

MAGNETOOPTICAL SPECTRA OF MnSb CRYSTAL BETWEEN 1.2 AND 6.4 eV

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Abstract—Magneto-optical spectra have been studied for photon energies between 1.2 and 6.4 eV in bulk crystals of undoped and Pt-doped MnSb. Kerr rotation of undoped crystals showed a prominent zero-crossing behavior around 4.5 eV. The zero-crossing energy moved to 4.1 eV in Pt-doped samples, which were explained by the averaged spectra of separated two phases of MnSb and PtMnSb.

KEYWORDS: Magneto-optical spectra, MnSb, Pt-doped MnSb, Bridgmann technique, bulk crystals

INTRODUCTION

Magneto-optical materials for short wavelength MO-recording have been extensively studied. Recently, Takahashi and his colleagues found that 6% Pt-substituted MnSb (hereafter referred to as MnSb(Pt)) shows a large magneto-optical Kerr rotation in visible wavelengths[1]. In order to elucidate the origin of magneto-optical effect of this material, basic researches in bulk crystals are required. However, only available data on MnSb is one measured by Buschow and his group [2], in which spectral region of measurement was limited to photon energies between 1 and 4 eV.

We prepared MnSb and MnSb(Pt) crystals by vertical Bridgmann technique and measured spectra of magneto-optical Kerr rotation and Kerr ellipticity for wider energy region of 1.2-6.4 eV.

This paper describes the results of the experiments and discusses on the electronic structures of the materials.

EXPERIMENTAL

Polycrystalline MnSb powder was synthesized in evacuated silica ampoule from elements of Mn(4N) and Sb(5N) by solid state reaction at 600°C. Slightly Mn-rich composition was necessary in order to get NiAs type MnSb phase.

Polycrystalline bulk crystals of MnSb were prepared by normal freezing method. The melt of the MnSb was cooled down slowly from the temperature slightly above the melting point of the material (825°C). Annealing was performed at 300°C for 2 hours in the vacuum of 10^{-5} Pa. Concerning MnSb(Pt), the starting composition (molar ratio) of Mn:Sb:Pt = 1:1:0.03 was employed for the sake of comparison with the reported film of MnSb(Pt).

Single crystals were grown by the Bridgmann technique, in which the polycrystalline MnSb powder was put in a crucible made of carbon-coated silica or pyrolytic boron nitride, sealed *in vacuo* into a silica ampoule and

placed in the furnace and kept at the temperature 880°C which is sufficiently above the melting point of the material for 20 hours. The ampoule was moved down across the melting point with the rate of 2.0 mm/hr in the temperature gradient of -13°C/cm for three days. The ingot obtained showed well-developed facets. The powder X-ray analysis showed that the bulk material was of a single phase of NiAs-type MnSb. The lattice constants were determined as $a=4.147\text{\AA}$ and $c=5.776\text{\AA}$. From the lattice constants the composition ratio of Mn and Sb was determined to be Mn:Sb=51.2:48.8.[3] According to Okita et al.[4] the magnetic anisotropy constant K_1 is negative for this composition, which means that easy axis of the magnetization is perpendicular to the c-axis of MnSb.

The faceted planes were polished by a successive use of lapping films with 3, 1 and 0.3 μm mesh. X-ray diffraction revealed that these planes were approximately parallel to either $\{110\}$ or $\{112\}$ plane.

Magneto-optical Kerr rotation and Kerr ellipticity were measured using photoelastic modulator technique for photon energies between 1.2 and 6.4 eV, the details of which have been published in ref. 5. The magnetic saturation was confirmed by the hysteresis measurements prior to the spectroscopic studies.

Reflectivity spectra were measured using a Hitachi type U-3410 spectrophotometer between 0.5 and 6.7 eV, to which Kramers-Kronig analysis was applied to obtain dielectric constants of the material with the help of the optical constants obtained by the ellipsometry.

Using the data of dielectric constants we obtained the off-diagonal terms of the conductivity tensor.

RESULTS AND DISCUSSION

Figure 1 shows spectra of Kerr rotation θ_K and Kerr ellipticity η_K in the polycrystalline bulk crystal of MnSb obtained by the normal freezing technique. The spectral shape of the bulk crystal is quite similar to that reported by Buschow [2] as plotted by dotted curves. Several humps are observed in both θ_K and η_K spectra for lower energies. A

prominent zero-crossing behavior of Kerr rotation appeared around 4.5 eV.

