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Araştırma Makalesi

An Analysis of Dielectric Response[†]

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Abstract: We perform a mathematical analysis of the dielectric spectrum within the framework of classical dispersion theory. The analysis is a complex plane analysis. With this analysis, a holistic analytical method is derived to decompose the imaginary part of dielectric function into its fundamental components. A complex plane is formed using the complex feature of the dielectric function. In this plane, each loop of the function $\varepsilon_2(\varepsilon_1 - 1)$, which completed to a circle, represents the linear optical response for a single Lorentz oscillator. The parameters of each Lorentz oscillator such as natural frequency, energy, and half-width are calculated by analyzing the circles.

Bir Dielektrik Tepki Analizi

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Anahtar Kelimeler

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Rezonans frekansı

Öz: Bu çalışmada, klasik dispersiyon teorisi çerçevesinde dielektrik spektrumun matematiksel bir analizi yapılmaktadır. Bu analiz, bir kompleks düzlem analizidir. Bu analizle dielektrik fonksiyonun sanal kısmını temel bileşenlerine ayırmak için bir bütüncül analitik yöntem türetilmektedir. Dielektrik fonksiyonun kompleks özelliği kullanılarak bir kompleks düzlem oluşturulmuştur. Bu düzlemde, bir çembere tamamlanan her bir $\varepsilon_2(\varepsilon_1 - 1)$ fonksiyon döngüsü, tek bir Lorentz osilatörü için lineer optik tepkiyi temsil eder. Her bir Lorentz osilatörünün, doğal frekans, enerji ve yarı genişlik gibi parametreleri, bu çemberler analiz edilerek hesaplanır.

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1. Introduction

The optical properties of a dielectric medium, which generally result from the interaction of the medium with the light, are classified in three main groups: reflection, propagation and transmission. Some of the incident light is reflected from the front surface of the medium and it leads to the optical properties of medium such as reflectivity and reflection. The fact that the rest of incident light enters the medium and interacts with the medium is the reason for optical properties such as absorption, scattering and refraction. While some part of the light reaching the back surface is reflected back from this surface, some part of it passes the surface and leaves the medium. This event leads to properties of the medium such as optical transmittance.

The dispersion theory explains how the light propagates in the medium and the process of interaction of light with the medium. Indeed, it is based on the oscillations of bound electrons inside the medium (Wooten 1972; Fox 2001; Moss 1959; Nye 1957; Peiponen et al., 1999; Hodgson 1970). The reason for these oscillations is the force which is exerted by the electric field component of the electromagnetic field of incident light on the bound electrons in the medium. In the other words, the electric field of the incident light acts as a coercive force on the bound electrons. This motion is described by the driven harmonic oscillator model called Lorentz oscillator (LO) (Wooten 1972; Fox 2001; Moss 1959; Lorentz 1916). Since the effect of the magnetic field of the light on the motion is very small, it is neglected in this model. The interaction of bound electrons with the field of light is governed by the laws of the classical electrodynamics, that is, Maxwell's laws.

All forces exerted on each bound electron modelled as a LO inside a dielectric medium are shown in Figure 1 (Figure 1 is partly taken from Peiponen et al., 1999). In this model, the bound electron is a driven harmonic oscillator which is driven by an external driving force F_d . The other forces exerted on the electron are the restoring force F_r and damping force F_s in Figure 1. Therefore, from Newton's second law, the equation of motion of the electron is written as follows:

$$m\ddot{x} + m\Gamma\dot{x} + m\omega_0^2x = -eE_0e^{-i\omega t}, \quad (1)$$

where, m , e , and ω_0 are mass, charge, and natural oscillation frequency of the electron, respectively; ω and E_0 are the frequency and the amplitude of the electrical field of incident light, respectively, and Γ is the damping parameter. The constant k in Figure 1, which comes from Hook's law, is the spring constant and defined as $k = m\omega_0^2$. As it is easily understood, in Eq. (1), $F_s = -m\Gamma\dot{x}$, $F_r = -m\omega_0^2x$, and $F_d = -eE_0e^{-i\omega t}$ which comes from Lorentz force.

