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The factorized form for dielectric relaxation

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Abstract. The factorized form of the dielectric function, introduced by Berreman and Unterwald to describe *reststrahlen*, is well know by infrared spectroscopists. In the present paper, we show that such form can be generalized to account for relaxational dispersion. After reviewing the fundamentals of this approach, we show that the factorized form proposed for the description of dielectric relaxation is flexible, founded on very general grounds and circumvents three basic limitations of the conventional sum model of Debye relaxors: the coupling between different polar units, the high frequency transparency problem and the non-inclusion of the effect of coupled non-polar degrees of freedom.

PACS. 77.22.Ch Permittivity (dielectric function) – 77.22.Gm Dielectric loss and relaxation

1 Introduction

A general microscopic description of the response of the polarization of a condensed medium to an external electric field requires the use of techniques of non-equilibrium statistical mechanics and represents a difficult task. In the linear response limit, the assumption that the polarization is capable of following the external driving field in a close vicinity of equilibrium allows us to express the average time evolution of a physical quantity A in terms of retarded correlation functions, calculated at equilibrium [1]:

$$\bar{A}(t) = \langle A \rangle_{eq.} + \frac{1}{kT} \int\limits_{-\infty}^{t} dt' \vec{E}_0(t') \left\langle A(t) \dot{\vec{P}}(t') \right\rangle . \qquad (1)$$

Even under these simplifying assumptions, a realistic evaluation of the equilibrium correlation functions $\left\langle A(t) \dot{\vec{P}}(t') \right\rangle$ for a particular material under study would require well adapted N-bodies microscopic models and a considerable effort of calculus.

Because of these difficulties, the description of dielectric dispersion of isolated materials still relies heavily on ideal or simplified models: the Debye model [2] for relaxation and the Helmholtz-Ketler-Lorentz (HKL) oscillator [3] for resonance. The experimental results of dielectric dispersion, which often deviate from these ideal models, are commonly described by assuming the superposition of independent Debye relaxors and HKL oscillators:

$$\varepsilon(\omega) = \varepsilon_\infty + \sum_j \frac{\Delta\varepsilon_j}{1 + i\omega\tau_j} + \sum_i \frac{\Delta\varepsilon_j \Omega_{0i}^2}{\Omega_{0i}^2 - \omega^2 - i\omega\Gamma_i} . \qquad (2)$$

However, linear dispersion relations are found in several branches of physics and verify some general properties that are, to a large extent, independent of the details of the particular problem under focus. As first recognized by Kramers and Krönig [4], they express some general properties of the system that stem from causality. It should therefore be possible to derive general dispersion relations without having a detailed knowledge of the basic interactions by which they arise or relying upon simplified or empirical models.

In the case of *reststrahlen* reflectance data, this phenomenological approach led to the factorized form of dielectric function proposed by Berreman and Unterwald [5], which reflects the contribution to $\varepsilon(\omega)$ of a discrete set of complex poles and zeros, located in the negative half complex plane near the real frequency axis and having mirror symmetry with respect to the imaginary frequency axis:

$$\varepsilon(\omega) = \prod_{j=1}^{N} \frac{\omega^2 + 2iz_j''\omega - (z_j'^2 + z_j''^2)}{\omega^2 + 2ip_j''\omega - (p_j'^2 + p_j''^2)} . \qquad (3)$$

This form generally allows a description of IR reflectance data to be made that is more accurate than that obtained from the use of a sum of independent Lorentz oscillators. This fact results from the ability to include

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non-harmonic effects by adjusting different imaginary components of paired zeros and poles. As shown by Gervais and Piriou [6], this corresponds to the incorporation (to some extent) of self-energy corrections arising from the perturbative treatment of phonon interactions. More recently, this form has been used to analyse the lattice dynamics and non-harmonic coupling effects between plasmon excitations and LO phonons in semiconductor films [7–10] and in a comprehensive analysis of the anisotropic polar-phonon spectrum of sapphire by spectroscopic ellipsometry [11].

