Retrieval of Optical Constants from Magnetoreflectance by Maximum Entropy Model

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(Rceived May 14, 1998; Accepted July 9, 1998)

Phase retrieval, and thereby retrieval of optical constants, from magnetoreflectance data is performed using the maximum entropy model for spectra analysis. The applicability of the analysis is tested by using the Lorentz model for the permittivity of a nonmagnetic insulator in a case of Faraday configuration.

Key words: magnetoreflectance, phase retrieval, maximum entropy model, Faraday configuration

1. Introduction

During this century the study of wavelength dependent reflection from highly absorbing materials—like metals or semiconductors—has provided the means to gain information about their optical properties. An important step for estimation of the optical constants of materials was the utilization of the Kramers-Kronig relation

\[ \theta(\omega) = -\frac{2\omega}{\pi} \int_{0}^{\infty} \frac{\ln|\rho(\omega')|}{\omega' - \omega} d\omega', \]

(1)

connecting the amplitude \(|\rho(\omega)|\) and phase \(\theta(\omega)\) of the complex reflectivity \(\rho(\omega) = \rho(\omega)e^{i\theta(\omega)}\). After \(|\rho(\omega)|\) is measured and \(\theta(\omega)\) is computed, the optical constants are obtained from Fresnel’s equations. Rigorous validity of relation (1) was studied by Velický, who treated the complex reflectivity as a holomorphic function in the upper half of complex angular frequency plane and used the symmetry property

\[ \rho^*(-\omega) = \rho(-\omega^*), \]

(2)

given at a complex angular frequency \(\omega^*\). The usage of natural logarithm in Eq. (1) caused some apparent problems—mainly because \(\ln|\rho(\omega)|\) tends to \(-\infty\) as \(|\rho(\omega)|\to0\) (which generally occurs when \(\omega\to\infty\)). Such a function cannot be treated as a square integrable function along the real axis. These problems were shown to be artifacts by Smith, who gave another type of simple proof of the validity of Eq. (1). In practice, however, the phase retrieval by Eq. (1) is problematic, because the extrapolations of \(|\rho(\omega)|\) outside its measurement range are critical due to the behaviour of \(\ln|\rho(\omega)|\) at high frequencies. To ease this difficulty, methods based on subtractive techniques have been developed.

When an external magnetic field is applied to a material, its optical properties are altered. Through the rise of Lorentz force, the magnetic field reduces the spatial symmetry of the electronic system. As a result, the system’s permittivity tensor contains asymmetric elements. The optical constants depend then both on the direction of propagation and on the polarization of the incident light. In the so-called Faraday configuration light propagates in parallel to the applied magnetic field. Then, the two allowed modes are right- (associated by “-” sign) and left-hand (“+” sign) circularly polarized. Accordingly, the absorption and dispersion of the material depend on the handedness of the circularly polarized light. The corresponding optical constants are the real refractive index \(n\) or \(n_\pm\), and the extinction coefficient \(k\) or \(k_\pm\). If the material is transparent, a spectropolarimeter can be used to measure transmittance to obtain the magnetic field induced circular dichroism, \(\Delta k = k_- - k_+\), and the related birefringence, \(\Delta n = n_- - n_+\). Unfortunately, such measurements are not possible in the case of highly absorbing materials. However, dispersion relations can also be written for the magneto-optical constants and, in particular, dispersion relations for magnetorelectivity can be obtained. Then, in principle, it is possible to estimate optical constants with the aid of magnetoreflectance measurements from highly absorbing materials.

Here, we present an alternative phase retrieval method, based on the maximum entropy model (MEM), which is applied for the case of magnetoreflectance. As far as we know this is the first time that the MEM has been applied in magneto-optics. We have shown that the MEM procedure can be a powerful method not only in linear reflection spectroscopy but also in nonlinear optical spectroscopy and even in cases where Kramers-Kronig relations cannot be applied. The merit of the MEM relies on the fact that data extrapolations are not needed.