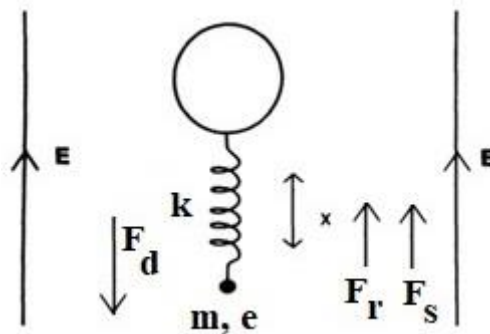


Figure 1. Lorentz oscillator (LO) model.

The solution of Eq. (1) gives the complex displacement (or amplitude) of LO:

$$x(t) = \frac{-eE_0/m}{(\omega_0^2 - \omega^2)^2 + (\Gamma\omega)^2} \left\{ [(\omega_0^2 - \omega^2) \cos \omega t + \Gamma\omega \sin \omega t] \right\}, \quad (2)$$

and the modulus of the complex displacement can be calculated easily as follows:

$$|x(t)| = \frac{eE_0/m}{\sqrt{(\omega_0^2 - \omega^2)^2 + (\Gamma\omega)^2}}. \quad (3)$$

The maximum value of amplitude $|x(t)|$ can be found taking the derivative of the expression in Eq. (3) with respect to frequency ω and equalling it to zero. Thus, the resonance frequency corresponding to this maximum amplitude of the LO becomes

$$\omega_r^2 = \omega_0^2 - \frac{\Gamma^2}{2}. \quad (4)$$

2. Material and Methods

2.1. Complex Linear Dielectric Susceptibility and Dielectric Function

Using the complex displacement given by Eq. (2), the electric dipol of LO induced by the electric field of light is written as follows:

$$d = \frac{e^2 E_0}{m} \frac{e^{-i\omega t}}{(\omega_0^2 - \omega^2) - i\Gamma\omega}. \quad (5)$$

If there are N LOs with the same natural frequency ω_0 in the unit volume, then we get the induced polarization

$$P = \frac{Ne^2 E_0}{m} \frac{e^{-i\omega t}}{(\omega_0^2 - \omega^2) - i\Gamma\omega}. \quad (6)$$

If the unit volume contains LOs which oscillate at different natural frequencies, then, the induced polarization per unit volume becomes (Wooten 1972):

$$P = \frac{e^2 E_0}{m} \sum_k \frac{N_k e^{-i\omega t}}{(\omega_k^2 - \omega^2) - i\Gamma_k \omega}. \quad (7)$$

As can be seen from this equation, since there exist N_k oscillators with damping parameter Γ_k and natural frequency ω_k in the unit volume, the total number of oscillators is (Wooten 1972)

$$\sum_k N_k = N. \quad (8)$$

It is well known from the linear dispersion theory that the linear dielectric susceptibility tensor $(\chi_d)_{ij}$, which is a 2-rank symmetric tensor, relates the induced polarization vector with the electrical field vector as follows (Nye 1957):

$$P_i = \varepsilon_0 (\chi_d)_{ij} E_j, \quad (9)$$

where ε_0 is the dielectric permittivity of vacuum. We can obtain a complex formula for each component of the linear dielectric susceptibility tensor by combining Eqs. (6) and (9),

$$\chi_d(\omega) = \frac{(Ne^2/m\varepsilon_0)}{(\omega_0^2 - \omega^2) - i\Gamma\omega}, \quad (10)$$

and it can be divided into real and imaginary parts easily:

$$Re(\chi_d(\omega)) = \frac{Ne^2}{m\varepsilon_0} \frac{(\omega_0^2 - \omega^2)}{(\omega_0^2 - \omega^2)^2 + (\Gamma\omega)^2}, \quad (11.a)$$

$$Im(\chi_d(\omega)) = \frac{Ne^2}{m\epsilon_0} \frac{\Gamma\omega}{(\omega_0^2 - \omega^2)^2 + (\Gamma\omega)^2} \quad (11.b)$$

We know that the relation between the dielectric permittivity tensor and the linear dielectric susceptibility tensor of a dielectric medium is (Nye 1957)

$$\epsilon_{ij} = \epsilon_0(\delta_{ij} + (\chi_d)_{ij}), \quad (12)$$

where δ_{ij} is the 2-rank unit tensor. The relative dielectric permittivity tensor of the medium can be easily written from expression (12):

$$(\epsilon_r)_{ij} = \frac{\epsilon_{ij}}{\epsilon_0} = (\delta_{ij} + (\chi_d)_{ij}). \quad (13)$$

