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Fog Droplets of Highly Absorbing Fluids Showing Unexpected Sign of Acceleration in a Laser Beam

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Highly absorbing fog droplets of a solution of Victoria Blue R in polyethylene glycol are accelerated into the direction of the source of a He-Ne laser. The reverse movement is observed with the more volatile diethylene glycol as a solvent. These qualitative results are discussed on the basis of known effects of photophoresis and on the hypothetical assumption of forces bound up with the conservation of photonic momentum in solutions of dyes, opening a link to the enhanced optical Kerr-effect in nematics.

Keywords: photophoresis; dyes; photonic momentum

INTRODUCTION

The conservation of the momentum of a photon being absorbed or scattered by isolated atoms or molecules is fundamental for photonics. However, it is generally accepted that the momentum of a photon absorbed by a dye molecule in a solvent surrounding is distributed randomly after the processes of excitation and nonradiative decay. This is also behind different interpretations of the enhanced optical Kerr-effect[1] which arises when a solution of a dichroic dye in a nematic

phase is exposed to a laser beam. The light-induced alignment can, in principle, be attributed to the stabilisation of the excited state of the dye in the anisotropic host imposing an angular momentum upon the nematic director. Based on the hypothetical principle of the maximum flow of energy[2] the idea came up that nevertheless the conservation of photonic momentum might be responsible for that phenomenon. This was supported by the finding that dichroic dyes forming a hydrogen-bond to the nematogenic molecules of the host showed another marked increase of the effect when hydrogen was replaced by deuterium[3]. The absorbances of the fluids used in all these investigations are very low. It seemed of general interest to see whether this hypothesis held when the absorbed power per volume was increased drastically. Because of the very large effects of thermal diffusion, an optical evaluation in stationary bulks was not considered practical. In the following it is shown that the motions of highly absorbing fog droplets in a laser beam permit an argument for this view, taking into account the well known effects of radiation pressure and radiometric forces.

EXPERIMENTAL PART

All dyes and solvents used in the experiments are commercially available: guaiazulene (1), Victoria Blue R (2), diethylene glycol, polyethylene glycol 400 (PEG400, Sigma-Aldrich) and paraffin oil of viscosity 115 to 135 mPas at 20 °C (Fluka). The solutions of 2 at 20 and 50 °C (see Table 1) were prepared by stirring a suspension of an excess of the dye for 20 hours, centrifugation at the appropriate temperature and

decanting. The values of A_1 (calculated absorbance of a 1 cm thick layer) were determined by diluting a small quantity of the supernatant solution in a volumetric flask and measuring the absorbances in a 1 cm cuvette using a spectrometer (Shimadzu UV-160A) at a wavelength λ of 633 nm. For convenience, in the case of guaiazulene, toluene and of Victoria Blue R, ethanol, respectively were used as diluents. Any effect of the solvent and a possible deviation from the Lambert-Beer law were neglected. The concentrations c (g dye per 100 ml solution) were calculated from the specific absorbances of the dyes. The fogs were generated with a glass sprayer (Rettberg, connected to a pressure bulb) and were directed into a cylindrical glass vessel (diameter 24 cm, height 30 cm) on the bottom of which was placed an open fluorescence cuvette (inner sizes 1 x 1 x 4 cm) on a 5 cm wooden cube. After 10 contractions of the bulb the system was kept for 5 min closed by a lid. The cuvette was then sealed by a stopper, cleaned outside by a piece of cloth and positioned under a microscope equipped with long-distance objectives. In the case of experiment 4 it had to be ensured that no crystallisation of the dye occurred. Before

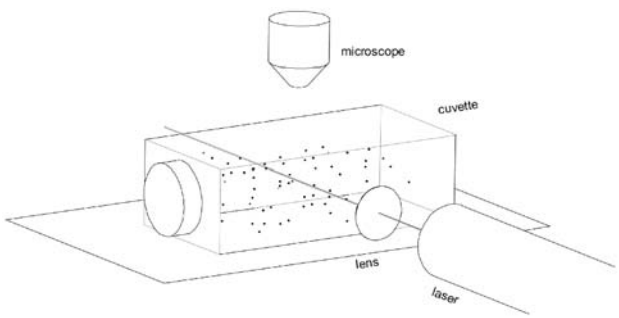

FIGURE 1 Arrangement for the microscopic observation of the movement of fog droplets in the beam of a He-Ne laser

generating the fog the sprayer containing the solution was heated to 100 °C and the air in the vessel warmed up to 70 °C by an electric heater. After cleaning, the cuvette was placed in a flat water basin of 70 °C for microscopy.

