A brief review of the relevant dielectric theories of solids

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Abstract

The purpose of the present paper is to outline in qualitative terms, the way in which the dielectric constant varies with frequency and temperature, and to indicate the type of information regarding the structure of materials which can be obtained from the study of the dielectric constant. The various types of polarizations, which have been demonstrated to exist, are listed, together with an outline of their characteristics. The development of the dielectric theory in recent years has been along such specialized lines that there is a need for some correlation between the newer and the older theories of dielectric behavior to keep clear what is common to both, though sometimes expressed in different terms. The theoretical concept of the dielectric constant (ε), which is an important dielectric property of matter, has been discussed by Clausius-Mossotti, Debye, Onsager.

Keywords: Dielectric constant, Dielectric loss, Clausius-Mossotti, Debye, Onsager.

Resumen

El propósito del presente trabajo es describir en términos cualitativos, la forma en que la constante dieléctrica varía con la frecuencia y la temperatura, y para indicar el tipo de información con respecto a la estructura de los materiales que se pueden obtener a partir del estudio de la constante dieléctrica. Se listan los distintos tipos de polarización, que se han demostrado que existen, junto con una descripción de sus características. El desarrollo de la teoría del dieléctrico en los últimos años ha sido a lo largo de líneas especializadas en las que hay necesidad de una cierta correlación entre la más nuevas teorías y las antiguas del comportamiento dieléctrico para mantenerse separado lo que es común a ambas, aunque a veces se expresa en términos diferentes. El concepto teórico de la constante dieléctrica (ε), que es una propiedad importante de la materia dieléctrica, ha sido discutido por Clausius-Mossotti, Debye, Onsager.

Palabras clave: Constante dieléctrica, pérdida dieléctrica, Clausius-Mossotti, Debye, Onsager.

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I. INTRODUCTION

In order to understand the dielectric ***behaviour*** of substances in the solid state, one has to essentially go through the conceptual background of the necessary theories of dielectric solids. The state of aggregation of molecules in a continuum depends on the type of chemical bond, molecular geometry, mutual ***effect*** between atomic groups, nature of complexes etc. A system of electric charges of molecules in the neighbourhood is involved in the process of molecular interactions. The spatial arrangement of the electrically charged atoms and molecules in the system is perturbed by the influence of physical conditions. These facts on the basis of certain theories, describe the bulk. Properties of the substances that exist in a physical state. Mostly the dielectric properties are described in terms of the **dielectric permittivity** and molecular dipole moments of the substances in local and non-local fields. The dielectric permittivity, often called the **dielectric constant,** is a characteristic measurable quantity. The molecular dipole moment measured from the dielectric constant of the bulk material depends on the interaction with its neighbour and hence gives valuable information about the molecular structure. The dielectric constant depends on how polarizable a material is and the frequency of the applied field. The fall of polarizability is related to the decrease of the dielectric constant and the occurrence of the absorption of electrical energy constituting dielectric dispersion. This behaviour is shown by the frequency dependent **dielectric loss**.

Electrical insulator materials, which will prevent the flow of current in an electrical circuit, have been used since from the beginning of the science and technology of electrical phenomena. Dielectrics are insulating materials that exhibit the property of electrical polarization; thereby they modify the dielectric function of a vacuum. The first capacitor was constructed by Cunaeus and Mussachenbroek in 1745, which was known as the Leyden jar [1]. But there were no studies about the properties of insulating materials until 1837. Faraday published the first numerical measurements on these materials, which he called dielectrics [2]. *He* has found that the capacity of a condenser was dependent on the nature of the material separating the conducting surface. This discovery encouraged further empirical studies of insulating materials, aimed at maximizing the amount of charge that can be stored by a capacitor. Throughout most of the 19th century, scientists searching for insulating materials for specific applications have become increasingly concerned with the detailed physical mechanism governing the behavior of these materials. In contrast *to* the insulation aspect, the dielectric phenomena have become more general and fundamental, as they originate from dielectric polarization.

Mossotti [3, 4] and Clausius [5] have done a systematic investigation about the dielectric properties of materials. They attempted to correlate the specific inductive capacity, a macroscopic characteristic of the insulator introduced by Faraday [2] which is now popularly termed as the dielectric constant with the microscopic structure of the material. Following Faraday in considering the dielectrics to be composed of conducting spheres in a non-conducting medium, Clausius and Mossotti succeeded in deriving a relation between the real part of the dielectric constant εr and the volume fraction occupied by the conducting particles in the dielectric. In the begning of the 20th century, Debye [6] realized that some molecules had permanent electric dipole moments associated with them, and this molecular dipole moment is responsible for the macroscopic dielectric properties of such materials. Debye succeeded in extending the Clausius -Mossotti theory to take into account the permanent moments of the molecules, which allowed him and others to calculate the molecular dipole moment from the measurement of the dielectric constant. His theory was later extended by Onsager [7] and Kirkwood [8, 9], and is in excellent agreement with the experimental results for most of the polar liquids. Debye's other major contribution to the theory of dielectrics is his application of the concept of permanent molecular dipole moment to explain the anomalous dispersion of the dielectric constant observed by Drude [10]. For an alternating field, Debye deduced that the time lag between the average orientation of moments and the field becomes noticeable, when the frequency of the field is within the same order of magnitude as the reciprocal relaxation time. In this way the molecular relaxation process leads to the macroscopic phenomenon of dielectric relaxation, i.e., the anomalous dispersion of the dielectric constant and the accompanying absorption of electromagnetic energy over a certain range of frequencies.