Thus, each components of the relative dielectric tensor of the medium has the form of

$$\epsilon_r(\omega) = 1 + \frac{(Ne^2/m\epsilon_0)}{(\omega_0^2 - \omega^2) - i\Gamma\omega}. \quad (14)$$

On the other hand, each component of the relative dielectric permittivity tensor is a function of frequency of incident light and it has a complex form of

$$\epsilon_r(\omega) = \epsilon_1(\omega) + i\epsilon_2(\omega), \quad (15)$$

thus, its real and imaginary parts become

$$\epsilon_1(\omega) = Re(\epsilon_r(\omega)) = 1 + \frac{Ne^2}{m\epsilon_0} \frac{(\omega_0^2 - \omega^2)}{(\omega_0^2 - \omega^2)^2 + (\Gamma\omega)^2}, \quad (16.a)$$

$$\epsilon_2(\omega) = Im(\epsilon_r(\omega)) = \frac{Ne^2}{m\epsilon_0} \frac{\Gamma\omega}{(\omega_0^2 - \omega^2)^2 + (\Gamma\omega)^2}, \quad (16.b)$$

respectively. The frequency dependencies of the real and the imaginary parts given by Eqs. (16) are plotted in Figure 2 using a online graphing program (Desmos), for the values of $(Ne^2/m\epsilon_0) = 100$, $\omega_0 = 3$, and $\Gamma = 2$, in arbitrary units. The resonance frequency calculated for these values is $\omega_r = 2.65$ in arbitrary units.

The difference of frequency values which correspond to the minimum and maximum points of the function $\epsilon_1(\omega)$ gives the value of Γ which is called the half-width of the LO because this value is also equal to the half-width of $\epsilon_2(\omega)$. This frequency region, where the real dielectric function decreases with the frequency, is called abnormal dispersion region. The regions where the real dielectric function increases with frequency are normal dispersion regions. The point at which the curve of $\epsilon_1(\omega)$ intersects with the vertical axis determines the static dielectric constant which is the relative dielectric function at low frequencies defined as

$$(\epsilon_r)_{st} = \epsilon_r(0) = 1 + \frac{Ne^2}{m\epsilon_0\omega_0^2}. \quad (17.a)$$

The calculated static dielectric constant for the example given in Figure 2 is $(\epsilon_r)_{st} = 12.11$. The value of relative dielectric function at high frequencies converges to unity,

$$(\epsilon_r)_\infty = \epsilon_r(\infty) = 1, \quad (17.b)$$

therefore, the difference of values of $(\epsilon_r)_{st}$ and $(\epsilon_r)_\infty$ is

$$(\varepsilon_r)_{st} - (\varepsilon_r)_\infty = \frac{Ne^2}{m\varepsilon_0\omega_0^2}. \quad (17.c)$$

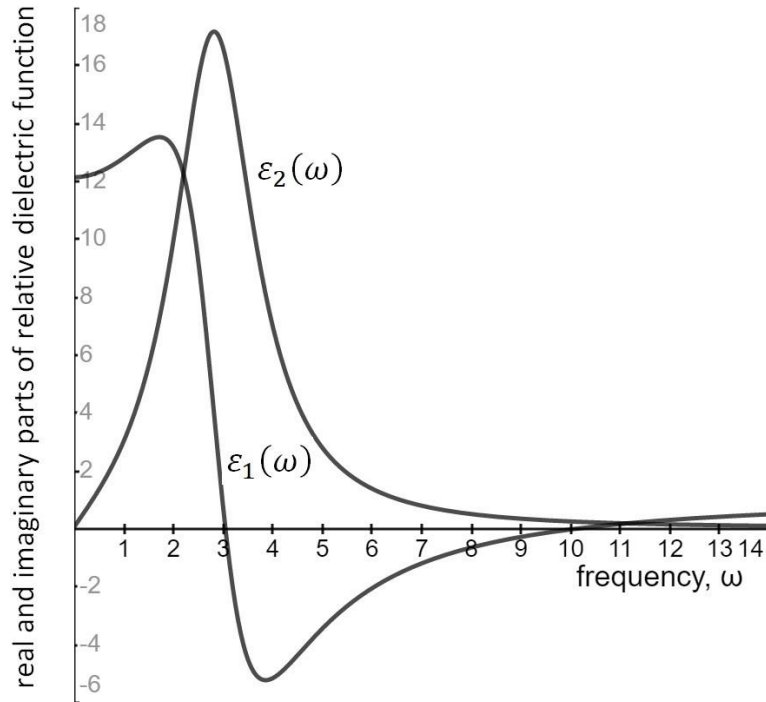


Figure 2. Frequency dependencies of the real and imaginary parts of relative dielectric function.

