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A Contribution to the Interpretation of Impedance Dispersion Curves of Powder Luminescent Materials

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Dispersion of the complex permittivity of powder luminescent materials in the region of acoustic and radio frequencies is caused both by the relaxation processes in phosphors themselves and by the relaxation processes determined by the geometry of the powder grains in the binder or in the air. In the presented paper relations have been derived that explain the deviations of the experimental curves from the Debye type dispersion curves and that enable to separate the mentioned two types of relaxation processes one from the other.

I. Introduction

It is useful to combine optical and luminescent measurements of luminescent materials with measurements of their electrical parameters e.g. conductivity and permittivity, by means of frequency dependence of the complex impedance. In the measurements of powder samples the behaviour of the studied material is influenced by the fact that the powder is embedded in an environment (binder or air) with other electrical parameters. The purpose of the paper is to find out the properties of the powder material itself without the influence of the environment.

It is well known that in some materials, e.g. in alkalihalides the complex permittivity has a frequency dependence not only in the IR region – connected with electron and ion polarisation — but also in the region of acoustic and radio frequences connected with lattice imperfections [1].

The dispersion of the complex permittivity $\varepsilon = \varepsilon' - j\varepsilon''$ in the mentioned frequency range (further only this region will be considered) is described by the Debye equations

$$\varepsilon' = \varepsilon_\infty + \frac{\varepsilon_0 - \varepsilon_\infty}{1 + \omega^2 \tau^2}, \tag{1}$$

$$\varepsilon'' = \frac{(\varepsilon_0 - \varepsilon_\infty) \omega\tau}{1 + \omega^2\tau^2}, \tag{2}$$

$$\operatorname{tg} \delta = \frac{\varepsilon''}{\varepsilon'} = \frac{(\varepsilon_0 - \varepsilon_\infty) \omega\tau}{\varepsilon_0 + \varepsilon_\infty \omega^2\tau^2} \tag{3}$$

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ε_0, ε_∞ are constants that equal the value of ε' at very low and very high frequencies. τ is a time constant of relaxations of dipoles present in the studied material, ω the angular frequency of the applied voltage.

Fig. 1 shows the grafic representation of the Debye equations. These equations were derived for the description of the behaviour of polar molecules but they are
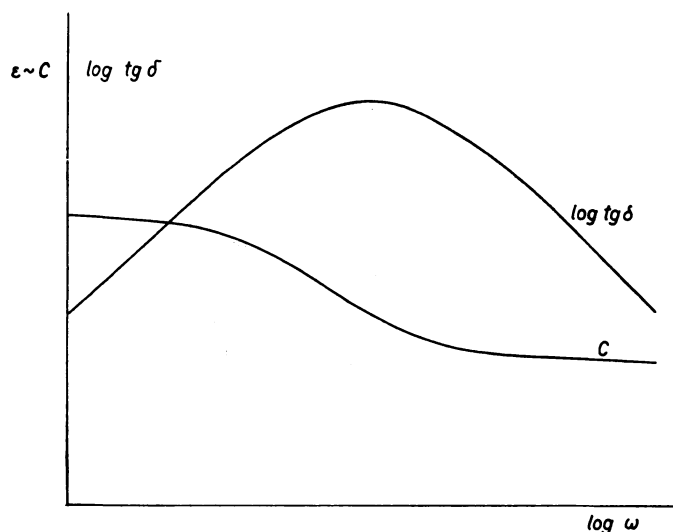