Debye's theory shows excellent agreement with the experiments for the polar liquids, while the dielectric behaviour for solids was found to deviate considerably. Several modifications and extensions of Debye's theory have been proposed to correct this. There are two major approaches in the extension of Debye's theory. The first approach, pioneered by Cole [11], Davidson [12] and William [13], interprets the non –Debye relaxation behavior of the material in terms of the superposition of an exponentially relaxing process, which then leads to the distribution of relaxation times. The second approach by Joncher [14] proposes that the relaxation behaviour at the molecular level is intrinsically non-Debye-like due to the cooperative molecular motions.

After more than eighty years of development, the theory of dielectrics is still an active area for research. Understanding the behaviour of dielectric materials with the variations of field, temperature and frequency is of particular importance for present day electronics. Modern day electronics demand dielectric materials with narrowly defined properties tailored for particular applications. The scaling of metal-oxide-semiconductor (MOS) devices for ultra large-scale integration (ULSI) applications has been placing an ever increasing burden upon the performance of gate dielectrics [15]. Durability has become an issue as the dielectric thickness is decreased leading to a search for dielectrics with better properties than the conventional SiO_2 dielectric. The gallium arsenaide (GaAs) based metal - insulator- semiconductor field effect transistor (MISFET) is still largely unavailable, due to the lack of a suitable dielectric material for the insulation layer [16].

Recent advances in wireless communication technologies have elevated the interest in materials with the unusual combination of properties like high dielectric constant, low dielectric loss and low values of temperature dependence of the dielectric constant [17]. The constant need for miniaturization provides the continuing driving force for the discovery and development of increasingly sophisticated materials to perform the same or improved functions with decreased size and weight. The dielectric materials mentioned above are used as the basis for resonators and filters for the microwaves carrying the desired information [18]. These materials are presently employed as bulk ceramics in microwave communication devices. This paper gives a qualitative account of the way in which dielectric properties and the behaviour of solid materials will be necessary to use some kind of theory to represent the dielectrics solids.

II. THEORIES OF DIELECTRICS

This section presents a brief description of the atomic interpretation of the dielectric and optical properties of insulator materials, on the basis of the classical theory. This section is essentially concerned with the static dielectric constant, the frequency dependence of the dielectric constant and dielectric losses.

A. Electric susceptibility and permittivity

It was Michael Faraday who first noticed, that when a capacitor of value C_0 under vacuum is filled with a dielectric material, its charge storage capacity (capacitance) increases to a value of C. The ratio χ' of the increase of capacitance $\Delta C = C - C_0$ to its initial capacitance- C_0,

$$\chi' = \frac{C - C_0}{C} = \frac{\Delta C}{C_0} \,, \tag{1}$$

χ' is called the electrical susceptibility of the dielectric. The most often used terminology is the dielectric permittivity or dielectric constant instead of susceptibility, which is defined as the ratio of the capacitance *C* of the capacitor filled with a dielectric to the value C_0 of the same capacitor under vaccum.

$$\varepsilon_r = \frac{C}{C_0} \,. \tag{2}$$

From the above equations the relationship between the electric susceptibility and the dielectric permittivity is given as:

$$\chi' = \varepsilon_r - 1 \,. \tag{3}$$

Thus, by definition, the electric susceptibility and permittivity are non-dimensional real quantities. The dielectric constant or permittivity of a material is the measure of the extent to which the electric charge distribution in the material can be distorted or polarized by the application of an electric field.

B. Mechanism of electric polarization

At the atomic level, all matter consists ultimately of positively and negatively charged particles, whose charges balance each other macroscopically in the absence of an electric field, giving rise to an overall charge neutrality. Once the electric field is applied, the balances of charges are perturbed by the following four basic polarization mechanisms [19].

Electronic polarization: It occurs in neutral atoms when an electric field displaces the nucleus with respect to the negative charge. Thus electronic polarization is an induced polarization effect.

Atomic/ionic polarization: It is observed when different atoms that comprise a molecule share their electrons asymmetrically, and cause the electron cloud to be shifted towards the stronger binding atom, the atoms acquire charges of opposite polarity and an external field acting on these net charges tends to change the equilibrium positions of the atoms themselves, leading to atomic polarization.

Dipolar/orientational polarization: When an ionic bond is formed between two molecules by the transfer of some valence electrons, a permanent dipole moment will originate in them. This permanent dipole moment is equal to the product of the charges of the transferred valence electrons, and the inter-atomic distance between them. In the presence of an electric field E, the molecules carrying a permanent dipole moment will orient to align along the direction of the electric field E. This process is called the dipolar or orientational polarization. This occurs only in dipolar materials possessing permanent dipole moments.

