat{\varepsilon}_2(\omega) - \sin^2(\theta)}}{\cos(\theta) + \sqrt{\hat{\varepsilon}_2(\omega) - \sin^2(\theta)}},
\end{align*}
\]

where $\hat{\varepsilon}_{r2}$ is the relative permittivity of medium 2 and is given by $\hat{\varepsilon}_2/\varepsilon_0$. We remark that the above equation is equivalent to Fresnel’s Equation with the exception that in (2.6) we have explicitly incorporated frequency dependence into the material parameter $\hat{\varepsilon}_2$. Finally we note that (2.6) will serve as the primary focus of our investigation.

### 2.3 Composition of Permittivity

Without loss of generality, we assume that the permittivity of the dielectric medium is described by either a Lorentz model or a Debye model where a probability measure is imposed on the relaxation times.

The Lorentz model for electronic polarization represents an oscillating restoring response due to an electric field. The relative permittivity of a single-resonance Lorentz model has the form

\[
\hat{\varepsilon}_2(\omega) = \varepsilon_{\infty} - \frac{\omega_p^2}{\omega^2 - i\omega/\tau - \omega_0^2}.
\]

In the above equation, $\varepsilon_{\infty}$ denotes the relative permittivity of the medium at infinite frequency, $\tau$ is the relaxation time, and $\omega_p = \omega_0 \sqrt{\varepsilon_s - \varepsilon_{\infty}}$ is called the plasma frequency of the medium, where $\omega_0$ is the resonance frequency, and $\varepsilon_s$ is the relative permittivity of the medium at zero frequency.

The Debye model is used to model the dielectric relaxation response of an ideal, noninteracting population of dipoles to an alternating external electric field. As described in [17, Section 4.6.3] and [3], the Debye model best represents solids or liquids composed of polar
molecules. The oft cited example of such a medium is water, which is well-described by the Debye model. The Debye model is expressed as

$$\varepsilon_{r2}(\omega) = \varepsilon_\infty + \frac{\varepsilon_s - \varepsilon_\infty}{1 + i\omega\tau}. \quad (2.8)$$

To allow for a distribution $F$ of relaxation times over an admissible set $\mathcal{T} \subset \mathbb{R}_+$, we generalize the relative permittivity for the Lorentz model to be

$$\tilde{\varepsilon}_{r2}(\omega; F) = \varepsilon_\infty - \int_{\mathcal{T}} \frac{\omega_p^2}{\omega^2 - i\omega/\tau - \omega_0^2} dF(\tau), \quad (2.9)$$

and for the Debye model to be

$$\tilde{\varepsilon}_{r2}(\omega; F) = \varepsilon_\infty + \int_{\mathcal{T}} \frac{\varepsilon_s - \varepsilon_\infty}{1 + i\omega\tau} dF(\tau), \quad (2.10)$$

where $F \in \mathcal{P}(\mathcal{T})$, the set of admissible probability measures on $\mathcal{T}$. Then the distribution-dependent reflection coefficient is given by

$$r_s(\omega, \theta; F) = \frac{\cos(\theta) - \sqrt{\tilde{\varepsilon}_{r2}(\omega; F) - \sin^2(\theta)}}{\cos(\theta) + \sqrt{\tilde{\varepsilon}_{r2}(\omega; F) - \sin^2(\theta)}}. \quad (2.11)$$

It is worth noting that this approach of imposing a probability measure on the model parameters was also considered in [6, 13], where the authors directly simulate Maxwell’s equations, attempting to estimate a distribution on relaxation times for a Debye medium.

## 3 Computational Framework

In this section we examine an inverse problem methodology for estimating the probability measure of relaxation times with simulated data at various noise levels. In these problems the observable considered is either the magnitude of the reflection coefficient, the reflection coefficient, or both the magnitude of the reflection coefficient and its derivatives. For all cases, we assume that the incident angle $\theta$ is zero.

### 3.1 Statistical Model

We consider a statistical model of the form

$$Y_j = h(\omega_j; F_0) + V_j, \quad j = 0, 1, 2, 3, \ldots, n. \quad (3.1)$$

In the above equation, $h(\omega_j; F_0)$ corresponds to the observed part of the system with the “true” probability measure $F_0$ at the measurement frequency $\omega_j$, $V_j$ denotes the measurement
error at the measurement frequency $\omega_j$, and $n+1$ is the total number of observations. For the current proof of concept discussion, we assume that $V_j, j = 1, 2, 3, \ldots, n$, are independent and identically distributed with zero mean and constant covariance matrix $\Sigma_0 = \sigma_0^2 I_\kappa$, where $I_\kappa$ is a $\kappa \times \kappa$ identity matrix with $\kappa$ being the dimension of $V_j$.

