Odd ($\propto M$) and even ($\propto M^2$) magneto-optic effects in linear and nonlinear reflected optical fields

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Asymmetries observed in the magneto-optic response of PtMnSb in both the linear and second harmonic reflected fields are analyzed in terms of the simultaneous detection of contributions odd ($\propto M$) and even ($\propto M^2$) in the sample magnetization. [S0163-1829(98)02546-6]

Pustogowa, Hubner, and Bennemann,[1] were the first to demonstrate that the magneto-optic response of a medium could be larger in the second harmonic field than in the linear-optical field. Several workers[2–4] have since provided additional experimental evidence of this possibility. The implications of this result with respect to the collection and analysis of some magneto-optic data are perhaps not yet fully apparent. In studies of the Heusler alloy, PtMnSb, we have observed a marked asymmetry in the second harmonic yield when the sample magnetization is reversed. These results suggest that the magneto-optic effects that are even ($\propto M^2$) in the sample magnetization (usually small and commonly neglected in the linear optical field) can be larger than those effects that are odd ($\propto M$). In such circumstances, it becomes difficult to achieve an experimental configuration to provide unambiguous measurements. We show here that the problem exists even in the transverse Kerr configuration, commonly regarded as the simplest configuration for magneto-optic measurement.

Following earlier extended studies[5–7] of thin films of PtMnSb, we report here a recent experiment to compare the linear and nonlinear magneto-optic behavior of thin PtMnSb films in the transverse Kerr configuration.

Magneto-optic measurements made in the linear optical field are usually related to the material properties using a phenomenological approach. The response of the magnetized medium to an incident optical field is described by a tensor permittivity $\varepsilon'[Q]$. It is also usual to consider any changes of the complex refractive index due to crystallographic effects to be small compared with changes produced by the magneto-optic interaction. It is then valid to permit the tensor symmetry to be determined solely by the magnetization when using either single crystal or polycrystalline samples.

If the magnetization is arbitrarily taken to be in the z direction and, following Sokolov[8] the function $\varepsilon'(Q)$ is expanded to include second-order terms in Q, the tensor becomes

$$[\varepsilon']=\varepsilon_0'\begin{bmatrix}(1+f_qQ^2) & \mp iQ & 0\\ \pm iQ & (1+f_qQ^2) & 0\\ 0 & 0 & 1\end{bmatrix}, \tag{1}$$

where Q is a complex, magnetization dependent, magneto-optic parameter and f_q is defined by Sokolov.[8]

Note that when terms involving the third and higher powers of Q are dropped, the behavior described by the diagonal and off-diagonal elements becomes separated to second order in M. The off-diagonal elements are seen to control only behavior linear (odd) in magnetization and the diagonal elements become responsible only for effects exhibiting a quadratic (even) dependence on magnetization.

The response odd in the magnetization ($\propto M$) is that commonly referred to as the transverse Kerr effect. In order to measure this effect, for the respective optical orders, it can be defined in terms of a fractional change in either the intensity of the reflected incident field or of the generated second harmonic field (SHG) when the magnetization is reversed. Adopting the usual convention we therefore define parameters δ and δ_{SHG} to be

$$\delta=\frac{I_{M_s}^{\uparrow}-I_{M_s}^{\downarrow}}{I_{M_s}^{\uparrow}+I_{M_s}^{\downarrow}},\quad \delta_{\mathrm{SHG}}=\frac{Y_{M_s}^{\uparrow}-Y_{M_s}^{\downarrow}}{Y_{M_s}^{\uparrow}+Y_{M_s}^{\downarrow}}, \tag{2}$$

where $I_{M_s}^{\uparrow}$, $I_{M_s}^{\downarrow}$, and $Y_{M_s}^{\uparrow}$, $Y_{M_s}^{\downarrow}$ are, respectively, the reflected intensities and detected SHG yields associated with opposite states of in-plane magnetic saturation, normal to the plane of incidence, i.e., $M_s^{\uparrow}$ and $M_s^{\downarrow}$.

In both the linear and the nonlinear cases the detected intensity changes associated with the response odd in the magnetization are expected to be symmetrical about the reflected intensity or SHG yield when the sample magnetization is zero along a direction (z) perpendicular to the plane of incidence, $I_{M=0}$ and $Y_{M=0}$, respectively.