The maximum value of $\varepsilon_2(\omega)$ is defined as

$$\varepsilon_2(\omega)_{max} = \varepsilon_{2m} = \frac{Ne^2}{m\varepsilon_0\Gamma\omega_0}, \quad (17.d)$$

and its calculated value is $\varepsilon_{2m} = 16.67$ for the example in Figure 2.

3. Results

3.1. Argand diagram for LO

Because the curve of the imaginary part of a complex function relative to its real part is called Argand diagram of this complex function, Argand diagrams for the relative dielectric function of a dielectric medium defined by (15) can be formed with the curves of

$$(\varepsilon_2)_k = f((\varepsilon_1)_k - 1)$$

where the index k represents a LO with the natural oscillation frequency ω_k . Using Eqs. (16), we can find the following equation which relates the functions $\varepsilon_1(\omega) - 1$ and $\varepsilon_2(\omega)$:

$$(\varepsilon_1 - 1)^2 + (\varepsilon_2)^2 - \left[\frac{a(\omega_0^2 - \omega^2)}{(\omega_0^2 - \omega^2)^2 + (\Gamma\omega)^2} \right]^2 - \left[\frac{a\Gamma\omega}{(\omega_0^2 - \omega^2)^2 + (\Gamma\omega)^2} \right]^2 = 0,$$

where $a = Ne^2/m\varepsilon_0$. With some rearrangement this equation becomes

$$(\varepsilon_1 - 1)^2 + (\varepsilon_2)^2 - (\varepsilon_1 - 1) \frac{a(\omega_0^2 - \omega^2)}{(\omega_0^2 - \omega^2)^2 + (\Gamma\omega)^2} - \varepsilon_2 \frac{a\Gamma\omega}{(\omega_0^2 - \omega^2)^2 + (\Gamma\omega)^2} = 0.$$

In this equation, if the constant a is taken as $a = \omega_0^2 \varepsilon_{10}$ in the third term (where $\varepsilon_{10} = \varepsilon_r(0) - 1$ from Eq. (17.a)) and as $a = \Gamma \omega_0 \varepsilon_{2m}$ in the fourth term (from Eq. (17.d)), then, we easily obtain the equation of

$$(\varepsilon_1 - 1)^2 + (\varepsilon_2)^2 - (\varepsilon_1 - 1) \varepsilon_{10} \frac{\omega_0^2(\omega_0^2 - \omega^2)}{(\omega_0^2 - \omega^2)^2 + (\Gamma \omega)^2} - \varepsilon_2 \varepsilon_{2m} \frac{\Gamma^2 \omega_0 \omega}{(\omega_0^2 - \omega^2)^2 + (\Gamma \omega)^2} = 0. \quad (18)$$

The imaginary dielectric function given by (16.b) gets its the maximum value of $\varepsilon_2 = \varepsilon_{2m}$ if the condition of

$$(\omega_0^2 - \omega^2)^2 + (\Gamma \omega)^2 = \Gamma^2 \omega_0 \omega \quad (19)$$

is provided. Thus, under this condition, Eq. (18) transforms into equation of

$$(\varepsilon_1 - 1)^2 + (\varepsilon_2)^2 - (\varepsilon_1 - 1) \varepsilon_{10} \frac{\omega_0}{\omega_0 + \omega} - \varepsilon_2 \varepsilon_{2m} = 0. \quad (20)$$

At frequencies that the frequency of light approaches to the natural oscillation frequency of the LO, the term $(\omega_0/\omega_0 + \omega)$ in the Eq. (20) goes to a half:

$$\lim_{\omega \rightarrow \omega_0} \left(\frac{\omega_0}{\omega_0 + \omega} \right) = \frac{1}{2}. \quad (21)$$

Under this limit, Eq. (20) is rewritten as follows:

$$(\varepsilon_1 - 1)^2 + (\varepsilon_2)^2 - \frac{(\varepsilon_1 - 1) \varepsilon_{10}}{2} - \varepsilon_2 \varepsilon_{2m} = 0. \quad (22)$$

