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Investigations of the nonhomogeneities in organic molecular crystalline materials using Kubelka-Munk theory

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This paper presents a study of the nonhomogeneities in doped meta-dinitrobenzene (m-DNB) crystalline films. These growth defects lying in the film plane with dimensions smaller than the films thickness are correlated with the effect of the dopant on the crystallization regime in the dendritic growth conditions. To evaluate the dimension of the growth nonhomogeneities we have used a model based on the Kubelka-Munk theory, emphasising the general constraints imposed to the sample and the experimental set-up, and developing a more complex model through a step-by-step relaxation of these constraints, that ensures a better approximation for our experimental configuration. We have deduced higher scattering coefficients in m-DNB doped with 1,3 dihydroxybenzene (resorcinol) than in m-DNB doped with 8-hydroxyquinoline (oxine) films prepared by a rapid thermal solidification process. We also have emphasised that for the films of m-DNB doped with oxine the size of the scattering centers is larger than the wavelength of the radiation and in films of m-DNB doped with resorcinol the radiation scattering corresponds to a multiple scattering process leading to the superposition of different scattering mechanisms on centers with different dimensions.

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1. Introduction

In the field of functional materials, organic crystals are important because of the wide variety of organic molecular structures obtained by molecular engineering and of the possibility to develop one or more specific properties for the crystalline material by crystal engineering. The interest for organic molecular crystalline materials derives from the numerous potential optical [1-3], electronic [4-7], and electro-optical [8-11] applications. The performances of the organic devices depend of the material quality determined by defects.

The investigation of the growth process of the crystalline organic material is important because there is a strong relation between the crystal growth process, defined by different experimental conditions, that induces defects, as compositional nonhomogeneities, determining the crystalline quality and the physical properties of these materials.

The behaviour of a material from the point of view of the optical properties is strongly influenced by the radiation absorption and scattering phenomena determined by the crystalline material quality as quantified by a good uniformity at the microscopic level in terms of composition and/or structure. Crystalline imperfections, structural defects, compositional inhomogeneities works as scattering and absorption centers, which affect the properties of the material.

Diffuse Reflectance Spectroscopy is a method for the investigation of the quality of the sample, but a special attention must be paid to the theoretical limitations imposed to the system under investigation to obtain a correct qualitative and quantitative interpretation of the experimental data [12, 13]. This method can offer information on the absorption properties of bulk and thin films samples [14] and also on the radiation multiple scattering process in strongly absorbing systems.

This paper presents an UV-VIS Reflectance Spectroscopy study of the non-homogeneity micro-defects in thin film samples of m-DNB doped with oxine and resorcinol, whose generation is governed by the dopants and the experimental parameters defining the crystallization regime.

2. Experimental

The samples for investigation were obtained by growing 20 μm organic films between two glass plates by a rapid thermal solidification process [15]. The thickness of the films has been estimated from geometrical considerations. The crystalline pieces from bulk doped m-DNB crystals ($T_{melting}$=89.9 °C) were firstly melted directly between two glass substrates and then rapidly frozen ensuring a high temperature gradient, $\Delta T>50\ ^{o}C$, to counteract the supercooling, which characterizes the organic compounds with large molecules. The films exhibit dendritic structure. Details about the purification and the bulk crystal growth process of pure m-DNB, m-DNB doped with 1 wt % oxine and m-DNB doped with 10 wt % resorcinol have been presented in previous papers [16-18].

These glass/doped m-DNB /glass structures have been studied by UV-VIS Transmission and Reflectance Spectroscopy using the standard transmission and diffuse reflectance sets-up of UV-VIS Spectrophotometer Specord M42. A special 45/0^o configuration was used for direct illumination and detection, respectively. The samples were fixed in the measurement position with a special system offering an ideal black non-reflecting background to eliminate the parameter characterizing the reflectance of the background [19]. We have used a $BaSO_4$ platelet as comparison white standard, but, because its absolute reflectance varies in time, we have utilized a surface–treated “milky-glass” sample as reference to calibrate the apparatus and ensure the reproducibility of the measurements.