To the best knowledge of the authors, the same methodological approach has not been adopted so far to describe dielectric relaxation. Díaz and Alexopoulos [12] analysed the restrictions imposed by causality to the form of dielectric dispersion and proposed that the complex permittivity of any physically realizable material could be expressed as a sum of Lorentzians. To reach this conclusion, these authors imposed a partial fraction expansion of a factorized form of the electric susceptibility. In this paper we follow the more general approach of Berreman and Unterwald and show that a factorized form of the dielectric function can be used to describe, in a more flexible and general way, the dispersion data of insulators.

2 The limits of the superposition of Debye relaxors

The traditional description of relaxation is based on the use of discrete or continuous sums of independent Debye terms (sum model). This approach has at least three important drawbacks:

i) the unphysical assumption that two polar Debye relaxors respond independently to the electric field, even if their frequencies are arbitrarily close to each other. The coupling between relaxing polar units is implicitly ignored and the dielectric strength of each relaxor does not depend on the local distribution of relaxation times;
ii) the failure in the high frequency limit. At high frequencies, the imaginary part of the Debye dielectric function, $\varepsilon_2(\omega)$, decays with ω^{-1}. In consequence, the absorption $\alpha = \frac{\omega\varepsilon_2(\omega)}{c}$ remains finite as $\omega \rightarrow \infty$. This would imply that the dielectric loss connected with orientational polarizability ($\nu < 10^9$ Hz) could originate absorption in the visible and ultraviolet range ($\nu > 10^{14}$ Hz). This result is a consequence of the fact that a Debye relaxor is essentially an inertialess system with a restoring force. The omission of inertia allows an effective response to an arbitrarily fast stimulus. Equivalently, in the time domain, it allows the polarization to respond instantaneously to an abrupt suppression of the electric field at $t = t_0$, which originates to a discontinuity of the time correlation function;
iii) the model does not allow the description of the coupling between the polar relaxing units and other non-polar degrees of freedom. Microscopically, relaxation is connected with rotational motions of polar molecules in a condensed medium. Manifold interactions are expected to induce a coupling between polar and non-polar degrees of freedom, which might affect the response of polarization to the external field. This effect cannot be included in the sum model.

3 Review of the fundamentals of the factorized form of $\varepsilon(\omega)$

Causality requires that the response of the polarization at a given time t cannot depend on the value of the electric field at a time $t' > t$. This implies that the Fourier transform of the electric susceptibility $\chi(\omega)$, the time correlation function $G(t - t')$, must be zero if $t' > t$. Titchmarsh [13] has shown that, if $\chi(\omega)$ is a (Lebesgue) square integrable function on the real axis, this implies that: a) $\chi(\omega)$ is the limit, as $\phi \rightarrow 0$, of an analytic function $\chi(\omega + i\phi)$ that is regular and bounded in modulus for $\phi > 0$, and b) the real and imaginary parts of $\chi(\omega)$are Hilbert transforms of each other. This last equivalence corresponds to the Kramers-Kronig theorem [4].

From Titchmarsh's theorem it follows that any singularity of $\chi(\omega + i\phi)$ must be located in the negative imaginary half plane ($\phi < 0$). As Berreman and Unterwald [5] have stressed, as $\chi^{-1}(\omega)$ also represents a linear response function, the zeros of $\chi(\omega)$ must also be located in the negative imaginary half plane. From the fact that $\varepsilon(\omega + i\phi) = 1 + \chi(\omega + i\phi)$ one can conclude that the same is true for the poles and zeros of the generalized dielectric function.

