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# Dispersion theory and phase retrieval of meromorphic total susceptibility

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**Abstract.** Meromorphic susceptibility is a material parameter that appears usually in pump-probe experiments, where so-called degenerate susceptibilities, as a function of angular frequency, describe the radiation and material interaction. Kramers–Kronig dispersion relations are invalid for meromorphic susceptibilities. Phase retrieval by the maximum-entropy model from the modulus of total susceptibility, with poles and zeros in the complex plane, is considered. Complex analysis is employed in the description of the nature of the meromorphic total susceptibility.

## 1. Introduction

During the past few decades, nonlinear optical spectroscopies have become important tools in material sciences. One major physical quantity that describes strong electric field interaction with material is the nonlinear susceptibility. The research on dispersion theory of nonlinear susceptibilities was started at the beginning of the 1960s and has continued to the present day [1–10]. However, it took a long time before the consistency between experiments and the theory could be shown [11]. Nevertheless, it has been shown that the Kramers–Kronig dispersion relations can fail in steady-state nonlinear spectroscopy as devised by Yariv for the case of a two-level atom [12]. In addition quite recently Tokunaga *et al* [13–15] have shown by theoretical and experimental studies that the Kramers–Kronig relations are not always valid in femtosecond spectroscopy, that is to say in time-resolved spectroscopy of short pump and probe light pulses. A main feature with such linear or nonlinear pathological susceptibilities is that they are meromorphic functions [16]. Meromorphicity is a property of a complex function that is holomorphic almost everywhere in the complex plane except on essential singular points called poles. Poles appear simultaneously in the upper and lower half planes.

With the aid of the maximum-entropy model we have examined meromorphic susceptibility by applying a phase retrieval procedure to yield the phase of the complex degenerate nonlinear susceptibility from its modulus [17]. Such a phase retrieval has been successfully applied also for data inversion of optical constants from linear reflectances [18–21], as well as resolving the real and imaginary parts of CARS spectra [22], and those related to the third-harmonic-wave generation from polysilanes [23].

In this paper we consider dispersion theory of meromorphic susceptibilities. We show by an example that phase retrieval by the maximum-entropy model (MEM) can resolve the real and imaginary parts of *total meromorphic susceptibility*. So far, our phase retrieval studies have been devoted to the nonlinear meromorphic susceptibilities. The total susceptibility that describes the overall material response to external electric fields includes both the linear and nonlinear contributions. A new aspect, compared with our previous phase retrieval studies, is that poles and also *zeros* of the total susceptibility appear simultaneously in the same half plane. Therefore, the applicability of the phase retrieval procedure is generalized to a new class of functions that possess zeros in the complex plane. Furthermore, we consider the mathematical concepts that are needed to estimate whether a total susceptibility is meromorphic or not. For this purpose we exploit the results of complex analysis.

## 2. Meromorphic susceptibility and phase retrieval

A meromorphic total susceptibility is simply a function that has poles in both halves of the complex angular frequency plane. It is holomorphic everywhere except at the poles. The appearances of the poles in the upper half plane mean that the conventional Kramers–Kronig dispersion relations have to be changed to take into account the nonzero residue terms [17, 24]. In this case an appropriate asymptotic fall of the complex susceptibility at infinite angular frequencies is needed and it is provided, in the classical description, by inspection of the Newtonian mechanics of the particle system. Unfortunately, we cannot usually gain information on the residues from measured spectra. Hence another type of data analysis is needed. Therefore, we consider the application of MEM for resolving the real and imaginary parts of meromorphic total susceptibility.

The causality conditions for meromorphic susceptibility were considered by Kircheva and Hadjichristov [10] who pointed out that causality is a necessary but not sufficient condition for the existence of the Kramers–Kronig relations. The breaking of the causality condition appears for instance in femtosecond spectroscopy, i.e. for cases where the pump pulse has caused polarization of the charge system before the incidence of the probe pulse. Such a physical phenomenon is governed by the simultaneous appearance of poles in both half planes [13].

