

Fig. 3. Geometry and transmission curves of structures A and B (the dark areas represent the conductor).  
 1) Type A, nickel,  $a/g = 0.3$ ,  $g = 50 \mu$ , 2) Type B, aluminum,  $a/g = 0.3$ ,  $g = 50 \mu$ , substrate - lavsan polyester  $5 \mu$  thick. 3) Type B, aluminum,  $a/g = 0.5$ ,  $g = 50 \mu$ , substrate - lavsan  $5 \mu$  thick. 4) Type B, aquadag,  $a/g = 0.1$ ,  $g = 500 \mu$ , substrate - paper  $70 \mu$  thick.

- [3] R.D. Rawcliffe and C.M. Rondoll, *Appl. Opt.* **6**, 1353 (1967).  
 [4] S.P. Varma and K.D. Moller, *Appl. Opt.* **8**, 1663 (1969).  
 [5] W. Desorbo and H.E. Cline, *JAP* **41**, 2099 (1970).

practically independent of the absorbing and reflecting properties of the material. This allows us apparently to state that there is a deep analogy between the transmission of the structure in the UXR and the far infrared regions. It follows also that in the UXR region one can use also structures of type A to cut off the long-wave background of the plasma. The procedure for preparing such structures is described in [5]. The filters proposed by us for UXR are essentially band filters. Both the individual filters A and B and their combinations can be useful in a number of problems of spectral plasma-discharge diagnostics.

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- [1] R.H. Huddleston and S.L. Leonard, *Plasma Diagnostic Techniques*, Academic, 1965.  
 [2] D.H. Tombolian and H.M. Pell, *Phys. Rev.* **83**, 1196 (1951).

#### DISPERSION OF NONLINEAR SUSCEPTIBILITY OF $\text{TeO}_2$ SINGLE CRYSTAL IN THE OPTICAL REGION

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By registering the second harmonic (SH) of light, we measured in the interval  $0.35 - 0.53 \mu$  the dispersion of the nonlinear susceptibility of  $\text{TeO}_2$ , in which SH generation is forbidden according to the Kleinman relation.

Kleinman's relation results from an analysis of the symmetry properties of the nonlinear-susceptibility tensor  $d_{ijk}$ , which is responsible for second harmonic (HS) generation in crystals. In the general case, without allowance for the crystal symmetry,  $d_{ijk}$  is symmetrical with respect to permutation of only two indices:  $d_{ijk} = d_{ikj}$ . Kleinman has proposed that far from the crystal absorption bands, when the dispersion of the nonlinear susceptibility can be neglected,  $d_{ijk}$  is symmetrical with respect to permutation of three indices:  $d_{ijk} = d_{kih} = d_{kji} = \dots$ . This symmetry relation proposed by Kleinman

decreases the number of independent elements  $d_{ijk}$  and causes some of its elements to vanish. Experiment has shown that Kleinman's symmetry relation is well satisfied. Small deviations from this relation have recently been reported, however, for lithium iodate [2, 3] and barium nitrite [3] crystals.

$\text{TeO}_2$  crystals belong to the non-centrosymmetrical class 422 [4]. According to Kleinman's ideas, SH generation in crystals of this class is forbidden. It was recently discovered, however, that  $\text{TeO}_2$  can double effectively the frequency of optical radiation at a wavelength  $\lambda = 1.064 \mu$  [5, 6]. Consequently, Kleinman's relation does not hold for  $\text{TeO}_2$ . We propose that this is due to the strong frequency dispersion of the nonlinear susceptibility of this crystal, which cannot be neglected as is done in the derivation of the Kleinman relation.

To check on this hypothesis, we measured the dispersion of the only independent element  $d_{123}$  of the nonlinear-susceptibility tensor, which is responsible for SH generation in crystals of class 422 (allowance for the Kleinman relation yields  $d_{123} = 0$ ). The SH was excited with ruby and neodymium lasers, and also by the first and second Stokes components of SRS in liquid nitrogen.

The experimental setup is shown in Fig. 1. To increase the accuracy of the relative measurements of the SH intensity we used a two-channel registration system. In the main channel, the SH was excited in  $\text{TeO}_2$  or in quartz (the comparison crystal), was separated from the fundamental frequency with a filter and monochromator, was registered with photomultiplier FEU-39, and was observed on the pulsed oscilloscope OK-17M. In the reference channel, the harmonic was excited in quartz, registered with a photomultiplier, and fed to the second channel of the oscilloscope.

The component  $d_{123}(\text{TeO}_2)$  was measured relative to  $d_{111}(\text{quartz}) = 1.29 \times 10^{-9}$  cgs esu [7], which was assumed to be constant in the investigated frequency range. The measurements were performed at room temperature, away from the synchronism directions, on wedge-shaped  $\text{TeO}_2$  samples, using the procedure described in [8]. The SH intensity registered by the photomultiplier is proportional in this case to

$$I_{2\omega} \propto \left[ \frac{d I_c I_\omega}{(n_\omega + 1)^2 (n_{2\omega} + 1)} \right]^2,$$

where  $I_\omega$  is the intensity of the exciting radiation,  $d$  is the component of the nonlinear tensor,  $I_c$  is the coherence length, and  $n_\omega$  and  $n_{2\omega}$  are the refractive indices at the fundamental and SH frequencies, respectively.