The factorized form of the dielectric function is based on the assumption that $\varepsilon(\omega+i\phi)$ does not possess essential singularities or continuous domains of singularities. From this basic simplifying assumption it follows that $\varepsilon(\omega)$ can be written as:

$$\varepsilon(\omega) = \frac{\prod_{m=1}^{M} (\omega - \tilde{z}_m)}{\prod_{n=1}^{N} (\omega - \tilde{p}_n)} \varphi(\omega) \ , \tag{4}$$

where M and N represent the number of complex zeros (z) and poles (p) in the negative imaginary half plane, respectively, and $\varphi(\omega)$ is the limit, as $\phi \rightarrow 0$, of a function $\varphi(\omega + i\phi)$ that is analytic and bounded in the whole complex plane. Moreover, as $\varepsilon(|\omega + i\phi|) \rightarrow 1$ as $|\omega + i\phi| \rightarrow \infty$, it follows from the theorem of Liouville [14] that $\varphi(\omega) = \text{const.} = 1$ and that the number of poles must be equal to the number of zeros ($M = N$). The simplest form of the dielectric function is therefore:

$$\varepsilon(\omega) = \prod_{n=1}^{N} \frac{\omega - \tilde{z}_n}{\omega - \tilde{p}_n}. \tag{5}$$

The physical nature of the medium can impose additional restrictions on the possible types of poles and zeros. If, as it is the case under analysis, the linear response of the

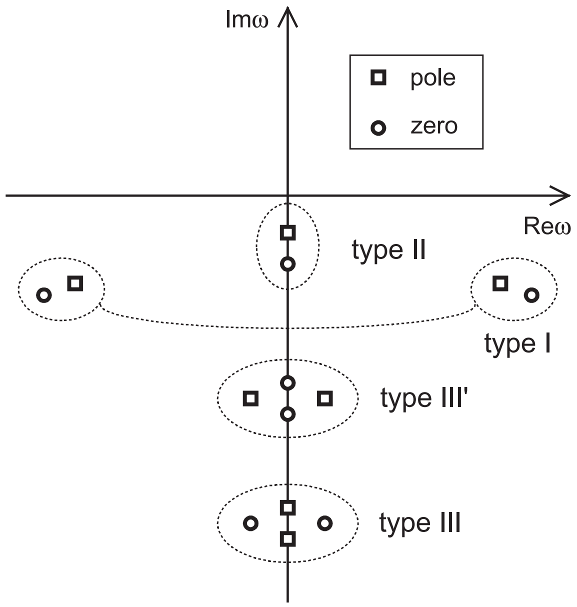

Fig. 1. Possible types of zeros and poles for a medium with a linear response characterized by a symmetric second order tensor.

system is characterized by a second order symmetric tensor, then the response of the medium must be the same for excitations propagating from the right to the left as from the left to the right (i.e. if $\omega \leftrightarrow -\omega$) [5,15]. In such a case, the map of zeros and poles must have mirror symmetry about the imaginary axis (*symmetry condition*).

Figure 1 depicts the possible types of paired zeros and poles compatible with the symmetry condition. We stress that the insulator condition [5] (the imaginary part of the dielectric function $\varepsilon_2 \to 0$ as $\omega \to 0$) is implicit in Titchmarsh's theorem ($\chi(\omega)$ is a square integrable function for any real frequency) and in the symmetry condition (there cannot be a single imaginary pole paired to a single complex zero, or vice versa). The four types of paired poles and zeros shown in the figure are therefore possible for dielectric materials.

The complex poles of type-I originate the Berreman and Unterwald model for resonance and their contribution to the dielectric function has been analyzed in [5]. In the following, we will focus on the other two possible types and show that they may give rise to natural descriptions of dielectric relaxation, providing an alternative to the sum model of Debye relaxors.

4 The product model for Debye relaxors

Type-II poles and zeros correspond to independent pairs of imaginary poles and zeros. The contribution of N pairs of this type to the dielectric function is

$$\varepsilon(\omega) = \varepsilon_\infty \prod_{k=1}^{N} \frac{\omega + iz_k}{\omega + ip_k} \ , \tag{6}$$

where ε_∞ is the contribution (real) to $\varepsilon(\omega)$ resulting from the set of all pairs of poles and zeros with real frequencies $\nu \gg \omega$. A single pair corresponds to a Debye relaxor:

$$\begin{aligned} \varepsilon(\omega) &= \varepsilon_\infty \frac{\omega + iz}{\omega + ip} \\ &= \varepsilon_\infty \left[\frac{\omega + iz}{\omega + ip} - 1 + 1 \right] \\ &= \varepsilon_\infty + \frac{\Delta\varepsilon}{1 + \frac{\omega}{ip}} \end{aligned} \tag{7}$$

with $\Delta\varepsilon = \varepsilon_\infty \left(\frac{z}{p} - 1 \right)$.

