Infrared and Millimeter Waves

VOLUME 8
ELECTROMAGNETIC WAVES IN MATTER,
Part I

Edited by
Kenneth J. Button

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PREFACE

This is the first volume in this treatise to deal exclusively with the millimeter and submillimeter wave properties of materials and the methods of measuring and interpreting these properties. A second volume on this topic is now in preparation, and plans for a third are in progress.

The table of contents speaks eloquently for the theme of this subseries on electromagnetic wave interaction in matter. G. W. Chantry's dielectric materials studies are the foundation of a branch of modern classical physics that he inspires with his semiannual Gordon conferences. This general treatment of dielectrics is naturally followed by the work of W. F. X. Frank and U. Leute on the far-infrared spectroscopy of high polymers and then by several semiconductor chapters. S. Perkowitz gives a general treatment of the spectroscopy of semiconductors; B. Jensen provides a most thorough treatment of free carrier behavior in semiconductors (which is most important in the far infrared); A. Hadni then reviews pyroelectric detectors.

We were most fortunate to obtain a contribution on cyclotron resonance by T. Ohyama and E. Otsuka, who have done a great deal of work in that area. F. Gervais contributed a chapter on his very innovative spectroscopic work. The chapter by Phillipe Goy appeared to me to be so timely that I designated it as the opening invited paper at the Miami Beach conference in December 1981. Finally, J. C. Maan was invited to prepare the chapter on semiconductor layered structures because he has the ability to make this specific example of a larger, emerging topic sound simple.

A second volume devoted to electromagnetic waves in matter is being readied for press. Volumes 9, 10, and 11 begin a subseries on millimeter wave components and techniques; all three of these volumes are in production and scheduled for publication very soon.

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CHAPTER 1

Properties of Dielectric Materials

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I. Introduction

The study of liquids and polymers by means of far-infrared and submillimeter spectroscopy forms part of the much larger topic of dielectric physics. A dielectric medium is one in which there are no free charges so the dc conductivity is zero, but the medium can sustain displacement currents and these may have lossy components. Thus dielectrics are all materials that are not metallic, semiconducting, or ionized. If an external field E is applied to a dielectric, the field inside the dielectric is given by

$$D = (\varepsilon/\varepsilon_0)E, \tag{1}$$

where ε is the permittivity of the dielectric and ε_0 the absolute permittivity of free space (8.85418×10^{-12} F/m). In nearly all dielectric work, however, it is customary to write $\varepsilon = \varepsilon/\varepsilon_0$ to avoid the constant apperance of the ε_0 factor; then ε so defined is the *relative permittivity of the medium,* i.e., the permit-

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tivity relative to that of the vacuum. The relative permittivity is complex because it will have a lossy component, and one therefore usually writes

$$\hat{\varepsilon} = \varepsilon' - i\varepsilon'', \tag{2}$$

where the caret is used to signify an explicitly complex quantity. The use of complex relative permittivities goes very well with the use of the complex representation to describe alternating fields. The dielectric medium will thus sustain two currents in response to an ac field of circular frequency ω, a quadrature or displacement current determined by ε' and an in-phase or lossy current determined by ε''. The complex relative dielectric conductivity $\hat{\sigma}$ is therefore also complex with components

$$\hat{\sigma} = j\omega\varepsilon' + \omega\varepsilon'', \tag{3}$$

and the current, in the complex plane, is not perpendicular to the voltage but departs from this position by an angle δ whose tangent is given by

$$\tan \delta = \varepsilon''/\varepsilon'. \tag{4}$$

In most practical cases, δ will always be small and it is permissible to write

$$\delta = \varepsilon''/\varepsilon', \tag{5}$$

which is the justification for the common practice of quoting the dimensionless ratio $\varepsilon''/\varepsilon'$ in microradians.

The presence of a slab of dielectric in a volume that would otherwise be vacuum leads to a polarization P whose magnitude is given by the field in the dielectric minus the applied field; in other words,

$$P = (\hat{\varepsilon} - 1)E. \tag{6}$$

This equation is not very useful as it stands because the frequencies at which P will fall to zero, and at which consequentially $\hat{\varepsilon}$ will equal unity, lie in the x-ray region, whereas all dielectric work is restricted to the region of millimeter and longer wavelengths. It is more usual, therefore, to write

$$P = (\hat{\varepsilon} - \varepsilon_\infty)E, \tag{7}$$

where ε_∞ is a high-frequency limiting permittivity. This is unfortunately a rather amorphous and poorly defined quantity because it is not possible, in fact, to find any measurable frequency at which dispersion does not exist. It is rather to be considered as a parameter: the permittivity in frequency regions so high that the particular dispersion mechanism under consideration will no longer have any effect. It is also helpful to normalize the polarization by writing

$$P_n = \frac{\hat{\varepsilon} - \varepsilon_\infty}{\varepsilon_s - \varepsilon_\infty} E, \tag{8}$$

where ε_s is the static relative permittivity. Then P_n will go from one at $\omega = 0$ to zero at $\omega = \infty$.