With some little rearrangement Eq. (22) can be easily rewritten in the form of

$$\left((\varepsilon_1 - 1) - \frac{\varepsilon_{10}}{4} \right)^2 + \left(\varepsilon_2 - \frac{\varepsilon_{2m}}{2} \right)^2 = \left(\frac{\varepsilon_{10}}{4} \right)^2 + \left(\frac{\varepsilon_{2m}}{2} \right)^2. \quad (23)$$

As can be easily seen, this equation is a circle equation in the complex plane of $(\varepsilon_1 - 1, \varepsilon_2)$. The center and the radius of this circle are given by $\left(\frac{\varepsilon_{10}}{4}, \frac{\varepsilon_{2m}}{2} \right)$ and $\sqrt{\left(\frac{\varepsilon_{10}}{4} \right)^2 + \left(\frac{\varepsilon_{2m}}{2} \right)^2}$, respectively.

Argand diagram for LO (ADLO) in the example of Figure 2 has been plotted in Figure 3 using Eq. (22) (or Eq. (23)). Taking $\varepsilon_1 - 1 = 0$ in Eq. (22), we find the points at which the ADLO intersects with ε_2 -axis as $\varepsilon_2 = 0$ and $\varepsilon_2 = \varepsilon_{2m} = 16.7$. Similarly, taking $\varepsilon_2 = 0$ in Eq. (22), the points at which the ADLO intersects with $(\varepsilon_1 - 1)$ -axis are found as $\varepsilon_1 - 1 = 0$ and $\varepsilon_1 - 1 = \frac{\varepsilon_{10}}{2} = 5.6$. As it is understood, if an ADLO can be drawn as in Figure 3, then, the maximum value of the imaginary dielectric function ε_{2m} , the static dielectric constant $(\varepsilon_r)_{st}$, the natural oscillation frequency ω_0 , and the half-width Γ of LO can be immediately determined from the ADLO. The natural oscillation frequency ω_0 is the frequency corresponding to the point that the ADLO intersects with vertical axis in Figure 3. It can be easily proved by taking $\varepsilon_2 = \varepsilon_{2m}/2$ in Eq. (23) that the upper half of the ALDO in Figure 3 takes place in the range of frequency from $\omega_0 - \frac{\Gamma}{2}$ to $\omega_0 + \frac{\Gamma}{2}$, that is, the half-width of the single LO is the Γ . The energy of the single LO, $E_0 = \hbar \omega_0$, also can be immediately calculated using the natural oscillation frequency ω_0 . While the real part of the complex relative dielectric function given by (16.a) is a measure of the response of dielectric medium to the electric field of incident light, the imaginary part given by (16.b) is a measure of the energy loss (or absorption) of incident light. Thus, if the energy of the LO is known, then, one of the absorption energies which corresponds to one of the interband transitions is found.

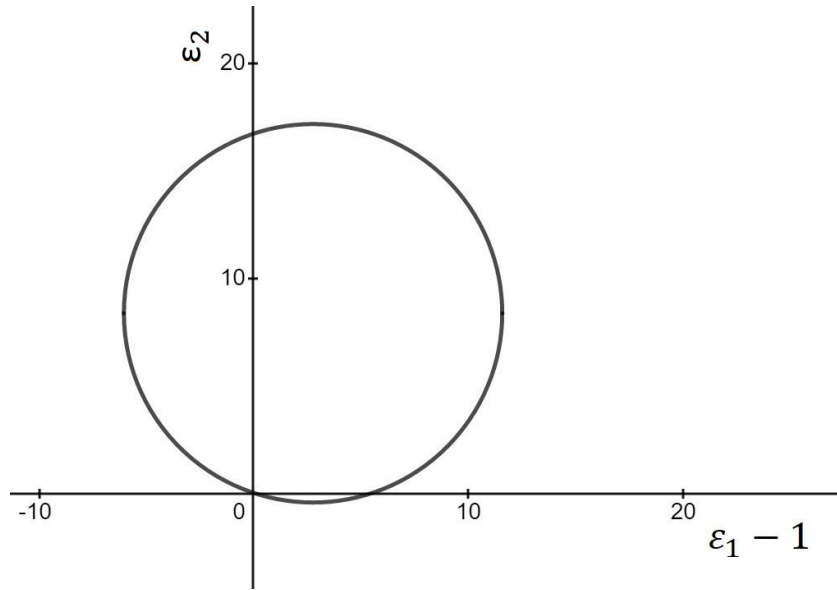


Figure 3. Argand diagram for LO (ADLO).