Fig. 1. Grafic representation of the Debye equations

used [2] for the description of relaxation processes in solids as well. The relaxation processes in solids are caused either by lattice imperfections having a dipole moment or by inhomogeneous conductivity. Besides, in powder materials the geometry of the sample configuration gives rise to a further type of relaxations (so called Maxwell--Wagner relaxations) that combine with the intrinsic relaxation processes of the investigated material. Dispersion relations are found also in luminescent materials exposed to light (so called photodielectric effect), while in the same materials in dark no dispersion is observed. In most materials this fact has been explained [3, 4, 5] on the base of inhomogeneous conductivity, whether macroscopical (dispersion of Maxwell-Wagner type) or microscopical (space-charge regions). In this case the time constant varies inversely as the conductivity and the dispersion region is determined by the reciprocal value of the time constant. It is therefore evident, that with decreasing conductivity the dispersion region shifts to low frequencies. The dark conductivity of the luminescent materials is very low and therefore dispersion may be found at frequencies lower than the acoustic and radio ones. On the other hand, the luminescent powders exhibit photoconductivity, their time constant shortens when they are exposed to light and consequently the dispersion region shifts to higher (acoustic and radio) frequencies.

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2. Dispersion Relations of Powder Samples

The material studied was AlN : Mn in the powder form situated in a condenser in the space between one full copper electrode and one mesh nickle electrode. The distance of the electrodes was 0,2 mm. Mercury 200 W lamp with filters served as the light source. A precise Schering bridge ballancing the sample with parallel combination of resistance R and capacity C was used for the measurements of the sample resistance (by means of the loss factor tg δ) and sample capacity in the region of frequencies from 300 Hz to 300 kHz and at the voltage of 10 V*.
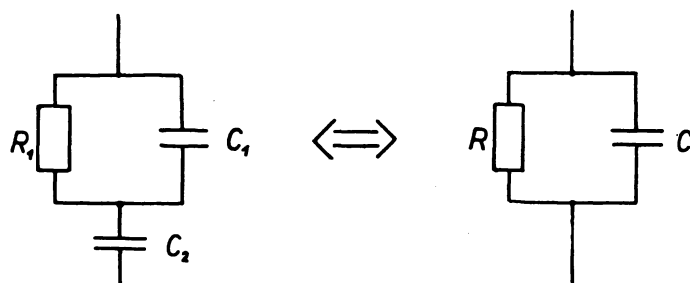

Fig. 2. The two-layer model of the sample (left side) and the equivalent compensatory circuit of the bridge (right side)

For describing the behaviour of our sample it must be taken into account that the sample consists of AlN grains separated by air so that a two-layer model (Fig. 2) must be applied. R is the ohmic resistivity of the AlN grains as a whole in the given geometrical configuration, C_1 their capacity and C_2 capacity of the air interspace. R_1 and C_1 are quantities proportional to specific resistivity and permittivity that can be used for describing the spectral response and temperature course of the photoconductivity, activation energies of traps etc. R and C refer to the compensatory circuit.

The following relations are valid: [6]

$$C = \frac{C_1 C_2}{C_1 + C_2} + \frac{C_2^2}{C_1 + C_2} \frac{1}{1 + \omega^2 \tau^2}, \tag{4}$$

$$\frac{1}{R} = \frac{C_2}{C_1 + C_2} \frac{\omega^2 R_1 (C_1 + C_2)}{1 + \omega^2 \tau^2}, \tag{5}$$

$$\text{tg } \delta = \frac{\omega R_1 C_2}{1 + \dfrac{C_1}{C_1 + C_2} \omega^2 \tau^2} \tag{6}$$

$$\tau = R_1 (C_1 + C_2). \tag{7}$$

The relations are evidently of Debye type, under the assumption that R_1, C_1 and C_2 are frequency independent. However, experimental dispersion curves show deviations from the Debye type (Fig. 3) and demonstrate thus that the frequency dependence of R_1 and C_1 must be taken into account. Let us do if formally by a new
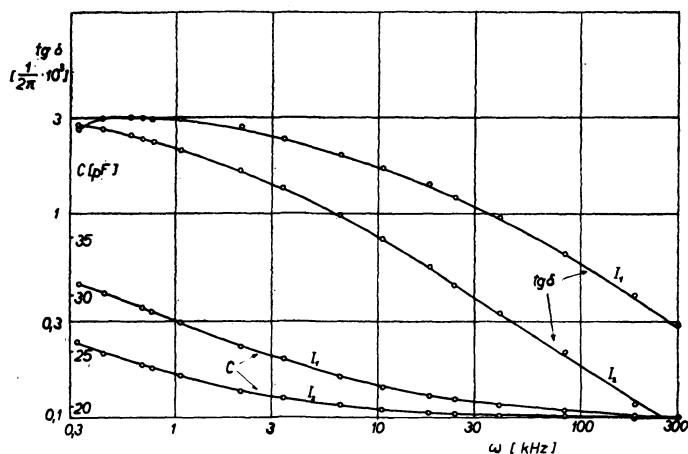

