https://doi.org/10.3176/phys.math.1998.4.07

Proc. Estonian Acad. Sci. Phys. Math., 1998, **47**, 4, 304–307

VISUALIZATION OF THE LASER COLORATION/DISCOLORATION INTENSITY DEPENDENCE IN KBr CRYSTALS

Viktor BICHEVIN and Henn KÄÄMBRE

Institute of Physics, University of Tartu, Riia 142, 51014 Tartu, Estonia; e-mail: henn@fi.tartu.ee

Received 9 April 1998, in revised form 12 May 1998

LASERVÄRVIMISE JA -PLEEGITUSE INTENSIIVSUSSÕLTUVUSE VISUALISEERIMINE KBr KRISTALLIDES. Viktor BITŠEVIN ja Henn KÄÄMBRE

Key words: XeCl excimer laser, crystal coloration, F centres, alkali halides.

In [1] we provided evidence that two-quantum laser coloration of ionic crystals is paralleled by one-quantum laser discoloration of the samples. This was revealed by the investigation of the coloration dynamics at different laser intensities using various uncoloured and preliminarily coloured alkali halide crystals. It turned out that all features of the experimental results could be consistently explained supposing that the two-quantum created excitons and/or electron-hole pairs are partially converted into lattice defects, namely F centres and complementary V centres. Further, it was shown that the discoloration occurs mainly via one-quantum photodissociation of the F-complementary V centres and annihilation of the dissociation products with the F centres. In these experiments we extracted from the laser beam with a small orifice a tiny central fraction to achieve a spatially homogeneous irradiation and to improve the signal-to-noise ratio. It was tempting to visualize the laser coloration/discoloration interplay in a single illustrative experiment. This is possible if one shines a laterally inhomogeneous laser beam (as it really is) onto the surface of a crystal sample.

According to [1], the F centre density n can be expressed as

$$n = N_o \exp(-BJt) + (A/B)\, J\, [1 - \exp(-BJt)] + (n_o - N_o) + (a - A) J^2 t \tag{1}$$

(formula (17) in [1]). Here N_o is the initial density of V centres, A and B are the empirical constants, J means pulse intensity of the laser radiation, t irradiation time, n_o initial density of F centres, and a is an empirical constant. Neglecting the minor terms $(n_o - N_o)$ and $(a - A)J^2t$, accordingly taking $n_o \approx N_o$, we have

$$n = n_o\exp(-BJt) + (A/B)\, J\, [1 - \exp(-BJt)]. \tag{2}$$

Thus, according to our model, a prolonged irradiation ($t >> 1/BJ$) must result in a saturation colour density allocation

$$n_s = n_s(x, y) = (A/B)\, J(x, y), \tag{3}$$

which depends solely on the spatial distribution of laser intensity $J(x, y)$. This holds both for uncoloured and preliminarily coloured samples. From (2)

$$\mathrm{d}n/\mathrm{d}t = BJ\, (n_s - n_o)\, \exp\, (-BJt). \tag{4}$$

Thus $\mathrm{d}n/\mathrm{d}t > 0$ if $n_s > n_o$ and $\mathrm{d}n/\mathrm{d}t < 0$ if $n_s < n_o$. In other words, the laser light induces additional coloration if the laser-induced saturation colour density n_s at a given intensity level J exceeds the preliminarily introduced colour density, and bleaching if it does not reach n_o.

Figure 1 displays a Mathcad-simulated colour map for preliminarily colourless (1*a*) and coloured (1*b*) crystals. The intensity distribution of the laser beam, in accordance with the experiment presented in [2], was assumed to be a Gaussian (see also Fig. 1*c*)

$$J(x, y) = J_o\, \exp\, [-(c_1x^2 + c_2y^2)], \tag{5}$$

where J_o, c_1, and c_2 are constants.

In our experiment we used, as in [1], an ELI-1 excimer laser (XeCl, 308.5 nm, ~20 ns pulses, ~25 mJ in a pulse, 10 Hz repetition rate) elaborated in Estonia. The test objects were KBr platelets cleaved out from the ingots melt-grown at our institute from "specpure" grade raw salt. Preliminary coloration was carried out with a commercial X-ray unit (W anode, 60 kV, 20 mA, 2 cm from the tube window, irradiation doses roughly 2×10^4 Gy).