Figure 1 shows the arrangement for the observation of the droplets a few of which were stable for several minutes due to convection streams before being precipitated onto the wall of the cuvette. The beam of the He-Ne laser (10 mW, beam diameter 0.68 mm, polarised) was focussed in the visual field of the microscope (magnification 40) by a lens of a focal length of 14 cm. The paths of the droplets could well be followed by the scattered laser light. The diameters of the droplets were estimated

Experiment No.	**Solvent**	**Dye**	**c**	**A_1**	**$1/\alpha$**	**Acceleration**
1	paraffin oil	1	10.0	$2 \cdot 10^2$	22	negative
2	paraffin oil	none	-	-	∞	positive
3	PEG 400	2	5.4[a)]	$6 \cdot 10^3$	0.72	negative
4	PEG 400	2	12.5[b)]	$14 \cdot 10^3$	0.31	negative
5	diethylene glycol	2	10.7[a)]	$12 \cdot 10^3$	0.36	positive

TABLE 1 Signs of acceleration of fog droplets of solutions of dyes 1 and 2 with concentration c and specific absorbance A_1 in the beam of a He-Ne laser; [a)]solution saturated at 20°C, [b)] at 50°C, $1/\alpha$ penetration depth in µm.

roughly as being between 2 and 5 μm by waving a microscope slide in the fog of experiment 3 and inspecting the precipitated flattened droplets under the microscope at a magnification of 350.

The time of the droplets between their appearance in the laser beam and their disappearance at the edge of the visible field (ca. 2,5 mm) were estimated to range from ca. 0.3 to 3 sec. Acceleration was very high in experiment 5, medium in 1, 3 and 4 and low in 2. It did not escape notice that a few droplets in experiments 3 to 5 showed an irregular motion just after having entered the beam before they followed the direction given in Table 1. Because of the great focal length, the qualitative observation of these experiments did not reveal a difference in acceleration before and behind the waist of the beam due to trapping forces.

DISCUSSION

Solid and liquid particles can be moved in liquids, gases and in vacuum by the action of light. Radiation pressure leads to an acceleration away from the light source but also trapping is possible [4]. Radiometric forces, on the other hand, are caused by temperature gradients surrounding the particle, due to absorption of light. Generally, fog droplets of highly absorbing liquids are accelerated away from the light source (positive photophoresis) whereas those of less absorbing liquids show a negative photophoresis due to the fact that light is focussed on their far side[5]. The complex dependence of sign and magnitude of acceleration on radius R of the droplets, absorbance and wavelength of

light λ has been refined by S. Arnold after the observation of droplets of glycerol in the beam of a CO_2-laser of comparatively low photonic momentum content per unit volume and reduced atmospheric pressure[6]. A positive photophoresis can be expected for the product $\alpha R \geq 1$ (bulk absorption coefficient $\alpha = 4\pi n\kappa/\lambda$, with bulk refractive index n, absorption index κ).

The observed motion in experiment 1 seemingly is in accordance with this. The droplets of pure paraffin oil 2 are moved by radiation pressure which can be understood by assuming that under the experimental conditions 10 per cent of the incident light is scattered back[4]. The observed negative sign of acceleration in the cases 3 and 4 does not fit into the given frame: taking α as the inverse of penetration depth (where intensity has decreased to 1/e of its initial value) and assuming a relatively low R of 1 μm one obtains for αR 1.4, 3.2, respectively. An indication that with these high concentrations of dye the front side of the droplets should be hottest, can be found in the positive sign of acceleration in 5, obviously due to the relatively high volatility of the solvent. An argument, apparently not considered in former photophoretic works is the induced rotation of droplets being moved into to the beam by convection. The deviation from known photophoretic behaviour suspected here should be verified by future work monitoring the exact masses of individual fog droplets and the forces exerted on them in a Millikan capacitor[6]. An estimation of the temperature reached on the surface of the droplets at the balance between the high density of heat evolved and the heat transmission cannot be given. For a rough estimate the fact may serve that droplets of the solution of 2 in

diethylene glycol (boiling point 245 °C at normal pressure) had not been evaporated completely during their flight time in the field of view of the microscope. The influence of surface-mode-enhanced local fields[7] found with particles smaller than the wavelength of the laser obviously can be neglected here. Possibly the unusual behaviour discussed here is observable only at high photonic momentum content i.e. high power density of the laser combined with high absorbances and low volatility of the fluid media.

