Kramers-Kronig relations and sum rules of negative refractive index media

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Abstract. Negative refractive index media have become a hot topic in physics due to their proposed revolutionary properties, which would have drastic consequences in design of novel optical devices. We show that Kramers-Kronig relations connecting the real and imaginary parts of the complex refractive index of absorbing media are valid even though the real refractive index may take negative value at some spectral range. In addition universal sum rules for linear optical constants of negative index media are also valid. This means that negative refractive index media are not fundamentally different from regular media. Hence, any spectrum measured from negative refractive index media can be analyzed using dispersion relations and sum rules, which have so far provided information on the optical properties of materials.

PACS. 77. Dielectrics, piezoelectrics, and ferroelectrics and their properties – 78.20.Ci Optical constants (including refractive index, complex dielectric constant, absorption, reflection and transmission coefficients, emissivity)

1 Introduction

Recently, the work of Veselago [1] has stimulated discussion of the possibility of negative linear refractive index materials. The issue of negative index has been a source of many studies of so-called metamaterials. Metamaterials, which are structured materials and which have been under experimental and numeric studies at microwave region, are beyond the scope of this study, since we are interested in media which may possess negative linear real refractive index at optical frequencies. Pendry [2] gave theoretical prediction for the existence of negative linear real refractive index medium, which could provide a perfect lens. Nevertheless, Garcia and Nieto-Vesperinas [3] pointed out that dispersiveless and lossless negative refractive index medium will not provide a perfect lens. Support to the Garcia and Nieto-Vesperinas was given by Valanju et al. [4] who claimed that causality and finite signal speed prohibit the existence of negative index medium. However, Pokrovsky and Efros [5] have pointed out some mistakes e.g. in the study of Valanju et al. [4]. Furthermore, Pokrovsky and Efros suggested that any medium may be described by both positive and negative linear refractive index. Recently, Zhang et al. [6] gave strong evidence on negative refraction at optical spectral region using crystals. Peiponen et al. [7] suggested that a negative index could be accomplished also using nonlinear optical properties of homogeneous or nanostructured media. In the regime of linear optics of positive refractive index the validity of Kramers-Kronig (K-K) relations [8] has been proved for nanocomposites [9]. Since causality is fundamental physical property that implies the validity of the K-K relations we may expect that K-K relations and sum rules [10,11] are valid also for negative linear real refractive index of homogeneous medium, which may be nanostructured. Since the K-K relations and sum rules are fundamental tools in practical analysis of optical spectra both from optically linear [12] and nonlinear media [13,14] one can expect that optical spectroscopy of negative index media will have importance in basic studies and engineering of such materials.

2 Kramers-Kronig relations and sum rules for negative linear real refractive index

We approach the problem of analyzing the optical properties of negative refractive index media from the perspective of basic classical theory of electromagnetism. We consider the angular frequency dependent permittivity $\varepsilon(\omega)$ and permeability $\mu(\omega)$ of a nanostructured medium. The material parameters have to be treated as complex valued functions, where the imaginary parts are related to energy loss processes. Our starting point is the definition of
the complex refractive index, by the assumption of time-
dependence exp(−iωt) of fields, as follows [8]:
\[
N(\omega) = n(\omega) + i\kappa(\omega) = \sqrt{\varepsilon(\omega)\mu(\omega)}
= \sqrt{[\varepsilon_1(\omega) + i\varepsilon_2(\omega)][\mu_1(\omega) + i\mu_2(\omega)]},
\]
where \(\omega\) is frequency, \(n(\omega)\) is the real refractive index, \(\kappa(\omega)\) is the extinction coefficient and \(\varepsilon_{1,2}(\omega)\) and \(\mu_{1,2}(\omega)\) have their usual meanings presenting the real and imaginary parts of the permittivity and permeability, respectively. An issue in the literature of negative index media has been the choice of the sign of the square root in equation (1), i.e. minus sign for negative index media. The sign convention of the real parts of the permittivity and permeability of homogeneous media [15] and uniaxial medium [16] have been discussed in the literature. Here we wish to treat a rather general case, namely we allow the switching of the sign of the real refractive index between negative and positive values as a function of wavelength. Therefore, we do not explicit spell out the sign in front of the square root in equation (1) since it may be positive for some spectral region and negative for another region, depending on the signs of the real parts of the permittivity and permeability. Actually, the property of switching the refractive index between negative and positive values would have engineering applications, for instance, in optical switching, and optical computing.