The reflectance property in these experimental conditions has been used to calculate the diffuse reflectance for a finite thickness sample of m-DNB doped with oxine or resorcinol and the Kubelka-Munk (K-M) function [14, 20]. By applying the Kubelka-Munk theory to the experimental transmission and reflectance data we have computed two material constants: the scattering coefficient and the absorption coefficient in reflection. There was evaluated the ratio between the absorption coefficient in reflection, respectively transmission, which provided information on the homogeneity of the material.

3. Model

To evaluate the dimensions of the growth (compositional and structural) nonhomogeneities in doped m-DNB film we have proposed a model based on the (K-M) theory taking into account two aspects:

1. the conditions in which the theory can be applied (the limitations of the model);

2. the subsequent developments of the model using successive levels of approximation relaxing the initial limitations and approaching the proposed theoretical system to the real investigated one.

In the Kubelka-Munk theory the sample is diffused irradiated and the backscattered radiation is presumed with an isotropic distribution being ignored any regular reflection. We can consider a single scattering process because the radiation is scattered on randomly distributed particles that are much smaller than the thickness of the sample. In the presence of the nonhomogeneities a reflection also occurs at the boundaries (as in polycrystalline material) and in this case the distribution is not isotropic, but it can be considered isotropic only for a big number of scattering particles (nonhomogeneities) and thick samples. As a consequence, for small scattering centers (particles, nonhomogeneities) in dense packed systems (similar to our samples) we can presume that the scattered radiation has an isotropic distribution as for a single scattering mechanism.

We have studied the transmittance and diffuse reflectance for nonhomogeneous, absorbant m-DNB doped with oxine or resorcinol layers, which present diffuse scattering (K=K(x) and S=S(x)) for a directed illumination [21-23]. The purpose of this paper is to draw the dependence of the scattering coefficient of our organic film that is a characteristic of the material resulting from the preparation conditions, as a function of the wavenumber in different hypotheses that approximate our experimental systems. The proposed successive approximations suggest a model that better fits the experimental configuration.

In the approximation “zero”, doped m-DNB wafer and the measuring configuration are subjected to the following hypotheses: 1. the sample is investigated in 45/0^o configuration (directed incident illumination at an angle of 45^o with the surface normal and detection of the diffused reemitted radiation in the surface normal direction; transmittance is measured perpendicularly to the surface); 2. surface reflection is neglected; 3. the reflected radiation is presumed to be isotropic; 4. single scattering process; 5. the interferences in reflected light inside the thin film are neglected.

In the next step, the first order correction, the above model is complicated applying the following corrections that remove some simplifying hypotheses of the approximation “zero”: 1. the top and back surface reflection contributions (r_1) are taken into account; 2. the layer is of finite thickness; 3. the upper order effects (as the reflection at the internal boundaries) are negligible (r_2=0).

Our experimental configuration can be approximated by a more general system of equations (Ryde-Cooper-Duntley model) for directed illumination and detection with an ideal black non-reflecting background [21-23], whose solution in particular conditions, as diffuse irradiation, can be converted to the Kubelka-Munk solution:

$$\tau = \frac{\left(1-\dot{r}_1\right)\cdot\left(1-\dot{r}_2\right)\cdot T_{32,1}}{\left(1-\dot{r}_2 R_{32,1;0}\right)^2-\dot{r}_2^2 T_{32,1}^2}$$

$$r_0 = r_1 + \left(1-r_1\right)\cdot\left(1-r_2\right)\cdot\frac{R_{32,1;0}\cdot\left(1-r_2 R_{32,1;0}\right)+r_2 T_{32,1}^2}{\left(1-r_2 R_{32,1;0}\right)^2-r_2^2 T_{32,1}^2} \tag{1}$$

where r_1= reflectance of the glass surfaces for an incidence angle α=45^o, $\dot{r}_1$=regulate reflection for an incident parallel radiation perpendicular to the surface and r_2 = reflection at the internal boundaries and on the back surface of the sample are calculated values (as presented bellow), and τ and r_0 are the transmittance and diffuse reflectance values measured experimentally. Solving this system we have obtained the transmittance ($T_{32,1}$) and diffuse reflectance with an ideal black non-reflecting background ($R_{32,1;0}$) for our samples (glass/organic film/glass structures).

