Generation of the second optical harmonic in liquid crystals

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The generation of the second optical harmonic has been investigated in the nematic and smectic phases of different liquid crystals for homeotropic and planar orientations of the samples. The generation of the second harmonic is interpreted within the framework of the multipole mechanism.

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Until now there has been no single point of view with regard to the presence of the generation of the second harmonic of light (GSH) in liquid crystals (LC)[1-3] and with regard to the GSH mechanism when it is observed.[4-7] On the other hand, a clearcut answer to this problem has fundamental importance, since it touches upon the question of the symmetry properties of an LC medium.

The GSH of light in LC was first discovered in cholesteryl carbonate[1] and was explained by the absence of an effective center of symmetry within the limits of so-called liquid-crystal "clusters." Later, more extensive and more thorough studies[2-3] of the GSH effect were conducted in nematic, cholesteric, and nonferroelectric smectic LC.[1)] No GSH was detected in any of the listed mesophases, and the special case of GSH in cholesteryl carbonate,[1] as was explained, is attributed to the presence of unmelted solid crystals of the substance in the mesophase.

The discussion of GSH of light in LC was reopened in Refs. 4-6, whose authors explain the presence of the GSH of light in MBBA by "the off-center symmetry of the oriented layers of nematic LC" (Ref. 5). Other authors,[7] however, disputing this explanation, attribute the GSH of light in MBBA to the appearance of surface polarization of the LC due to the flexoelectric effect.

We had reported elsewhere[9] that the GSH of light was induced by an electric field in the nematic and smectic phases of LC. The purpose of this work is to look for the GSH effect in the absence of an inducing field in order to resolve the existing conflict.

Our experiment can be summarized as follows: By applying an electric field to the LC layer to remove the center of symmetry, we have determined the directions of phase synchronism (PS) for interactions of the type *ee-o* and *oe-o*. Then the signal of the second harmonic without the inducing field was measured in these same directions. This approach to the task of observing the GSH effect makes it possible to increase considerably the sensitivity of the experiment and to reduce the power density of the pumping of the sample, as compared with the situation in Refs. 4-6; this reduces the likelihood of secondary effects, which are produced by the action of high-intensity laser radiation on LC, influencing the GSH process.

The experiments were carried out on the apparatus described in Ref. 9. The power density of the pumping radiation on the sample was 100-200 MW/cm^2. The sensitivity of our apparatus is estimated at a level of ~30 photons of the second harmonic in each laser pulse. The accuracy of the measurement of the intensity $I_{2\omega}$ of the second harmonic of light was ~10%. The cell for observing phase-synchronous GSH was comprised of two right-angle glass prisms.[9] The experiments were performed in cells with homeotropic and planar orientation of the LC director with respect to the glasses in order to determine the role of surface orientation of the LC in the GSH of light. In both cases the inducing electric field was directed perpendicularly to the director, and the electric vector of the pumping wave was oriented at a 45° angle to it.

In this work we have investigated the GSH of light in MBBA, in p-n-hexyloxy-p′-n-amyl-α-cyanostilbene (α-CS), in p-n-pentyl-p′-cyanobiphenyl (5 CB), and p-n-octyl-p′-cyanobiphenyl. GSH was observed in all the mentioned LC in the directions of the above-stated types of PS. The results of the measurements of the MBBA for the planar and homeotropic orientations are shown in Figs. 1 and 2, from which we see that PS of the *ee-o* type appears only when an inducing electric field is applied to the sample, whereas PS of the *oe-o* type occurs without the field. We note that GSH is observed both for the planar and for the homeotropic orientations of the LC layers. To understand the nature of the dependence of $I_{2\omega}$ on the thickness of the LC layer in the *oe-o* type synchronism, we have fabricated a planar cell with a wedge-shaped LC layer; this made it possible to vary the optical path length of the radiation in the LC layer while satisfying the PS condition by scanning the laser beam at right angles. The intensity of the second harmonic increases monotonically with increasing

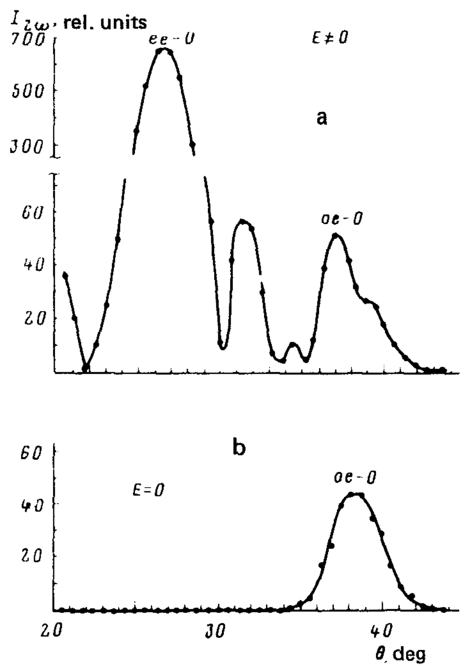

FIG. 1. Dependence of the intensity of the second optical harmonic on the angle θ between the director and its wave vector for MBBA for homeotropic orientation in the presence (a) of of an inducing electric field and in its absence (b). a–Electric-field intensity is $E = 14$ kV/cm, b–$E = 0$. The thickness of the liquid-crystal layer is 50 μm and $t = 23.5$ °C.