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Effect of unmodulated laser light on the nanostructure of a thin solid AD-1 azo dye film

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Abstract. **Exposure to light uniform in intensity and polarisation causes marked changes in the surface topography of a thin (320 nm) nanostructured AD-1 low molecular weight azo dye film. Linearly polarised incoherent light with a wavelength of 470 nm and intensity of 1 mW cm^{-2} produces numerous teardrop-shaped hillocks of the order of 200 nm in radius over most of the film surface.**

Keywords: *nanophotonics, azo dye, mass transport, nanotopography*.

One uncommon manifestation of light – matter interaction is photoinduced molecular mobility in solid polymers and amorphous materials [1]. The physicochemical properties inherent in azo dye molecules ensure them an exceptional photoinduced mobility, which has caused researchers to pay special attention to azo dyes [2]. In particular, they can be used in engineering photoactuated mechanical nanomachines [3].

In recent studies [4 – 6], a new type of thin nanostructured film has been proposed, consisting of a pure AD-1 azo dye [2] and differing in properties from other azo-containing materials. In particular, in studies of the behaviour of such films exposed to laser light, both one- and two-photon scattering processes were found to be highly dependent on the incident light polarisation [5]. This effect may find application in optical memory technology. Jung et al. [4] were the first to demonstrate photoalignment of azo dye molecules under two-photon excitation [6]. In this paper, we report the observation of another manifestation of photoinduced molecular mobility in AD-1 films: photoinduced mass transport that leads to changes in the surface topography of the film.

The film was irradiated using a light-emitting diode (LED) at a wavelength near 470 nm, lying within the absorption range of the dye. The LED beam was collimated to a divergence angle of about 20°. Linearly polarised light was obtained using a thin-film polariser. The beam was normal to the film surface, and the incident light intensity was about 1 mW cm^{-2}. The film surface was imaged by atomic force microscopy (AFM) with a Solver PRO instrument (Russia) in intermittent contact mode using standard silicon cantilever probes (NSG01 Golden series, Russia).

To adequately compare AFM images obtained at different instants in time and determine the film thickness, two intersecting straight cuts were made in the film down to the glass substrate. Uncoated glass areas in AFM images were used to determine and subtract the tilt of the sample. The surface topography of the unexposed film is illustrated by the AFM image in Fig. 1a, where one can see a flat glass substrate and a film about 320 nm in thickness. Preliminary measurements showed that multiple AFM scans had no effect on the surface topography of the film. Prior to illumination of the sample, the cantilever was brought away from the film surface in order not to distort the incident optical field.

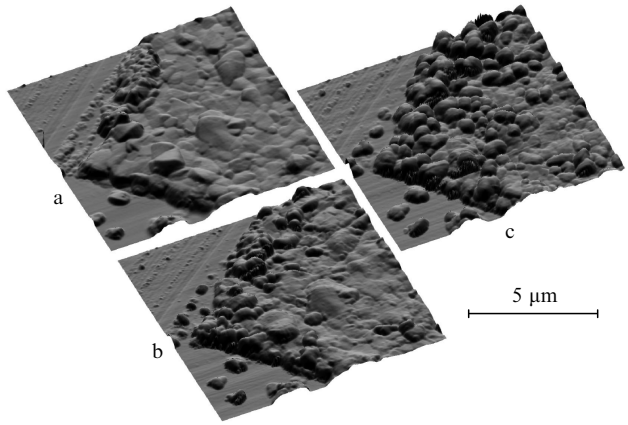

Figure 1. 3D AFM images of the film (scan size, 7 × 7 μm) (a) before exposure, (b) after exposure to linearly polarised light and (c) after exposure to light with an orthogonal polarisation.

Exposure to linearly polarised light for 1 h produced significant changes in the surface topography of the film, as illustrated by Fig. 1b, where teardrop-shaped bumps on the film surface are well seen. Our experiments showed that a nearly steady state was reached within 1 h: the mobile molecules aligned themselves with the normal to the plane of polarisation, and the process stopped. At the same time, when the polarisation direction was changed by 90° and the sample was exposed for another 1 h, more marked changes in surface topography were observed almost throughout the sample surface (Fig. 1c).

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Surface height variations can be quantified by the rms deviation $\sigma_z = (\langle z^2 \rangle - \langle z \rangle^2)^{1/2}$. Prior to exposure, σ_z was 35 nm. After exposure to linearly polarised light for 1 h, it was 41 nm. Subsequent exposure to light with an orthogonal polarisation for another 1 h increased it to 58 nm, which exceeded the initial value by more than a factor of 1.5.

A unique feature of the effect under consideration is that the incident light was not intensity- or polarisation-modulated along the film surface. The observed topography changes cannot be accounted for in any of the theoretical models proposed in the literature [7–10], which assume incident light modulation along the surface to be basic to the process.

The most intriguing feature of the effect is the shape of the growing hillocks. The AFM images in Fig. 1 demonstrate that the unexposed film surface is relatively smooth and that exposure produces bumps, which take the shape of $\sim$ 200-nm-radius hemispherical domes while growing. This is well illustrated by Fig. 2, where a hillock produced on the film surface is shown at a higher magnification.

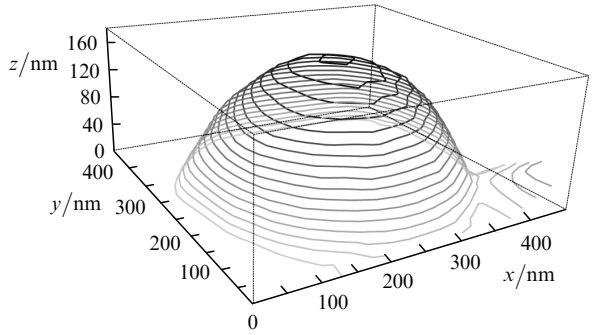

Figure 2. 3D AFM image of a hillock produced on the film surface.

It seems likely that a new mechanism is responsible for the observed photoinduced molecular motion, similar to the formation of liquid droplets on a poorly wettable surface. The effect may find practical application, e.g. as an optical nanojack actuator in nanotechnology. There is every reason to expect that such a nanojack will offer greater forces in comparison with its possible analogues based on other azo materials. This is suggested by the strong interaction between the AD-1 molecules in the nanostructured film and by their high photoinduced mobility demonstrated in our experiments.

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