In Fig. 1*c* the intensity distribution (after [2]) in the ELI-1 laser beam (curve 1) is shown together with the colour distribution (2, dots) for a preliminarily uncoloured sample. The colour profile repeats within the error limits the Gaussian intensity profile, in accordance with our assumptions. The colour profile was measured with a 0.04 mm slit with monochromatic light at the F band maximum (620 nm). A photomultiplier tube FEU-62 was used. Figure 1*d* demonstrates the coloration profile for an X-ray precoloured sample. The case $n_s < n_o$ was chosen. The picture also crudely corresponds to the laser intensity distribution: a more intense colour occurs in the middle of the spot and heavy bleaching at the wings. However, the curve is distinctively flattened as compared to the Gaussian. Obviously, our simple model only roughly reflects the details of the coloration/discoloration process. For example, it does not take into account the progressive in-depth decrease in the laser light intensity in the crystal during irradiation (due to absorption at the V-type centres), diffusion of the mobile quasiparticles, etc. Unfortunately, exact calculations are mathematically rather complicated. Nevertheless, we are just attempting to develop an advanced model.

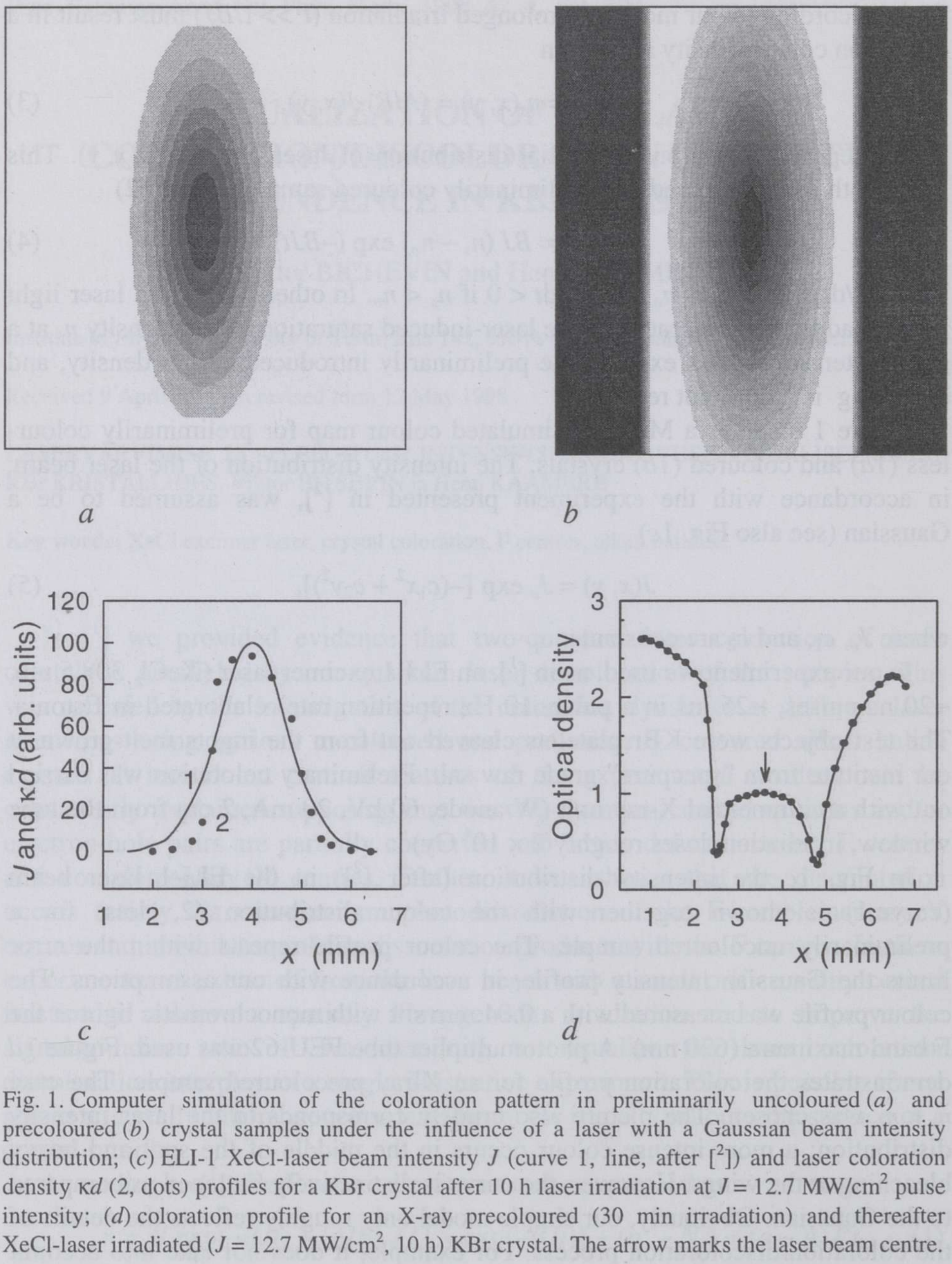

Fig. 1. Computer simulation of the coloration pattern in preliminarily uncoloured (*a*) and precoloured (*b*) crystal samples under the influence of a laser with a Gaussian beam intensity distribution; (*c*) ELI-1 XeCl-laser beam intensity J (curve 1, line, after [2]) and laser coloration density κd (2, dots) profiles