Nonlinear optical effects of fourth order in the field in a lithium formiate crystal

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The effective fourth-order nonlinearity was determined by measuring the relative fourth-harmonic powers generated in different directions in a lithium formiate crystal. Particular attention was paid to the influence of cascade processes on fourth-harmonic generation.

1. We report in this article the results of experiments in which nonlinear optical effects of fourth order in the field were registered for the first time in crystals. We obtained in a lithium-formiate (LFM) crystal a synchronous process of fourth-harmonic generation, connected with nonlinear polarization of the type $\mathbf{P}_{nl} = \chi^{(4)}\mathbf{E}^4$. We determined the effective value of the nonlinear fourth-order susceptibility, which was found to be $\chi^{(4)}_{eff} = 2.8\times10^4\,[d_{32}]^3$; since $d_{32} = 2.8\times10^{-9}$ cgs esu for LFM, we get $\chi^{(4)}_{eff} = 0.5\times10^{-21}$ cgs esu.

2. Besides being of fundamental interest, experimental research on optical nonlinearities of higher order (which have recently been stimulated by progress in the theory of nonlinear polarizability) have also acquired practical significance, namely, these nonlinearities can be used in high-power fields of picosecond pulses to generate higher harmonics.[1]

The experimental data published to date on nonresonant nonlinearities do not go beyond the frame work of the the definitions $\mathbf{P}_{nl} = \chi^{(3)}\mathbf{E}^3$ of the nonlinear polarization and of the corresponding susceptibility $\chi^{(3)}$. It is impossible to determine $\chi^{(3)}$ correctly in crystals without central symmetry without taking into account the competition on the part of cascade processes; it is only most recently that attention has been paid to this important circumstance.[2,3] Naturally, for high-order processes, the problem of competition on the part of cascade processes produced by lower nonlinearities becomes even more acute. One of the principal tasks of the present study was the evaluation and experimental investigation of this problem in the case of fourth-harmonic generation in an LFM crystal.

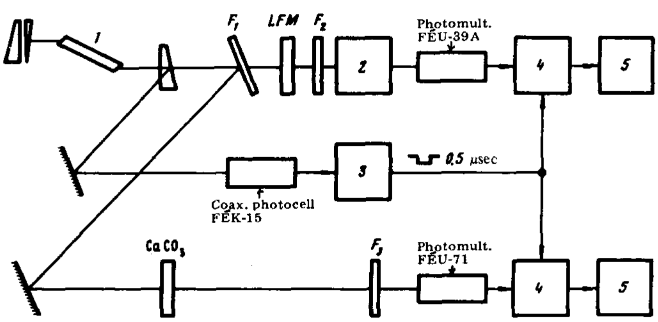

FIG. 1. Block diagram of experimental setup: 1—neodymium-glass picosecond laser, 2—monochromator, 3—gating-pulse shaper, 4—amplitude-time converter, 5—counter, F_1, F_2, F_3—filters.

3. In crystals without inversion center, such as LFM, the nonlinear polarization can be expressed, accurate to fourth-order terms in the field, in the form

$$\mathbf{P}_{nl} = \chi^{(2)}\mathbf{E}^2 + \chi^{(3)}\mathbf{E}^3 + \chi^{(4)}\mathbf{E}^4.$$

Fourth-harmonic generation, including synchronous generation, can result either directly from the nonlinearity $\chi^{(4)}$, or from different cascade processes due to the nonlinearities $\chi^{(2)}$ and $\chi^{(3)}$. Since the cascade processes are synchronous in the synchronism direction $4k_1 = k_4$, it is in this direction that the effective susceptibility is actually measured

$$\chi^{(4)}_{eff} = \chi^{(4)} + a_1\chi^{(3)}_{\omega+\omega+\omega}\chi^{(2)}_{3\omega+\omega} + a_2\chi^{(2)}_{\omega+\omega}\chi^{(3)}_{2\omega+\omega+\omega} + a_3[\chi^{(2)}_{\omega+\omega}]^2\chi^{(2)}_{2\omega+2\omega}\,. \quad (1)$$

The constants α_1, α_2, and α_3 are determined by the orientation of the crystal and by the refractive indices for the interacting waves.[4]

In addition to the direction $4k_1 = k_4$, there are also several directions of synchronous fourth-harmonic generation, corresponding to pure cascade processes, including $2k_2 = k_4$, $k_1 + k_3 = k_4$, and $2k_1 + k_2 = k_4$. Mea-

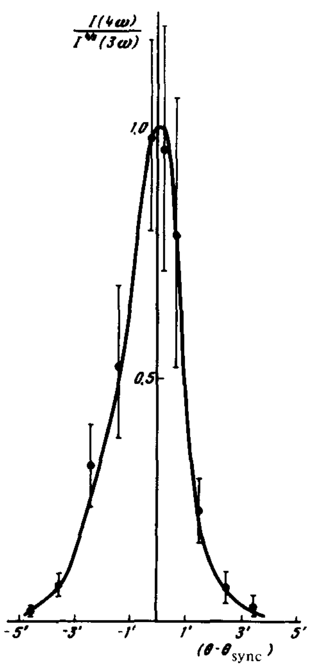

FIG. 2. Angular dependence of the intensity of the fourth harmonic generated near the synchronous-interaction direction $4k_1 = k_4$.