The form (6) is not equivalent to the sum model, essentially because the contribution of each polar relaxor (i.e. a pair of one imaginary zero and one imaginary pole) to the static dielectric constant depends on the global distribution of poles and zeros. In fact, if $\Delta\varepsilon = \varepsilon_0 - \varepsilon_\infty = \varepsilon_\infty \left(-1 + \prod_{i=1}^{N} \frac{z_i}{p_i} \right)$ is expressed as a sum of the contributions of the different relaxing units $\Delta\varepsilon = \sum_{i=1}^{N} \Delta\varepsilon_i$, then the dielectric strength of each unit must be:

$$\Delta\varepsilon_i = \varepsilon_\infty \frac{1}{p_i} \frac{\prod_{k=1}^{N} (z_k - p_i)}{\prod_{k \neq i} (p_k - p_i)}. \tag{8}$$

The sum model, in which two relaxing units i and k respond independently, corresponds to the limit case where $\frac{z_k - p_i}{p_k - p_i} \approx 1$, i.e. is valid only for a contribution well separated in frequency. The product model of Debye relaxors, expressed by equation (6), is therefore more general, as it includes coupling effects that are unnecessarily neglected in the conventional model.

As referred to above, the Debye model must also be seen as a low frequency approximation, as it produces a finite absorption as $\omega \to \infty$. This limitation is not overcome by the product model, since it is originated in the inertialess nature of a Debye relaxor. In fact, for high frequencies:

$$\begin{aligned} \varepsilon(\omega) = \varepsilon_\infty \frac{\omega + iz''}{\omega + ip''} &\approx \varepsilon_\infty \left[\left(1 + \frac{iz''}{\omega} \right) \left(1 - \frac{ip''}{\omega} + ... \right) \right] \\ &= \varepsilon_\infty \left[1 + \frac{i}{\omega} \left(z'' - p'' \right) + ... \right] \end{aligned}$$

the imaginary part of $\varepsilon(\omega)$ decays with $\frac{1}{\omega}$ unless $z'' = p''$, i.e. unless the dielectric strength of the relaxor is zero. In addition, the coupling between polar and non-polar relaxing units cannot be taken into account by using equation (6).

5 The Berreman-Unterwald model and relaxation

The omission of inertia is justified in the limit when damping (viscous or resistive forces) dominates. It is well known