Space charge polarization: It is present in dielectric materials which contain charge carriers that can migrate for some distance through the bulk of the material (via diffusion, fast ionic conduction or hopping, etc.) thus creating a macroscopic field distortion. Such a distortion appears to an outside observer as an increase in the capacitance of the sample and may be indistinguishable from the real rise of the dielectric permittivity. Space charge polarization is the only type of electrical polarization that is accompanied by a macroscopic charge transport (and in the case when the migrating charge carriers are ions a macroscopic mass transport as well). In general, space charge polarizations can be grouped into hopping polarization and interfacial polarization. In dielectric materials, localized charges (ions and vacancies, or electrons and holes) can hop from one site to another, which creates the hopping polarization. Similarly, the separation of the mobile positive and negative charges under an electric field can produce an interfacial polarization.

C. Polarization and dielectric constant

The ability of a dielectric material to store electric energy under the influence of an electric field, results from the field-induced separation and alignment of electric charges. Polarization occurs when the electric field causes a separation of the positive and negative charges in the material. The larger the dipole moment arms of this charge separation in the direction of a field and the larger the number of these dipoles, the higher the material's dielectric permittivity. In the presence of electronic, ionic and dipolar polarization mechanisms, the average induced dipole moment per molecule P_{av} will be the sum of all the contributions in terms of the local field (effective field) acting on each individual molecule.

$$P_{av} = \alpha_e E_{loc} + \alpha_i E_{loc} + \alpha_d E_{loc} \,. \tag{4}$$

Here, α_e, α_i, α_d are the electronic, ionic and dipolar polarizabilities. E_{loc} is the local field or the effective field at the site of an individual molecule that causes the individual polarization. Each effect adds linearly to the net dipole moment of the molecule. Interfacial polarization cannot be simply added to the total polarization as $\alpha_{ij}E_{loc}$, because it occurs at the interfaces and cannot be put into an average polarization per molecule in the bulk. Moreover, the fields are not well defined at the interfaces.

For simple dielectrics (eg. gases) one can take the local field to be the same as the macroscopic field. This means that E_{loc}=E the applied field, and therefore the polarization is,

$$P = \chi_e \varepsilon_e E = (\varepsilon_r - 1)\varepsilon_0 E \,, \tag{5}$$

P= *N*. Pav where *N* is the number of atoms or molecule per unit volume [20].

$$\varepsilon_r = 1 + N\alpha / \varepsilon_0 \,, \tag{6}$$

α is the polarizability of the molecule.

D. Clausius and Mossotti relation for dielectric permittivity

Consider a molecule of a dielectric medium situated in a uniform electric field E. The total electric field acting on this molecule Eloc will have three main components- E_1, E_2, and E_3. Here E_1 is the applied electric field E, E_2 is the field from the free ends of the dipole chain, and E_3 is the near field arising from the individual molecular interactions. In solids we have to consider the actual effective field acting on a molecule in order to estimate the dielectric permittivity. For electronic and ionic polarization, the local field for cubic crystals and isotropic liquids can be given by the Lorenz field, as

$$E_{loc} = \frac{1}{3\varepsilon_0} P \cdot \tag{7}$$

By assuming that the near field E_3 is zero, Clausius and Mossotti derived a relation for the dielectric constant of a material under electronic and ionic polarization [21].

$$\frac{\varepsilon_r - 1}{\varepsilon_r + 2} = \frac{1}{3\varepsilon_0}\left(N_i \alpha_i + N_e \alpha_e\right) \cdot \tag{8}$$

Here, ε_r is the relative permittivity at low frequencies, α_i is the effective ionic polarizability per ion pair, Ni is the number of ions pair per unit volume, α_e is the electronic polarizability and Ne is the number of ions (or atoms) per unit volume exhibiting electronic polarization. The atomic/ionic polarizability αi and the electronic polarizability αe cannot be separated at low frequencies, and hence, they are represented together as the induced polarizability α_{ind}. Hence, equation 8 can be written as:

$$\frac{\varepsilon_r - 1}{\varepsilon_r + 2} = \frac{1}{3\varepsilon_0}\left(N_m \alpha_{ind}\right) \cdot \tag{9}$$

This is known as the Clausius – Mossotti equation for non polar dielectrics. Above the frequencies of ionic polarization relaxation, only electronic polarization will contribute to the relative permittivity, which will be lowered to $\varepsilon_{r\infty}$ (relative permittivity at optical frequencies).

$$\frac{\varepsilon_{r\infty} - 1}{\varepsilon_{r\infty} + 2} = \frac{N_e \alpha_e}{3\varepsilon_0} \cdot \tag{10}$$

By using the Maxwell relation for a lossless (non-absorbing), non magnetic medium,

$$n^2 = \varepsilon_{r\infty} \ , \tag{11}$$

where n is the index of refraction of the material, equation (10) can be rewritten as:

$$\frac{n^2 - 1}{n^2 + 2} = \frac{N_e \alpha_e}{3\varepsilon_0} \cdot \tag{12}$$

In this form, it is known as the Lorentz-Lorenz equation. It can be used to approximate the static dielectric constant εr of non polar and non magnetic materials from their optical properties. In the case of dipolar materials we cannot use the simple Lorentz field approximation, and hence the Clausius–Mossotti equation cannot be used in the case of dipolar materials.

