THIRD-ORDER NONLINEAR OPTICAL COEFFICIENTS OF LiIO3

Masakatsu Okada

Citation: Applied Physics Letters **18**, 451 (1971); doi: 10.1063/1.1653490
View online: http://dx.doi.org/10.1063/1.1653490
View Table of Contents: http://scitation.aip.org/content/aip/journal/apl/18/10?ver=pdfcov
Published by the AIP Publishing

THIRD-ORDER NONLINEAR OPTICAL COEFFICIENTS OF $LiIO_3$

Masakatsu Okada
Broadcasting Science Research Laboratories of NHK (Japan Broadcasting Corporation)
1-10-11, *Kinuta, Setagaya, Tokyo*, 157, *Japan*
(Received 9 February 1971)

The third-order nonlinear optical coefficients of $LiIO_3$ have been measured by third harmonic generation. It has been found that the third-order nonlinear optical coefficient c_{35} is large. The coefficient c_{12} has a finite value, which shows that Kleinman's symmetry relations are not strictly valid in $LiIO_3$.

Since $LiIO_3$ was found to show a large optical nonlinearity,[1,2] it is used for the phased-matched second harmonic generation, optical mixing, and parametric oscillation. Moreover, $LiIO_3$ has been pointed out[1] to be appropriate for a check on the Kleinman's symmetry relations[3] on nonlinear optical coefficients. We have shown[4] that the Kleinman's relations are not perfectly valid in the second-order nonlinear optical coefficients of $LiIO_3$. In this letter we report that the third-order nonlinear optical coefficients have large values and do not satisfy the Kleinman's relations.

Third-order nonlinear optical coefficients c_{ij} have been measured through the third harmonic generation using a repetitively Q-switched Nd:YAG laser, which radiates an output with a peak power of 2 MW and a pulse width of 22 nsec at 1.064 μ.

$LiIO_3$ belongs to the point group 6. Nonlinear coefficients are given as

$$\begin{matrix} c_{11} & c_{12} & 0 & c_{14} & 0 & c_{16} & 0 & c_{11}/3 & c_{12}/3 & 0 \\ -c_{12} & c_{11} & 0 & c_{16} & 0 & -c_{14} & 0 & -c_{13}/3 & c_{11}/3 & 0 \\ 0 & 0 & c_{33} & 0 & c_{35} & 0 & c_{35} & 0 & 0 & 0. \end{matrix}$$

If the Kleinman's relations are applied, coefficients c_{12} and c_{14} vanish and c_{35} is equal to c_{16}.[5]

Third-order nonlinear phase-matchable processes are classified into three types,[5] that is, (I) o + o + o → e, (II) o + o + e → e, and (III) o + e + e → e (o and e refer to the ordinary and extraordinary waves, respectively). In $LiIO_3$ the third-order nonlinear polarizations for each type of process are written as

$$P(3\omega; \mathrm{o}+\mathrm{o}+\mathrm{o}\rightarrow \mathrm{e}) = c_{12}\cos\theta\,[E^{\mathrm{o}}(\omega)]^3,$$

$$P(3\omega; \mathrm{o}+\mathrm{o}+\mathrm{e}\rightarrow \mathrm{e}) = (c_{11}\cos^2\theta/3 + c_{35}\sin^2\theta) \times [E^{\mathrm{o}}(\omega)]^2\,E^{\mathrm{e}}(\omega),$$

$$P(3\omega; \mathrm{o}+\mathrm{e}+\mathrm{e}\rightarrow \mathrm{e}) = (c_{12}\cos^3\theta/3 + c_{14}\cos\theta\sin^2\theta) \times E^{\mathrm{o}}(\omega)[E^{\mathrm{e}}(\omega)]^2,$$

where θ is the phase-matching angle, and $E^{\mathrm{o}}(\omega)$ and $E^{\mathrm{e}}(\omega)$ are amplitudes of ordinary and extraordinary components of fundamental wave, respectively. However, if the Kleinman's relations are applied, the type I and III processes are forbidden, while the polarization in the type II process is unchanged.

Phase-matching angles for each process have been calculated by using refractive indices $n_1^{\mathrm{o}} = 1.860$, $n_1^{\mathrm{e}} = 1.719$, $n_3^{\mathrm{o}} = 1.982$, and $n_3^{\mathrm{e}} = 1.815$. In the type I process the phase-matching angle is 57° 02′ and in the type III process it does not exist. The phase-matching angle for the type II process is near 90°, but only several symmetrical sideband structures have been observed around 90°, as shown in Fig. 1, in the experiment of angular variation since the perfect phase-matching angle is not within 90°.

Two pieces of crystal have been prepared for the type I and II processes to satisfy the condition of nearly normal incidence in the phase-matched state. Cross sections of crystals are large compared with the beam diameter of 2 mm, and lengths are 4.56 and 290 mm for the type I and II processes, respectively. The optical transmission of a crystal with a thickness of 4.56 mm has been measured from 0.28 to 1.5 μ for the two polarizations, $E \parallel c$ and $E \perp c$, and shown to be sufficiently transparent at 0.355 μ as shown in Fig. 2.

For the type I process, which is forbidden by the Kleinman's relations, a sharp central peak with a width of about 10′ has been observed in the experiment of dependence of the third harmonic power on the angular variation around the phase-matching angle, which has been measured to be 57° 46′. Moreover, in order to ascertain the process to be the third order, the dependence of third harmonic intensity on the polarization of fundamental wave has been measured by rotating the direction of polarization of fundamental wave. The third harmonic power is expected to be proportional to $\cos^6\phi$ in the type I process, where ϕ is the rotating angle of the direction of polarization of the incoming fundamental wave from that of the ordinary wave. The experimental result has been consistent with the prediction.