In our experimental conditions, for an incidence illumination angle α=45^o and β determined from the refraction law, the regular reflectance of the natural radiation ($r_{1;r}$) at the top surface of the structure, interface 1 (Fig. 1), given by the formula:

$$r_{1;r} = \frac{1}{2}\cdot\left[\frac{\sin^2(\alpha-\beta)}{\sin^2(\alpha+\beta)}+\frac{tg^2(\alpha-\beta)}{tg^2(\alpha+\beta)}\right] \text{ [23]} \tag{2}$$

has the value $r_{1;r}$=5.3 %, and the evaluated total reflection coefficient [19] at the back (glass/air) surface is $r_{1;t}$=61 %.

The regulate reflections for perpendicular illumination for each of the glass surface characterised by n=1.52, has been evaluated to $\dot{r}_{1;1}$=4.25 % using the relation:

$$\dot{r}_{1;1} = \left(\frac{n-1}{n+1}\right)^2 \tag{3}$$

Because r_1=66.3 % and $\dot{r}_1 = 8.5\%$ it is obvious we cannot neglect the effect of the first order correction introduced by the surface reflection.

In the first order correction (r_2) is neglected and the values of the diffuse reflectance and transmittance coefficient (resulted from Eqs. (1)) for the investigated structure are:

$$R_{32,1;0} = \frac{r_0 - r_1}{1 - r_1} \tag{4}$$

and

$$T_{32,1} = \frac{\tau}{1 - \dot{r}_1} \tag{5}$$

where reflectance (r_0) has been measured for direct illumination at 45°, transmission (τ) has been measured for direct perpendicular illumination of the sample, and r_1 and $\dot{r}_1$ are the values calculated above.

The next development of the model, second order correction, was to introduce the "internal scattering" for direct illumination at an angle of 45°, because the radiation is scattered at the interfaces between the layers. Within the sample the radiation can be theoretically partially totally reflected emerging from the more dense medium m-DNB (n=1.625) or glass (n=1.52) to the less dense medium glass (n=1.52) respectively air (n=1). At the m-DNB-glass interface, the condition associated with the critical-angle is not satisfied, so the radiation is not totally reflected. The sum of the directed or diffuse regular reflection at each interface (air-glass, glass-m-DNB, m-DNB-glass, glass-air) has at direct 45° illumination a total value r_2=2 % and cannot be neglected in Eqs. (1). The regular component of the reflection at direct illumination for interface 2 (glass/m-DNB) was calculated with Eq. (2) for the angles β and γ' (Fig. 1) and at diffuse illumination for interface 3 (m-DNB/glass) for the angles γ and δ (Fig. 1) with the Eq. (6) and Eq. (7):

$$R_{reg(dif)} = 2 \cdot \int_0^{critical-angle} \sin(\gamma) \cdot \cos(\gamma) \cdot f(n,\gamma,\delta)\, d\gamma \tag{6}$$

where

$$f(n,\gamma,\delta) = \frac{1}{2} \cdot \left[\frac{\sin^2(\gamma-\delta)}{\sin^2(\gamma+\delta)} + \frac{tg^2(\gamma-\delta)}{tg^2(\gamma+\delta)}\right] \tag{7}$$

$n_{glass}\sin\beta = n_{org\ film}\sin\gamma'$, $n_{org.film}\sin\gamma = n_{glass}\sin\delta$, β and γ, and γ' and δ, are the incidence angles and respectively the refraction angles at the glass/organic film, organic film/glass interfaces and the refraction indices of the materials are n_{glass}=1.52, $n_{org.film}$=1.625, n_{air}=1.