As discussed above, we consider three types of data. In the case that the observed part of the system is the magnitude of the reflection coefficient (or reflectance), we have

$$h(\omega_j; F) = |r_s(\omega_j; 0; F)|,$$  \hspace{1cm} (3.2)

In the case that the reflection coefficient can be observed (that is, both the real and imaginary part of the reflection coefficient can be observed), $h$ has two components (i.e., $\kappa = 2$)

$$h(\omega_j; F) = (\text{Real}(r_s(\omega_j, 0; F)), \text{Imag}(r_s(\omega_j, 0; F)))^T,$$  \hspace{1cm} (3.3)

where $\text{Real}(r_s(\omega_j, 0; F))$ and $\text{Imag}(r_s(\omega_j, 0; F))$ denotes the real and imaginary part of the reflection coefficient $r_s(\omega_j, 0; F)$, respectively. For the case in which the data contains both the reflectance and the derivative of the reflectance, $h$ again has two components and is given by

$$h(\omega_j; F) = \left( |r_s(\omega_j, 0; F)|, \frac{d}{d\omega_j} |r_s(\omega_j, 0; F)| \right)^T.$$  \hspace{1cm} (3.4)

We remark that the reason for choosing the second component of $h$ as above is because the magnitude of the derivative of the reflectance is extremely small (for the parameter values that we choose for our proof of concept computations) compared to the value of the reflectance.

### 3.2 Inverse Problem

Under the assumptions for the measurement errors in the statistical model, the estimate $\hat{F}$ of $F$ can be obtained using the ordinary least squares formulation (e.g., see [10] for details)

$$\hat{F} = \arg\min_{F \in \mathcal{P}(\mathcal{T})} J(F).$$  \hspace{1cm} (3.5)

In the above equation, the cost functional $J$ is defined as

$$J(F) = \sum_{j=0}^{n} (h(\omega_j; F) - y_j)^T (h(\omega_j; F) - y_j),$$  \hspace{1cm} (3.6)

and $y_j$ is a realization of $Y_j$, that is,

$$y_j = h(\omega_j; F_0) + \nu_j, \hspace{0.5cm} j = 0, 1, 2, 3, \ldots, n,$$  \hspace{1cm} (3.7)

where $\nu_j$ is a realization of $\mathcal{V}_j, j = 0, 1, 2, \ldots, n$. The existence of $\hat{F}$ can be established under the Prohorov Metric Framework as developed in [1, 2, 5, 6, 7, 8, 9]. Specifically, if we assume
that $\mathcal{T}$ is compact, then it is well known that $\mathcal{P}(\mathcal{T})$ is a compact metric space when taken with the Prohorov [16] metric $\rho^*$ (e.g., see [1, 2, 7, 8] for details). Note that $h$ is continuous with respect to $F$. Hence, $J$ is continuous with respect to $F$. Thus, we know that there exists a solution to the least squares problem (3.5).

We note that (3.5) is an infinite dimensional optimization problem (as $\mathcal{P}(\mathcal{T})$ is an infinite dimensional space). Hence, we need to approximate the infinite dimensional space $\mathcal{P}(\mathcal{T})$ with finite dimensional space $\mathcal{P}^N(\mathcal{T})$ so that we have a computational tractable finite-dimensional optimization problem given by

$$\hat{F}^N = \arg\min_{F \in \mathcal{P}^N(\mathcal{T})} J(F).$$

(3.8)

Of course, one needs to choose $\mathcal{P}^N(\mathcal{T})$ such that $\mathcal{P}^N(\mathcal{T}) \to \mathcal{P}(\mathcal{T})$ in some sense as $N \to \infty$ so that $\hat{F}^N$ approaches to the solution to (3.5) as $N$ goes to infinity. One such approximation approach involves the use of Dirac measures to approximate probability measures $F$; that is,

$$F \approx F^N = \sum_{k=1}^{N} \alpha_k \Delta_{\tau_k},$$

(3.9)

where the weights $\alpha_k$, $k = 1, 2, \ldots, N$, are non-negative real numbers such that $\sum_{k=1}^{N} \alpha_k = 1$, and $\Delta_{\tau_k}$ is a Dirac measure with atom at node $\tau_k$. With this approximation, the least squares problem that we wish to solve is (3.8) with

$$\mathcal{P}^N(\mathcal{T}) = \left\{ F \in \mathcal{P}(\mathcal{T}) \left| F = \sum_{k=1}^{N} \alpha_k \Delta_{\tau_k}, \sum_{k=1}^{N} \alpha_k = 1 \right. \right\}.$$  