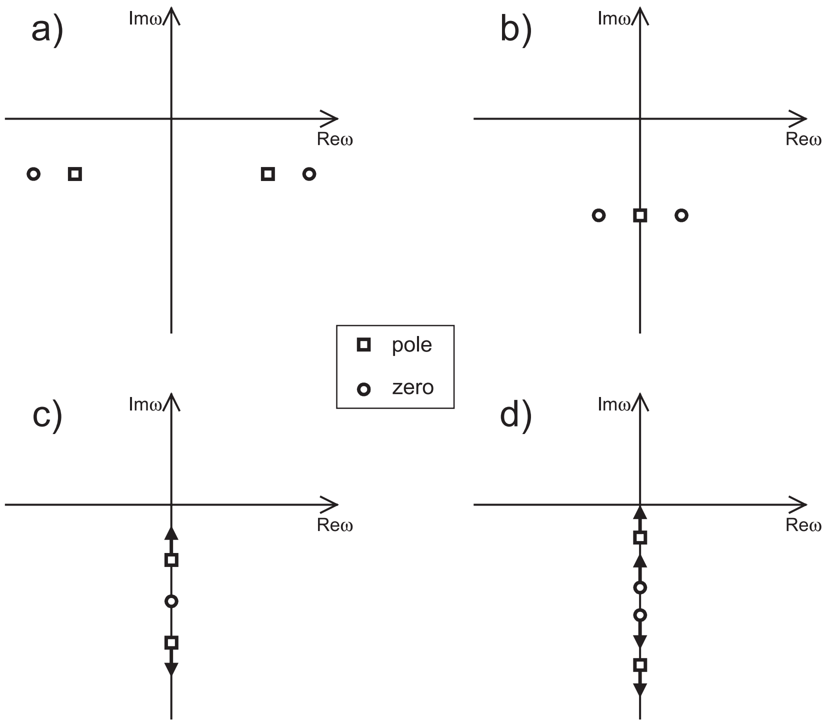

Fig. 2. a) Single Lorentz oscillator described by two zero-pole pairs located symmetrically with respect to the imaginary axis, having the same imaginary component (condition of high frequency transparency); b) critically damped pole (imaginary pole of order 2) and a sub-damped zero (two symmetrical complex zeros) located near the imaginary axis; c) overdamped pole and critically damped zero; d) overdamped poles and zeros (overdamped oscillator).

that a Debye relaxor behaviour can be obtained from a Lorentz oscillator with a mass m, damping γ and elastic force constant k if $m \rightarrow 0$ and $\gamma \rightarrow \infty$ in such a way that $m\frac{\gamma}{k} = \tau$ remains finite [16].

The limiting conditions of zero mass and infinite damping are essentially unphysical. Consider alternatively two zero-pole pairs located symmetrically with respect to the imaginary axis (type-I). By writing $z'' = p'' = \Gamma$ (condition of *high frequency transparency* [5]), $z'^2 + z''^2 = \Omega_z^2$ and $p'^2 + p''^2 = \Omega_p^2$, one can express their contribution to $\varepsilon(\omega)$ (given by Eq. (3)) as:

$$\varepsilon(\omega) = \varepsilon_\infty \frac{\omega^2 + 2i\Gamma\omega - \Omega_z^2}{\omega^2 + 2i\Gamma\omega - \Omega_p^2}. \tag{9}$$

If the damping is progressively increased at constant polar strength (i.e. at constant Ω_z and Ω_p; see Fig. 2), these two symmetrically located zeros and poles approach the negative imaginary axis. A critically damped pole (transversal optical mode) and a sub-damped zero would correspond to an imaginary pole of order 2 and two symmetrical complex zeros located near the imaginary axis. Further increasing of the damping transform the pole of order 2 into two separated and correlated poles of order 1 (type-III in Fig. 1 [1]). Similarly, above the critical damping of the zeros ($\Gamma > \Omega_z$), we obtain two correlated pairs of imaginary poles and zeros (overdamped oscillator) corresponding to the frequencies:

$$\omega_{\pm}^{z,(p)} = -i\Gamma \pm i\Gamma\sqrt{1 - \frac{\Omega_z^2, (\Omega_p^2)}{\Gamma^2}}. \tag{10}$$

Note that if the damping forces dominate, i.e. if $\frac{z''}{\Gamma} \ll 1$, the contribution of the overdamped oscillator to the dielectric function (equation 9) is approximately given by:

$$\Delta\varepsilon = \varepsilon_\infty \frac{\left(\omega + i\frac{z''^2}{2\Gamma} + ...\right)\left(1 - i\frac{\omega}{2\Gamma} + \left(\frac{z''^2}{4\Gamma^2}\right) + ...\right)}{\left(\omega + i\frac{p''^2}{2\Gamma} + ...\right)\left(1 - i\frac{\omega}{2\Gamma} + \left(\frac{p''^2}{4\Gamma^2}\right) + ...\right)}, \tag{11}$$

showing that equation (6) (Debye relaxor) actually represents a low frequency approximation of equation (9) $\left(\omega \approx \frac{z''^2}{2\Gamma} \ll \Gamma\right)$. The essential difference in the use of an overdamped oscillator instead of a Debye relaxor is that, as inertia is included in the former case, the correct behaviour at high frequency is assured (the imaginary part of $\varepsilon(\omega)$ decays as $\frac{1}{\omega^3}$). This can also be clearly seen in the

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[1] Type-III poles and zeros could also describe the contribution of free charges (Drude model) if one of the poles were located at $z'' = 0$. However, this situation will not be discussed because it is not relevant for insulating materials and it is not compatible with the conditions imposed by Titchmarsh theorem (that requires $\epsilon(\omega)$ to be squared integrable on the real axis).

