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TRANSMISSION SECOND HARMONIC GENERATION IN $Cd_{1-x}Mn_xTe$ at 1.064 μm*

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Second harmonic generation, created by nanosecond Nd:YAG laser pulses at 1.064 μm with relatively low intensity in $Cd_{1-x}Mn_xTe$ bulk crystals and thin layers was measured in transmission geometry. The effect practically occurs in a very thin surface layer of the material and it is used as a relatively straightforward method of layer quality characterization. It is shown that the angular dependence of the second harmonic generation intensity in thin layers of CdTe with good crystallographic (and optical) quality agrees very well with the theory in contrary to the samples with some distortions from the ideal structure which exhibits large distortions from the theory.

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Recently, nonlinear properties of thin layers and quantum structures of various II–VI and III–V semiconductors attracted considerable attention due to possible application in a variety of fields. Relatively high electro-optic coefficient of these materials and relatively fast optical response make them attractive for devices used in optical communication, data processing, real time optical processing and optical limiting. Recent studies [1–3] show that cadmium manganese telluride exhibits strong nonlinear response in the visible and in the near-infrared. These materials may be of interest since it is possible to control its forbidden gap by the manganese content thus enhancing a nonlinear response due to band-to-band optical transitions.

It has been recently shown that the second order nonlinear susceptibility coefficients for CdMnTe crystals are higher than these for CdTe [4] which makes this material even more attractive for applications.

CdMnTe crystals were found to generate efficiently second harmonic in the infrared, excited by CO_2 laser radiation at 10.6 μm. In this paper we show that CdMnTe crystals, grown by the Bridgman technique and molecular beam epitaxy

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(MBE) grown layers also generate efficiently second harmonic when excited by the fundamental beam from the nanosecond (6 ns pulse) Nd:YAG laser with a relatively low pulse energy below 1 mJ. The second harmonic light is strongly absorbed by CdMnTe, independent of the composition, therefore the green light at 532 nm wavelength is emitted only from a very thin surface layer of the crystal of thickness about a few tens of μm. The second harmonic photons generated inside the crystal are immediately absorbed, contributing to a creation of electron–hole plasma, and never reach the surface of the sample.

The theory of Bloembergen and Pershan [5] was found to describe accurately the properties of second-harmonic radiation generation in piezoelectric crystals in the spectral regions where samples are transparent and absorbing for the frequency-doubled beam. According to this theory, the second harmonic light intensity is proportional to the square of the second-harmonic polarization, P. Intensity of the second-harmonic light polarized parallel to the fundamental beam, I_σ, and perpendicular, I_π, depends on the plane of incidence of the fundamental beam on the sample surface and relative orientation of polarization of the beam and main crystallographic axes of the crystal. The angular dependencies of the π and σ component second harmonic generation (SHG) signal intensities for incidence on the (100), (110) and (111) planes for crystals with zinc blende structure are given in Table.

TABLE

The angular dependencies of the π and σ component SHG signal intensities for incidence on the (100), (110) and (111) planes for crystals with zin blende structure [6].

Incidence on a plane	I_π	I_σ
(111)	$(2/3)E_{\rm p}^2(\omega)\chi^2\sin^2 3\phi$	$(2/3)E_{\rm p}^2(\omega)\chi^2\cos^2 3\phi$
(110)	$(1/2)^2E_{\rm p}^2(\omega)\chi^2\sin^2\phi(1-3\cos^2\phi)^2$	$(3/2)^2E_{\rm p}^2(\omega)\chi^2\sin^4\phi\cos^2\phi$
(100)	0	$2E_{\rm p}^2(\omega)\chi^2\sin^2\phi\cos^2\phi$

where χ — the value of the nonvanishing elements of the nonlinear susceptibility tensor for tetrahedral symmetry, $E_{\rm p}$ — the amplitude of the fundamental electric field inside the nonlinear crystal, ϕ — angle between the electric field $E_{\rm p}$ and the appropriate crystallographic axis of the crystal ([110], [100], [100], respectively).