(3.10)

Under this computational framework the optimization problem (3.8) is reduced to a standard optimization problem over $\mathbb{R}^N$ in which we seek to estimate the finite set of values $\{\alpha_k\}_{k=1}^{N}$. The theoretic foundation for such approximation relies on the Prohorov Metric Framework. Specifically, it was shown in [2] that if $\mathcal{T}^\infty = \{\tau_k\}_{k=1}^{\infty}$ is an enumeration of the rational numbers of $\mathcal{T}$, then for any element $F \in \mathcal{P}(\mathcal{T})$ there exists a sequence $\{F^{N_j}\}$ with $F^{N_j} \in \mathcal{P}^{N_j}(\mathcal{T}^\infty)$ such that $\rho^*(F^{N_j}, F) \to 0$ as $N_j \to \infty$. Thus, we see that this Dirac measure approximation method can be used regardless of the smoothness of $F$. This is particular important in practice as one often has no information of the smoothness of the probability measure. It is worth noting that the Dirac measure approximation method has been successfully used to estimate probability measures in a number of applications (e.g., see [4, 5, 6, 9]).

Given the approximate distributions in (3.9), the relative permittivity for the Lorentz model (2.9) is

$$\tilde{\varepsilon}_2(\omega; F^N) = \varepsilon_\infty - \sum_{k=1}^{N} \frac{\omega_p \alpha_k}{\omega^2 - i\omega/\tau_k - \omega^2_0},$$

(3.11)
and the relative permittivity for the Debye model (2.10) is given by
\[
\hat{\varepsilon}_{r2}(\omega; F^N) = \varepsilon_{\infty} + (\varepsilon_s - \varepsilon_{\infty}) \sum_{k=1}^{N} \frac{\alpha_k}{1 + i\omega \tau_k}.
\] (3.12)

### 3.3 Parameter Values and Simulated Data

#### 3.3.1 Parameter Values

The permittivity and permeability in free space are respectively given by (e.g., see [13])
\[
\varepsilon_0 = 8.854 \times 10^{-12}, \quad \mu_0 = 1.2566 \times 10^{-6}.
\]
For all the simulations below, the values for all the other parameters are given as follows.

**Parameter values for the Lorentz model:** For the Lorentz model, we choose \( \mathcal{T}^N = \{\tau_k\}_{k=1}^{N} \) to be \( N \) evenly spaced nodes over the interval \([1 \times 10^{-17}, 2 \times 10^{-14}]\) in the approximations, and the values for the parameters \( \varepsilon_s \) and \( \varepsilon_{\infty} \) are chosen as (adapted from [11])
\[
\varepsilon_s = 2.25, \quad \varepsilon_{\infty} = 1.0.
\] (3.13)

Preliminary data sets from available initial experiments involve excitation in the frequency range of \( 10^{13} \) to \( 10^{15} \) Hz. Because of this we choose a value of \( \omega_0 = 1 \times 10^{14} \) Hz as a baseline value for our computational examples.

**Parameter values for the Debye model:** For the Debye model model, we choose \( \mathcal{T}^N = \{\tau_k\}_{k=1}^{N} \) to be \( N \) evenly spaced nodes over the interval \([1 \times 10^{-13}, 2 \times 10^{-10}]\) in the approximations, and the values for the parameters \( \varepsilon_s \) and \( \varepsilon_{\infty} \) are chosen as (adapted from [13])
\[
\varepsilon_s = 80.1, \quad \varepsilon_{\infty} = 5.5.
\] (3.14)

#### 3.3.2 Simulated Data

To generate the simulated data (that is, \( \{y_j\}_{j=0}^{n} \) with \( y_j \) given by (3.7)), the measurement error was chosen from a multivariate normal distribution with \( \sigma_0 = 0.01 \) or \( \sigma_0 = 0.05 \). The true measure and the measurement frequencies are varied from model to model, and will be given below.