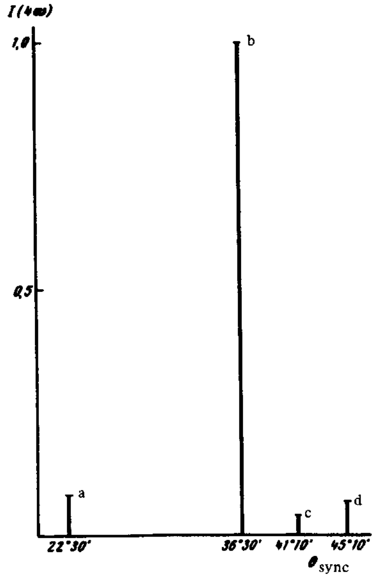

FIG. 3. Relative intensities of the fourth harmonic generated in the LFM crystal for different synchronous interactions: a) $2k_1+k_2=k_4$; b) $4k_1=k_4$; c) $2k_2=k_4$; d) $k_1+k_3=k_4$.

surement of the relative powers of the fourth harmonic generated in different synchronous directions makes it possible to express $\chi^{(4)}_{eff}$ in terms of the lower nonlinearities and, in addition, if the signs of the different terms in expression (1) are known, it becomes possible to determine $\chi^{(4)}$.

4. A block diagram of the experimental setup is shown in Fig. 1. Its principal elements are an Nd^{3+}-glass picosecond laser operating in a regime with a zero transverse mode, and a sensitive recording system. Gating of the amplitude-time converted, with a gate duration 0.5 μsec, was used to cut off the photomultiplier noise. To decrease the effect of the instability of the pump-pulse parameters, we used a standard channel in which we registered the third harmonic excited in a calcite crystal. The LFM crystal were 5 mm long. The laser radiation propagated in the XZ plane of the crystals.

Figure 2 shows the angular dependence of the intensity of the fourth harmonic near the direction $4k_1=k_4$. The experimental synchronism width was 2.5′.

To determine $\chi^{(4)}_{eff}$ we compared the fourth-harmonic signals in the synchronism directions a and d (Fig. 3). The angle between the vector $\mathbf{E}$ and the Y axis was 45° in this case. It was found in the experiment that $\chi^{(4)}_{eff} = (2.8\pm0.7)\times10^4 d_{32}^2$: The value of $\chi^{(4)}_{eff}$ was determined from the equation $\mathbf{P}(4\omega)=\chi^{(4)}_{eff}\ (4\omega,\omega,\omega,\omega,\omega)\ E_y^4(\omega)$; the field E was determined in the same manner as in[5].

If we use the value given in[6] for the component d_{32} of LFM, we obtain $\chi^{(4)}_{eff}=(0.6\pm0.3)\times10^{-21}$ cgs esu.

To estimate the contribution of the cascade terms in expression (1) for $\chi^{(4)}_{eff}$, we compared the intensities of the fourth harmonic in the directions, a, b, and c (Fig. 3). In these measurements the laser radiation was polarized parallel to the Y axis. The lack of information on the signs of the components $\chi^{(3)}$ which enter in $\chi^{(4)}_{eff}$ introduces an uncertainty in the estimate of the component $\chi^{(4)}_{zyyy}$ that is responsible for the fourth-harmonic generation on the nonlinearity $\chi^{(4)}$ at a given crystal orientation. It can be stated on the basis of the obtained data that $\chi^{(4)}_{zyyy}$ lies in the range $(0.2-2)\times10^{-21}$ cgs esu.

5. The problem of separating the direct process from the cascade processes becomes much simpler when a fifth harmonic is generated in crystals with an inversion center. In the $CaCO_3$ crystal it is possible to obtain direct generation of a synchronous fifth harmonic of a neodymium laser; the only competing process in this case is the cascade process on the cubic nonlinearity.

6. The results characterize primarily the fourth-order nonlinearity in LFM. If we are interested in polarizations of higher orders for the generation of higher optical harmonics, it is necessary to strive to increase $\chi^{(n)}_{eff}$. Account must then be taken of the possible interference of the cascade and direct processes.

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