For perpendicular illumination the internal reflection composed only by the regular reflectance at direct illumination for the glass/organic film, organic film/glass can be evaluated with the Eq. (3) at $\dot{r}_2$ =0.22 % and can be neglected in Eqs. (1).

To evaluate the transmittance (T) and reflectance (R) for a system formed from three homogeneous layers glass (3)/organic film (2)/glass (1) having an ideal black non-reflecting background, we have firstly studied the structure glass (3)/organic film (2), characterized by

$$T_{3,2} = \frac{T_3 \cdot T_2}{1 - R_3 \cdot R_2}$$

and

$$R_{3,2} = R_3 + \frac{T_3^2 \cdot R_2}{1 - R_3 \cdot R_2} \tag{8}$$

and the result has been combined with the values T_1 and R_1 for the glass layer (1):

$$T_{32,1} = \frac{T_{32} \cdot T_1}{1 - R_{32} \cdot R_1}$$

$$R_{32,1} = R_{32} + \frac{T_{32}^2 \cdot R_1}{1 - R_{32} \cdot R_1} \tag{9}$$

where the layers "1" and "3" are identical and homogeneous glasses.

In the first step we have considered the particular structure glass/air/glass considering all the layers homogeneous. Because T_2=T_A=1 and R_2=R_A=0, where T_A and R_A are the characteristics of the air, we have obtained

$$T_{3A,1} = \frac{T_G^2}{1 - R_G^2} \text{ and } R_{3A,1} = R_G + \frac{R_G \cdot T_G^2}{1 - R_G^2} \tag{10}$$

where $T_{3A,1}$ and $R_{3A,1}$ are measures determined experimentally, and

$$R_G = \frac{R_{3A,1}}{1 + T_{3A,1}} \text{ and}$$

$$T_G = \sqrt{T_{3A,1}\left[1 - \left(\frac{R_{3A,1}}{1 + T_{3A,1}}\right)^2\right]} \tag{11}.$$

In the second step we have investigated the system glass/nonhomogeneous organic film/glass, using a similar procedure, and we have evaluated the values T_S (transmittance of the organic film) and R_{S0} (coefficient of diffuse reemission of the organic film with an ideal black non-reflecting background) knowing T_G and R_G and considering no difference in transmittance and reflectance coefficients depending of the illumination direction:

$$R_{S\,0} = \frac{\left(-T_{32,1}^2 \cdot R_G + T_{32,1}^2 \cdot R_G^3 + T_G^2 \cdot R_{32,1;0} - T_G^2 \cdot R_G\right)}{\left(-T_{32,1}^2 \cdot R_G^2 + T_{32,1}^2 \cdot R_G^4 - T_{32,1}^2 \cdot R_G^2 \cdot T_G^2 + T_G^2 \cdot R_{32,1;0} \cdot R_G - T_G^2 \cdot R_G^2 + T_G^4\right)}$$

$$T_S = -T_G^2 \cdot T_{32,1} \cdot \frac{\left(-1 + R_{32,1;0} \cdot R_G\right)}{\left(-T_{32,1}^2 \cdot R_G^2 + T_{32,1}^2 \cdot R_G^4 - T_{32,1}^2 \cdot R_G^2 \cdot T_G^2 + T_G^2 \cdot R_{32,1;0} \cdot R_G - T_G^2 \cdot R_G^2 + T_G^4\right)} \tag{12}$$

where T_S and R_{S0} were determined from the corrected values $T_{3,2,1}$ and $R_{3,2,1;0}$ of the experimental data τ and r_0, for the transmission and reflection of the glass/organic film/glass structures and the computed values of the same parameters for glass, T_G and R_G using experimental values for the transmission and reflection of the glass/air/glass structure.