The very basic principle of causality of the response of any material to an external electromagnetic field (in the case of electromagnetic radiation both electric and magnetic field propagate simultaneously in the medium) ensure that both permittivity and permeability, which both are holomorphic functions in the upper half of the complex frequency plane, obey K-K relations. The appearance of the poles in the lower half of the plane is due to the choice of the present time dependence of the field. If the field is chosen to oscillate as exp(\(i\omega t\)), then the poles are located in the upper half plane, and then \(\varepsilon\) and \(\mu\) are treated as holomorphic functions in the lower half plane. In the present case we assume an isotropic insulating effective medium. Then, one may ask if K-K relations can also be given for the complex refractive index? The answer is yes and this matter has been dealt with in the literature [17,18]. Actually, a great deal of the present knowledge of complex linear refractive index of transparent and opaque media is based on the utilization of transmission and reflection spectroscopies, and appropriate K-K relations that couple the real refractive index and extinction coefficient, and also reflectance and phase. In the case of an anisotropic medium the expressions for K-K relations that involve complex refractive index were studied by Ginzburg and Meiman [19]. The critical point in the derivation of the K-K relations for the complex refractive index of isotropic media is the holomorphicity of the function given in equation (1) (the sign convention either plus or minus has no role with regard to the holomorphism of the complex refractive index). It is important that the function \(\sqrt{\varepsilon\mu}\) has no zeros in the upper half plane, which would essentially be the branch points of this function. Fortunately, there are no such branch points, and Räty et al. [20] have given a rigorous proof of the holomorphy of the complex refractive index and the validity of the K-K relations for the complex refractive index for nonmagnetic media. The argumentation of the absence of branch points in the case of \(\sqrt{\varepsilon\mu}\) follows the same outlines as those presented in the book of Räty et al. [20].

Next we deal with the validity of K-K relations and sum rules for the peculiar linear real refractive index which takes both negative and positive values at the infinite angular frequency space. For the sake of simplicity and clarity we assume that the relative permittivity and permeability of the medium (for metals the DC conductivity term has to be taken into account) can be described by single resonance Lorentzians as follows:
\[
\varepsilon(\omega) = 1 + \frac{\omega_{pe}^2}{\omega_0^2 - \omega^2 - i\gamma_\varepsilon\omega}
\]
and
\[
\mu(\omega) = 1 + \frac{\omega_{pm}^2}{\omega_0^2 - \omega^2 - i\gamma_\mu\omega},
\]
where \(\omega_{pe/m}\) are the plasma frequencies, \(\omega_0/m\) are the resonance frequencies and \(\gamma_{\varepsilon/\mu}\) are the corresponding line widths. The symbol “e” is related to electric and the symbol “m” to magnetic. The fundamental reason why we use the above Lorentzian model is because of the crucial properties of holomorphy in the upper complex \(\omega\)-plane, because of the sufficiently fast asymptotic fall off (\(\omega^2\)) of the quantities \(\varepsilon(\omega) - 1\) and \(\mu(\omega) - 1\), and because of its symmetry properties with respect of conjugation of the variable \(\omega\) [8,11,18,21]. We emphasize that the theory below is not restricted to the Lorentzian model but holds more generally, and to systems that may involve multiple resonances instead of a single resonance. Rigorous theory of the sufficient asymptotic behavior and the holomorphicity of the optical constants is based on the quantum mechanical treatment and the use of Kubo theory [13,22].

From equations (2) and (3) we can find under which conditions the real parts of the permittivity \(\varepsilon_1(\omega)\) and permeability \(\mu_1(\omega)\) are negative. Reminding that the index “\(e\)” refers to the quantities related to the permittivity and the index “\(m\)” to those related to the permeability, we obtain that under the condition:
\[
\omega_{pe/m}^2 > \gamma_{e/m}^2 + 2\gamma_{e/m}\omega_0/e/m,
\]
that the physical quantity under investigation is negative in the interval \([\omega_{le/m}, \omega_{ue/m}]\), where:
\[
\omega_{le/m} = \left[\frac{2\omega_{pe}^2/\omega_0^2 + \omega_{pe/m}^2 - \gamma_{e/m}^2}{2} - \frac{(\omega_{pe/m}^4 + \gamma_{e/m}^4 - 2\omega_{pe}^2/\omega_0^2\gamma_{e/m}^2 - 4\gamma_{e/m}^2\omega_{pe/m}^2)^{1/2}}{2}\right]^{1/2},
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