As an example of a meromorphic total susceptibility we consider the classical model of a two-level atom as treated by Yariv. Accordingly, the total susceptibility that is related to the saturation of the probe, which is used also for pumping, can be written as

$$\chi(\omega) = C[(\omega_0 - \omega) + i/T_2][(\omega_0 - \omega)^2 + (1 + I/I_s)/T_2^2]^{-1} \quad (1)$$

where  $C$  is a constant,  $\omega_0$  the transition (angular) frequency,  $T_2$  the dephasing time,  $I$  the pump irradiance and  $I_s$  the saturation irradiance. This function has interesting properties that include the simultaneous appearance of two symmetric poles, in opposite half planes, and also a zero in the upper half plane. Then the logarithm of the susceptibility,  $\ln|\chi|$ , which is the crucial function in phase retrieval by the Kramers–Kronig relations, explodes at such singular points of the logarithm and can no longer be considered as a regular function in the complex plane. A function that is holomorphic in the upper half plane, but has zeros in the upper half plane and poles in the lower half plane, in a manner such that there is a symmetry among the zeros and the poles, can be described by the Blaschke product [25], which is then a meromorphic function [26] in the whole complex plane. Evidently the total susceptibility of (1), which could be described by a single Blaschke factor, does not possess symmetry among the zero and the poles. In addition, it is meromorphic already in the upper half plane. Almost the same behaviour, but related to Raman excitation profiles, has

been recently described by Lee [27]. However, in the case of phase retrieval from Raman excitation profiles the poles lie strictly in one half plane and the zeros in the other half plane. In such a case the related complex function can be treated as a holomorphic function in the half plane where the zeros lie. As a result of the zeros the phase change of the electric field can exceed the value  $2\pi$ . Thus, a so-called non-minimum phase retrieval problem arises. Nevertheless, dispersion relations for the phase retrieval, whose logical foundations have already been devised by Toll [25], can be applied for non-minimum phase retrieval from Raman excitation profiles. These relations include the conventional Kramers–Kronig integral and additional new terms. The situation here is somewhat different since the total susceptibility has simultaneously a pole and a zero in the same half plane. Generally we cannot apply a simple model like the one of (1). Then we usually do not know exactly the number of poles or zeros from any intensity data. Especially, resolving the number and location of zeros [25], which defines the multiplicity of the phase angle in non-minimum phase problems, may be problematic with the aid of the measured data. Therefore, the Kramers–Kronig analyses have been done in non-minimum phase retrieval, so far, only with theoretical curve models [27].

When working with phase retrieval by MEM, which we abbreviate from now on as PRMEM, we do not use any logarithm function. This means that we can neglect an ill behaved function at singular points. Indeed, PRMEM deals only with the modulus of general type total susceptibility in the present case. The decomposition of the real and imaginary parts of the degenerate total susceptibility is based on its maximum-entropy model [19]

$$\chi(\nu) = |\beta| \exp[-i\phi(\nu)] \left/ \left[ 1 + \sum_{k=1}^M a_k \exp(-2\pi i k \nu) \right] \right. \quad (2)$$

where the unknown MEM coefficients  $a_k$  and  $|\beta|$  are obtained by fitting the measured modulus spectrum  $|\chi(\omega)|$ ,  $\omega_1 \leq \omega \leq \omega_2$ , by its maximum-entropy model,

$$|\chi(\nu)| = |\beta| \left/ \left| 1 + \sum_{k=1}^M a_k \exp(-2\pi i k \nu) \right| \right. \quad (3)$$

The variable  $\nu$  is a normalized frequency,  $\nu = (\omega - \omega_1)/(\omega_2 - \omega_1)$ . The detailed (phase retrieval) procedure using (2) and (3) is described elsewhere [21, 23]. The idea of using MEM in resolving  $\chi(\omega)$  from its modulus is that now the problem of finding the phase of  $\chi$  is reduced to a problem of finding the so-called error phase,  $\phi$ , in (2). Namely, it has turned out that  $\phi$  is a ‘slowly varying’ function compared to the actual phase. Therefore, it can be estimated, as done in PRMEM, by a polynomial interpolation,

$$\phi(\nu) = B_0 + B_1\nu + \dots + B_L\nu^L = \sum_{l=0}^L B_l\nu^l \quad (4)$$

such that the optimum degree  $L_{opt}$  of the polynomial is usually low (typically  $L_{opt} = 1$ ). The additional information that is now needed to get an estimate to  $\phi(\nu)$ , and, thus, for phase recovery, is  $L + 1$  discrete values of  $\phi(\nu_l)$ . In practice this means that either the value of  $\text{Im } \chi$  or  $\text{Re } \chi$  must be known at  $L + 1$  discrete frequencies inside the measurement range. The merit of this type of phase retrieval procedure is that there is no need for data extrapolations beyond the measured range if compared with the phase retrieval by Kramers–Kronig relations. In figure 1(a) we show the modulus of the total susceptibility of (1) for various values of the light intensity ratios. The corresponding error phases,  $\phi$ , are shown in figure 1(b). It is notable that low intensity ratio  $I/I_s$  results in an almost linear line shape to

