Giant low-temperature enhancement of magneto-optic Kerr effects in PtMnSb

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In PtMnSb the magneto-optic Kerr effect exhibits extreme temperature dependence in the range 100–320 K. The large polar Kerr rotation of about $-1.5°$ at photon energies around 1.85 eV at room temperature, is anomalously enhanced to almost $-5°$ as the temperature reaches 80 K. This extreme value is shown to arise because the diagonal elements of the permittivity tensor change with temperature.

In 1983 van Engen *et al.*[1] discovered that the Cl_0 Heusler-type compound PtMnSb exhibits the largest magneto-optic Kerr effects of any metallic system at room temperature. Consequently, the material became the subject of extensive theoretical and experimental study.

Early attempts to understand the origin of the large magneto-optic (MO) effects produced the theoretical discovery[2] that the material has a unique band structure with a gap in the density-of-states of the minority carriers at the Fermi energy. The material was thus characterized as a half-metallic ferromagnet in which the functions of the majority and minority carriers are separated into channels that display metallic and semiconductor behavior, respectively.

The precise origin of the large MO effects has continued to be the subject of extensive study[3–7] and a comprehensive summary of the most credible current theoretical models is offered by Antonov *et al.*[8] Somewhat surprisingly, given the unusual nature of the material and its potential application as a MO recording medium, few attempts have been made to characterize its properties as a function of temperature. The few available reports[9,10] are confined to the measurement of transport properties such as the resistivity and Hall effect; both show some unique but undramatic features.

In this paper, we report measurements of both the optical and MO properties of PtMnSb as a function of temperature. The MO behavior, observed in reflection, exhibits an unexpected and extreme temperature dependence. The permittivity also changes, in a phase-transition-like manner, from metallic behavior at room temperature to dielectric behavior at 80 K.

We have studied the preparation and properties of PtMnSb films extensively since 1990.[11] In the present paper, the MO behavior of PtMnSb films, in reflection, is shown to be extremely temperature dependent in the range 80 to 400 K. Between 100 and 320 K both the polar and transverse Kerr effects depart significantly from the expected temperature variation (that follows the magnetization). Cooling PtMnSb films from ambient temperature to around 80 K increases their saturation magnetization ($\mathbf{M_s}$) by about 16%. The polar Kerr rotation ($\phi_\mathbf{K}$) however, already large and similar to other reported values, increases by some 240% to nearly $-5°$ as shown in Fig. 1. The extreme temperature dependence of $\phi_\mathbf{K}$ in this range becomes very clear in the inset where $|\phi_\mathbf{K}|$ is shown with $\mathbf{M_s}$ from Ref. 9 (both data sets normalized to their 300 K values). Above 320 K the polar rotation follows the magnetization, as expected, but below this temperature it shows an extreme linear change as the temperature is decreased.

Similar behavior is also observed in the transverse Kerr effect as is shown in the inset of Fig. 2. Although access to the cryostat is limited in this configuration, fractional changes in the reflectance, *del* exceeding 15% (on reversal of the magnetization) are observed at temperatures below 150 K. This phenomenon is not confined to the 670-nm wavelength. Spot checks at other wavelengths in the visible and near infrared produce similar behavior. However, in apparent contradiction to these observations in reflection, similar changes are not detected in transmission. The large Faraday rotation [$\approx 8.5°$ for a 180-nm-thick film at $\lambda = 825\,\text{nm}$ (1.5 eV)] exhibits only weak temperature dependence corresponding closely with changes in the magnetization, as shown in Fig. 2.

For analytical purposes, the observed parameters of the effects, in both reflection and transmission, are usually expressed as functions of the diagonal (ε_{xx}) and off-diagonal