The response that is even in the magnetization ($\propto M^2$) is the phenomenon of linear magnetic birefringence (LMB) and is seen as a change in either the reflected intensity or the generated SH intensity when the sample is switched between the states $M_z=0$ and $M_z=M_s^{\uparrow}$ or $M_s^{\downarrow}$. Such changes produce a similar magneto-optic response because of the quadratic dependence on M_z.

We can again characterize this effect in both the linear and nonlinear optical fields in terms of a fractional intensity change, associated with magnetization reversal, i.e.,

$$\Delta=\frac{I_{M_s}^{\uparrow}-I_{M_z=0}}{I_{M_z=0}},\quad \Delta_{\mathrm{SHG}}=\frac{Y_{M_s}^{\uparrow}-Y_{M_z=0}}{Y_{M_z=0}}. \tag{3}$$

In the transverse Kerr configuration, at any angle other than normal incidence, the total magneto-optic response, in both the linear and nonlinear fields, is the sum of the contributions from both the diagonal and the off-diagonal elements. Clearly it is not necessary to measure either $I_{M=0}$ or $Y_{M=0}$ to evaluate either δ or δ_{SHG} and these parameters are rarely

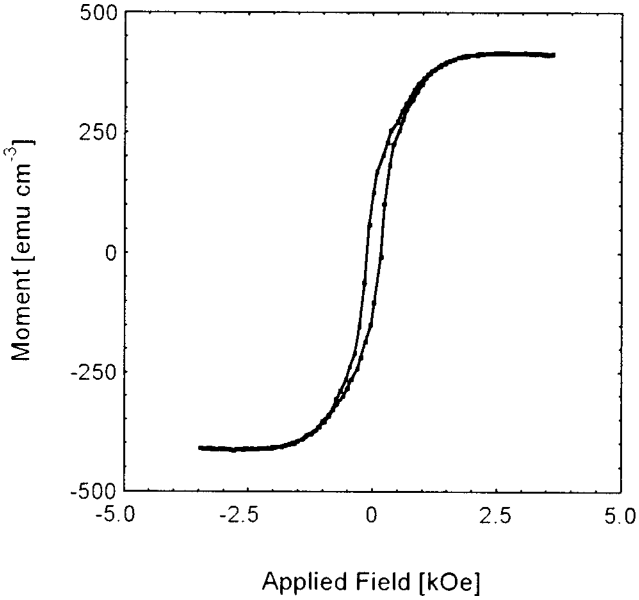

FIG. 1. Magnetic hysteresis loop (vibrating-sample magnetometer) of the sample used in these studies.

FIG. 2. Asymmetry in SHG yield under magnetization reversal in transverse Kerr configuration.

measured. When measuring δ in the linear optical regime the contribution of the diagonal elements is generally small enough to be ignored. Δ is usually so small that it becomes difficult to record, except at normal incidence when the contribution of the off-diagonal elements vanishes.

In these cases, linear magneto-optic effects can therefore be used with confidence to display hysteresis loops or to image magnetic domain structure. Nonlinear magneto-optic effects however are now being used more frequently to characterize both the hysteretic properties and domain structure of magnetic surfaces and interfaces. In such applications, problems will arise when Δ_{SHG} has a magnitude that is comparable to or greater than δ_{SHG}.

The PtMnSb films used in this study were prepared by magnetron sputtering from individual targets of Pt and MnSb (50:50 at %) on to a rotating substrate. The films reported on here were 190 nm thick. As deposited they were amorphous and nonferromagnetic. After rapid thermal processing in vacuum using a 60-W CO_2 laser they crystallized and became magnetic, with the magnetization isotropic in the film plane as shown in Fig. 1. X-ray diffraction analysis showed the resulting samples to be composed of very fine crystallites with the (111) surface predominant.

Prior to making any magneto-optic measurements, the optical behavior of the sample in the demagnetized state was studied in both the linear and nonlinear (SHG) fields. In the linear field the response was standard and the optical constants measured by ellipsometry will be reported elsewhere. Complete ellipsometric measurement of the SHG field was not possible but simple measurements, recording the polarization of the SHG (532 nm) signal as a function of the polarization of the fundamental (1064 nm) beam, were straightforward.

