

Inertial and interference effects in optical spectroscopy

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Abstract. Interference between free-space and material components of the displacement current plays a key role in determining optical properties. This is illustrated by an analogy between the Lorentz optical model and a-c circuits. Phase shifts in material-polarization currents, which are *inertial*, relative to the *non-inertial* vacuum-polarization current cause interference in the total displacement current and, hence, variation in E-M wave propagation. If the displacement-current is reversed, forward propagation is inhibited yielding the semi-metallic reflectivity exhibited by intrinsic silicon. Complete cancellation involves material currents offsetting free-space currents to form current-loops that correspond to plasmons.

1. Introduction

An analogy between the optical response of matter and currents in a-c circuits suggests a novel interpretation of optical properties as the result of interference between free-space (vacuum) and matter displacement currents. The analogy reflects the similarity of the differential equations for the response of optical materials and a-c circuits. However, the underlying physics initially appears different: The Lorentz model rests on Newton's laws; electric-circuit analysis assumes Kirchhoff's laws. Further, optics focuses on electric-dipole moments; the focus in circuits is on currents.

Yet, these approaches are basically the same. Their relationship parallels that between Newtonian and energy-based methods in mechanics [1-3], but with emphasis on different dynamical variables. Significant insights into optical properties can be had by considering displacement current as well as polarization. This emphasizes the roles of vacuum and matter currents in E-M wave propagation. As with circuit branch currents, these displacement currents interfere to determine the optical response.

Interference is pronounced near material resonances and may lead to large excursions in total-current phase relative to an applied field. In the extreme, displacement-current reversal yields reflection not transmission. This may occur even in the absence of free electrons, as in some intrinsic semiconductors, e.g., silicon. Moreover, the analogy provides a picture of plasma oscillations in terms of the complete destructive interference of vacuum and matter currents. The analogy emphasizes the role of inertia in optics and optical sum rules, e.g., the f -sum rule [4, 5], and their circuit analogues [6].

2. Optical properties and circuits

Optical properties are commonly discussed in terms of electric susceptibilities. In the dielectric-continuum model a material's susceptibility is the sum of the susceptibilities of free space and of a uniform distribution of the matter present, $\chi(\omega)$ [7]. The free-space component is assumed to be unperturbed by the matter. The matter's susceptibility is commonly approximated in the Lorentz



model [8] by a density $\mathcal{N}$ of mechanical oscillators with mass m , charge q , spring constant k and viscous damping constant Γ . The dielectric function for a single-resonance is then [8, 9]

$$\varepsilon(\omega) = 1 + 4\pi\chi(\omega) = 1 + \frac{\omega_p^2}{\omega_0^2 - \omega^2 + i\gamma\omega}, \quad (1)$$

where $\omega_0 = (k/m)^{1/2}$, $\omega_p = (4\pi\mathcal{N}q^2/m)^{1/2}$ and $\gamma = \Gamma/m$. Eq. (1) is based on Newton's dynamical equation for the oscillator's displacement, x , caused by an applied electric field $E(t)$, namely

$$m \frac{d^2x}{dt^2} + \Gamma \frac{dx}{dt} + kx = qE(t), \quad (2)$$

Many systems obey similar equations; the series LCR circuit of Figure 1 is of special interest here. In contrast to the mechanical oscillator, this circuit is treated using Kirchoff's potential law [10]. The result is an equation in charge, Q , and current, $\mathcal{J} = dQ/dt$, for the response to an EMF, $\mathcal{E}(t)$,

$$L \frac{d\mathcal{J}(t)}{dt} + R\mathcal{J}(t) + \frac{Q}{C} = \mathcal{E}(t). \quad (3)$$