2. Dispersion Relations for Magnetoreflectance

For simplicity, let us consider a case where circularly polarized light propagates along the \(z\) axis in a nonmagnetic insulator with \(\mathbf{C}_1\) symmetry, i.e., whose permittivity tensor is

\[ \epsilon = \begin{pmatrix} \epsilon_{xx} & \epsilon_{xy} & 0 \\ -\epsilon_{xy} & \epsilon_{xx} & 0 \\ 0 & 0 & \epsilon_{zz} \end{pmatrix}. \]

(3)
This tensor describes an isotropic solid in an external magnetic field, \( H = H_z \), along the \( z \) axis. Thus, we limit our treatment to a case of Faraday configuration. The complex refractive index is then defined as
\[
N^2(\omega) = \varepsilon_\perp(\omega) + i \varepsilon_\parallel(\omega). \tag{4}
\]
In contrast, \( N_z(\omega) \) can be given by the magnetorefractivity, \( r_z(\omega) = r_z(\omega) \exp(i \theta_z(\omega)) \), as
\[
N_z(\omega) = \varepsilon_z(\omega) + ik_z(\omega) = 1 + r_z(\omega) \exp(i \theta_z(\omega)) = 1 - r_z(\omega) \exp(i \theta_z(\omega)). \tag{5}
\]
The symmetry relation for magnetorefractivity is now given by
\[
r_z^*(\omega) = r_z(-\omega^*), \tag{6}
\]
and the Kramers-Kronig relation connecting the phases \( \theta_z(\omega) \) and the amplitudes \( |r_z(\omega)| \) is
\[
\theta_z(\omega) + \theta_z(\omega) = -\frac{2\omega}{\pi} \int_0^\infty \frac{\ln|r_z(\omega)| + \ln|\omega^2 - \omega^2|}{\omega} d\omega'. \tag{7}
\]
Also, a non-Kramers-Kronig dispersion relation can be given as
\[
\theta_z(\omega) - \theta_z(\omega) = -\frac{2\omega}{\pi} \int_0^\infty \omega \ln|\omega^2 - \omega^2| d\omega'. \tag{8}
\]
Note that in Eqs. (7) and (8) the phases and amplitudes describing left-hand "+" and right-hand "−" polarization modes of the light appear simultaneously. Therefore, to get the optical constants, \( n_z(\omega) \) and \( k_z(\omega) \), for example, both \( |r_z(\omega)| \) and \( r_z(\omega) \) must be measured, and both Eqs. (7) and (8) must be successfully computed.

Next, we show how the maximum entropy model can be used to solve the phase retrieval problem from a magnetoreflectance measurement. The phase \( \theta_z(\omega) \) can then be obtained directly from the corresponding magnetoreflectance \( R_{\parallel} = |r_z(\omega)|^2 \) (or \( R_{\perp} = |r_z(\omega)|^2 \)) without any data extrapolations. Thus, the scheme is completely different from the dispersion relation formalism of Eqs. (7) and (8).

3. Phase-Retrieval by Maximum Entropy Model

Suppose that a magnetoreflectance, \( R_{\parallel}(\omega) = |r_z(\omega)|^2 \), where \( \parallel \) is either + or −, is measured within a frequency interval of \( \omega_1 \leq \omega \leq \omega_2 \). Now, defining a normalized frequency by
\[
\nu = \frac{\omega - \omega_1}{\omega_2 - \omega_1}, \tag{9}
\]
we can fit the measurement by its maximum entropy model
\[
\tilde{R}_\parallel(\nu) = \frac{|\beta|^2}{1 + \sum_{k=1}^M a_k \exp(-i2\pi k\nu)} \tag{10}
\]
The unknown MEM coefficients \( a_k \) and \( |\beta|^2 \) can be obtained from the set of linear (Yule-Walker) equations:
\[
\sum_{k=0}^M a_k C(n-k) = \frac{|\beta|^2}{1 + \sum_{k=1}^M a_k \exp(-i2\pi k\nu)}, n = 0, 1, \ldots, M \tag{11}
\]
where the autocorrelations, \( C(m) \), can be computed by a