$\phi$ , whereas a high ratio results a curved line shape. Therefore, in the case of a low intensity ratio successful phase recovery can be achieved by using a linear estimate for  $\phi$ ; hence, only two values for  $\text{Im } \chi$  are needed (i.e.,  $L + 1 = 2$ ) in addition to the measured spectrum. In the case of high intensity ratios more additional information is needed. Figures 1(c) and (d) illustrate this as  $\text{Re } \chi$  and  $\text{Im } \chi$  are computed by PRMEM in case of low ( $I/I_s = 0.1$ ) and high ( $I/I_s = 1.0$ ) intensity ratios, where corresponding integers are  $L + 1 = 2$  and  $L + 1 = 6$ . However, since the intensity ratio does not affect to the actual phase function,  $\chi(\omega; I/I_s)$  is best obtained by measuring a low-intensity spectrum  $|\chi(\omega); I/I_s \approx 0|$  in addition to a high-intensity  $|\chi(\omega); I/I_s|$  spectrum, and by computing the phase using the low intensity data  $|\chi(\omega); I/I_s \approx 0|$ . Obviously PRMEM succeeds well in a phase retrieval where the Kramers–Kronig relation cannot be applied.

### 3. Existence of poles and zeros

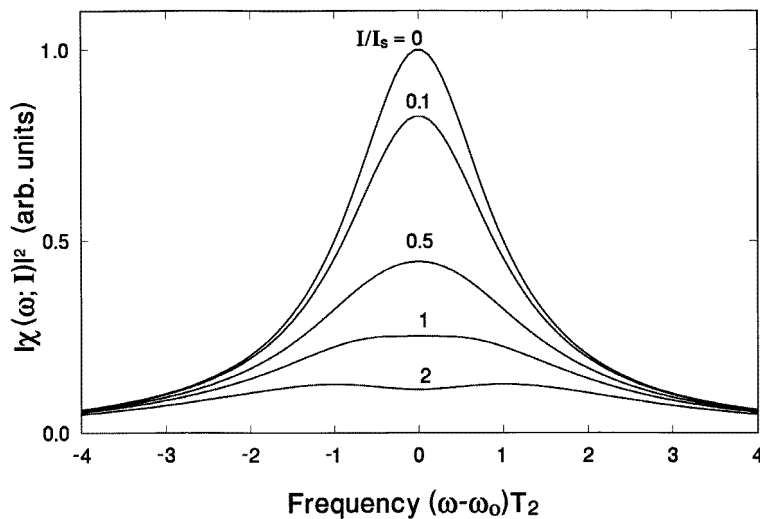
Quite often we can judge from the knowledge of the material properties and experimental set-up whether the total susceptibility is expected to be meromorphic or not. However, the information about the correct number of poles and/or zeros is not usually revealed by the intensity spectra. This problem can be present also in the spectra analysis of non-meromorphic Raman excitation profiles. The worst case might be that we have no idea at all whether the total susceptibility, whose modulus spectrum may resemble a familiar spectral Lorentzian line shape and therefore be misleading, has a meromorphic nature. Remembering that all that we may have available is a measured spectrum then the estimation of the number of poles (or zeros) may become an overwhelming problem. Of course, there is a mathematical method, namely the argument theorem, that involves a complex function and its derivative and also complex contour integration in order to estimate the number of poles and zeros as in the case of the Nyquist stability criterion of amplifiers [28]. Then, one has to know the function and the derivative as a function of complex variable. In practice the sole measurable quantity is the modulus of a meromorphic total susceptibility as a function of a *real* variable. Therefore, the argument theory does not work in practice. Fortunately the existence of poles and zeros of a meromorphic function can be estimated using another type of mathematical model. This model is based on Jensen's formula [29], which is well known in complex analysis but has received little attention in dispersion theory of physics. Jensen's formula is obtained here by applying a conformal mapping of the upper half plane (the lower half plane would do as well) onto a unit disc by the transformation