We note that in the case that the observed part of the system contains both the reflectance and the derivative of the reflectance, we need to calculate \( \frac{\partial}{\partial \omega} \) \( r_s(\omega, \theta; F) \). However, a closed
form analytical solution of this derivative has proved to be very difficult to obtain due to the form of \( |r_s(\omega, \theta; F)| \). Because of this, we take the forward difference to approximate the derivative; that is,

\[
\frac{\partial}{\partial \omega} |r_s(\omega_j, \theta; F)| \approx \frac{|r_s(\omega_{j+1}, \theta; F)| - |r_s(\omega_j, \theta; F)|}{\omega_{j+1} - \omega_j}.
\]

**Simulated data for the Lorentz model:** The measurement frequencies are taken at \( \{\omega_j\}_{j=0}^{100} = \{10^7 + 0.09 j\}_{j=0}^{100} \). The true probability measure \( F_0 \) is chosen as either a discrete measure

\[
F_0 = 0.6 \Delta_{7.14 \times 10^{-16}} + 0.4 \Delta_{10^{-14}},
\]

or a Gaussian distribution with the corresponding probability density function given by

\[
f_0(\tau) = \frac{1}{2.5 \times 10^{-15} \sqrt{2 \pi}} \exp \left\{ -\frac{(\tau - 10^{-14})^2}{2(2.5 \times 10^{-15})^2} \right\},
\]

or a Bi-Gaussian distribution with the corresponding probability density function given by

\[
f_0(\tau) = \frac{1}{2 \times 10^{-16} \sqrt{2 \pi}} \exp \left\{ -\frac{(\tau - 10^{-15})^2}{2(10^{-16})^2} \right\} + \frac{1}{2 \times 10^{-16} \sqrt{2 \pi}} \exp \left\{ -\frac{(\tau - 1.2 \times 10^{-14})^2}{2(10^{-16})^2} \right\}.
\]

**Simulated data for the Debye model:** The measurement frequencies are taken at \( \{\omega_j\}_{j=0}^{100} = \{10^6 + 0.09 j\}_{j=0}^{100} \). The true probability measure \( F_0 \) is chosen as either a discrete measure

\[
F_0 = 0.6 \Delta_{8.1 \times 10^{-12}} + 0.4 \Delta_{7 \times 10^{-11}},
\]

or a Gaussian distribution with the corresponding probability density function given by

\[
f_0(\tau) = \frac{1}{9 \times 10^{-12} \sqrt{2 \pi}} \exp \left\{ -\frac{(\tau - 5 \times 10^{-11})^2}{2(9 \times 10^{-12})^2} \right\},
\]

or a Bi-Gaussian distribution with the corresponding probability density function given by

\[
f_0(\tau) = \frac{1}{4 \times 10^{-12} \sqrt{2 \pi}} \exp \left\{ -\frac{(\tau - 10^{-11})^2}{2(2 \times 10^{-12})^2} \right\} + \frac{1}{10^{-11} \sqrt{2 \pi}} \exp \left\{ -\frac{(\tau - 7 \times 10^{-11})^2}{2(5 \times 10^{-12})^2} \right\}.
\]

4 Numerical Results for the Case Where the Data is Generated by a Discrete Distribution

In this section we consider the case where the true distribution of relaxation times is assumed to be a discrete measure given by (3.15) for a Lorentz model and (3.18) for a Debye model.
We will use the above discussed Dirac measure approximation method to find an estimate \( \hat{F}^N \) for \( F_0 \), where the observable is either the reflectance, the reflection coefficient, or both the reflectance and the derivative of the reflectance.

### 4.1 Observable: the Reflectance

We first considered the case that only the reflectance can be observed (that is, \( h \) is given by (3.2)). Figure 2 depicts the results obtained for the Lorentz model with \( \sigma = 0.01 \) (left column) and \( \sigma_0 = 0.05 \) (right column) using \( N = 30 \) nodes in the approximation. We see from this figure that in the case of lower noise level \( \sigma_0 = 0.01 \) the estimated reflectance is a good approximation to the true reflectance, however the estimated distribution is not a good approximation to the true distribution. With an increased noise level \( \sigma = 0.05 \), the estimated reflectance begins to deviate from the true reflectance, and the estimated distribution is a poor approximation of the true distribution.