Experiments have been performed for the type II

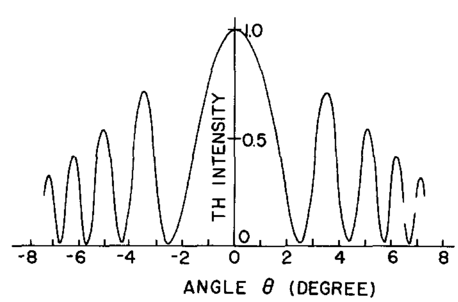

FIG. 1. Dependence of third harmonic intensity on the angular variation of crystal near the phase-matching angle in the type II process.

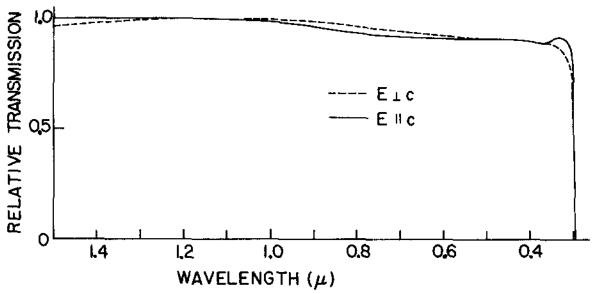

FIG. 2. Optical transmission characteristics of $LiIO_3$ from 0.28 to 1.5 μ.

process in the same way as for the type I process. The dependence of third harmonic power on the polarization of fundamental wave, which is proportional to $\cos^4\phi \sin^2\phi$, had a good agreement with the experimental results. In the angular dependence of the third harmonic power, several sidebands have been observed around 90°. From the ratios of peak values of successive sidebands, the first measured sideband in Fig. 1 is estimated to correspond to the 5th or 6th one in the regular phase-matching characteristics.

Nonlinear coefficients of $LiIO_3$ have been calculated by comparing with those of KDP and ADP.[6] The third harmonic generation in KDP and ADP have been done through the type I process. KDP and ADP crystals with length of 11 mm and cross section of 4×5 mm have been prepared so that the condition of normal incidence might be satisfied when the azimuthal angle was chosen as 22.5° in the phase-matched state. An expression for the dependence of the third harmonic power S_3 on the crystal length l and the effective nonlinear optical coefficients[7] c_{eff} has been derived by modifying the procedure for the second harmonic generation.[7] In the case of small diffraction and double refraction, S_3 is given as

$$S_3 = 96 S_1^3 \left(\frac{128\pi^2\omega^2 c_{eff}^2}{c^4 n_1^3 n_3}\right)\frac{l^2}{w_0^4},$$

where S_1 is the laser intensity, w_0 the radius of the beam waist, and $c_{eff} = (c_{11} - 3c_{18})\cos\theta \sin(4\times 22.5°)/4$ for KDP and ADP, $c_{eff} = c_{12}\cos\theta$, and $c_{eff} = (c_{11}\cos^2\theta + c_{35}\sin^2\theta)$ for the type I and II processes of $LiIO_3$, respectively. By using data for refractive indices and phase-matching angles, the third-order nonlinear coefficients are estimated as follows:

$$(c_{11} - 3c_{18})^{KDP} = 2.1(c_{11} - 3c_{18})^{ADP},$$

$$c_{12}^{LiIO_3} = 12(c_{11} - 3c_{18})^{ADP},$$

$$c_{35}^{LiIO_3} = 300 \sim 400(c_{11} - 3c_{18})^{ADP}.$$

It is concluded that $LiIO_3$ shows a large third-order nonlinear optical coefficient c_{35}, and may be useful in the third-order nonlinear optical processes. Moreover, it is noted that c_{12} is a few percent of c_{35}, and thus the Kleinman's symmetry relations are not strictly valid in $LiIO_3$.

The author thanks Dr. S. Ieiri for helpful discussions and T. Umezawa for supplying $LiIO_3$ crystals.

[1]G. Nath and S. Haussühl, Appl. Phys. Letters **14**, 154 (1969).
[2]F. R. Nash, J. G. Bergman, G. D. Boyd, and E. H. Turner, J. Appl. Phys. 40, 520 (1969).
[3]D. A. Kleinman, Phys. Rev. 126, 1977 (1962).
[4]M. Okada and S. Ieiri, Phys. Letters (to be published).
[5]J. E. Midwinter and J. Warner, Brit. J. Appl. Phys. 16, 1667 (1965).
[6]C. C. Wang and E. L. Baardsen, Appl. Phys. Letters 15, 396 (1969).
[7]G. D. Boyd and D. A. Kleinman, J. Appl. Phys. **39**, 3597 (1968).

VISUAL OBSERVATION OF SUBMILLIMETER WAVE LASER BEAMS

F. Keilmann
Max-Planck-Institut fuer Plasmaphysik, 8046 Garching, Germany

and

K. F. Renk
Physik Department der Technischen Universitaet, Muenchen, Germany
(Received 15 March 1971)

The operation of a thermal image converter for infrared and submillimeter wave laser radiation is reported. An energy density of 0.004 J/cm^2 is sufficient for full contrast, independent of wavelength. Photographs taken with HCN laser radiation are shown.

For the detection of submillimeter laser radiation, only pointlike detectors have been used, e.g., a Golay cell integrating over a small area of, say, 0.1 cm^2. It is, however, desirable to develop a simple means of visualizing two-dimensional intensity patterns for many applications, where a scanning type of image synthesis[1] seems inadequate or too complicated.

Our approach is based on a thermal effect, as first demonstrated for detecting near-ir radiation by Hansen *et al.*[2]: Through absorption in a thin film, a temperature image is formed which can be reversibly read out by using the optical properties of a layer of cholesteric liquid crystals. This material shows a sharply peaked back-scattering spectrum if illuminated with white light; the wavelength