Linear magneto-optic measurements were made using a chopped, highly stabilized quartz halogen source with wavelength selection being achieved using narrow band interference filters and a solid-state detector. The nonlinear measurements were made with a Minilite Nd:YAG (yttrium aluminum garnet) ($\lambda = 1064$ nm) laser emitting Q-switched 5 ns, 25 mJ pulses at a repetition rate of 10 Hz to produce an irradiance of 7×10^7 W cm^{-2} at the film surface. Appropriate filters were used to block SHG signals produced by the input optical train and also to isolate the photomultiplier from the fundamental 1064 nm beam. In both cases reflected or generated intensity values were obtained for the sample states $M_z = 0$ and $M_z = M_s^{\uparrow}$ or $M_s^{\downarrow}$. A Lecroy digital oscilloscope was used to average the detected signals over 500 laser pulses for each data point recorded.

The order in which the results are presented follows the history of their measurement. The nonlinear results were taken first. The subsequent, linear measurements were obtained to confirm the origin of the observed asymmetries in the nonlinear data.

The initial nonlinear studies on the demagnetized sample confirmed fully the results of Reif, Rau, and Mathias,[2] the only previous know nonlinear work on PtMnSb. They reported that the SHG output beam remained p polarized for all linear polarizations of the fundamental 1064 nm beam and that the efficiency of the SHG process was greatly reduced when the input beam was s polarized.

When measuring the magnetization dependence of the SHG yield in the transverse Kerr configuration, we noted significant departures from symmetry of the response about the yield when the magnetization is zero along the z direction. Moreover, the degree of asymmetry showed a marked dependence on the angle of incidence, shown in Fig. 2. Measurements made over a limited range of angles, in the longitudinal configuration, revealed similar asymmetry. Apart from a reversal of sign, essentially identical results were obtained for both left and right circularly polarized, fundamental 1064 nm input beams. Close examination of the longitudinal measurements of Reif, Rau, and Matthias[2] made at a single angle of incidence on a single crystal sample revealed a similar but very small asymmetry presented without com-

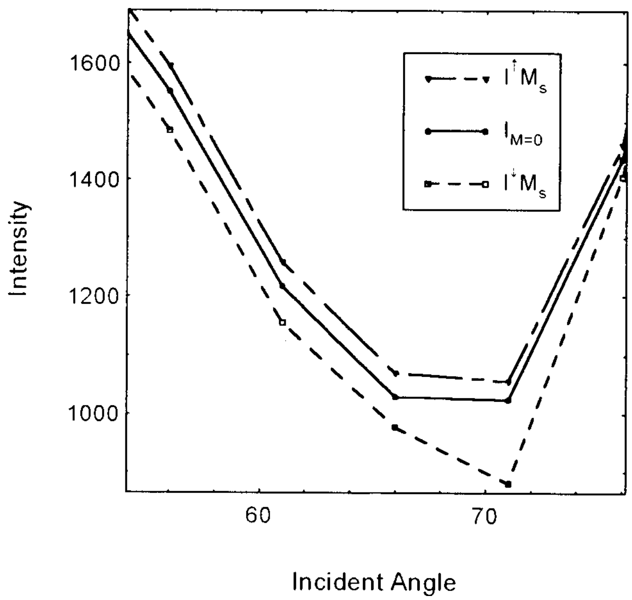

FIG. 3. Asymmetry observed in the linear response in the transverse Kerr configuration (expanded scale).