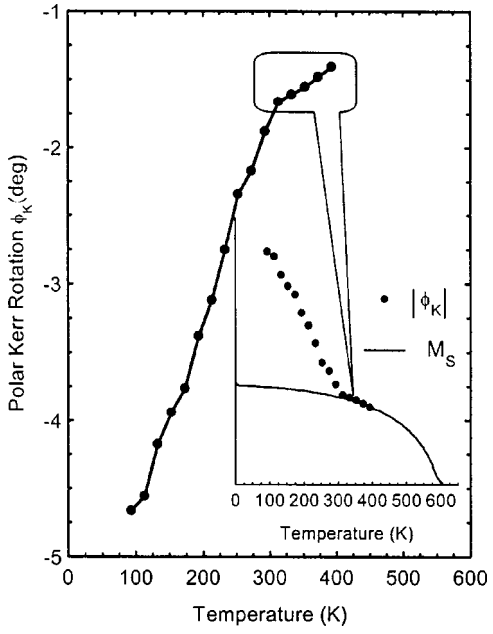

FIG. 1. Temperature dependence of the polar Kerr rotation at normal incidence for PtMnSb films at a wavelength of 670 nm (1.85 eV). Inset: $|\Phi_K(T)|$ compared with $M_s(T)$ Ref. 9. Values normalized at 300 K.

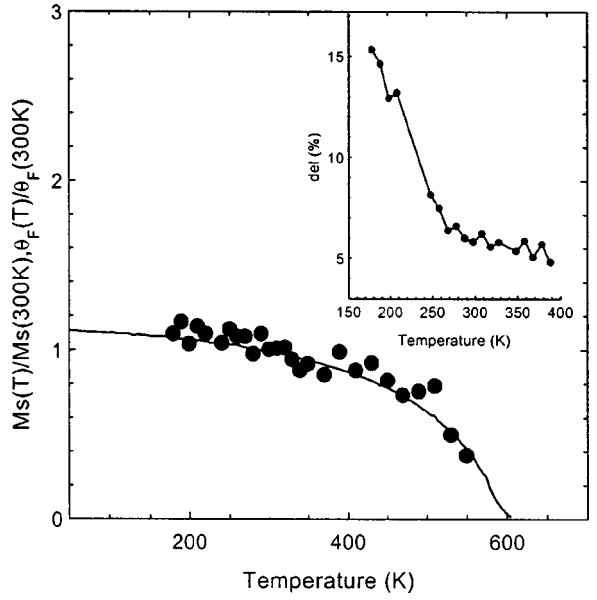

FIG. 2. The reduced Faraday rotation of a 180 nm PtMnSb film at wavelength of 825 nm compared with $M_s(T)$ Ref. 9. Inset: Temperature dependence of the transverse Kerr effect at 670 nm.

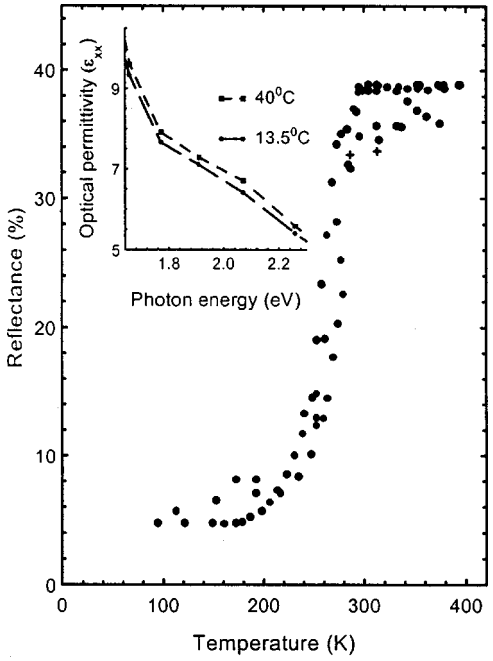

FIG. 3. The normal incidence reflectance as a function of temperature at a wavelength of 670 nm (1.85 eV). Inset: shows the variation with photon energy of $|\varepsilon_{xx}|$ determined from ellipsometric measurements made in air for two temperatures close to ambient.

(ε_{xy}) elements of either the permittivity or the conductivity tensors. In expressions describing both the Faraday and polar Kerr effects,[12] (ε_{xx}) and (ε_{xy}) appear in the denominator and numerator, respectively, as shown below. The same is true for the transverse effect but the expressions are more complicated.

$$\Theta_F - i\varepsilon_F = \frac{\pi \cdot l}{\lambda_0} \frac{\varepsilon_{xy}}{\sqrt{\varepsilon_{xx}}}, \tag{1}$$

$$\Theta_K - i\varepsilon_K = \frac{i\varepsilon_{xy}}{\sqrt{\varepsilon_{xx}}(\varepsilon_{xx}-1)}. \tag{2}$$