Fourier transform of \( R_{\parallel}(\nu) \) as
\[
C(m) = \int_0^1 R_{\parallel}(\nu) \exp(i2\pi mv) d\nu. \tag{12}
\]
The phase retrieval can now be based on the maximum entropy model for the (complex) magnetoreflectivity
\[
r_{\parallel}(\nu) = \frac{|\beta|^2 \exp(-i\phi_{\parallel}(\nu))}{1 + \sum_{k=1}^M a_k \exp(-i2\pi k\nu)}, \tag{13}
\]
where \( \phi_{\parallel}(\nu) \) is a so-called error phase. This is the only quantity in Eq. (13) that cannot be directly obtained from the measurement of \( R(\nu) \).

The idea of using the MEM in phase retrieval is that the problem of finding the phase \( \theta(\nu) \) reduces to a problem of finding the error phase \( \phi_{\parallel}(\nu) \). Although not obvious, it is typically the case that \( \phi_{\parallel}(\nu) \) is a much simpler function than \( \theta(\nu) \). Nevertheless, to find an estimate for \( \phi_{\parallel}(\nu) \), we must have some additional information on \( r(\nu) \) (besides its modulus \( |r(\nu)| \)). For instance, if we know the value of the real (or imaginary) part of \( r(\nu) \) or the value of the phase \( \theta(\nu) \) at \( L+1 \) discrete frequency values \( \nu_l \) within the measurement range, we can compute the corresponding error phase values \( \phi_{\parallel}(\nu_l) \) and estimate the error phase by a polynomial interpolation as
\[
\tilde{\phi}_{\parallel}(\nu) = B_0 + B_1 \nu + \cdots + B_L \nu^L = \sum_{l=0}^L B_l \nu^l, \tag{14}
\]
where \( B_l \) can be obtained from a Vandermonde system
\[
\begin{bmatrix}
1 & \nu_0 & \nu_0^2 & \cdots & \nu_0^L \\
1 & \nu_1 & \nu_1^2 & \cdots & \nu_1^L \\
\vdots & \vdots & \vdots & \ddots & \vdots \\
1 & \nu_L & \nu_L^2 & \cdots & \nu_L^L
\end{bmatrix}
\begin{bmatrix}
B_0 \\
B_1 \\
\vdots \\
B_L
\end{bmatrix}
= \begin{bmatrix}
\phi(\nu_0) \\
\phi(\nu_1) \\
\vdots \\
\phi(\nu_L)
\end{bmatrix}. \tag{15}
\]
In practice, the optimum degree \( L = L_{\text{opt}} \) of the polynomial in Eq. (14) is low, i.e., the number of required \textit{a priori} known values of \( \phi_{\parallel}(\nu) \) is low. Clearly, the ideal situation would be that the error phase could be given by a linear estimation, i.e., \( L_{\text{opt}} \approx 1 \). In fact, we have found that it is possible to reduce the optimum degree \( L_{\text{opt}} \) of the polynomial by a simple "squeezing" procedure. Namely, instead of using the measured spectrum \( R_{\parallel}(\nu) \) as in Eq. (12), we can use a modified spectrum \( R_{\parallel}^\nu \) given by
\[
R_{\parallel}^\nu(\nu;K) = \begin{cases}
R_{\parallel}(\omega), & 0 \leq \nu \leq z_K(\omega) \\
R_{\parallel}(\omega), & z_K(\omega) \leq \nu \leq z_{K+1}(\omega) \\
R_{\parallel}(\omega), & z_{K+1}(\omega) < \nu \leq 1
\end{cases}, \tag{16}
\]
where
\[
z_K(\omega) = (2K+1)^{-1} \left( \frac{\omega - \omega_1}{\omega_2 - \omega_1} + K \right), \tag{17}
\]
and
\[
\nu = \frac{z_K(\omega) - z_K(\omega)}{z_{K+1}(\omega) - z_K(\omega)}. \tag{18}
\]
Now, by choosing the squeezing parameter \( K=0 \), Eq. (16) restores the original reflectance spectrum in \( R_{\parallel}^\nu \) as a function of the normalized frequency \( \nu \). However, when \( K > 0 \) the original spectrum is transformed (squeezed) into a narrower range. For example, by setting \( K = 2 \) the spec-
trum $R_{\ell}(\nu)$ is squeezed from the interval $\nu \in [0, 1]$ into interval $\nu \in [0.4, 0.6]$. The rest of the squeezed spectrum is obtained by adding constant wings having the same values as the original spectrum at its end points $\nu=0$ and $\nu=1$. Quite commonly, this procedure reduces $L_{\text{max}}$ enough that a linear approximation ($L=1$) provides a good estimation for the error phase.