From the expression of transmittance as a function of the organic film of finite thickness (d) and reflectance of a sample of infinite thickness from the same material $R_{S\infty}$:

$$T_S = \frac{\left(1-R_{S\infty}^{2}\right)\cdot e^{-bSd}}{1-R_{S\infty}^{2}\cdot e^{-2bSd}} \quad [19] \qquad (13)$$

we have deduced in a simplified way the scattering power (S·d) from the experimental data. To avoid the graphical evaluation of the Kubelka-Munk equations we have used the hyperbolic functions and a logarithmic expression

$$Sd = \left(\frac{1}{2b}\right)\cdot \ln\frac{1-a\cdot R_{S0}+b\cdot R_{S0}}{1-a\cdot R_{S0}-b\cdot R_{S0}} \qquad (14)$$

where T_S=transmittance and R_{S0}=diffuse reflectance of the organic layer of finite thickness, d, with an ideal black non-reflecting background and are determined with equation (12) from experimental data.

4. Results

In our experimental conditions the values determined from optical measurements are transmittance coefficient (T_S) and diffuse reflectance coefficient (R_{S0}) for the organic layer and, a and b are defined from the Kubelka-Munk theory as:

$$a = \frac{1+R_{S0}^{2}-T_S^{2}}{2R_{S0}^{2}} = \frac{1}{2}\cdot\left(\frac{1}{R_{S\infty}}+R_{S\infty}\right) \text{ and}$$

$$b=\sqrt{a^2-1} \quad [19] \qquad (15)$$

The experimental data were used to estimate $R_{S\infty}$ (the diffuse reflectance of an layer of infinite thickness from the same material) and the Kubelka-Munk function, $F\left(R_{S\infty}\right)\equiv\frac{\left(1-R_{S\infty}\right)^2}{2R_{S\infty}}$, which are tabulated as a function of the wavelength [19] and are necessary to evaluate the absorption coefficient in reflection $K_R = F\left(R_\infty\right)\cdot S$.

In the simplifying conditions mentioned above we can apply the Kubelka-Munk theory to calculated a and b. S·d, r_0 and τ are determined experimentally and r_1, r_2, $\dot{r}_1$ and $\dot{r}_2$ are calculated as presented above.

In agreement with the Rayleigh theory for single scattering process on small particles characterised by $(1/\lambda^{\alpha})$ with α=4 dependence between the scattering coefficient and the wavenumber, it is expected a dependence of the scattering coefficient of the third power of the particle dimension (S~d^3).

In our case $S=(1/\lambda^{\alpha})$ where $\alpha<4$ is the slope of the curve and offers a qualitative measure of the scattering process, being correlated with the dimension of the particles. When the particle dimension decreases the power coefficient is increasing but it will never attend the value α=4 corresponding to a single scattering process on small particles for which the Rayleigh law can be used. When slope of the curve (α) is decreasing $\alpha \ll 4$ the scattering process can be associated with an increased dimension of the particles. For each sample the measured reflectance has been divided with the reference reflectance. The logarithmic plot of the reference sample reflectance ($BaSO_4$/"milky" sample) versus the wavenumber shows four regions with different slopes to characterize the scattering mechanism in $BaSO_4$ compared to the "milky" sample (Fig. 2).

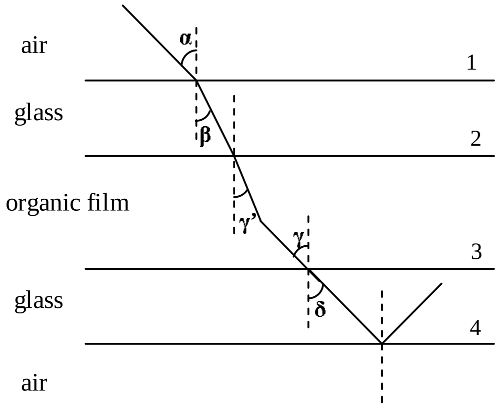

Fig. 1. Experimental configuration.