However, (ε_{xy}) may be expressed as $Q(\varepsilon_{xx})$, where Q is the magnetization dependent Voigt parameter, which is small and exhibits the temperature dependence of the magnetization. Equations (1) and (2) become

$$\Theta_F - i\varepsilon_F = \frac{\pi \cdot l}{\lambda_0} Q\sqrt{\varepsilon_{xx}}, \tag{3}$$

$$\Theta_K - i\varepsilon_K = \frac{iQ\sqrt{\varepsilon_{xx}}}{(\varepsilon_{xx}-1)}. \tag{4}$$

For a given sample and wavelength therefore, the expression for the polar Kerr effect differs from that for the Faraday effect by the term $(\varepsilon_{xx}-1)$ in the denominator. This, together with the experimental observations, suggests strongly that it is the diagonal elements of the permittivity or conductivity tensor that are responsible for the observed temperature dependence in the polar Kerr effect.

This analysis is confirmed in Fig. 3 where the normal-incidence optical reflectance at a photon energy of 1.85 eV is seen to undergo a rapid, phaselike, transition from a value typical of a poorly conducting metal to that more typical of a dielectric after a 100 K change in temperature. The data presented are the result of several excursions of the sample between 80 and 400 K and are typical of the several samples studied. Like the polar and transverse effects, these measurements were made in a vacuum cryostat. The reflectance is computed from the measured change in the reflected intensity for constant input intensity and referenced to the known reflectance at ambient temperature. The inset shows further evidence. By mounting a Peltier heating/cooling stage on the ellipsometer table it was possible to measure the spectral dependence of the optical constants, in air, at two temperatures 30 K apart and close to room temperature. Even for this limited temperature excursion, it can be seen that the diagonal elements of the optical permittivity tensor ($|\varepsilon_{xx}|$) increase by between 5% and 10% and that the change is largely independent of photon energy.

Such anomalous behavior indicated the possibility that the optical properties of PtMnSb films are likely to be dependent on the incident optical intensity, i.e., they will exhibit nonlinear behavior as a consequence of optical heating. Figure 4 presents the results of an experiment to investigate this possibility using a high power (3 W) semiconducting laser with a wavelength of 801 nm. For this experiment the PtMnSb film is coated with a layer of silicon nitride and the angle of incidence adjusted for minimum reflectance. The laser power was increased incrementally with approximately a 20 s interval between each data point measurement. It can be seen that below and immediately beyond laser threshold the transmitted intensity follows the linear increase in laser power before turning and eventually saturates as heating of the film raises the refractive index. The overall hysteresis is believed to arise as a consequence of the thermal inertia of the sample. These results are presented to emphasize the temperature sensitivity of ε_{xx} in the immediate temperature range above ambient. The experiment is to be published in full elsewhere.

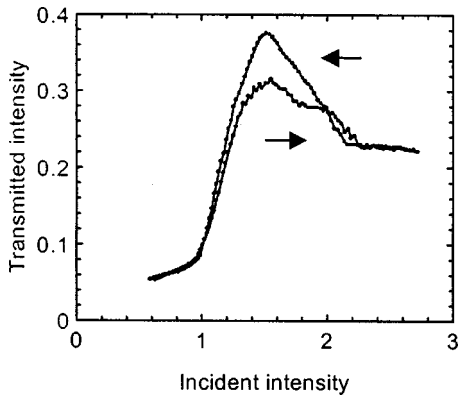

FIG. 4. Nonlinear behavior of the transmitted intensity through an overcoated 180-nm-thick PtMnSb film as a function of incident intensity at angle of incidence for minimum reflectance (→ intensity increasing, ← intensity decreasing).

In all other respects, the behavior of the films under discussion is typical of that for PtMnSb films reported in the literature. In particular they have the large room temperature polar Kerr rotation typical of this alloy.