The results of SHG measurements in the transmission geometry for CdTe and CdMnTe samples are presented in Fig. 1. Angular dependence of the SHG conversion efficiency for CdTe single crystal of very good crystallographic and optical quality is shown in Fig. 1a [7]. The fundamental infrared red was perpendicular to the (110) surface of the sample. The Bloembergen and Pershan theory, shown by lines in Fig. 1, describes the observed results with very good accuracy.

The results of SHG investigations for $Cd_{0.8}Mn_{0.2}Te$ crystal, grown by the Bridgman method, are shown in Fig. 1b. The fundamental beam was perpendicular to nominally (111) surface of the crystal. The best fits of the Bloembergen-Pershan theory to the data are also shown in Fig. 1b. The theory does not describe the data well. Relatively poor crystallographic quality of the crystal is responsible for

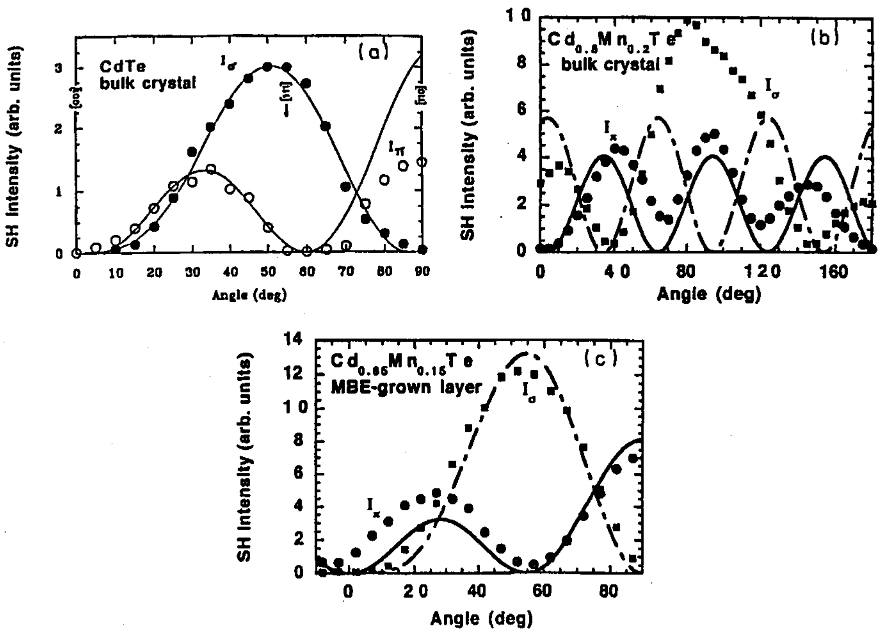

Fig. 1. Angular dependence of experimentally observed SHG from a (a) a (110) slab of CdTe of good crystallographic quality (taken from Ref. [7]); (b) a nominally (111) slab of $Cd_{0.8}Mn_{0.2}Te$ crystal, grown by the Bridgman method; (c) a nominally (100) $Cd_{0.85}Mn_{0.15}Te$ MBE grown layer. The lines are computer fits of the Bloembergen–Pershan theory to the data.

observed discrepancies between the data and the theory. The X-ray measurement of the crystal structure confirmed that the crystal contains several crystallographic defects and cannot be considered as a single crystal. Similar results were observed for a few other CdMnTe crystals with different manganese content.

The data for $Cd_{0.85}Mn_{0.15}Te$ MBE grown layer of a few μm thick are shown in Fig. 1b. Although the sample surface was (100), the angular dependence of SHG exhibits a large contribution of light generation on the (110) surface. The Bloembergen–Pershan theory with the fundamental beam perpendicular to such a surface fits the data quite well. The SHG data for $Cd_{0.35}Mn_{0.65}Te$ MBE grown layer exhibits a larger distortion and it is difficult to estimate the orientation of this layer. A possibly small (2°) distortion from (100) orientation of the substrate for the MBE grown layers induces a large distortion from the ideal structure for such thick layers.

In conclusion, we showed that the angular dependence of SHG with a frequency-doubled beam strongly absorbed in the sample can provide a quite straightforward test of surface crystallographic quality of crystals and layers. Appropriate formulas for any crystallographic structure and sample orientation can be easily calculated.

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