At extra-high frequencies and beyond ($\nu > 30$ GHz) it is not possible, with the currently available techniques, to measure field parameters such as the vectorial electric field strength and its phase. Rather, one measures energy flow; and in this situation one is led naturally to introduce a refractive index n which is the ratio of phase velocity in free space (i.e., c) to that in the medium. The space dependence of the field is then given by

$$E = E_0 \exp(-i\omega n x/c), \tag{9}$$

but the measurable quantity, the intensity, is given by

$$I = \tfrac{1}{2}\varepsilon_0 c E_0^2. \tag{10}$$

The propagating medium will usually be lossy, so one has a progressive attenuation of the field given by Lambert's law

$$E = E_0 \exp(-\tfrac{1}{2}\alpha x), \tag{11}$$

which also can be written as

$$I = I_0 \exp(-\alpha x). \tag{12}$$

Here α is the power absorption coefficient (usually measured in nepers per centimeter). Equations (9) and (11) can be combined to give

$$E = E_0 \exp(-i\omega \hat{n} x/c), \tag{13}$$

where the complex refractive index $\hat{n}$ is defined by

$$\hat{n} = n - i(\alpha/4\pi\bar{\nu}), \tag{14}$$

in which the wave number

$$\bar{\nu} = \nu/c = \omega/2\pi c \tag{15}$$

is introduced. The two formalisms, in terms of either $\hat{\varepsilon}$ or $\hat{n}$, are readily connected by means of Maxwell's celebrated relationship

$$\hat{\varepsilon} = \hat{n}^2. \tag{16}$$

Identifying real and imaginary components in Eq. (16) gives the simple relations

$$\varepsilon' = n^2 - (\alpha/4\pi\bar{\nu})^2 \tag{17a}$$

and

$$\varepsilon'' = 2n(\alpha/4\pi\bar{\nu}), \tag{17b}$$

which have the not-so-simple converses

$$n = (\varepsilon'/2)^{1/2}\{[1 + (\varepsilon''/\varepsilon')^2]^{1/2} + 1\}^{1/2} \tag{18a}$$

and

$$\alpha/4\pi\bar{\nu} = (\varepsilon'/2)^{1/2}\{[1 + (\varepsilon''/\varepsilon')^2]^{1/2} - 1\}^{1/2}. \tag{18b}$$

It is a fairly widespread practice in dielectric work to introduce the absorption index k and to write

$$\hat{n} = n - ik, \tag{19}$$

from which it immediately follows that

$$k = \alpha/4\pi\bar{\nu}. \tag{20}$$

This is a rather unsatisfactory practice because confusion with the wave vector, also designated by k, arises; nevertheless it is firmly established, especially in solid-state work. Another convention, fortunately now tending to become obsolete, is the use of the extinction index κ defined by

$$\hat{n} = n(1 - i\kappa). \tag{21}$$

It is also worth mentioning that there is no universally agreed convention for the sign of the imaginary components in these complex quantities. One can find $\hat{\varepsilon}$, for example, defined with either sign. There is no physical difference; the two signs merely represent the sense of the conventional rotation of the vectors in the Argand diagram: negative going with clockwise and positive with anticlockwise. However, as soon as one has defined any one of these quantities, all the rest become fixed. Thus, because it seems more natural to consider the vector $\hat{\mathbf{E}}(t)$ rotating anticlockwise into the first quadrant, one writes $\hat{\mathbf{E}}(t) = E_0 \exp(i\omega t)$; then the sign for the imaginary component of $\hat{\varepsilon}$ will be negative because the charge will lag behind the driving field.