Figure 2 shows the magneto-optical Kerr spectra of undoped MnSb before and after annealing. The fine structures become prominent after annealing. Furthermore well-defined peak of Kerr rotation appears at 5.5 eV. This seems to be due to an effect of changes in diagonal dielectric constants by annealing, since in annealed samples considerable decrease in the reflectivity was observed.

Magneto-optical spectra of single crystals of MnSb grown by the Bridgmann technique measured on {110} and {112} crystal planes are shown in Fig. 3 by solid and dotted curves, respectively. Spectral shapes and peak values were slightly different between two crystal planes. Magnetization curves measured by Kerr hysteresis showed that saturation was easier in {110} than in {112}.

Figure 4 shows spectra of real and imaginary parts of conductivity tensor multiplied by frequency deduced from the magneto-optical spectra of single crystal of MnSb for two different orientations. Although the <110> direction is easier to be saturated than <112>, the off-diagonal conductivity of the {110} plane has a smaller value than that of {112} as illustrated in Fig. 4. This result suggests an existence of a strong anisotropy of electronic transitions, indicating in turn, the strong direction-dependence of the electronic structures in this material. Similar anisotropy of magneto-optical spectra was also found in MBE-grown MnSb epitaxial films.[6]

Prominent magneto-optical structure (peak of $\omega\sigma''_{xy}$) is observed around 6 eV in both orientations. Another structure is seen around 3.2-3.5 eV with the opposite polarity to the 6 eV peak.

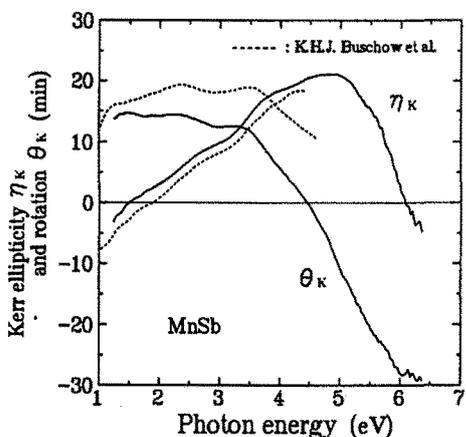


Fig. 1 Spectra of magneto-optical Kerr rotation and ellipticity in polycrystalline bulk crystal of MnSb (solid curve). Those reported by Buschow et al. are plotted by dotted curves.

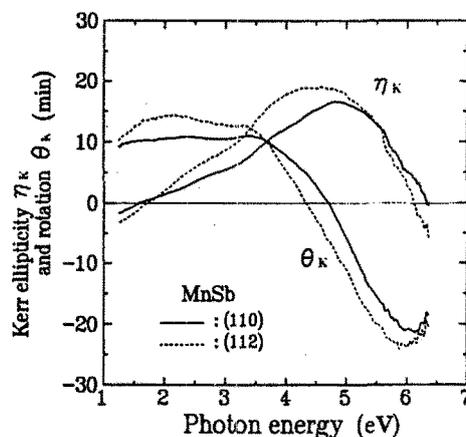


Fig. 3 Spectra of magneto-optical Kerr rotation and ellipticity of a single crystal of MnSb for two crystal planes, solid curves corresponding to {110} plane and dotted curves {112} plane.

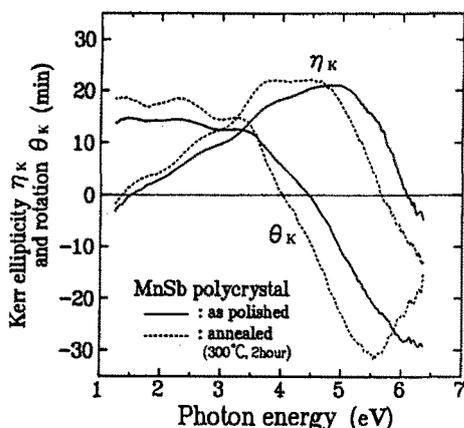


Fig. 2 Spectra of magneto-optical Kerr rotation and ellipticity in polycrystalline bulk crystal of MnSb before (solid curve) and after (dotted curves) annealing in vacuo at 300°C for 2 hours.