This is traditionally solved by introducing the complex impedance, $\hat{Z}$, and admittance, $\hat{Y}$. The result is

$$\hat{Y}_{\text{series}}(\omega) = \frac{1}{\hat{Z}(\omega)} = \frac{\mathcal{J}(\omega)_{\text{series}}}{\mathcal{E}(\omega)} = i\omega \left[\frac{1/L}{\omega_{\text{series}}^2 - \omega^2 + i\omega\gamma'} \right], \quad (4)$$

where $\omega_{\text{series}} = (1/LC)^{1/2}$ and $\gamma' = R/L$. The point to note is that the quantity in brackets, $\hat{Y}_{\text{series}}/i\omega$, has the same form as $\chi(\omega)$ the material part of $\varepsilon(\omega)$. This similarity may be made explicit by substituting dQ/dt for the current. Then the dynamical equation becomes the second-order differential equation

$$L \frac{d^2Q}{dt^2} + R \frac{dQ}{dt} + \frac{Q}{C} = \mathcal{E}(t), \quad (5)$$

which is identical to Eq. (2), but with $Q(t)$ the analogue of $x(t)$. (See [11] for the quantum case.)

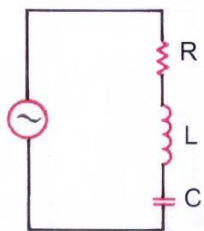


Figure 1. Analogue of $\chi(\omega)$.

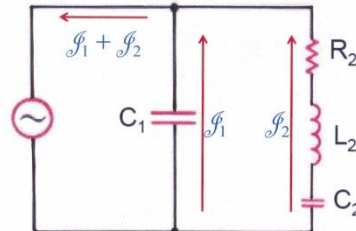


Figure 2. Analogue of $\varepsilon(\omega)$.

A full circuit analogue of $\varepsilon(\omega)$ is suggested by the rule that admittances of parallel circuits add. The result is the parallel combination of capacitive and LCR branches of Figure 2 with admittance

$$\hat{Y}_{\parallel}(\omega) = \hat{Y}_{\text{capacitor}} + \hat{Y}_{\text{series}} = i\omega \left[C_1 + \frac{1/L_2}{\omega_{\text{series}}^2 - \omega^2 + i\omega\gamma'} \right]. \quad (6)$$

C_1 has the value of the free-space susceptibility; the LCR branch that of matter susceptibility $\chi(\omega)$.

The ease of visualizing circuit currents suggests an extension of the analogy between $\hat{Y}_{\parallel}$ and $\varepsilon(\omega)$ to optical polarization currents. From Maxwell's equations [7] the displacement current density is

$$J_{\text{displacement}}(\omega) = \frac{1}{4\pi} \frac{\partial D}{\partial t} = \frac{1}{4\pi} i\omega \varepsilon(\omega) E(\omega) = i\omega \left[\frac{1}{4\pi} + \chi(\omega) \right] E(\omega). \quad (7)$$

Here the current has been explicitly divided into its two parts:

1 A (virtual) vacuum-polarization current, $(1/4\pi) \partial E/\partial t$. The factor $i\omega$ shows it leads the electric field by 90° . Its circuit analogue is the capacitive-branch current, $\mathcal{J}_1$.

2 A (real) material current associated with the rate of change of the polarization, $\partial P/\partial t$. This has the same frequency-dependent phase as $i\omega\chi(\omega)$. Its circuit analogue is the series LCR branch current, $\mathcal{J}_2$.

Since the magnetic field of a propagating E-M wave arises from the displacement current, the frequency-dependence of optical properties is seen to result from interference between vacuum- and matter-polarization currents. This is a direct analogue of the frequency-dependent variations in external current for the circuit of Figure 2 caused by branch-current interference. Specifically:

a. At frequencies below resonance, $\omega \ll \omega_{\text{series}}$, both circuit branch currents are primarily “capacitive” and so in phase. [Capacitive impedance, $1/\omega C_2 \gg$ inductive impedance, ωL_2 , in the LCR branch.] Similarly, in a dielectric-continuum, when $\omega \leq \omega_0$ both vacuum and matter displacement currents are largely in phase: Near resonance the matter leads; the *inertial* term in the equation for matter polarization is negligible compared to the spring’s contribution, so the oscillator’s motion is largely in phase with the non-inertial vacuum polarization over most of the range $\omega \leq \omega_0$.