Figure 2: The results obtained for the Lorentz model with data having noise level \( \sigma_0 = 0.01 \) (left column) and \( \sigma_0 = 0.05 \) (right column) using \( N = 30 \) nodes in the approximation: the model fit to the data (first row), and the true and estimated probability distributions (second row).
The results obtained for the Debye model are shown in Figure 3. This figure indicates that the estimated reflectance agrees with the true one in both low and high noise level. However, the estimated distribution is a poor approximation to the true one in both cases.

Figure 3: The results obtained for the Debye model with data having noise level $\sigma_0 = 0.01$ (left column) and $\sigma_0 = 0.05$ (right column) using $N = 30$ nodes in the approximation: the model fit to the data (first row), and the true and estimated probability distributions (second row).

Similar conclusions were also reached using $N = 50$ nodes in the approximation. Overall these two figures suggest that if only the reflectance can be observed, then we are unable to recover the true distribution even in the low noise level case (even though the estimated reflectance may agree with the true reflectance). This indicates that the solution to the optimization problem is not unique (that is, the corresponding inverse problem is not well-posed).
4.2 Observable: the Reflection Coefficient

We next considered the case where the complex reflection coefficient can be observed (that is, \( h \) is given by (3.3)). Figure 4 depicts the results obtained for the Lorentz model using \( N = 30 \) nodes in the approximation with \( \sigma_0 = 0.01 \) (left column) and \( \sigma_0 = 0.05 \) (right column). We

![Graphs showing the true and estimated real and imaginary parts of the reflection coefficient along with the data.](image)

Figure 4: Results obtained for the Lorentz model with data having noise level \( \sigma_0 = 0.01 \) (left column) and \( \sigma_0 = 0.05 \) (right column) using \( N = 30 \) nodes in the approximation: the true and estimated real part of the reflection coefficient along with the data (first row), the true and estimated imaginary part of the reflection coefficient along with the data (second row), and the true and estimated probability distributions (third row).
see from this figure that in the case of lower noise level $\sigma_0 = 0.01$ the estimated reflection coefficient agrees with the true reflection coefficient, and the estimated distribution is a good approximation to the true distribution. However, with an increased noise level $\sigma = 0.05$, the estimated reflection coefficient deviates from the true reflection coefficient, and the estimated distribution is a poor approximation of the true distribution.

Figure 5 shows the results for the Debye model. This figure indicates that the estimated reflection coefficient provides an excellent approximation to the true reflection coefficient in both low and high noise level cases. However, the estimated distribution is a poor approximation to the true one in both cases.

Overall these two figures suggest that we are not able to consistently recover the distribution form for the all cases even in the low noise level.
Figure 5: Results obtained for the Debye model with data having noise level $\sigma_0 = 0.01$ (left column) and $\sigma_0 = 0.05$ (right column) using $N = 30$ nodes in the approximation: the true and estimated real part of the reflection coefficient along with the data (first row), the true and estimated imaginary part of the reflection coefficient along with the data (second row), and the true and estimated probability distributions (third row).
4.3 Observable: the Reflectance and the Derivative of the Reflectance

We next considered the case where both the reflectance and the derivative of the reflectance can be observed (that is, $h$ is given by (3.4)). Figure 6 depicts the results obtained for the Lorentz model using $N = 30$ nodes in the approximation with $\sigma_0 = 0.01$ (left column) and $\sigma_0 = 0.05$ (right column). We observe from this figure that in the case of the lower noise level $\sigma_0 = 0.01$ the estimated reflectance and the derivative of the reflectance agree with the true reflectance and derivative of the reflectance, and that the estimation for the distribution is very good. This figure also reveals that even in the case of higher noise levels the fits are still very good and the estimated distribution is reasonably close to the true one. Similar conclusions were also reached when we tried to estimate the probability distribution for the Debye model (shown in Figure 7).