3.2. An application: Decomposition of dielectric spectrum into single LOs

The dielectric spectrum of a dielectric crystal consists of the combination of a large number of single LOs and the decomposition of the spectrum into single LOs is one of the problems of the optical spectroscopy of solids. There exist some works in the literature about the solution of this problem (Aleynikova et al. 2004; Cabuk and Mamedov 2004; Kalugin and Sobolev 2005; Piccard 1986; Sobolev et al. 2000). The curve of $\epsilon_2(\omega)$ versus $\epsilon_1(\omega) - 1$ for the cubic $RbNbO_3$ crystal has been plotted in Figure 4 for the purpose of application of the analytical methods derived in Section 3. The data for this graphic were produced from Erzen and Akkus (2018).

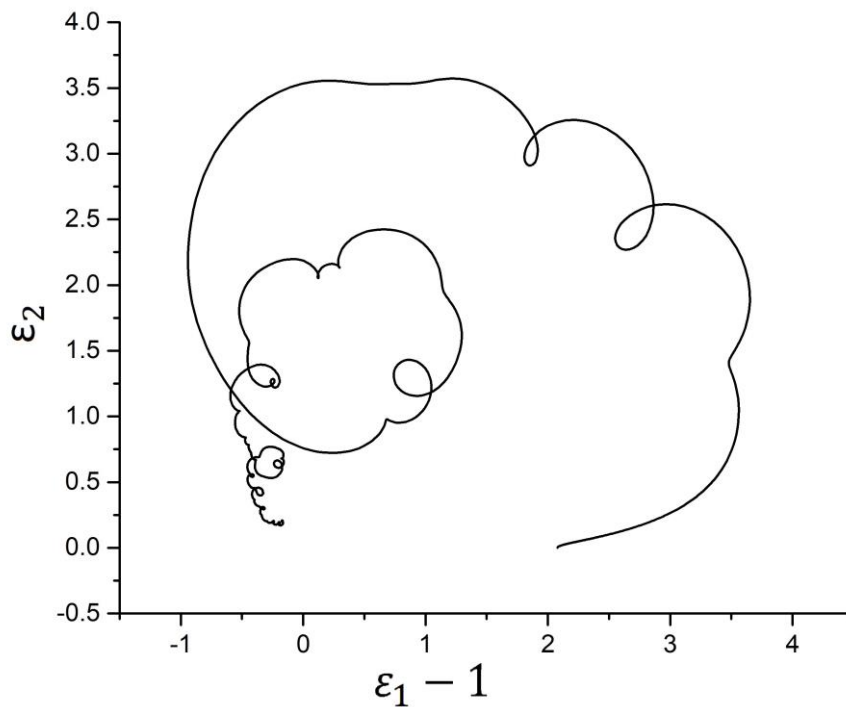


Figure 4. Argand diagram of multiple LOs for cubic $RbNbO_3$.

Each of the tangent circles which are drawn to the Argand curve at certain points in Figure 4 corresponds to a single LO. The drawn tangent circles for five different single LOs are shown in Figure 5. These circles have been redrawn as ADLO in Figure 6.