D. Debye theory for polar dielectrics

In addition to the induced polarization present in all dielectrics, the polar dielectrics possess an orientational polarization that exists even in the absence of an applied electric field. It should be noted that the polarizability α_o corresponding to the orientational polarization is related to the orientation of the molecules, which are heavier than atoms or electrons that are involved in induced polarization. Hence, the α_o contributes to the total molecular polarizability α, at much lower frequencies than αind does. So the dielectric constant that remains after the relaxation of the orientationally polarization (the dielectric constant due to the induced polarization) can be designated separately, and it is usually represented by ε_∞ in the case of dipolar dielectrics. So the equation (9) can be written as:

$$\frac{\varepsilon_\infty - 1}{\varepsilon_\infty + 2} = \frac{N_m}{3\varepsilon_0}\alpha_{ind} \cdot \tag{13}$$

To account for the orientational contribution to the dielectric constant, Debye [22] used the classical Boltzmann statistics and the Langevin function *yL(y) =coth y − 1/y* from the theory of paramagnetism, to estimate the temperature dependence of a permanent dipole orientation. Assuming that these dipoles do not interact with each other, Debye derived the following equation for the orientational polarizability.

$$\alpha_o = \frac{\mu^2}{3KT} \cdot \tag{14}$$

Using Clausius-Mosotti's internal field argument discussed above, this additional polarization contributes to the static dielectric constant according to the following formulae:

$$\frac{\varepsilon' - 1}{\varepsilon' + 2} = \frac{N_m}{3\varepsilon_0}\alpha_{ind} + \frac{N_d}{3\varepsilon_0}\alpha_o \cdot \tag{15}$$

Here N_d is the number of dipolar molecules per unit volume which is the same as Nm. This equation can be rewritten in the following form using equation (13).

$$\frac{\varepsilon' -1}{\varepsilon' +2} - \frac{\varepsilon_\infty -1}{\varepsilon_\infty +2} = \frac{N_d \mu^2}{9\varepsilon_0 KT}. \qquad (16)$$

This result, from Debye [23], has been used successfully to predict the static dielectric constant of many polar gases and polar liquids. However, when applied to the condensed state of matter, Debye's theory breaks down while predicting the infinite dielectric susceptibility (Mosotti catastrophe). The reason for this breakdown lies in the assumption that is made in the expression for the Clausius-Mosotti local field. The near field in this case is assumed to be zero. In the condensed phase, permanent dipoles tend to lose their individual freedom of orientation through association and steric hindrance. Their interaction with their surroundings has to be taken into account, and the near field cannot be ignored.

F. Onsager theory

To avoid the Mossotti catastrophe, Onsager modified the Debye theory by introducing a cavity. In his new approach to the problem, the electric field was represented by the sum of a 'cavity field' and a 'reaction field'. If the surroundings of each molecule are considered to be a homogeneous continuum, having the macroscopic properties of the substance, then the 'cavity field' is the field inside the cavity of molecular dimensions, due to a uniform external field. This cavity field is the field in the cavity resulting from the polarization induced in the surrounding medium by the molecule in the cavity. This part of the field exerts no torque on the molecule. Onsager's molecular model consisted of a sphere with a permanent dipole moment and an isotropic polarizability. Based on this model he arrives at the following expression, linking the molecular dipole moment with the static dielectric constant:

$$\frac{\varepsilon' -1}{\varepsilon' +2} - \frac{\varepsilon_\infty -1}{\varepsilon_\infty +2} = \frac{3\varepsilon'(\varepsilon_\infty +2)}{(2\varepsilon' + \varepsilon_\infty)(\varepsilon' +2)} \frac{N_d \mu^2}{9\varepsilon_0 KT}. \qquad (17)$$

Onsager's relation is very satisfactory for non associated polar liquids [24, 25] and can also be applied to weakly bound Van der Waals solids. In general, most of the solid dielectrics do not obey any of the local field expressions at sufficiently low frequencies, due to the charge carriers present in these materials, mostly ions, but possibly also electrons. This renders any meaningful measurement of the low frequency dielectric permittivity very difficult, making the comparison with the local field theory rather doubtful.

G. Debye Theory of Dielectric Behavior

Debye [26] gave the classical picture of the relaxation of polarization with a single relaxation time. In his work he considered a set of non-interacting dipoles free to rotate in opposition to some viscous resistance in a fluid like medium. The equation for complex permittivity is

$$\varepsilon^* = \varepsilon_\infty + \frac{\varepsilon_0 - \varepsilon_\infty}{1 + i\omega\tau}, \qquad (18)$$

where ε_0= Dielectric constant at low frequency, ε_∞ = Dielectric constant at high frequency, ω = Angular frequency, and τ = Relaxation time. According to Frohlich, the real and imaginary parts of the dielectric constant are given by

$$\varepsilon^* = \varepsilon_\infty + \frac{\varepsilon_0 - \varepsilon_\infty}{1 + i\omega^2\tau^2}, \quad \varepsilon'' = \left(\frac{\varepsilon_0 - \varepsilon_\infty}{1 + i\omega\tau}\right)\omega\tau. \qquad (19)$$