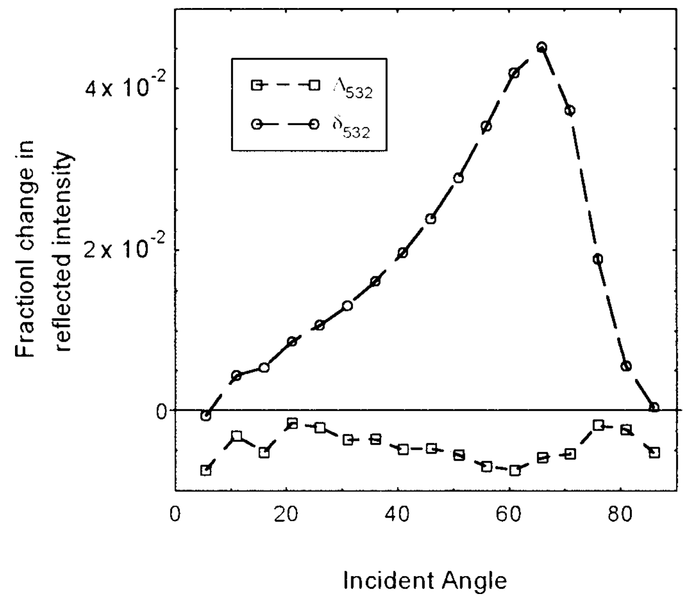

FIG. 4. TKE and LMB components separated from the total observed response in the linear optical field at a wavelength of 532 nm.

ment. Further comparison with the present results is difficult, however, since their SHG Kerr measurements made using circularly polarized light are presented without direct reference to the SHG signal from the demagnetized state.

Careful examination of the linear magneto-optic measurements in the transverse Kerr configuration revealed a similar, but much smaller, asymmetry, shown in Fig. 3. Systematic analysis of the all the results, assuming throughout that the observed response is the sum of contributions both linear and quadratic in the magnetization and that the linear contribution is symmetrical about the condition $I_{M=0}$ permits relatively easy separation of the transverse Kerr effect (TKE) ($\propto M$) and LMB ($\propto M^2$) effects. The respective magnitudes of the TKE and LMB contributions at any given angle of incidence are, of course, wavelength dependent. Here, we show only the results relevant to the present nonlinear studies, i.e., at wavelengths of 532 nm, Fig. 4 and 1066 nm, Fig. 5. The wavelength dependence of the linear results will be presented elsewhere.

This analysis is confirmed for the linear field in Fig. 6, which shows the magnetization-dependent change in reflected intensity as a function of applied magnetic field measured in the transverse Kerr configuration at normal incidence when the TKE ($\propto M$) is identically zero. The loop obtained is clearly the square of that shown in Fig. 1.

Nearly all nonlinear magneto-optic measurements, reported to date, are for single-crystal samples with a centrosymmetric structure. In such cases a nonlinear response arises only from the loss of symmetry in the immediate vicinity of the surface.

When analyzing magneto-optic effects in these materials it is usual to assume quadrapole interactions to be negligible and follow the approach of Pan, Wei, and Shen.[9] The geometry-dependent elements of the dipole nonlinear susceptibility tensor are separated into two classes, odd and even with respect to magnetization (**M**) reversal about a given direction. This approach is not really tenable for the case of polycrystalline samples that are noncentrosymmetric and generate both surface and volume contributions to the SHG field. Moreover, it should be noted that, when the asymmetry is ignored, the calculated δ_{SHG} values are typically of the order of 25–35 %. Such large, magnetically induced, changes in the SHG field are probably greater than any effects attributable to geometric, crystallographic, or surface structure. It seems reasonable, therefore, to assume that the phenomenological approach used in the linear regime can similarly be applied in the analysis of the nonlinear behavior of polycrystalline PtMnSb.

Figure 7 shows the result of applying this analysis to the data from Fig. 2. These results can be compared directly with the linear results shown in Fig. 4. Pavlov *et al.*[10] have demonstrated the existence of a new transversal nonlinear

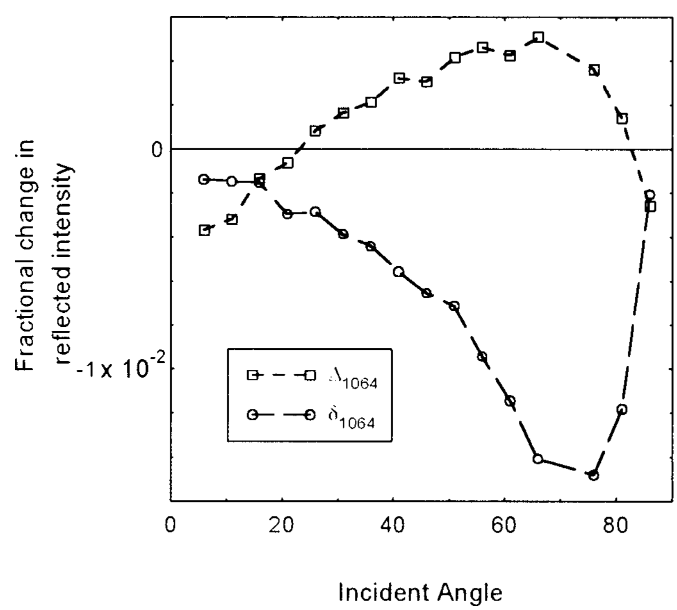

FIG. 5. TKE and LMB components separated from the total observed response in the linear optical field at a wavelength of 1064 nm.

