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Publisher: Taylor & Francis

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## Molecular Crystals and Liquid Crystals Science and Technology. Section A. Molecular Crystals and Liquid Crystals

Publication details, including instructions for authors and subscription information:

<http://www.tandfonline.com/loi/gmcl19>

### Enhanced Back Scattering of Light in Polycrystalline Organic Films

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Version of record first published: 04 Oct 2006.

To cite this article: Jan Godlewski, Jan Kalinowski, Sergio Stizza & Ivab Davoli (1992): Enhanced Back Scattering of Light in Polycrystalline Organic Films, Molecular Crystals and Liquid Crystals Science and Technology. Section A. Molecular Crystals and Liquid Crystals, 218:1, 159-164

To link to this article: <http://dx.doi.org/10.1080/10587259208047033>

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## ENHANCED BACK SCATTERING OF LIGHT IN POLYCRYSTALLINE ORGANIC FILMS

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**Abstract** The phenomenon of enhanced back scattering of light due to constructive interference effects has been, for the first time, observed in polycrystalline films of organic compounds. The results obtained on tetracene films are explained in terms of the isotropic theory of multiple (elastic) scattering of waves, assuming that microcrystallites of the tetracene layers form discrete individual scattering centres. From the exact isotropic light-scattering theory based on diffusion approximation, and the resulting angular-dependent enhanced back scattering intensity, the full width at half maximum of the back scattering cone could be related to the transport mean free path and conclusions concerning the film morphology drawn on its basis.

### INTRODUCTION

The phenomenon of enhanced back scattering (EBS) of light from random medium due to constructive interference effects has been recently a subject of a great deal of attention.<sup>1-12</sup> The effect known as weak localization of light is pointed out to correspond to Anderson localization of electrons.<sup>1-4,7,13-17</sup> In all recent experimental

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works<sup>10,12</sup> the random media were intentionally prepared suspensions of dielectric particles in a liquid. Parameters which varied in those studies were concentration, particle size and thickness of the cell. In such a way the theory could be profoundly tested. Much insight in the field of EBS allows at present to undertake search for its new modifications and possible applications in various fields of scientific interest.

In this communication we report on the results of a preliminary study of the weak localization of light in thin layers of vacuum-evaporated organic materials, tetracene ( $C_{12}H_{10}$ ) chosen as an example. Thin films of organic materials evaporated on glassy substrates are generally known to form more or less ordered polycrystalline layers which can be considered as finite slabs of the medium composed of microcrystallites standing for light scattering centres. We have chosen tetracene (Tc) films because they were widely studied with respect to their structural,<sup>18,19</sup> photoelectrical<sup>20</sup> and optical<sup>18,21</sup> properties, showing highly oriented polycrystalline structure<sup>18</sup> and asymmetry of photoelectrical<sup>20</sup> and optical<sup>21</sup> features dependent on whether their substrate (S) or non-substrate (NS) surfaces were illuminated. These effects should be correlated with film morphology and, in particular, with microcrystallites shape and dimensions, the latter being determined by the conditions of film preparation such as the evaporation rate or the surface mobility of the evaporated molecules on the substrate. The total enhancement factor and shape of EBS cones may then be employed as a new tool to characterize the film structure.

## RESULTS AND DISCUSSION

With our experimental set up similar to other experimental arrangements<sup>5,12</sup> scans have been performed perpendicular to the direction of the incoming polarization of the scattered light, alternatively placing a parallel or crossed polarizer in front of the detector. Resolution was about 4 mrad. Two sets of experimental data for tetracene samples are shown in Fig.1. The results can be explained in terms

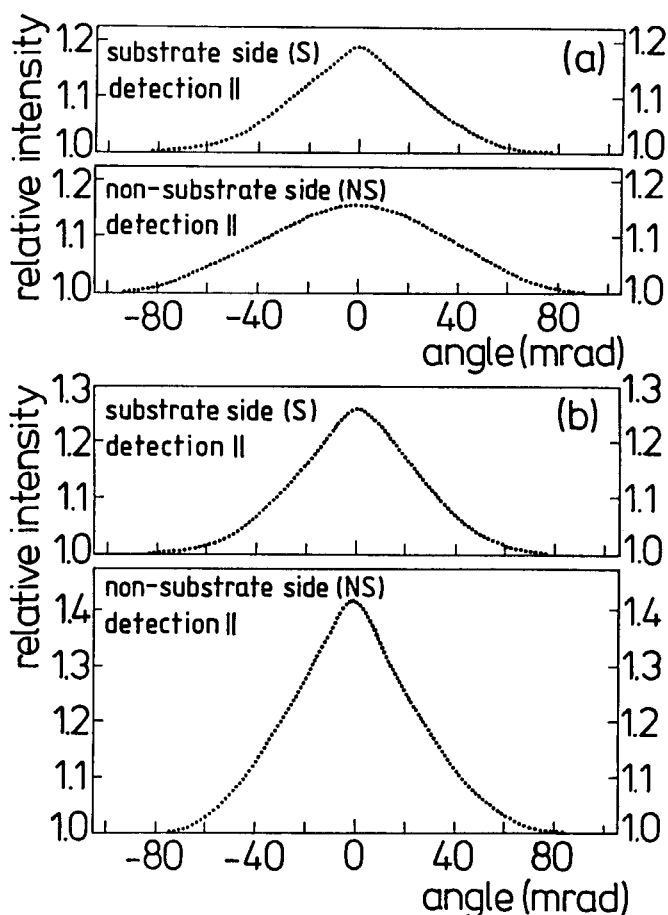


FIGURE 1 Enhanced backscattering of the parallel light component from Tc films deposited by vacuum-evaporation onto the substrate of a  $150\mu\text{m}$ -thick glass slide as a function of the scattering angle (Total Backscattering Cones - abbr. TBC). (a) TBC from a Tc film of thickness  $d = 3.0\ \mu\text{m}$ , and (b) TBC from a Tc film of thickness  $d = 11.0\ \mu\text{m}$ . In the both cases upper curve obtained with the light entering the tetracene slab through the substrate side (S) of the sample and lower one with the light penetrating the tetracene slab from the non-substrate side (NS) of the sample. All the curves obtained by averaging the data for many runs with the angle accuracy  $\pm 3\%$ .