The maximum values of ε' and ε'' are,

$$\varepsilon' = \frac{\varepsilon_0 - \varepsilon_\infty}{2}, \quad \varepsilon'' = \frac{\varepsilon_0 - \varepsilon_\infty}{2}, \qquad (20)$$

$$\varepsilon_0 - \varepsilon_\infty = \left(\frac{3\varepsilon_0}{2\varepsilon_0 + \varepsilon_\infty}\right)\left(\frac{\varepsilon_0 + 2}{3}\right)\frac{4\pi n g \mu^2}{3kT}, \qquad (21)$$

where n = Dipole Moment, g = Parameter related to dipole interaction and T = Temperature. Every molecular dipole in a given chain is coupled to the neighboring dipole of the same chain by primary valence bands so that the motion of any dipole affects the motion of its neighbors, and they in turn, influence its response to a torque. Further, in various configurations, which in chain molecules can assume, we can find another segment of the chain acting effectively as a co-operative electrical unit, and these segments will of course vary in length between the improbable extremes of a single non-numeric unit and the whole extended chain. Such a state leads to the distribution of the relaxation time. On eliminating the parameter ωτ between the two equations and rearranging the two parameters (ε' and ε") we get,

$$\left[\varepsilon' - \frac{\varepsilon_0 - \varepsilon_\infty}{2}\right]^2 + \varepsilon''^2 = \left[\frac{\varepsilon_0 - \varepsilon_\infty}{2}\right]^2. \qquad (22)$$

The above equation is of a circle of radius $\frac{\varepsilon_0 - \varepsilon_\infty}{2}$. Only the semicircle over which ε'' is positive has physical significance. Materials with a single relaxation time yield a semicircle in the ε' and ε'' plane. Cole [27] modified the Debye equation by an empirical equation for complex permittivity.

$$\varepsilon^* - \varepsilon_\infty = \frac{\varepsilon_0 - \varepsilon_\infty}{1 + (j\omega\tau)^{1-\alpha}}, \qquad (23)$$

where α is an empirical parameter. It lies between 0 and 1, and it denotes the angle of tilt of the circular arc from the real axis. The modified expression measures the small deviation from the ideal Debye behavior, but some materials deviate very much from the Debye behavior. Havirilik and Negami [28] gave an expression, which is of the form,

$$\varepsilon^{*}-\varepsilon_{\infty}=\frac{\varepsilon_{0}-\varepsilon_{\infty}}{\left[1+(j\omega\tau)^{1-\alpha}\right]^{1-\beta}} \ . \tag{24}$$

The dielectric constant and losses were calculated using the equation valid for a parallel plate capacitor,

$$\varepsilon^{'}=\frac{C_{x}d}{\varepsilon_{0}A}, \quad \varepsilon^{''}=\varepsilon^{'}\tan\delta \ , \tag{25}$$

where C_x is the sample capacitance in Farad, A is area of the sample, d is the thickness of the sample and ε_0 is a constant representing the permittivity of free space. The variation of the dielectric constant and loss tangent, suggest the net effect of some internal field within the crystal along with the external AC electric field. The dipole-dipole interactions between the different groups or many body interactions suggest lower losses with a higher frequency range. The dependence of the dielectric constant on frequency can be determined from the equation,

$$C=C_{g}+\frac{S\tau}{1+\tau^{2}\omega^{2}} \ , \tag{26}$$

where C_g is the geometrical capacitance, S the conductance corresponding to the absorption current, τ is the dipole relaxation time and ω the angular frequency. The above equation shows that C should diminish with increasing frequency. The increase in losses at a low frequency could be associated with the polarization of the trapped charge carriers. With an increase in the frequency, polarization decreases and becomes vanishing small at high frequencies. The expression for the dielectric loss is given by the equation [29, 30],

$$\tan\delta=\frac{\omega^{2}\tau^{2}\left(G_{in}+S\right)+G_{m}}{\omega\left\{S\tau+C_{g}\left(\omega^{2}\tau^{2}+1\right)\right)} \ , \tag{27}$$

where G_{in} is the conductance for the residual current. Obviously, from the equation,

$$Lim_{\omega\to 0}\tan\delta=\infty \ . \tag{28}$$

Differentiating the equation with respect to ω, and equating the derivative to zero, it is possible to obtain the value of ω of the frequency corresponding to the maximum loss.

H. Dielectric Theory of Optical Properties

In most cases, crystals are transparent to visible and/or infrared light. The interaction of the electromagnetic radiation with these crystals is treated, by applying the boundary conditions to the solutions of the Maxwell equations at the boundary between the different media. In the field of optical crystals, the wavelength of the light is always much larger than the inter atomic dimensions. Thus, the interaction of light and matter is averaged over many unit cells. As a consequence, the optical properties within each layer can be described macroscopically, in terms of phenomenological parameters, the so-called optical constants or optical parameters. As shown below, these are the real and imaginary parts of a complex index of refraction $\tilde{n}$. The real part, $n(\lambda)$, is the ratio of the velocity of light in vacuum to the velocity of the light of the wavelength (λ) in the material. The imaginary part, $-\kappa(\lambda)$, is an attenuation coefficient measuring the absorption of light with distance. Using the Maxwell equations, it is possible to relate these frequency-dependent "constants" to other optical parameters, such as the dielectric constant and conductivity. The crystals are composed of charged particles: bound and conduction electrons, ionic cores, impurities, etc. These particles move differently with oscillating electric fields, giving rise to polarization effects. At visible and infrared light frequencies, the only contribution to polarization comes from the displacement of the electron cloud, which produces an induced dipole moment. The parameters describing these optical effects, that is, the dielectric constant ε, the dielectric susceptibility χ, and the conductivity σ, can be treated as scalars for isotropic materials.