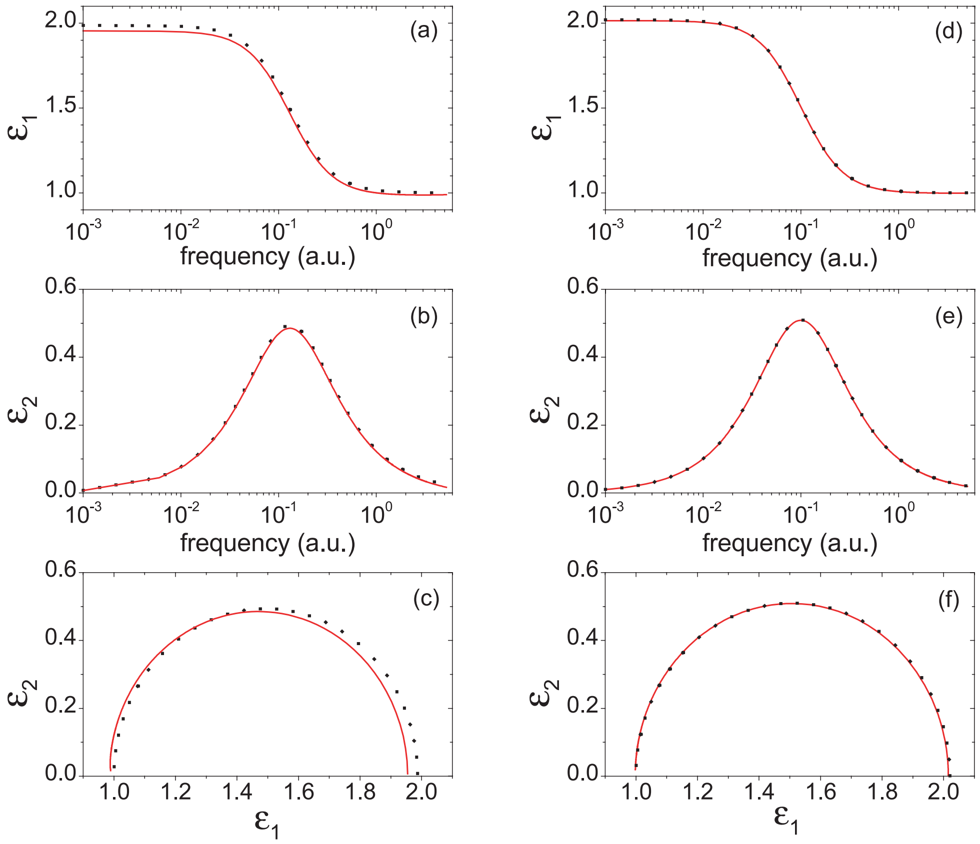

Fig. 3. Debye relaxor (dots) or overdamped oscillator (line)? The dispersion of the real (ε_1) and imaginary (ε_2) parts of the dielectric function originating from overdamped oscillators ($p = 1$, $z = 2^{1/2}$ and a damping frequency $\Gamma = 4$ ((a)-(c)) or $\Gamma = 10$ ((d)-(f))) and from the corresponding Debye relaxors obtained by neglecting the higher frequency zero-pole pair. The corresponding Cole-Cole plots are also shown.