In the following an attempt is made to find an explanation for an overcompensation of radiometric forces based on the idea that initiated the experiments presented here. As a working hypothesis for the correlation of chemical structure and molecular order in fluid systems it was suggested that molecules which are free to move, align in such a way that the sum of any energy E accepted and donated by all individual molecules in an interval of time is maximum whereby expressly elastic collisions in the thermal pool are regarded as being accompanied by an exchange of energy[2] which can be seen as being real or virtual. An exchange can be formulated in the classical[8] view as being one between kinetic and potential energy, as well as in the quantum theoretical[9] view seeing the collision partners united in a wave field as centres of repulsive forces, not as hard spheres. Nevertheless, the treatment of molecular ensembles as a centre-of-mass system, mostly used in physical chemistry, neglects an energy exchange during an elastic collision. The variational principal of the maximum flow of energy the concept of which is outlined elsewhere [2] permits to connect the view favoured here to electrodynamics and quantum-electrodynamcis.

Particular useful was the introduction of an anisotropic "exchange pressure" as an analogy to radiation pressure, comprising all kinds of energy of the thermal pool as well as external electromagnetic energy, which permits one to predict the heat of transition between the nematic and the isotropic state as a function of heat content and volume change[2,10]. In this context it can be seen that in a liquid or a liquid crystalline state the photonic momentum is conserved during the absorption of electromagnetic energy and the subsequent nonradiative decay. This occurs so that, for absorption and any deactivation step of the excited state, an accompanying elastic collision can be postulated in the thermal pool of a liquid with its almost continuous quantum description. The acceleration of the droplets reverse to the direction of photonic propagation may be imagined to be caused by the transition of momenta from the droplets to the gas molecules on their far side comparable to a classic elastic collision between a small (droplet) and a big (atmosphere) mass.

SUMMARY and OUTLOOK

The observed acceleration of highly absorbing fog droplets consisting of Victoria Blue R dissolved in polyethylene glycol in the focussed beam of a He-Ne laser cannot be explained satisfactorily by existing theories of photophoresis. It is suggested that the extension of the validity of the principle of conservation of photonic momentum to elastic collisions within the solution of the dye causes overcompensation of the known radiometric forces exerted onto the droplets. Clarification can be

expected only after quantitative measurements of the acceleration taking into account the exact masses of the droplets. Measurements under normal pressure and in vacuum under microgravitational conditions are especially promising.

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References

[1] I. Jánossy, A. D. Lloyd, B. S. Wherrett, Mol. Cryst. Liq. Cryst. **179**, 1 (1990).
[2] R. Eidenschink, Kontakte (Darmstadt) **1980**(3), 12; **1987**(1), 15; **1994**(1), 49.
[3] M. Kreuzer, F. Hanisch, R. Eidenschink, D. Paparo, L. Marrucci, Phys. Rev. Lett., accepted
[4] A. Ashkin, Phys. Rev. Lett. **24**, 156 (1970).
[5] O. Preining in Aerosol Science, edited by C. N. Davies, Academic, New York, 1966, pp. 111-135.
[6] S. Arnold, M. Lewittes, J. Appl. Phys. **53**, 5314 (1982).
[7] S. Arnold, A. B. Pluchino, K. M. Leung, Phys. Rev. A **29**, 654, (1984).
[8] E. A. Moelwyn-Hughes, Physical Chemistry, 2nd Ed., Pergamon Press, Oxford 1961, p. 325.
[9] S. Chapman, T. G. Cowling, The Mathematical Theory of Non-Uniform Gases, Cambridge at the University Press, 1952, pp. 295.
[10] S. Asahina, M. Sorai, R. Eidenschink, Liq. Cryst. **10**, 675 (1991); **24**, 201 (1998).