$$W = (\omega - i)(\omega + i) \quad (5)$$

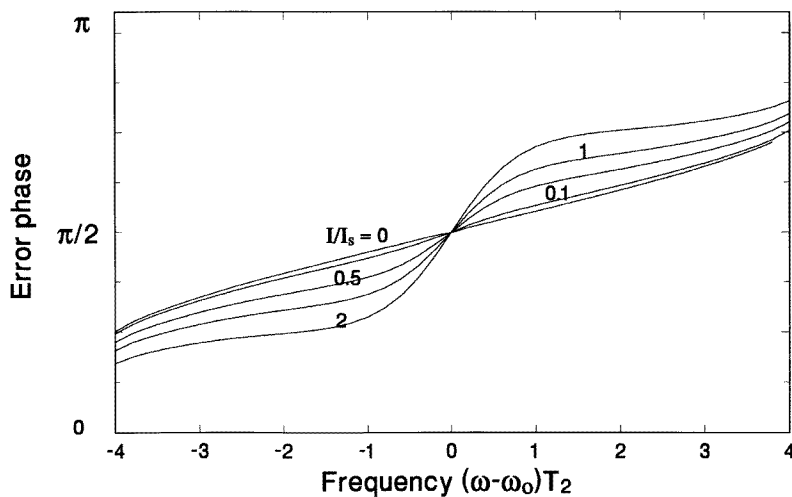
where  $i$  is the imaginary unit and  $\omega$  is the angular frequency, which is a real number. Now the real axis is mapped onto the boundary of the unit disc. We have to consider here the meromorphic permittivity of the material. The permittivity has the mathematical properties that are required to fulfil the assumptions imposed on the existence of Jensen's formula. The permittivity,  $\epsilon = 1 + \chi$ , where  $\chi$  is the total susceptibility, can have zeros  $z_k$ , and poles  $p_j$ , inside the unit disc. Note carefully that the zeros will be at different locations with  $\epsilon$  and  $\chi$ . If we denote the multiplicity of the poles by  $n_j$  and the multiplicity of zeros by  $m_k$ , then we can write Jensen's formula for the meromorphic permittivity as follows:

$$(2\pi)^{-1} \int_0^{2\pi} \ln |\epsilon(\exp(i\theta))| d\theta = \ln |\epsilon(0)| + \sum_k m_k \ln |z_k|^{-1} - \sum_j n_j \ln |p_j|^{-1} \quad (6)$$

where  $W = \exp(i\theta)$ ,  $|z_k| < 1$  and  $|p_j| < 1$ , and furthermore  $\epsilon(0)$  is neither zero nor infinite. The integrand of (6) involves data that are obtained by measurement. At first



(a)



(b)

**Figure 1.** (a) Total susceptibility spectra  $|\chi(\omega; I)|^2$  with intensity ratios  $I/I_s = 0, 0.1, 0.5, 1.0$  and  $2.0$ . (b) Corresponding error phase functions. Recovered (c)  $\text{Re}\chi$  and (d)  $\text{Im}\chi$  curves in cases of low ( $I/I_s = 0.1$ ) and high ( $I/I_s = 1.0$ ) intensity ratios.

the estimation of the value  $\epsilon(0)$  seems to be problematic, since the permittivity calculated at the centre of the unit disc corresponds to the imaginary frequency,  $i$ , in the original angular frequency plane. Fortunately, the purely imaginary frequency matches quite well the zero-frequency value of the permittivity in the original angular frequency plane. Indeed the features of the optical spectra of the permittivity are very far away from the ‘static value’ of the permittivity at  $i$ . This static value is practically speaking governed by the linear susceptibility of a material. Then it is possible to estimate the permittivity at the