of the isotropic theory of multiple (elastic) scattering of waves,<sup>12</sup> assuming that microcrystallites of tetracene layers form discrete individual scattering centres. From the exact isotropic

light-scattering theory based on a diffusion approximation, and the resulting angular-dependent enhanced backscattering intensity, the full width at half maximum (FWHM) of the backscattering cone can be related to the transport mean free path ( $\lambda_{tr}$ ) as follows<sup>1,2</sup>

$$FWHM \approx \frac{0.7}{2\pi} \frac{\lambda}{\lambda_{tr}}, \tag{1}$$

where  $\lambda$  is the wavelength of the scattered light component polarized parallel to the incident beam and  $\lambda_{tr}$  is the transport mean free path (to be distinguished from the scattering mean free path  $\lambda_{sc}$  defined as the reciprocal of turbidity). In Table I we present part of our results comprising FWHM,  $\lambda_{tr}$ , and enhancement factors for a number of Tc layers of different thicknesses with light entering the samples

TABLE I Experimental values for width (FWHM) and enhancement factors, and transport free paths determined from the exact isotropic theory of multiple scattering of waves<sup>1,2</sup> according to (1) for different Tc samples. All the parameters have been obtained for light entering the sample from substrate (S) and non-substrate (NS) side at  $\lambda_{vac} = 632.8\text{nm}$ .

Sample thickness d ( $\mu\text{m}$ )	Enhancement		FWHM(mrad)		$\lambda_{tr}(\mu\text{m})$	
	S side	NS side	S side	NS side	S side	NS side
3.0	1.19	1.15	55	80	1.26	0.87
3.2	1.23	1.46	66	67	1.05	1.03
8.0	1.34	1.26	47	64	1.47	1.08
11.0	1.26	1.42	54	54	1.28	1.28
Average	1.26 $\pm 0.03$	1.33 $\pm 0.03$	58	70	1.27	1.07

from two opposite sides. Enhancement factors are defined as the ratio of total back-scattering at exactly  $180^\circ$  and background. The samples were studied over a total scan width up to  $120\text{mrad}$  so that all the experimental curves were corrected for the angular dependence of the response. For each sample, the recorded enhancement factor depends on the relative orientation of the front-detector polarizer. Only the parallel light component was recorded since the applied isotropic theory

is expected to hold in this case. In addition, the low values of the enhancement factor and the broad cone width made the results for the perpendicular component highly unreliable.

Upon going from thin to thick samples (slabs) (i) the parallel enhancement factor increases reflecting an increasing value of the bistatic scattering coefficient ( $\gamma$ ) for the interference terms which initially increases with the optical thickness  $b = d/\lambda_{tr}$  and for  $b > 10$  saturates independent of albedo.<sup>12</sup> It is seen that in our case for the thinnest sample slab  $d = 3\mu\text{m}$ ,  $b \ll 10$ , and its enhancement factor seems to be diminished the most. (ii) the parallel enhancement factors, for the samples with  $d > 3\mu\text{m}$  and  $b$  approaching 10 (or  $b > 10$ ), do not show any regular characteristic behaviour suggesting the differences in albedo (note that only 1% decrease in albedo leads to over 25% decrease in  $\gamma$ <sup>12</sup>). This effect which has seemed to be ignored up to date experiments on modelling random media (albedo  $a = 1$  has been strictly assumed) can in fact be of essential importance for the enhancement factor due to light absorption by residual impurities in the components forming the testing media. (iii) FWHM's and corresponding light paths (expressed by transport free paths  $\lambda_{tr}$ ) obtained for the incident light entering the Tc slabs through the S side are not larger and smaller, respectively, than those obtained for the incident light entering the slabs from the NS side. In average  $[\lambda_{tr}(S) - \lambda_{tr}(NS)]/\lambda_{tr(NS)} \approx 20\%$ . This asymmetry in the enhanced back scattering behaviour corresponds to the asymmetry of photoelectrical and optical properties of vacuum-evaporated layers of tetracene. By their comparison we can relate the low-reflectivity broad spectra originated from high-roughness NS surfaces with lower values of transport paths and, on the other hand, the thickness-independent moderate reflectivity spectra from the smooth S surface with larger values of free paths (cf. Ref. 21). The transport free path is then correlated with type of defects created at S and NS sides of the tetracene layers: the NS side defects relatively deeply trapping charge carriers and excitons but of weak gradient distributions in space and energy occur at the short transport free path, and the S

side higher concentration defects forming rather shallow charge carrier and exciton traps with stronger distribution gradients accompany the long transport free path of the film.<sup>20</sup>

## CONCLUSIONS

In summary we conclude that enhanced back scattering can be observed in vacuum-evaporated layers of organic materials, the effect being dependent on the samples morphology. An asymmetry in the enhanced back scattering from substrate and non-substrate side of the layers corresponds to asymmetries in optical and photoelectrical properties of such layers, and can be used as a new feature characterizing properties of organic polycrystalline materials.

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