Fig. 3. Dispersion curves of AlN: Mn (1%) measured at two different intensities of light ($I_1 > I_2$; HBO 200 mercury lamp)

set of Debye relations as all types of relaxations (see before in the mentioned frequency region can be described by them:

$$C_1 = C_{1\infty} + \frac{C_{10} - C_{1\infty}}{1 + \omega^2 \vartheta^2} = C_{10} \frac{1 + \dfrac{C_{1\infty}}{C_{10}} \omega^2 \vartheta^2}{1 + \omega^2 \vartheta^2} = C_{10} F, \qquad (8)$$

$$\frac{1}{R_1} = \frac{1}{R_{\text{ohm}}} + \frac{(C_{10} - C_{1\infty}) \omega^2 \vartheta^2}{1 + \omega^2 \vartheta^2}. \qquad (9)$$

Function F is defined as

$$F = \frac{1 + \dfrac{C_{1\infty}}{C_{10}} \omega^2 \vartheta^2}{1 + \omega^2 \vartheta^2}. \qquad (10)$$

Symbols C_{10} and $C_{1\infty}$ equal the value of capacity of AlN at very low and very high frequencies and ϑ is a time constant that determines the relaxation processes in AlN itself.

In the equation (9) the first term on the right side means the ohmic conductivity and the other term the contribution of the relaxation processes to the conductivity.

Inserting (8) and (9) into (4) and (6) we get

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$$C = \frac{C_2}{1 + \dfrac{C_2}{C_{10}F}}\left[1 + \frac{C_2}{C_{10}F}\cfrac{1}{1 + \omega^2\tau^2\left(\cfrac{1}{1 + \dfrac{C_{1\infty}}{C_2}}\cfrac{1 + \dfrac{C_{10}}{C_2}F}{1 + R_{\mathrm{ohm}}C_{10}G}\right)^2}\right] \quad (11)$$

$$\operatorname{tg}\delta = \frac{\omega\tau\;\cfrac{1}{1 + \dfrac{C_{1\infty}}{C_2}}\cfrac{1}{1 + R_{\mathrm{ohm}}C_{10}G}}{1 + \omega^2\tau^2\left(\dfrac{C_{10}}{C_{1\infty}}\right)^2\cfrac{1}{1 + \dfrac{C_2}{C_{1\infty}}}\left(1 + \dfrac{C_2}{C_{10}F}\right)\left(\dfrac{F}{1 + R_{\mathrm{ohm}}\;C_{10}G}\right)^2} \quad (12)$$

where function G is defined by

$$G = \left(1 - \frac{C_{1\infty}}{C_{10}}\right)\frac{\omega^2\vartheta}{1 + \omega^2\vartheta^2}\;.$$

At very low frequencies $C \to C_2$ and $\operatorname{tg}\delta \to 0$, at very high frequencies $C \to \dfrac{C_2 C_{1\infty}}{C_2 + C_{1\infty}}$ and $\operatorname{tg}\delta \to 0$. At intermediate frequencies the time constant τ in the relations for C and $\operatorname{tg}\delta$ is multiplied by a factor containing the frequency ω (through the functions F and G) so that the measured dispersion curves are flatter than the ideal Debye curves.

3. Conclusion

Fitting of the experimental curves to the relations [11] and [12] enables to pick out the intrinsic dispersion curves [8] and [9]. A particular attention to the comparison of the experimental data and theoretical formulae will be paid in an other paper.

Acknowledgment

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