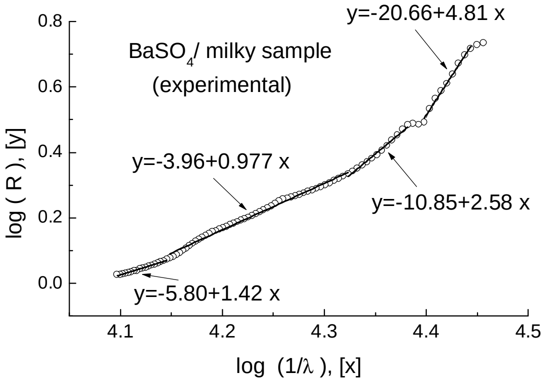

Fig. 2. Logarithmic dependence of the reference reflectance (Ba_2SO_4 standard compared with „milky" sample) of the wave number.

For each sample (thin organic film) and each spectral range, the reflectance deduced from experimental data has been divided to the reference reflectance and we have obtained for the power coefficient describing the scattering mechanism using the Rayleigh formula the expression: $\alpha_{\exp} = \alpha_S - \alpha_{ref}$, that compares the slopes of the sample and reference curves. The dependence log(S)=f(wavenumber) is presented for m-DNB doped with oxine (Fig. 3) and m-DNB doped with resorcinol (Fig. 4).

The scattering coefficients values were greater for m-DNB doped with resorcinol (200 cm^{-1}) compared to m-DNB doped with oxine (~100 cm^{-1}).

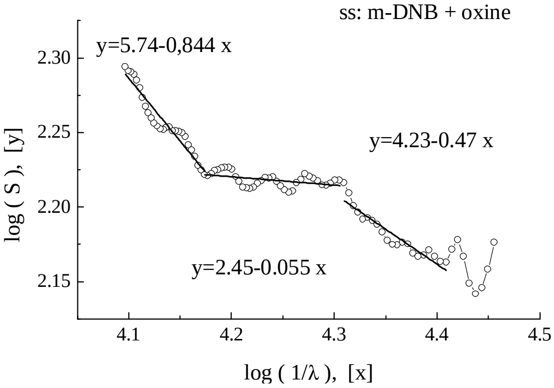

Fig. 3. Logarithmic plots of the scattering coefficients (S) as a function of the wave number in the approximation of a m-DNB doped with 1 % oxine film with finite thickness (20 μm) and r_2 correction.

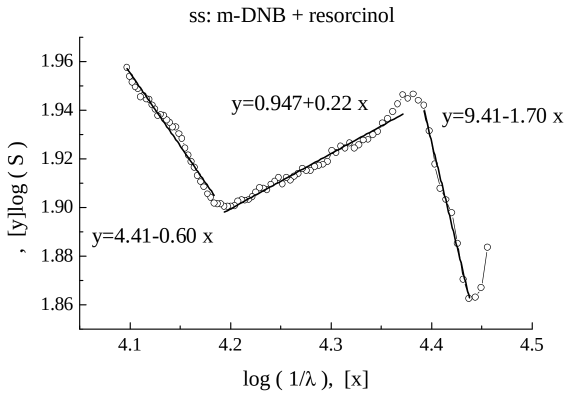

Fig. 4. Logarithmic plots of the scattering coefficients (S) as a function of the wave number in the approximation of a m-DNB doped with 10 % resorcinol film with finite thickness (20 μm) and r_2 correction.

For m-DNB doped with oxine we have obtained for α_s values situated between 0 and 1 sustaining a scattering process on nonhomogeneities with higher dimension than the wavelength and for m-DNB doped with resorcinol the values for α_s suggesting the presence of different scattering centers leading to the superposition of many scattering mechanisms (Table 1).

Table 1. Scattering mechanisms in films of m-DNB doped with oxine respectively resorcinol.