To demonstrate the applicability of the present MEM procedure in the analysis of magnetoreflectance, we use here the Lorentz oscillator model. The permittivity elements of Eq. (3) are given as

$$
\varepsilon_{xx}(\omega) = 1 - \frac{\omega_c^2 (\omega_c^2 - \omega^2 - i\Gamma\omega)}{(\omega_c^2 - \omega^2 - i\Gamma\omega)^2 - \omega_c^2 \omega_c^2},
$$

$$
\varepsilon_{xy}(\omega) = -\frac{i\omega_c^2 \omega}{(\omega_c^2 - \omega^2 - i\Gamma\omega)^2 - \omega_c^2 \omega_c^2},
$$

where the cyclotron frequency, $\omega_c$, is defined by

$$
\omega_c = eH/m_e
$$

and the plasma frequency, $\omega_p$, by

$$
\omega_p^2 = \frac{4\pi e^2}{m_e}. \quad (21)
$$

Here, $\eta$ is the density of Lorentz oscillators, i.e., valence electrons. In Fig. 1(a) are shown the magnetoreflectance for right- and left-hand modes, and in (b) the corresponding phases, $\theta_\pm$, and error phases, $\phi_\pm$. The error phases were obtained using the squeezing procedure of Eqs. (16)–(18) with $K=2$. It is notable that not only are both $\phi_+$ and $\phi_-$ almost linear, but also that they are practically the same, i.e., $\phi_+ \approx \phi_-$. In Figs. 1(c) and (d) we show the corresponding magneto-optical constants $n_\pm$ and $k_\pm$, i.e., the real and imaginary parts of the complex refractive indices $N_\pm$.

The arrows in Figs. 1(c) and (d) denote the two frequencies where the additional phase information was taken for the MEM procedure. Thus, we have used a linear estimation for the error phases $\phi_\pm$. Obviously, we would get better results by using additional information and/or by taking the additional points further apart from each other. In practise, however, it is most likely that the frequency region of the potential additional information is limited to a narrow frequency range. Therefore, in the simulation herein we have assumed the phase to be known only at two closely located frequencies. Note that the calculations involved either right- or left-hand mode, whereas in the dispersion relation analysis both modes are required. In addition, the MEM procedure can be applied using data in a finite spectral range, whereas the dispersion relations always need data extrapolations for successful phase retrieval.

4. Conclusions

We have applied, as far as we know for the first time, the MEM procedure to the context of magnetoreflectance. The calculations were based on the exploitation of the well-known Lorentz oscillator model and Faraday configuration. We can draw the conclusion that the MEM is well suited for reflectance spectra analysis in magnetooptics.

Acknowledgments

K.-E.P. wishes to express his gratitude to the Academy of Finland for financial support of his research.

References