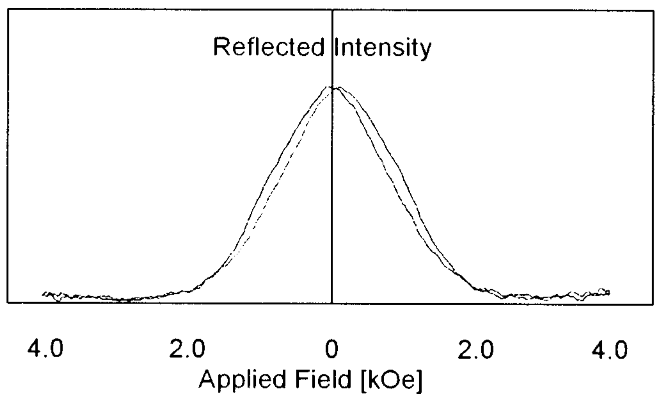

FIG. 6. Hysteresis loop taken magneto-optically using the linear field in the transverse Kerr configuration at normal incidence.

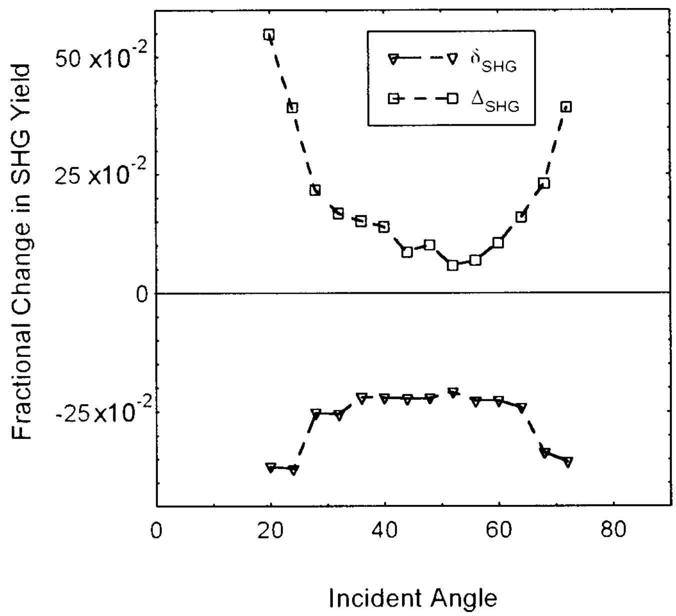

FIG. 7. TKE and LMB components separated from the total observed response in the SHG field (532 nm).

magneto-optic effect. In materials like PtMnSb, in which time-reversal and space-inversion symmetries are broken simultaneously, this effect is linear in M and would not produce the results we observed. We therefore ascribe the asymmetry observed in the detected SHG yield under magnetization reversal to the same mechanism that describes the linear results, i.e., to the simultaneous detection of TKE and LMB effects. The following points should be noted. (i) The sign of both the TKE and LMB changes between linear and SHG fields, (ii) in general, the effects observed in the SHG field are at least an order of magnitude larger than the corresponding effects observed in the linear field and finally, (iii) in the SHG field, the magnitude of the LMB is broadly similar to that of the TKE and makes a significant contribution to the detected signal at most angles of incidence.

Nonlinear magneto-optic effects are increasingly being used to characterize the magnetic behavior of surfaces and interfaces and to image domain structures and some domain wall motion. It has already been pointed out by Osgood *et al.*[11] that the occasionally reported asymmetries in magneto-optic hysteresis loops, recorded using the linear optical field, can be explained in terms of magneto-optic effects even in magnetization. The work reported here indicates that similar caution is required when employing the SHG field for these purposes.

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