Dielectric spectroscopy is the determination of the variation of $\hat{\varepsilon}$ or $\hat{n}$ with frequency. In principle there is no upper frequency limit to dielectric spectroscopy; but to keep the concept useful, it is usual to introduce a division that separates phenomena involving long-range cooperative motion from phenomena involving highly localized, essentially independent motion. The former is the class to be treated by dielectric concepts, whereas the latter is treated by the methods of, for example, molecular spectroscopy. This division is helpful, but it is arbitrary, and physical phenomena may well transgress its boundaries. Thus the positions and intensities of lines in molecular spectra can be calculated by purely local models; but calculating their shapes demands a more global treatment involving long-range interactions. The division does make sense spectroscopically, because at frequencies below 6 THz (200 cm^{-1}), one is often talking of dielectric phenomena whereas at higher frequencies one seldom is. So, for the purposes of this

review, the upper limit to dielectric spectroscopy will be taken to be 200 cm^{-1}.

One can approach dielectric physics from two directions: one can consider the macroscopic theory, which essentially stems from the application of universally valid physical laws such as Maxwell's equations; or one can consider the microscopic theory, which shows how the motion of the constituent entities leads to the observed dielectric properties. The macroscopic theory gives results that are exact or nearly so but of somewhat limited information content. The microscopic theory is much more interesting because it can, in principle, give a wealth of information; however, the computational difficulties involved in its full elaboration are formidable and can be side-stepped only by means of rather drastic approximations. Therefore the microscopic theory does not give quite as much insight as one might hope; nevertheless considerable progress has been made.

II. The Macroscopic Theory

The complex permittivity $\hat{\varepsilon}$ is the natural quantity for those who have coherent sources and detectors and can measure field parameters. Typically this means those who work below 30 GHz, but the rapid development of techniques for measuring the higher-frequency regions is tending to extend this limit upward. The complex refractive index $\hat{n}$ is the natural quantity for those limited to optical techniques with incoherent black body sources and thermal detectors. Rather interestingly, the closing up of the millimeter–submillimeter gap has enabled direct tests of Maxwell's relation Eq. (16) to be carried out by comparing $\hat{\varepsilon}$ with $\hat{n}^2$ at a fixed frequency; but the results, as expected, entirely support the theory. Therefore one can use either formalism; but because that involving $\hat{\varepsilon}$ is simpler, it is usually preferred. We now consider those properties of $\hat{\varepsilon}$ that are independent of particular microscopic models, calling this branch of the topic the *macroscopic theory.*

A. The Response Function

The behavior of dielectric media can be modeled quite well by simple RC circuits. There are many possible combinations, but they all contain a common element, a resistor R and a capacitor C in series. It is therefore important to analyze the response of this simple circuit combination to various time dependences of the applied field. The situation is illustrated in Fig. 1. A driving voltage $V(t)$ is applied across the resistor and capacitor in series, and we are interested in the resulting voltage across the capacitor $V_c(t)$. The basic differential equation is

$$V(t) - [dV_c(t)/dt]RC = V_c(t), \tag{22}$$

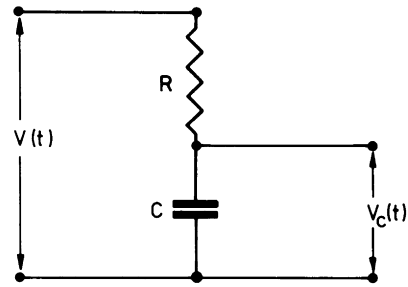

FIG. 1 A simple RC circuit used to model a lossy dielectric.

and we seek solutions valid in the long time limit (i.e., $t \gg RC$) when starting transients will have died out. Taking Laplace transforms throughout gives

$$V(s) = V_c(s) + RCsV_c(s) \tag{23}$$

from which

$$V_c(s) = V(s)/(1 + RCs). \tag{24}$$

Inverting the Laplace transform and invoking the convolution theorem gives

$$V_c(t) = \int_0^t V(t')\tau^{-1} \exp[-(t - t')/\tau]\, dt', \tag{25}$$

which is a special case of the more general relation

$$V_c(t) = \int_0^t V(t')\dot{R}(t - t')\, dt. \tag{26}$$

The function $R(t)$ whose time derivative appears in this equation is the response function. For the simple RC circuit it takes the elementary form

$$R(t) = \exp(-t/\tau), \tag{27}$$

where $\tau = RC$. It is called the response function because, if $V(t)$ changes abruptly, $R(t)$ measures the subsequent response. Thus suppose that $V(t)$ is constant for a time t_0 ($\gg \tau$) but then drops to zero. What will be the voltage across the capacitor at a subsequent time t ($> t_0$)? Substituting in Eq. (26) and noting that $\exp(t_0/\tau)$ is much greater than unity gives the answer

$$V_c(t) = V_0 \exp(-(t - t_0)/\tau). \tag{28}$$

It t_0 is allowed to decrease toward zero, this becomes