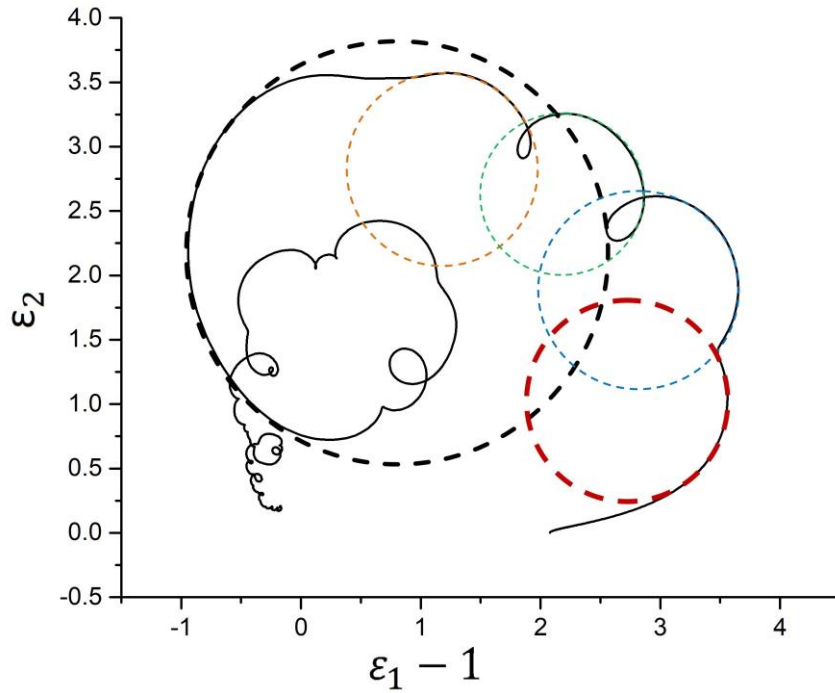


Figure 5. Five tangent circles (ADLOs) drawn on the Argand diagram of cubic $RbNbO_3$.

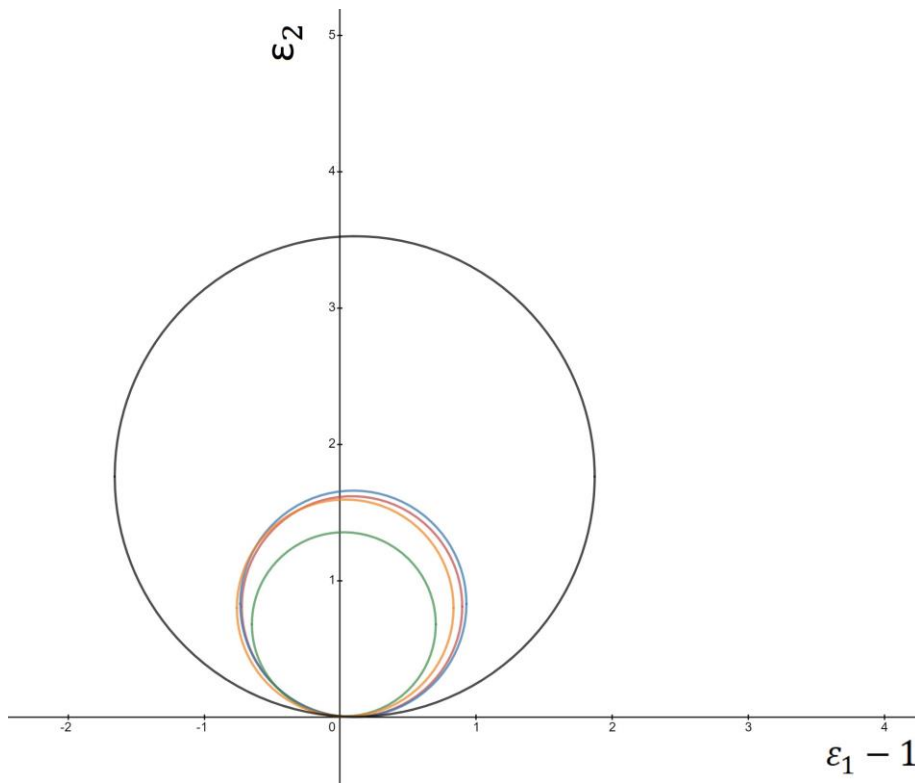


Figure 6. Five ADLOs for cubic $RbNbO_3$.

The analysis of five ADLOs drawn in Figure 6 has been performed with the help of analytical formulae derived in Section 3. The calculated parameters of the five ADLOs obtained from the

analysis are given in Table 1. The first column in Table 1 contains the colours of single ADLOs drawn in Figures 5 and 6. The second column contains the maximum values of imaginary dielectric functions which are defined by the Eq. (17.d) and calculated for each single LO. The ε_{2m} values were calculated from Figure 5 and Figure 6. The value of ε_{2m} for each LO is the value of the point that the ADLO intersects with vertical axis (except for the point that the circle intersects with vertical axis at the origin). The ε_{10} values, which associated with the static dielectric constant of each oscillator, are given in the third column. The value of ε_{10} for each single LO is twice of the value of the point that the ADLO intersects with horizontal axis (except for point that the circle intersects with horizontal axis at the origin).