b. At frequencies above resonance, $\omega \geq \omega_{\text{series}}$, inductance dominates the LCR branch. Its current lags, while that through C_1 still leads. Destructive interference determines the external current and, when the magnitude of the LCR branch current is large (small R_2) the external current may reverse. In the dielectric analogue, when $\omega \geq \omega_0$, *inertia* dominates the Lorentz-oscillator motion and the material current interferes *destructively* with the vacuum current. For large matter currents the *total* current may also reverse above resonance. This reverses the magnetic field of an E-M wave and propagation is inhibited. The result is semi-metallic reflectivity as exhibited by intrinsic silicon (reflectivity of 50 to 70% from 5 eV to 15 eV.) Figure 3 shows the reactive currents, phase and reflectivity for intrinsic silicon [12] and for a circuit with parameters matching silicon’s, but with negligible resistance, $R_2 \sim 0$.

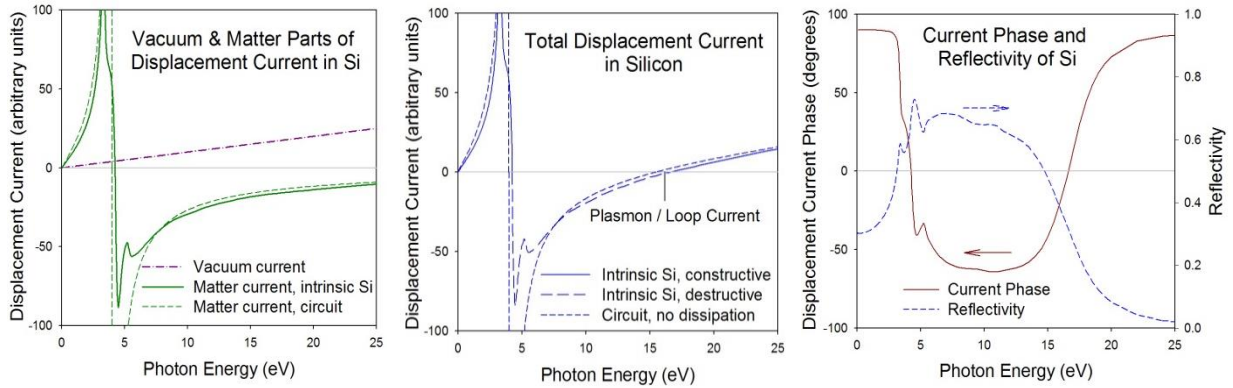


Figure 3. Displacement currents, phase and reflectivity for silicon, and currents for an analogous circuit model.

3. Excitations

The a-c circuit also provides insight into a dielectric’s E-M excitations. Rearranging Eq. (6) yields

$$\hat{Y}_{\parallel} = i\omega \left[C_1 + \frac{1/L}{\omega_{\text{series}}^2 - \omega^2 + i\omega\gamma'} \right] = i\omega C_1 \left[\frac{\omega_{\text{collective}}^2 - \omega^2 + i\omega(R_2/L_2)}{\omega_{\text{series}}^2 - \omega^2 + i\omega(R_2/L_2)} \right] \quad (8)$$

Here $\omega_{\text{series}} \approx (1/L_2 C_2)^{1/2}$ and $\omega_{\text{collective}} \approx (1/L_2 C_1 + 1/L_2 C_2)^{1/2}$. Mathematically, the poles and zeros characterize $\hat{Y}_{\parallel}$ [13]. Physically they represent the extreme responses to electrical or optical probes.

a. Poles. The pole of $\hat{Y}_{\parallel}$ (an admittance maximum for $\omega > 0$) occurs at

$$\omega_{\text{pole}} = \sqrt{1/L_2 C_2 - (R_2/2L_2)^2} + i(R_2/2L_2). \quad (9)$$

This is a resonance in the LCR branch; it lies at $\omega_{\text{series}} \approx (1/L_2 C_2)^{1/2}$ (for no dissipation). Both $\mathcal{J}_2$ and $\mathcal{J}_{\text{external}} = \mathcal{J}_1 + \mathcal{J}_2$ have maxima since $\mathcal{J}_1$ is negligible. Thus, a circuit pole represents the *single-branch* excitation of Figure 4. The dielectric-continuum analogue is a *single-particle* optical resonance at ω_0 .