for a KBr crystal after 10 h laser irradiation at $J = 12.7$ MW/cm^2 pulse intensity; (*d*) coloration profile for an X-ray precoloured (30 min irradiation) and thereafter XeCl-laser irradiated ($J = 12.7$ MW/cm^2, 10 h) KBr crystal. The arrow marks the laser beam centre.

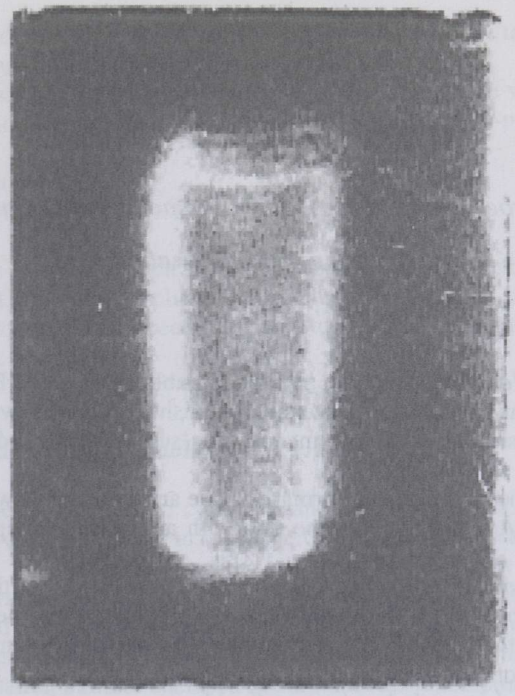

Fig. 2. A KBr crystal, preliminarily X-rayed and successively exposed to excimer laser ELI-1 radiation. Note the ring-shaped, the most extensively laser-bleached area. Dark surrounding: X-coloured unexposed region.

Figure 2 displays a photograph of the crystal whose colour profile is presented in Fig. 1*d*. The effect of the laser "ring burning", according to the intensity distribution in the beam, is distinctively seen. The overall resemblance with the expectance displayed in Fig. 1*b* is apparent. As a result, we have shown that the validity of our concept of laser coloration/discoloration action can be convincingly visualized in a single unsophisticated experiment, needing only availability of a proper laser and X-ray facility. Indeed, exclusive laser colouring should not induce any bleaching, whereas a mere discoloration effect should be maximal at the laser spot centre.

We acknowledge the financial support of the Estonian Science Foundation, grant No. 1930.

REFERENCES

1. Bichevin, V. and Käämbre, H. Laser-induced coloration and discoloration processes in alkali halides. *Phys. Stat. Sol. (b)*, 1994, **186**, 1, 57–70.
2. Treshchalov, A. B. and Peet, V. E. Dynamics of the discharge, pumping and lasing in a XeCl excimer laser. *IEEE J. Quantum Electron.*, 1988, **24**, 2, 169–176.