In addition, we found that the estimates are consistent across simulated data sets. Using the derivative information alone to estimate the probability measure was also considered. However, we found that this information is not enough to provide consistency in the estimation across simulated data sets.
Figure 6: Results obtained for the Lorentz model with data having noise level $\sigma_0 = 0.01$ (left column) and $\sigma_0 = 0.05$ (right column) using $N = 30$ nodes in the approximation: The model fit to the data (first row), the derivative fit to the derivative data (second row), and the true and estimated probability distributions (third row).
Figure 7: Results obtained for the Debye model with data having noise level $\sigma_0 = 0.01$ (left column) and $\sigma_0 = 0.05$ (right column) using $N = 30$ nodes in the approximation: The model fit to the data (first row), the derivative fit to the derivative data (second row), and the true and estimated probability distributions (third row).
5 Numerical Results for the Case Where the Data is Generated by a Gaussian Distribution

In this section we consider the case where the true distribution of relaxation times is assumed to be a Gaussian distribution with probability density function given by (3.16) for the Lorentz model and (3.19) for the Debye model. We will continue to use the Dirac measure approximation method to estimate the distribution.

5.1 Observable: the Reflectance

Figures 8 summarizes the results obtained for the Lorentz model using $N = 30$ nodes in the approximation with $\sigma = 0.01$ (left column) and $\sigma_0 = 0.05$ (right column). We see from this figure that in the case of lower noise level $\sigma_0 = 0.01$ the estimated reflectance agrees with the true reflectance, but the estimated distribution is not a good approximation to the true distribution.
distribution. With an increased noise level $\sigma = 0.05$, the estimated reflectance still agrees with the true reflectance, but the estimated distribution is a poor approximation of the true distribution.

Figure 9 depicts the results for the Debye model. This figure indicates that the estimated reflectance agrees with the true reflectance in both low and high noise level. However, the estimated distribution is a poor approximation to the true one for both cases. This is consistent with the conclusion obtained for the discrete distribution case (demonstrated in Section 4.1).

Figure 9: The results obtained for the Debye model with data having noise level $\sigma_0 = 0.01$ (left column) and $\sigma_0 = 0.05$ (right column) using $N = 30$ nodes in the approximation: the model fit to the data (first row), and the true and estimated probability distributions (second row).

5.2 Observable: the Reflection Coefficient

Figure 10 depicts the results obtained for the Lorentz model using $N = 30$ nodes in the approximation with $\sigma_0 = 0.01$ (left column) and $\sigma_0 = 0.05$ (right column). We see from this figure that in the case of lower noise level $\sigma_0 = 0.01$ the estimated reflection coefficient
agrees with the true reflection coefficient, however the estimated distribution is not a good approximation to the true distribution. With an increased noise level $\sigma = 0.05$, the estimated reflection coefficient still agrees with the true reflection coefficient, but the estimated distribution is a poor approximation of the true distribution. Similar conclusions were also reached when we tried to estimate the probability distribution for the Debye model (shown...
in Figure 11).

Figure 11: Results obtained for the Debye model with data having noise level $\sigma_0 = 0.01$ (left column) and $\sigma_0 = 0.05$ (right column) using $N = 30$ nodes in the approximation: the true and estimated real part of the reflection coefficient along with the data (first row), the true and estimated imaginary part of the reflection coefficient along with the data (second row), and the true and estimated probability distributions (third row).
5.3 Observable: the Reflectance and the Derivative of the Reflectance

Figure 12 depicts the results for the Lorentz model using $N = 30$ nodes in the approximation with $\sigma_0 = 0.01$ (left column) and $\sigma_0 = 0.05$ (right column). We observe from this figure that...
in the case of the lower noise level $\sigma_0 = 0.01$ the estimated reflectance and the derivative of the reflectance agree with the true reflectance and derivative of the reflectance, and that the estimation for the distribution is good. This figure also reveals that even in the case of higher noise levels the fits are still excellent and the estimated distribution is reasonably close to the true one. Similar conclusions were also reached when we tried to estimate the probability distribution for the Debye model (shown in Figure 13).

Figure 13: Results obtained for the Debye model with data having noise level $\sigma = 0.01$ (left column) and $\sigma_0 = 0.05$ (right column) using $N = 30$ nodes in the approximation: The model fit to the data (first row), the derivative fit to the derivative data (second row), and the true and estimated probability distributions (third row).
6 Numerical Results for the Case Where the Data is Generated by a Bi-Gaussian Distribution

We now repeat the investigation of using a Bi-Gaussian distribution for the true distribution of relaxation times, where the probability density function is given by (3.16) for the Lorentz model and (3.19) for the Debye model.