time domain, if we compare the time correlation function resulting from (6) and (9):

$$G(t) = \frac{\chi_0}{\tau} e^{-\frac{t}{\tau}}\ ; \qquad (12)$$

$$G(t) = e^{-\frac{\Gamma}{2}t} \frac{\Delta\Omega^2}{\Gamma\sqrt{1-\frac{\Omega_p^2}{\Gamma^2}}} \sinh\left(\Gamma t\sqrt{1-\frac{\Omega_p^2}{\Gamma^2}}\right). \qquad (13)$$

While the Debye time-correlation function presents a discontinuity at $t = 0$ $\left[G(t \to 0^-) = 0;\ G(t \to 0^+) = \frac{\chi_0}{\tau}\right]$, the overdamped oscillator produces a time correlation function that is continuous at $t = 0$.

Figure 3 compares the real and imaginary parts of a Debye relaxor with those obtained from equation (9) (with $\Omega_p^2 = 1$ and $\Omega_z^2 = 2$). If for $\Gamma = 4$ the Cole-Cole plot is still slightly deformed, for $\Gamma = 10$ the two curves nearly coincide except at a very high frequency, not usually covered by the experimental data.

For a poly-dispersive system, equation (9) must be generalized to a factorized form, which is similar to that introduced by Berreman and Unterwald for the description of the infrared *reststrahlen* [5]:

$$\varepsilon(\omega) = \varepsilon_\infty \prod_i \frac{\omega^2 + 2i\omega\Gamma_{zi} - \Omega_z^2}{\omega^2 + 2i\omega\Gamma_{pi} - \Omega_p^2}. \qquad (14)$$

In this case, the *transparency condition* at high frequency requires the sum rule $\sum(\Gamma_{zi} - \Gamma_{pi}) = 0$ rather than $\Gamma_{zi} = \Gamma_{pi}$. This ability to accommodate slightly different values of the damping of paired zeros and poles is a well known source of flexibility in the description of experimental IR results. Similar to the case found in the Berreman-Unterwald model, the contribution of each relaxing unit to the static dielectric constant is of the form:

$$\Delta\varepsilon_i = \varepsilon_\infty \frac{1}{p_i^2} \frac{\prod_{k=1}\left(z_k^2 - p_i^2\right)}{\prod_{k\neq i}\left(p_k^2 - p_i^2\right)}. \qquad (15)$$

Notice that, contrary to the case of equation (6), which represents coupled Debye relaxors, non-polar relaxing modes can be included in equation (14). As can be seen, the inclusion of non-polar relaxing modes ($\Omega_{zi} = \Omega_{pi}; \Delta\epsilon_i = 0$) alters the dispersion $\varepsilon(\omega)$ if the non-polar units are coupled to polar degrees of freedom, i.e. if different dampings are ascribed to the zero and pole of a non-polar relaxor in such a way that the condition resulting from the sum rule $\sum(\Gamma_{zi} - \Gamma_{pi}) = 0$ is verified.

The effect of coupled non-polar relaxations to the dielectric dispersion is usually ignored in the analysis of experimental data. However, this effect can be important, as illustrated in the example depicted in Figure 4.