Sample	Spectral range λ (nm)	Slope	Spectral range λ (nm)	Slope	Nonhomogeneities sizes (nm)
m-DNB+oxine	470-500	$0<\alpha_s<1$	670-800	$0<\alpha_s<1$	>λ
m-DNB+resorcinol	300-470	$2<\alpha_s<4$	600-800	$0<\alpha_s<1$	multiple scattering centers

We have also calculated the ratio, r, between the absorption coefficient in reflection (K_R) and transmission (K_T), which offers information about the homogeneity of the material. K_T and K_R are proportional but not identical. This ratio has a theoretical value of 2 [19] for a homogeneous medium, because the absorption and scattering coefficients are not function of the position and the radiation distribution is isotropic. For r<2 it is suppose that the material is more dense in transmission than in reflection absorbing more in transmission than in reflection. For r>2 the radiation distribution become non isotropic because of the modifications in the real path of the wave determined by structural (crystallites boundaries) or compositional nonhomogeneities (zone with high values for the absorption coefficient is alternating with zones with low values).

For all the samples the scattering power (S·d) have a linear dependence of (R_{S0}) the reflectance of the same sample measured with an ideal black non-reflecting background (Fig. 5, 6), as was previously anticipated for non-absorbing materials [19]. This conclusion is in concordance with our results, r<<1, the investigated samples having a greater absorption coefficient in transmission (is less densely packed in reflection than in transmission).

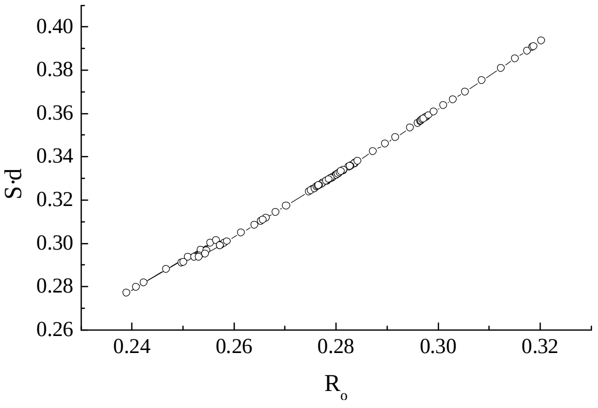

Fig. 5. The dependence of the scattering power (S·d) of R_{S0} (diffuse reflectance of the sample with an ideal black non-reflecting background) for 20 μm film of m-DNB doped with oxine between two plan parallel substrates of glass.

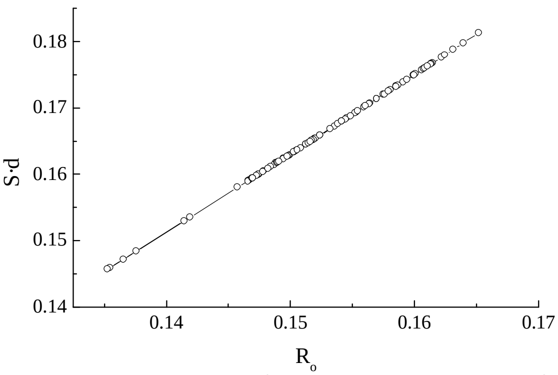

Fig. 6. The dependence of the scattering power (S·d) of R_{S0} (diffuse reflectance of the sample with an ideal black non-reflecting background for 20 μm film of m-DNB doped with resorcinol between two plan parallel substrates of glass.

The investigated material has a structure characterised by the presence of nonhomogeneities in the film plane with dimensions much lower than the film thickness (20 μm), and these defects are generated by the preparation conditions. Only in the case of a high number of particles or nonhomogeneities and thick samples (>10 μm), the scattered radiation inside the sample can have an isotropic distribution and the Kubelka-Munk theory can be applied.

5. Conclusions

This paper present a development of the model based on the Kubelka-Munk two constant theory, considering some higher order corrections as the surface reflection and the reflection at the internal interfaces of the structure to calculate the scattering power of some glass/ doped m-DNB/glass samples. The viability and limitations of the model have been experimentally tested through the evaluation of the homogeneity of the organic films.