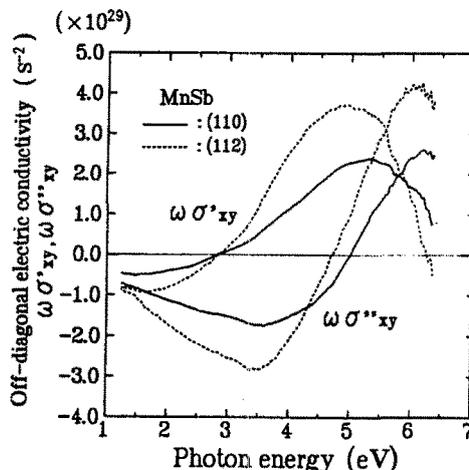


Fig. 4 Spectra of conductivity tensor multiplied by frequency in MnSb for two different orientations: solid curves correspond to {110} plane and dotted curves {112} plane.

These structures may be assigned to band-to-band transitions in the band diagram calculated by Motizuki [7].

X-ray diffraction patterns of polycrystalline bulk crystals of MnSb(Pt) could be indexed by mixtures of NiAs-type hexagonal MnSb phase and C1b-type cubic PtMnSb phase. A map of Pt distribution obtained by an area analysis of EPMA (electron-probe microanalyzer) of the MnSb(Pt) is shown in Fig. 5, in which the Pt distribution is displayed by light area. This figure clearly suggests a phase separation of the crystal into two phases. The analyzed composition of the light area was approximately of PtMnSb and that of the dark zone was MnSb.

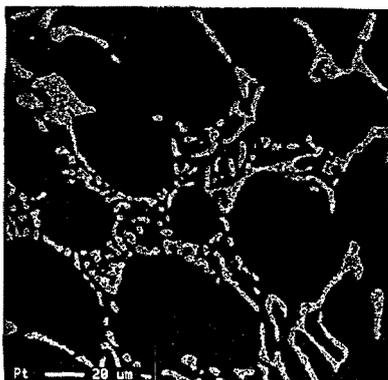


Fig. 5 A distribution map of Pt in polycrystalline bulk material of MnSb(Pt) observed by an area analysis of EPMA.

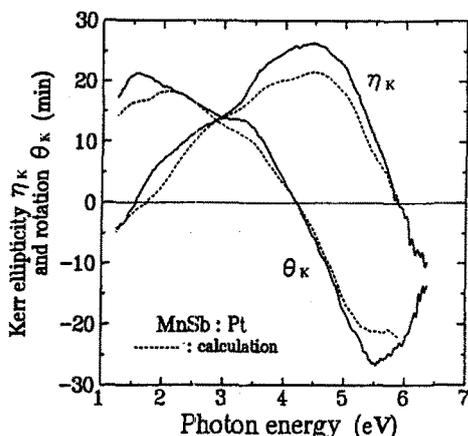


Fig. 6 Spectra of magneto-optical Kerr rotation and ellipticity in polycrystalline bulk crystal of MnSb(Pt). Solid curves denote experimental spectra and dotted curves represent the calculated "averaged" spectra.

The magneto-optical spectra of the polycrystalline bulk crystal of MnSb(Pt) are given by a solid curve in Fig. 6. The Kerr rotation spectrum crosses zero at 4 eV and makes a negative peak at 5.5 eV.

We simulated the magneto-optical spectrum of the "mixed-phase" crystal by calculating a weighted average of diagonal and off-diagonal parts of dielectric constants of MnSb and PtMnSb [8]. The weight was 90% for MnSb and 10% for PtMnSb judging from the EPMA map. The resulted magneto-optical spectra are plotted by dotted curves in Fig. 6, which provide considerably good fit to the experimental curves.

The spectral features are much different from those observed by Takahashi [1] in the sputtered MnSb(Pt) films, suggesting that the crystal structures of the film was only realized by thermally non-equilibrium conditions, while mixed phase crystals are obtained in the equilibrium condition.

CONCLUSION

Magneto-optical investigations were carried out in polycrystalline bulk crystals and single crystals of MnSb. The spectra showed strong magneto-optical transition in 5-6 eV region, which have not been reported. The spectral features could be correlated with the electronic structure of the material. The single crystal of MnSb showed an orientation dependence of magneto-optical spectrum. The melt-grown Pt-substituted MnSb was found to show a phase separation into MnSb and PtMnSb. The spectra of magneto-optical effect could be simulated by the weighted average of magneto-optical spectra of the two phases. The sputter-deposited MnSb(Pt) film which was reported to show a strong magneto-optical effect seems to have different crystal structure from the bulk crystal obtained in this study.

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