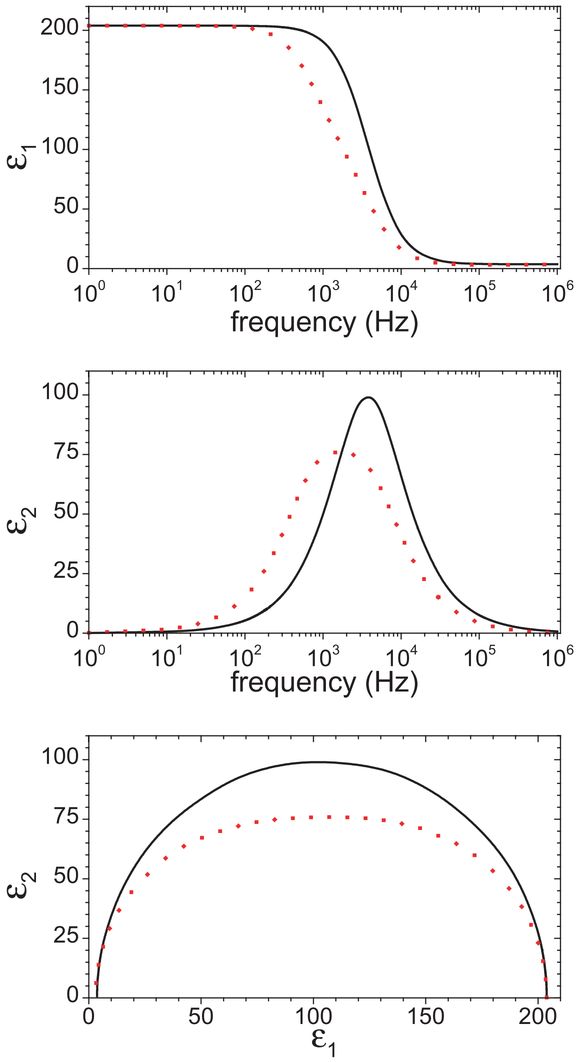

Fig. 4. The effect of coupling between an overdamped oscillator and non-polar oscillator. Parameters of the single overdamped oscillator (line): $\Omega_p = 1.3\times 10^5$ Hz, $\Omega_z = 9.5\times 10^5$ Hz, $\Gamma_p = 2.25\times 10^6$ Hz, $\Gamma_z = 2.25\times 10^6$ Hz. Parameters of the coupled oscillators (dots): $\Omega_p = 1.3 \times 10^5$ Hz, $\Omega_z = 9.5 \times 10^5$ Hz, $\Gamma_p = 2.2 \times 10^6$ Hz, $\Gamma_z = 2.3 \times 10^6$ Hz (polar oscillator); $\Omega'_p = 2 \times 10^4$ Hz, $\Omega'_z = 2 \times 10^4$ Hz, $\Gamma'_p = 2.5 \times 10^5$ Hz, $\Gamma'_z = 1.5 \times 10^5$ Hz (non-polar oscillator).

Here, a typical polar relaxing unit (characterized by $\Omega_p = 1.3 \times 10^5$ Hz, $\Omega_z = 9.5 \times 10^5$ Hz, $\Gamma_p = 2.25 \times 10^6$ Hz and $\Gamma_z = 2.25 \times 10^6$ Hz) can display a dispersion, which can not be practically distinguished from a single Debye relaxor (notice the symmetry corresponding Cole-Cole diagram). If this relaxor is considered coupled to a non-polar overdamped mode (with $\Omega'_p = \Omega'_z = 2 \times 10^4; \Gamma_z = \Gamma'_p = 2\times 10^5$ Hz), different damping coefficients must be ascribed to the two pairs of zeros and poles. A small coupling, expressed by slightly changes of the original coefficients ($\Gamma_p = 2.2 \times 10^6$ Hz, $\Gamma_z = 2.3 \times 10^6$ Hz, $\Gamma'_p = 2.5 \times 10^5$ Hz and $\Gamma'_z = 1.5\times 10^5$ Hz) can induce important modifications in the observed dispersion. In this example, the Cole-Cole plot obtained is strongly deformed and shows characteristics resulting from the empirical Cole-Cole function.

6 Conclusion

The factorized form of dielectric dispersion is founded on general and fundamental grounds which are not dependent on any specific details of the system. From the consequences of causality and from the simplifying assumption of absence of essential singularities of the dielectric function generalized to the complex plane, a very general form for dispersion can be derived.

The general method presented has been applied successfully to the description of *reststrahlen* since the pioneer work of Berreman and Unterwald. In this paper we have shown that a similar method can be also applied to the description of dielectric relaxation. This alternative description of relaxation circumvents three basic limitations of the conventional sum model of Debye relaxors: the coupling between different polar degrees of freedom, the high frequency transparency problem and the inclusion of the effects of coupled non-polar relaxation mechanisms. It therefore represents a simple method to analyze the experimental data by adjusting the location of the most relevant correlated imaginary poles and zeros and mapping their temperature or field dependence.

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