Table 1. Calculated parameters of single LOs

ADLO	ε_{2m}	ε_{10}	ω_0 (1.52×10^{15}) rad/s	Γ (1.52×10^{15}) rad/s	E_0 (eV)
Red	1.61	0.37	4.41	1.00	4.41
Blue	1.65	0.40	4.44	1.08	4.44
Green	1.35	0.11	5.54	0.40	5.54
Orange	1.59	0.15	6.25	0.60	6.25
Black	3.52	0.44	6.69	0.84	6.69

The fourth column in Table 1 consists of the natural oscillation frequency values which calculated for each single LO. These frequency values are the frequency values corresponding to the points that each ADLO intersects with vertical axis (excluding the origin) in Figure 6. In the fifth column, the half-width values calculated for each single LO are given. The half-width parameter Γ is also the damping coefficient of each LO, which take place in Eq. (1). The Γ values have been calculated from the difference of frequency values correspond to the end points of the horizontal diameters of the ADLOs in Figure 6. In the last column, the calculated photon energy values corresponding to the natural oscillation frequency of each LO are given.

Finally, the imaginary part of the relative dielectric function drawn in Figure 4 and the imaginary dielectric functions of the five single LOs have been plotted in a narrow frequency range in Figure 7 using Eq. (16.b) and the calculated parameters in Table 1.

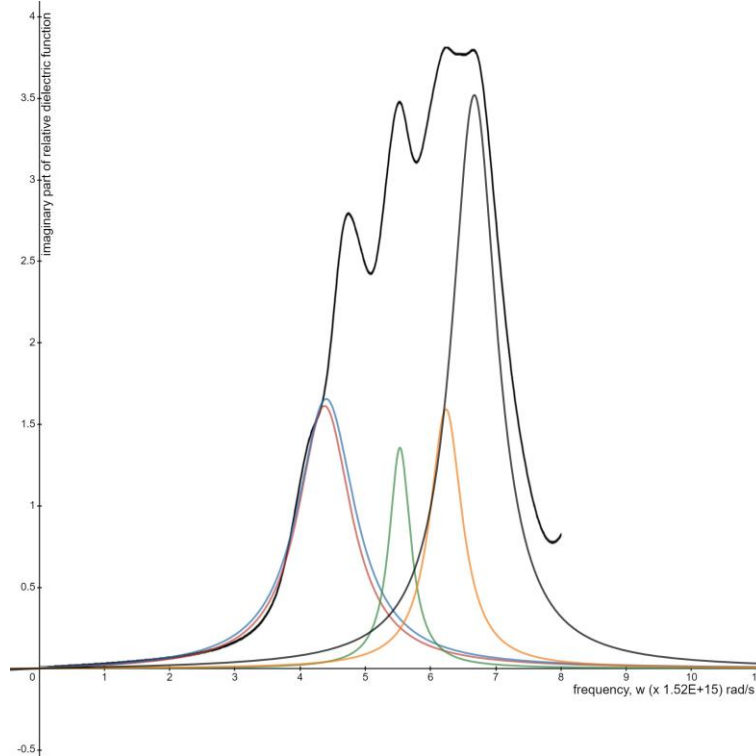


Figure 7. Five single LOs on the $\varepsilon_2(\omega)$ spectrum of cubic $RbNbO_3$.

4. Discussion and Conclusion

As a main theory of optics the dispersion theory explains the interactions of the light propagating in a medium. Although the optical properties are reduced to constant physical quantities in some special cases depending on the structural symmetry of the medium, they are generally tensor quantities whose all components are the function of the frequency of the light. One of the optical properties of the medium is the dielectric tensor of the medium. This tensor is a 3-dimensional and 2-rank tensor and has generally nine components. As each complex component depicts the dielectric spectrum in a special crystallographic direction, the dielectric spectrum determines all the optical properties of the medium. The classical dispersion theory of dielectrics is based on the harmonic motion model of the bound electrons affected by the electric field of incident light. Each bound electron is a LO in this model and the dielectric spectrum consists the response of a large number of LOs. Decomposition of the dielectric spectrum into its elementary components is one of the difficult and important issues of optics. In the present study, a detailed and holistic analytical method has been derived to solve the decomposition problem and an application of this method to the dielectric spectrum of cubic $RbNbO_3$ crystal has been presented.

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