b. Zeros. The zero of $\hat{Y}_{\parallel}$ (impedance maximum for $\omega > 0$) occurs at

$$\omega_{\text{zero}} = \sqrt{(1/L_2)(1/C_1 + 1/C_2) - (R_2/2L_2)^2} + i(R_2/2L_2). \quad (10)$$

Here, the *external current is zero* (minimum for real ω) regardless of EMF. This seemingly counter-intuitive condition involves a collective-resonance: a loop current circulating through *both* parallel branches, see Figure 5. This may be seen since ω_{zero} is the resonant frequency for the *series* combination of capacitive (vacuum) and LCR (matter) branches, $(1/L_2 C_1 + 1/L_2 C_2)^{1/2}$ (no dissipation).

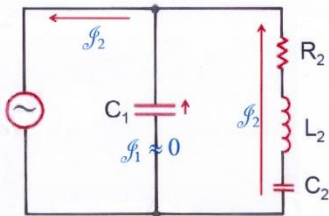


Figure 4. Single-branch resonance, the analogue of a single-particle resonance.

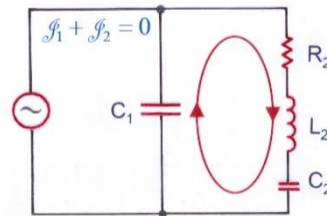


Figure 5. Circuit-loop resonance, the analogue of a plasmon resonance.

This resonance loop-current mode circulates *entirely* within the parallel network. It is not usually considered since an external voltage applied to the network terminals does not create an EMF within the loop that would excite the loop current (transients would dissipate). However, the mode can be easily excited by a changing magnetic field passing *through* the loop; say by moving a magnet into the loop. (Or, by coupling one of the elements to a signal. Cf. the tank circuit of a radio receiver [14, 15].)

Since $\hat{Y}_{||}/i\omega$ is the analogue of $\varepsilon(\omega)$, a zero of $\hat{Y}_{||}$ corresponds to a zero of the dielectric function, the hallmark of plasma modes [16]. Hence, the internal loop-current resonance is the circuit analogue of a bulk plasma excitation. The traditional picture of plasma excitations is of a fluctuation in an electron gas. The resulting charge imbalance yields a collective oscillation in the electron density. Such excitations are easily observed as quantized energy-loss by charged particles passing through condensed matter, especially in good metals and some semiconductors. However, in insulators the zeros of $\varepsilon(\omega)$ generally lie high in the complex-frequency plane. Then the plasmon lifetime is too short (line widths too broad) for the excitation to be well defined. The circuit analogue for this is a large value of R_2 that damps out any internal loop currents before a full oscillation takes place.

In summary, there are two distinct circuit excitations, the single-branch and loop resonances. Analogously, in a dielectric continuum there are also two distinct excitation classes: A Helmholtz decomposition [17] of the electric field yields a transverse component corresponding to optical excitations; and a longitudinal component corresponding to plasma modes.

4. Circuit sum rules analogies

We have used circuit analogies to clarify E-M processes in dielectrics. A reverse case is the prediction of integral circuit rules from optical sum rules: The f sum rule [4, 5] for electron density, $\mathcal{N}$, is

$$\int_0^\infty \omega \varepsilon_2(\omega) d\omega = 4\pi \mathcal{N} e^2 \frac{\pi}{2m} . \quad (11)$$

This forms the basis of Smakula's equation [18] for defect density and for much quantitative optical spectroscopy. However, using the analogies between $\varepsilon(\omega)$ and $Y(\omega)$ and between m and L , one finds a similar rule for the dissipative part of the admittances of the circuits of the sorts considered above,

$$\int_0^\infty \text{Re} \hat{Y}(\omega) d\omega = \frac{\pi}{2L} . \quad (12)$$

The physical basis of the f sum rule is the inertia of the optical response [19]. By analogy, the basis of the circuit rule is inductance. Remarkably, while both rules involve the dissipative response, they are *independent* of the dissipative parameters Γ and R , but depend only on m and L , respectively.

Acknowledgements

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