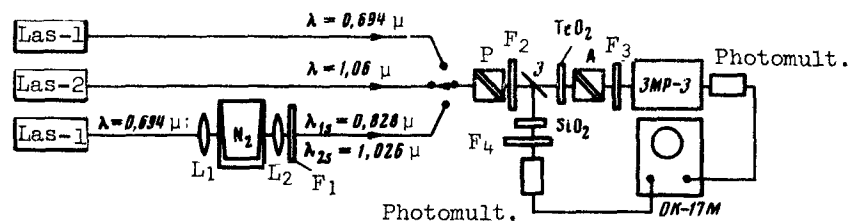


Fig. 1. Experimental setup: Las-1, Las-2 - ruby and neodymium lasers, respectively,  $\text{N}_2$  - Dewar with liquid nitrogen,  $L_1$ ,  $L_2$  - lenses for SRS excitation, P - polarizer, A - analyzer,  $F_1$  - KS-19 filter,  $F_2$  - S-13 filter,  $F_3$ ,  $F_4$  - filters to separate the SH, 3 - semitransparent mirror, 3MR-3 - quartz monochromator.

Nonlinear optical properties of TeO<sub>2</sub>

$\lambda, \text{\AA}$	$d_{123},$ $10^{-9}$ cgs esu	$\ell_c, \mu$	$\delta_{123},$ $10^{-8}$ cgs esu
10600	1.7	8.4	5.5
10260	1.6	6.9	5.4
8280	3.7	1.7	7.0
6943	4.7	0.61	6.9

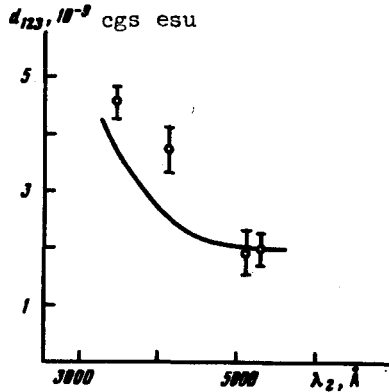


Fig. 2. Dependence of the nonlinear susceptibility of TeO<sub>2</sub> on the second-harmonic wavelength.

The TeO<sub>2</sub> crystal was oriented with a type URS-25 x-ray goniometer relative to the crystallographic axes x, y, and z (z is the optical axis) with accuracy not worse than 0.5 deg and was cut perpendicular to the [011] direction. The fundamental radiation was normally incident on the crystal surface. The electric-field vector E made an angle of 45° to the axes y and z. The nonlinear polarization vector was parallel to the x axis and was equal to  $P_{nl} = d_{123}E^2$ .

The experimental results are listed in the table. The coherence length  $\ell_c$  and the Miller coefficient  $\delta_{123}$  were calculated from the data on the dispersion of the refractive indices of TeO<sub>2</sub> [9].

The measured values of  $d_{123}$  are compared in Fig. 2 with the theoretical curve calculated in accordance with formula  $d_{123} = \delta_{123}(n_{2\omega}^2 - 1)$

$(n_{\omega}^2 - 1)/(4\pi)^3$ , assuming  $\delta_{123}$  to be independent of the wavelength of the light.

The observed discrepancies between theory and experiment show that the coefficient  $\delta$  of TeO<sub>2</sub> has frequency dispersion.

The results reveal a rather appreciable violation of the Kleinman relation in TeO<sub>2</sub>, particularly in the short-wave region of the spectrum, where the nonlinear coefficient of TeO<sub>2</sub> is much larger than, say, in KDP. At the same time, as shown by our measurements, the presence of a strong frequency dispersion of the nonlinear susceptibility of TeO<sub>2</sub> casts doubts on the validity of using the Kleinman relation for this crystal.

- [1] D.A. Kleinman, Phys. Rev. 126, 1977 (1962).
- [2] M. Okada and S. Jeiri, Phys. Lett. 34A, 63 (1971).
- [3] V.M. Koval'chuk, Abstracts of Papers at Sixth All-union Conference on Non-linear Optics, Minsk, 1972.
- [4] J. Leciejewicz, Z. Kristal. 116, 345 (1961).
- [5] S. Singh, W.A. Bonner, and L.G. Van Uitert, Phys. Lett. 38A, 407 (1972).
- [6] D.S. Chemla and J. Jerphagnon, Appl. Phys. Lett. 20, 222 (1972).
- [7] R. Bechman, S.K. Kurtz, and Landolt-Bornstein, Numerical Data and Functional Relationships, Group 3, Vol. 2, Springer, Berlin, 1969.
- [8] A. Savage, J. Appl. Phys. 36, 1496 (1965).
- [9] N. Uchida, Phys. Rev. B4, 3736 (1971).