To find out what kind of electromagnetic waves exist inside the dielectric films, we take $\rho = -\nabla\ \mathbf{P}$ and $\mathbf{j} = \partial\mathbf{P}/\partial t$, where ρ is an effective charge, **P** is the polarization induced by the electromagnetic wave, assumed to be proportional to the electric field, and **j** is the corresponding current density averaged over a small volume. Under these conditions, the average field Maxwell equations in MKS units read:

$$\nabla.E=-\frac{\nabla.P}{\varepsilon_{0}} \ , \tag{29}$$

$$\nabla\times E=-\frac{\partial B}{\partial t} \ , \tag{30}$$

$$\nabla.B=0 \ , \tag{31}$$

$$c^{2}\nabla\times B=\frac{\partial}{\partial t}\left(\frac{P}{\varepsilon_{0}}+E\right), \tag{32}$$

where the symbols have their usual meaning. Note that the normal component of the electric field **E** is not conserved at the interface between materials of different polarizability. Instead, **D** = ε_0 + **P**, called electrical displacement, is conserved across such interfaces. The solutions to these equations have the form of harmonic plane waves with the wave vector k:

$$E=E_{0}\exp\left[i(\omega t-k\cdot r)\right], \tag{33}$$

$$H=H_{0}\exp\left[i(\omega t-k\cdot r)\right], \tag{34}$$

and represent a wave travelling with a phase velocity $\omega/k = c/n$, where c is the speed of light in vacuum and n is the index of refraction. When optical absorption is present, the wave vector and the index are complex quantities. From the Maxwell equations, a dispersion relation $k_2 = \varepsilon(\omega/c)^2$ is obtained relating the time variation with the spatial variation of the perturbation. In general, then, the wave vector k and the dielectric constant ε are complex quantities, that is, $k = k_1 - ik_2$ and $\varepsilon = \varepsilon_1 - i\varepsilon_2$. It is useful to define a complex index of refraction:

$$\tilde{n} = \tilde{k}\left(\frac{c}{\omega}\right) = n - ik. \tag{35}$$

For isotropic materials, k_1 and k_2 are parallel and

$$\varepsilon_1 = n^2 - k^2,\ \varepsilon_2 = 2nk, \tag{36}$$

$$n^2 = \frac{1}{2}\left[\varepsilon_1 + \left(\varepsilon_1^2 + \varepsilon_2^2\right)^{1/2}\right],\ k^2 = \frac{1}{2}\left[-\varepsilon_1 + \left(\varepsilon_1^2 + \varepsilon_2^2\right)^{1/2}\right]. \tag{37}$$

In the photon energy region where ε is real, $n = \varepsilon^{1/2}$ is also real and the phase (ω/k) and group ($\partial\omega/\partial k$) velocities are equal to c/n. In general, the velocity is reduced to $v(\lambda) = 1/\sqrt{\varepsilon_c(\lambda)}$ in the medium of a complex dielectric constant ε_c. The real part of n determines the phase velocity of the light wave, the imaginary part determining the spatial decay of its amplitude. The absorption coefficient α measures the intensity loss of the wave. For a beam travelling in the z direction, $I(x) = I(0)\ exp(-\alpha z)$, which means $\alpha = 2\omega\kappa/c = 4k\kappa/\lambda$.

III. DIELECTRIC LOSS

The permittivity of a dielectric material has both real and imaginary mathematical representations. The imaginary part of permittivity is represented in mathematical equations as ε''. This imaginary part of permittivity describes the energy loss from an AC signal as it passes through the dielectric. The real part of permittivity εr is also called the dielectric constant and relative permittivity. The permittivity of a material describes the relationship between an AC signal's transmission speed and the dielectric material's capacitance. When the word "relative" is used in front of permittivity, the implication is that the number is reported relative to the dielectric properties of a vacuum**.** The imaginary part of the dielectric permittivity which is a measure of how much field is lost as heat during the polarization of a material by an applied alternating electric field, is also termed as dielectric loss. The characteristic orientation of the dipoles in an electric field results in a frequency variation of the dielectric constant and loss, over a broad band of frequencies. The typical behavior of the real and imaginary parts of permittivity as a function of frequency is shown in Figure 1. [31].

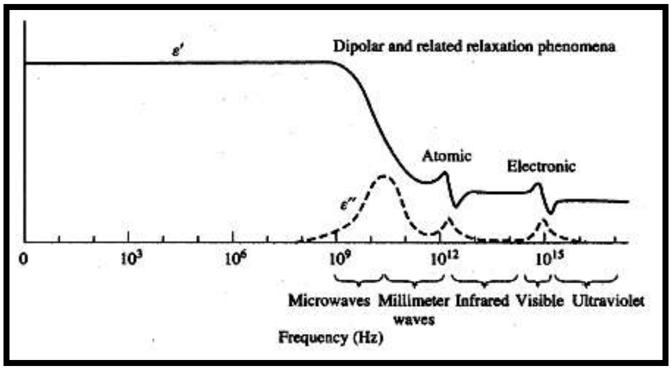

FIGURE 1. Frequency dependence of dielectric permittivity of an ideal dielectric material.