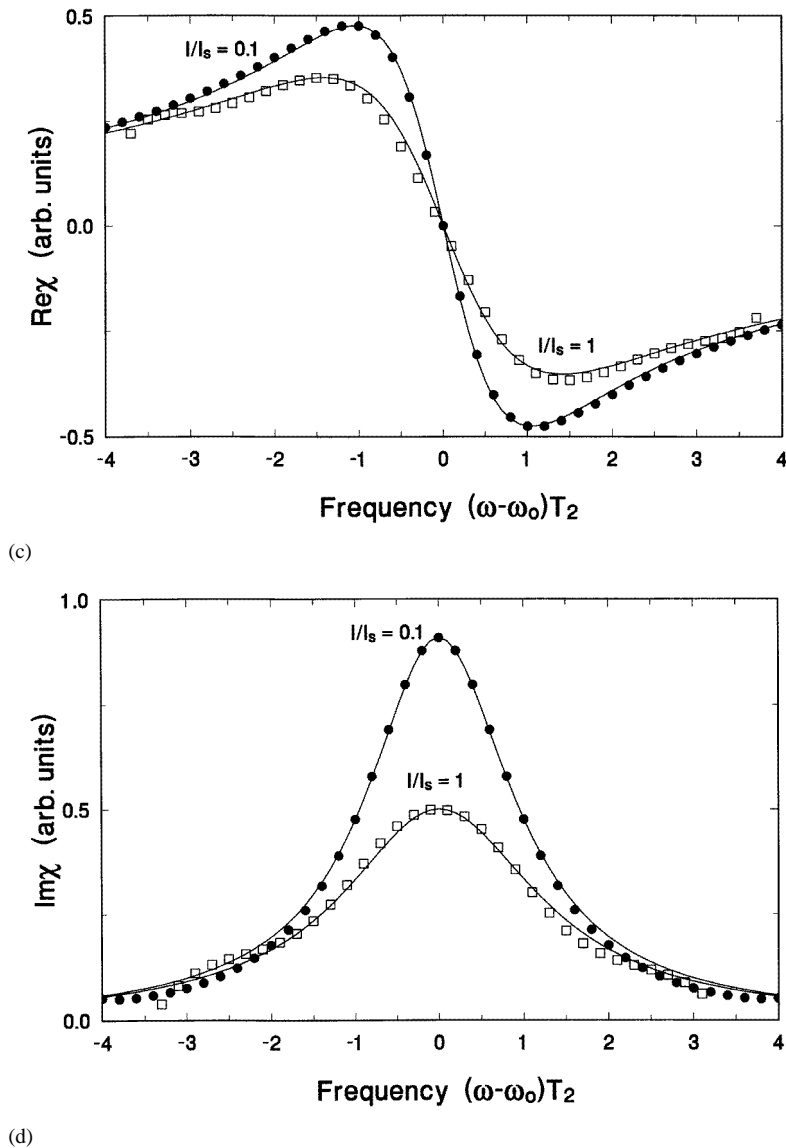


Figure 1. (Continued)

centre of the unit disc with the sum rule given by King [30] as follows:

$$\epsilon(0) = 1 + \frac{2}{\phi} \int_0^{\infty} \omega \operatorname{Im} \chi^{(1)}(\omega) d\omega / (\omega^2 + 1) \quad (7)$$

where  $\operatorname{Im}$  stands for the imaginary part and  $\chi^{(1)}$  is the linear complex susceptibility. Note that we first wish to construct the meromorphic total susceptibility by the PRMEM procedure and thereafter try to estimate the existence of zeros and poles by equation (6). In the event that the total susceptibility has strong asymptotic fall-off, integrations at limited integration intervals in equations (6) and (7) are sufficient. If the total susceptibility has a weak asymptotic fall-off, then the data extrapolations are certainly needed. A solution for the

last-mentioned case could be the coupling of PRMEM procedure with the method of spectra estimations using the model of Hulthen [31]. According to Hulthen it is possible to estimate a spectrum  $|\chi|$  beyond the measurement range provided that the real and imaginary parts of  $\chi$  are known within some limits.

#### 4. Conclusions

We have presented brief notes about the generalization of the phase retrieval from the modulus of a meromorphic total susceptibility, with poles and zeros, by the maximum-entropy model. In addition, a mathematical basis for the estimation of zeros and poles of meromorphic permittivity was introduced. The estimation is based on Jensen's formula, which is a well known result in complex analysis but has not received, as far as the authors known, attention in dispersion theory. Jensen's formula may find applications also in spectrum analysis of the Raman emission profiles.

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