6.1 Observable: the Reflectance

Figure 14 summarizes the results obtained for the Lorentz model with $\sigma = 0.01$ (left column) and $\sigma = 0.05$ (right column). We see from this figure that the estimated reflectance agrees with the true one in both low and high level noise cases, however the estimated distribution is a poor approximation to the true distribution. We also obtain similar conclusion when we attempt to estimate the probability distribution for the Debye model (shown in Figure 15).
Figure 15: The results obtained for the Debye model with data having noise level \( \sigma_0 = 0.01 \) (left column) and \( \sigma_0 = 0.05 \) (right column) using \( N = 30 \) nodes in the approximation: the model fit to the data (first row), and the estimated probability distribution (second row).

Figures 14 and 15 again suggest that if only the reflectance can be observed, then we are unable to recover the true distribution even in the low noise level case (even though the estimated reflectance agrees with the true one).

### 6.2 Observable: the Reflection Coefficient

Figure 16 depicts the results obtained for the Lorentz model using \( N = 30 \) nodes in the approximation with \( \sigma_0 = 0.01 \) (left column) and \( \sigma_0 = 0.05 \) (right column). We see from this figure that in the case of lower noise level \( \sigma_0 = 0.01 \) the estimated reflection coefficient agrees with the true one, however the estimated distribution is not a good approximation to the true distribution. With an increased noise level \( \sigma = 0.05 \), the estimated reflection coefficient still agrees with the true one, but the estimated distribution is a poor approximation of the true distribution.

Figure 17 depicts the results for the Debye model. This figure indicates that the estimated reflection coefficient agrees with the true one in both the low and high noise level cases.
Figure 16: Results obtained for the Lorentz model with data having noise level $\sigma_0 = 0.01$ (left column) and $\sigma_0 = 0.05$ (right column) using $N = 30$ nodes in the approximation: the true and estimated real part of the reflection coefficient along with the data (first row), the true and estimated imaginary part of the reflection coefficient along with the data (second row), and the true and estimated probability distributions (third row).

However, the estimated distribution is a poor approximation to the true one.
Figure 17: Results obtained for the Debye model with data having noise level $\sigma_0 = 0.01$ (left column) and $\sigma_0 = 0.05$ (right column) using $N = 30$ nodes in the approximation: the true and estimated real part of the reflection coefficient along with the data (first row), the true and estimated imaginary part of the reflection coefficient along with the data (second row), and the true and estimated probability distributions (third row).
6.3 Observable: the Reflectance and the Derivative of the Reflectance

Figure 18 depicts the results obtained for the Lorentz model using $N = 30$ nodes in the approximation with $\sigma_0 = 0.01$ (left column) and $\sigma_0 = 0.05$ (right column). We observe from

Figure 18: Results obtained for the Lorentz model with data having noise level $\sigma = 0.01$ (left column) and $\sigma = 0.05$ (right column) using $N = 30$ nodes in the approximation: The model fit to the data (first row), the derivative fit to the derivative data (second row), and the true and estimated probability distributions (third row).
this figure that in the case of the lower noise level $\sigma_0 = 0.01$ the estimated reflectance and the
derivative of the reflectance agree with the true reflectance and derivative of the reflectance,
and that the estimation for the distribution is very good. This figure also reveals that even
in the case of higher noise level the fits are still excellent and the estimated distribution
is reasonably close to the true one. Similar conclusions were also reached when we try to
estimate the probability distribution for the Debye model (depicted in Figure 18).

Figure 19: Results obtained for the Debye model with data having noise level $\sigma = 0.01$ (left
column) and $\sigma_0 = 0.05$ (right column) using $N = 30$ nodes in the approximation: The model
fit to the data (first row), the derivative fit to the derivative data (second row), and the true
and estimated probability distributions (third row).
7 Summary Remarks

In this note we use a Lorenz model and a Debye model as examples to investigate the type of data that might be needed to accurately estimate a probability measure imposed on the model parameters through simulated data. The numerical results demonstrate that using the reflectance alone is not sufficient to consistently recover the true distribution form even at low noise levels (even though the fits to the data are good). Similar conclusions were also reached for the cases where the observable is the complex coefficient.

However, if both the reflectance and the derivative of the reflectance can be observed, then the estimated distribution is reasonably close to the true distribution even in the case where the noise level is relatively high. We note that the method used to simulate the data for the derivative of the reflectance is somewhat artificial. However, we have preformed an important investigation and proof of concept which demonstrates that if reliable derivative data can be obtained in practice, then one may be able to successfully determine the distribution of relaxation times.

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