The relative permittivity of a material is related to a variety of physical phenomena that contribute to the polarization of the dielectric material. In the low frequency range the ε'' is dominated by the influence of ion conductivity. The variation of permittivity in the microwave range is mainly caused by dipolar relaxation, and the absorption peaks in the infrared region and above, are mainly due to atomic and electronic polarizations. The dielectric properties of solid dielectrics at microwave and radio frequencies are highly influenced by the ionic positions and changes, caused by the lattice vibrations. Two types of dielectric losses are identified in crystalline solids at high frequencies, namely, intrinsic and extrinsic losses. The dielectric dispersion in solids depends on factors such as ionic masses, electric charge/valence state of the ions, spring constant of the bond, lattice imperfections etc. The dielectric losses close to the lattice vibration frequencies are generally estimated in terms of the anharmonicity of the lattice vibrations. The low frequency phonons are responsible for the intrinsic dielectric losses in solid dielectrics. The intrinsic loss mechanism occurs due to the interaction between the phonons and the microwave field, or due to the relaxation of the phonon distribution function. The lattice phonon modes will determine the intrinsic limits of the high frequency dielectric losses in crystalline solids. The extrinsic losses occur due to the interaction between the charged defects and the microwave fields.

A. Complex dielectric permittivity and Maxwell equations

In the case of dielectric polarization, the polarization of the material is related to the electric field by:

$$P = \varepsilon_0 \chi_e E\ . \tag{38}$$

This leads to:

$$D = \varepsilon_0 (1 + \chi_e) E = \varepsilon_0 \varepsilon_r E\ . \tag{39}$$

For real materials D can be described as [32]:

$$D = (\varepsilon - j\varepsilon_p) E\ . \tag{40}$$

Here, $\varepsilon=\varepsilon_0\varepsilon_r$, the real part of permittivity, and $\varepsilon_p=\varepsilon_o\varepsilon''$ is a factor describing the dielectric (polarization) losses. For a region filled with a homogeneous isotropic material, the first Maxwell equation can be written as:

$$\nabla\times H=\frac{\partial D}{\partial t}+\sigma E\,. \tag{41}$$

Here, σ is the conductivity of the material. Substituting for D from equation (40) equation (41) becomes:

$$\nabla\times H=i\omega(\varepsilon-i(\varepsilon_p+\sigma/\omega))E\,. \tag{42}$$

The complex dielectric constant is defined as below:

$$\varepsilon^*=\varepsilon-i(\varepsilon_p+\sigma/\omega)\,. \tag{43}$$

Here, ε is the real part of the permittivity and is defined as:

$$\varepsilon=\varepsilon_r\varepsilon_0\,. \tag{44}$$

Here ε_r is known as the relative permittivity or dielectric constant, and ε_0 is the permittivity of free space. Here the first and second terms in the imaginary part of the complex permittivity, represent the dielectric and ohmic losses respectively [33]. The loss tangent is given as:

$$\tan\delta=\frac{\varepsilon''}{\varepsilon'}\,. \tag{45}$$

In this ε_r is used throughout to represent the relative permittivity of the materials and tanδ is used to represent a measure of the dielectric loss.

IV. CLASSIFICATIONS OF DIELECTRIC MATERIALS

Dielectric materials can be classified into two major categories: Linear (normal dielectric) materials and non linear dielectric materials. The linear dielectric materials can again be subdivided into three classes, based on the mechanism of electric polarization as non-polar and dipolar materials.

A. Linear dielectric materials

The dielectric materials, which exhibit a linear relationship between the polarization and applied electric field are known as linear dielectrics. This class of materials gets polarized with the application of the field, and depolarized on the removal of the field. Based on the nature of the polarization mechanism, the linear dielectrics can be grouped as follows [34]:

Non polar materials: In materials of this class, an electric field can cause only elastic displacement of the electron cloud (mainly the valence electron cloud). So they have only electronic polarization. Such materials are generally referred to as elemental materials.

Polar materials: In materials of this class, an electric field can cause only an elastic displacement of the electron clouds as well as elastic displacement of the relative positions of ions. These materials have both electronic and ionic polarization. The material may be composed of molecules and each of the molecules is made of more than one kind of atom, without any permanent dipole moment. Examples of such materials are ionic crystals; in this case the total polarizability is the sum of the ionic and electronic polarizabilities.

$$\alpha=\alpha_e+\alpha_i\,. \tag{46}$$

Dipolar materials: The materials of this class have all three fundamental polarizations: electronic, ionic and orientation. Thus the total polarizability for them is

$$\alpha=\alpha_e+\alpha_i+\alpha_o\,. \tag{47}$$

B. Non linear dielectric materials

The materials which have a spontaneous polarization even in the absence of an external field are grouped into the class of non linear dielectrics. The spontaneous polarization appears in these classes of materials due to their crystalline structure. A necessary condition for a solid to fall in the class of non linear dielectrics is the absence of a center of symmetry. Among the 32 crystal classes, 11 have a center of symmetry, and hence, they will not exhibit spontaneous polarization. Out of the remaining 21 classes of crystals without a centre of symmetry, 20 are piezoelectric, i.e., these crystals can be polarized under the influence of an external stress. Ten out of the 20 piezoelectric crystals exhibit the pyroelectric effect, i.e., the polarization of these classes of materials can be changed with the change of temperature. The ferroelectric materials discussed below, are part of the spontaneously polarized pyroelectrics.