Only for high number of scattering centers and thick samples an isotropic distribution of the scattered radiation can be achieved and the Kubelka-Munk theory can be applied for the evaluation of the dimension of the nonhomogeneities. The scattering coefficient is higher in m-DNB doped with resorcinol ($S=200\ cm^{-1}$) compared to m-DNB doped with oxine ($S\sim100\ cm^{-1}$) and the values obtained for the slope of the $\log(S)=f(1/\lambda)$ curve are suggesting in m-DNB doped with resorcinol the superposition of different mechanism for scattering ($0<\alpha_S<1$ and $2<\alpha_S<4$), correlated with the generation, during the preparation of the film, of different scattering centers as a result of an non-rigorous control of the rapid solidification process.

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References

[1] Ch. Bosshard, K. Sutter, Ph. Prętre, J. Hullinger, M. Flôrsheimer, P. Kaatz, P. Günter, Organic Nonlinear Optical Materials" in "Advances in Nonlinear Optics, vol. 1, Gordon Breach Publishers, Switzerland (1995).
[2] J. H. Schön, Ch. Kloc, A. Dodabalapur, B. Batlogg, Science **288**, 2338 (2000).
[3] J. H. Schön, Ch. Kloc, A. Dodabalapur, B. Batlogg, Science **289**, 599 (2000).
[4] H. Klauk, D. J. Gundlach, S. F. Nelson, T. N. Jackson, IEEE Trans. Electron Devices **46**, 1258 (1999).
[5] G. Horowitz, R. Hajlaoui, R. Bourgouiga, M. Hajlaoui, Synth. Met. **101**, 401 (1999).
[6] L. Torsi, A. Dodabalapur, L. Sabbatini, P. Zambonoin, Sens. And Act. **B 67**, 312 (2000).
[7] S. E. Shaheen, C. J. Brabec, N. S. Sariciftci, F. Padinger, T. Fromherz, J. C. Hummelen, Appl. Phys. Lett. **78**, 841 (2001).
[8] R. Fried, J. Burroughes, T. Shimoda. Phys. Word **12**, 35 (1999).
[9] S. Forrest, P. Burrows, M. Thompson, IEEE Spectr. **37**, 29 (2000).
[10] G. Yu, Y. Cao, J. Wang, J. McElvain, A. J. Heeger, Synth. Met. **102**, 904 (1999).
[11] P. Peumans, V. Bulovic, S. R. Forrest, Appl. Phys. Lett. **76**, 3855 (2000).
[12] M. Milosevic, S. L. Birets, Appl. Spectrosc. Rev. **37**, 347 (2002).
[13] R. Nagarajan, D. Gupta, Infrared Phys. Tech. **43**, 377 (2002).
[14] D. J. Dahm, K. D. Dahm, K. H. Norris, J. Near Infrared Spectrosc. **10**, 1 (2002).
[15] J. Kommandeur, Physics and chemistry of the organic solid state, Eds. D. Fox, M. M. Labes and A. Weissberger, Interscience Publishers, vol. **II**, 1 (1965).
[16] A. Stanculescu, A. Popina, Proceedings SPIE, **2700**, 93 (1996).
[17] A. Stanculescu, F. Stanculescu, H. Alexandru, J. Cryst. Growth, **198/199,** 572 (1999).
[18] A. Stanculescu, F. Stanculescu, J. Optoelectron. Adv. Mater., **2**, 536 (2000).
[19] G. Kortüm, Reflectance Spectroscopy: Principle, Methods, Applications, Springer-Verlag (1969).
[20] P. Kubelka, F. Munk, Z. Tech. Physik **12**, 593 (1931).
[21] J. W. Ryde, Proc. Roy. Soc. London **A131**, 451 (1931).
[22] B. S. Cooper, Proc. Roy. Soc. London **A131**, 464 (1931).
[23] S. Q. Duntley, J. Opt. Soc. Am. **32**, 61 (1942).

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