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ELECTROMODULATION OF LIGHT SCATTERING IN SINGLE CRYSTALS OF ANTHRACENE*

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Abstract Modulation of light scattering by electric field in anthracene crystals is reported. Scattered light was that obtained from a He-Ne laser. Effective modulation of scattered light was possible only when concentration of trapped charge carriers was changed by an external electric field under space-charge-limited conditions. External voltage characteristics of electromodulation signal were structured, the signal height was up to ten percent. The observed effect is discussed in terms of the modulation of the population of the filled up charge trapping domains responsible for the light scattering. The change in the local complex refractive index due to the charge captured by a spatially extended defect (macrotrap) is considered as the most probable microscopic mechanism of the effect.

INTRODUCTION

Scattering of light occurs whenever a change in refractive index or absorption coefficient of the medium is encountered¹. In real organic crystals such a change arises on a variety of chemical and physical imperfections which thus act as scattering centres. The scattering properties of these centres are modified when become charged by capture of the charge carriers introduced to/or produced in some way inside of the crystal. Therefore, the charge evolution in the crystal can be followed by observing the pattern of the scattered light, provided the charge concentration is sufficient to bring detectable effects.

The purpose of the present work was to check such a possibility, using a model organic crystal, anthracene, contained between two strongly injecting CuI electrodes.

In the present communication we report the first experimental observation of electric-field modulation of light scattering in this system and suggest a physical explanation for the voltage dependence of the scattering intensity.

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RESULTS AND DISCUSSION

Electromodulation of light scattering from single anthracene crystals was detected after external voltage modulation of charge carriers concentration in the bulk of the crystals. The crystals were illuminated with He-Ne light ($\lambda = 632.8$ nm) and the scattered light was observed at angle 30° relative to the direction of the incident beam. The scattered light was detected by a photomultiplier connected to lock-in amplifier. Steady-state voltage (U_{DC}) and sinusoidal voltage ($U = U_0 \sin(\omega t)$) with $\omega = 1100 \text{ s}^{-1}$ could be applied to the crystal, separately or simultaneously with U_{DC} . Electrical contacts were prepared by vacuum evaporation of Cu or Au on (ab) planes of anthracene crystals and the Cu contacts exposed then to iodine vapour to form CuI electrodes. Thicknesses of the crystal used in the experiment were between $50 \mu\text{m}$ and $100 \mu\text{m}$. For steady-state voltage, crystals with CuI electrodes gave typical space-charge-limited currents.

For the system Au/Anthracene/Au there was practically no modulation of scattered light observed. For the system CuI/Anthracene/CuI, under similar physical conditions, strong electromodulation of scattered light, up to 10%, has been observed. An example is presented in Fig. 1.

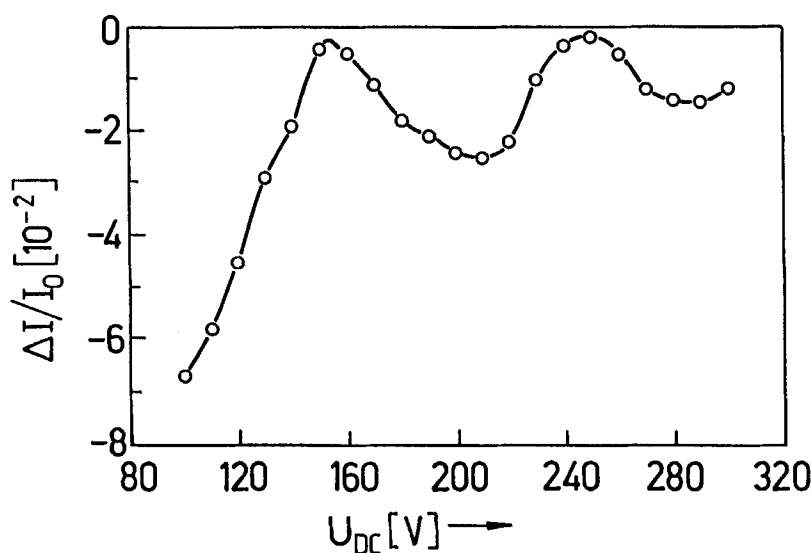


FIGURE 1 Electromodulation of the He-Ne laser light scattered by CuI/Anthracene/CuI system as a function of steady-state voltage, U_{DC} . Amplitude of the modulating voltage $U_0 = 70 \text{ V}$, $\omega = 1100 \text{ s}^{-1}$. Thickness of the crystal $d = 60 \mu\text{m}$.

Though a field-induced decrease in the scattered light intensity is seen from the figure, a positive effect could be observed as well. A "quasi-periodic" structure of the electromodulation signal versus U_{DC} and U has been observed for all of the crystals. However, the height and field behaviour of the signal have been dependent of history of the sample.

These general features of the effect suggest that it must be assigned to the presence of localized charge carriers in the crystal. The holes injected from CuI electrodes can modify light scattering properties of the structural defects of real anthracene crystal or lead to a scattering of 632.8 nm light by ionic radicals of anthracene. Such radicals reveal an absorption maximum for the light used in the experiment.³ At present no unambiguous decision between both explanations can be made. However, an argument for the former hypothesis is the appearance of a "quasi-periodicity" in the signal versus applied voltage, the effect similar to cascade-like voltage evolution of the triplet exciton lifetime.⁴ It is unlikely that it has to be associated with field-modulated concentration of mono-positive anthracene radicals, since then the energy separation between consecutive trapping sites falls in the range below kT . Sequential filling of a set of discrete spatially extended trapping domains (macrotraps) by increasing concentration of holes would explain the observed effect. By virtue of quantization of charge, the macrotrap energy takes discrete values reached at suitable concentrations of charge. They correspond to a sequence of discrete values of applied voltage between which the concentration of the trapped charge remains constant⁴ (or become diminished in case of alternating voltage used in the modulation experiment).

In principle, sequential formation of multi-positive anthracene radicals, leading to sequential drops in the scattering efficiency of the He-Ne laser light on mono-positive radicals, cannot be excluded as an alternative explanation. The observation of minima in the evolution of the scattered light intensity is compatible with both alternatives. Though an experiment with light out of remarkable absorption by anthracene radicals would give a conclusive decision regarding the microscopic mechanism of the scattering, formation of multi-positive radicals in anthracene crystal seems to be unlikely and, therefore, sequential filling of macrotraps by injected charge carriers provides the most probable explanation for the observed voltage evolution of light scattering, at present.

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