C. Ferroelectric Materials

A ferroelectric material is a non-linear dielectric that exhibits a remanent polarization in the absence of an external electric field, and its direction can be switched by an applied electric field [35]. The name ferroelectricity comes from the similarities between polarizations of ferroelectric materials with the magnetization of ferromagnetic materials. Ferroelectric materials display a hysteretic effect of polarization with an applied field. The hysteretic loop is caused by the existence of permanent electric dipoles in the material. When the external electric field is initially increased from zero value, the polarization increases. as more of the dipoles are lined up along the direction of the field. When the field is strong enough, all dipoles are lined up with the field, so the material is in a saturation state. If the applied electric field decreases from the saturation point, the polarization also decreases. However, when the external electric field reaches zero, the polarization does not reach zero. The polarization at the zero fields is called the remanent polarization. When the direction of the electric field is reversed, the polarization decreases. When the reverse field reaches a certain value, called the coercive field, the polarization becomes zero. By further increasing the field in this reverse direction, the reverse saturation can be reached. When the field is decreased from this saturation point, the sequence just reverses itself.

In a ferroelectric material a transition occurs from a centro symmetric to a noncentro symmetric unit cell at the Curie point Tc. The shift in the structural symmetry affects both the structural and physical properties of the crystal. Ferroelectricity can be maintained only below the Curie temperature. When the temperature is higher than Tc, a ferroelectric material is in its paraelectric state. Ferroelectric materials have great application potential in developing smart electromagnetic materials, structures, and devices, including miniature capacitors, electrically tunable capacitors, filters and phase shifters, in recent years. Their application in the microwave frequencies are still under intensive investigation.

V. TUNABLE DIELECTRICS

Dielectric materials, which have a voltage-dependent dielectric constant, are termed as tunable dielectric materials [36]**.** Generally, this class of materials exhibits a large change in the dielectric constant, with an applied DC electric field. The major classes of materials being considered for tunable dielectric applications are ferroelectrics in their paraelectric state. The ferroelectric materials (FE) have been investigated in the microwave range since the 1950s. Only recently, the monolithically compatible processing of certain ferroelectric thin-film compounds has become possible, and has generated great interest and promise for designing a new class of tunable microwave devices. For a microwave engineer the main attraction of a tunable material is the strong dependence of its dielectric permittivity ε on the applied bias electric field E_0. This characteristic is commonly described by a parameter named, tunability n, defined as the ratio of the permittivity of the material at zero electric field $\varepsilon_{(0)}$ to its permittivity at some non-zero electric field $\varepsilon_{(E)}$ as given by equation (28). The relative tunability nr is defined by equation (49) [36].

$$n=\frac{\varepsilon(0)}{\varepsilon(E)}, \tag{48}$$

$$n_r=\frac{\varepsilon(0)-\varepsilon(E)}{\varepsilon(0)}. \tag{49}$$

The dielectric loss of a tunable dielectric material is also dependent on the applied DC electric field. Experiments show that a ferroelectric material with a higher loss tangent usually has a larger tunability. Since the loss tangent of a material is an important factor affecting the performances of the electric circuit, in the development of electrically tunable ferroelectric microwave devices, a figure of merit *K* (*K*-factor), defined by *K*=Tunability/tanδ .

$$K=\frac{\varepsilon(0)-\varepsilon(E)}{\varepsilon(0)}\times\frac{1}{\tan\delta}, \tag{50}$$

is often used to indicate the quality of the tunable dielectric materials. Usually, in the calculation of *K*, the loss tangent at the maximum external DC electric field is used [37].

A. Tunable materials for microwave devices

Microwave materials have been widely used in a variety of applications ranging from communication devices to satellite services, and the study of their properties at microwave frequencies, and the development of functional microwave materials, have always been among the most active areas of solid-state physics, materials science, and electrical and electronic engineering. In recent years, the increasing need for the development of high speed and high frequency circuits and systems has made a thorough understanding of the properties of materials at microwave frequencies, a necessity [38].

Wireless systems operating in the microwave region are required to be lightweight, compact and of low cost, which could be addressed by miniaturization and integration. Meanwhile, the need of frequency agile applications demands the use of low loss, and highly tunable devices to allow multi-bandwidth operation with little impact on the component count. Microwave tunable passive devices mainly include filters, phase shifters, delay lines and matching circuits in connection with applications, such as reconfigurable antennas, software defined radios, etc [39, 40]. Implementing several separate transceiver circuits in a single hardware device increases the component count, and hence, the overall cost. Therefore, in terms of RF front end circuitry, significant cost saving can be achieved, by using electronically tunable components. In this scenario a single tunable component is employed to replace several fixed components. For example, a band pass filter (BPF) with a tunable pass band could replace several fixed filters or a tunable delay line could replace a set of fixed delay lines in the beam-forming network of a phased array antenna [41].

VI. CONCLUSION

Theoretical discussions in respect of the dielectric constant (ε), which is an important physical quantity determining the dielectric properties of matter, have been discussed by Clausius-Mossotti, Debye, Onsager and others. These models have provided an insight into the complex polarization mechanisms in solids. Some new applications and results have been discussed.

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