Energy dispersive x-ray analysis shows the samples to be slightly antimony rich. X-ray diffraction studies indicate the presence of single phase PtMnSb crystallites with a predominant (111) orientation. Moreover, x-ray diffraction curves at two temperatures (278 and 343 K) show no evidence of any phase or structural transition that might give rise to the observed anomaly in the optical behavior. The spectral reflectance curve for the material, calculated from the ellipsometric optical constants and shown in Fig. 5, agrees closely with the curves for bulk samples by Takanashi *et al.*[13] and Krillova *et al.*[4] as well as with those from capped epitaxial films reported recently by Bobo *et al.*[14] Comparison with the

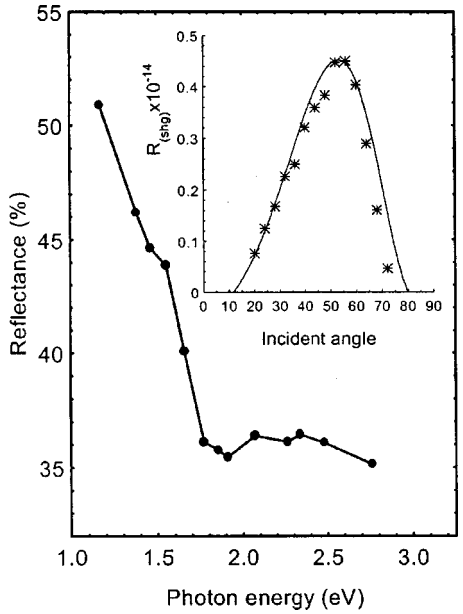

FIG. 5. Reflectance of the PtMnSb films at room temperature as a function of photon energy. Values calculated from ellipsometric measurements. Inset: Experimental SHG reflectance data (*) fitted to a theoretical curve calculated using the free-electron model of Jha (Ref. 17).

results presented by Saito *et al.*[15] for rf sputtered samples also shows very close agreement in both the diagonal and off-diagonal elements of the permittivity tensor.

By fitting the spectral dependence of the room-temperature optical data to Drude theory we obtain the general form of the interband structure and can determine values for the plasma frequency (ω_p) and minority-carrier bandgap E_g. There is some uncertainty in the fitting process since there are relatively few data points and only limited extension into the infrared. Nevertheless, the values so obtained, $\omega_p = 1 \times 10^{16}\,\mathrm{s}^{-1}$ and $E_g = 1.7\,\mathrm{eV}$, are in excellent agreement with those determined by Kautzky[16] from a more extensive data set. In addition we have determined a further, independent, estimate of the plasma frequency from measurement of the angular dependence of the second-harmonic radiation generated (SHG) from the films using a fundamental wavelength of 1064 nm. By fitting these experimental results, shown in Fig. 5 (inset), to the free-electron theory of Jha[17] we obtained a value for ω_p ($4.6 \times 10^{15}\,\mathrm{s}^{-1}$) identical to that calculated theoretically by Antonov *et al.*[8] This lower value is consistent with our temperature-dependent observations. The SHG measurements were made in an air-conditioned laboratory where the ambient temperature is constant while that in the laboratory, where the ellipsometric and magneto-optic measurements were made, is often as much as 10 °C more.

Recent literature contains reports of materials exhibiting a similar temperature response of the polar Kerr effect. Di and Uchiyama[18,19] record that, over a similar temperature range, the polar Kerr effect in Ni films increases by 35% with only a 4% increase in magnetization and in MnBi the corresponding changes are 35% and 50%. However, they also conclude that a similar behavior is expected in the Faraday effect. They suggest that these results are a consequence of spin-orbit (SO) coupling strength varying with temperature because of changes in the electronic structure. This temperature sensitivity is associated primarily with the off-diagonal rather than diagonal elements of the permittivity/conductivity tensor; the optical constants being only weakly temperature dependent.

The temperature sensitivity in the polar Kerr effect reported here is nearly an order of magnitude greater than that reported by Di and Uchiyama and it seems unlikely that the same explanation can be applied. Oppeneer *et al.*[5] identify the importance of the SO on Pt to the giant MO effects in PtMnSb and their sensitivity to the position of the Fermi energy. However, if either of these were strongly temperature dependent it would also be apparent in the behavior of the off-diagonal elements of the permittivity/conductivity. Feil and Haas[3] believe the large room-temperature MO behavior in PtMnSb to derive from interaction of the plasma edge with a resonant transition at around 1.85 eV. Clearly, the very large values of the polar and transverse Kerr effects observed at low temperatures occur because the diagonal permittivity is somehow suppressed.

The mechanism underlying this anomalous optical behavior is puzzling and we can only speculate about its origin at present. Clearer understanding requires, as a minimum, a full spectral study of the optical and magneto-optical behavior at temperatures around 100 K. It is as though the majority electrons in the metallic channel are frozen out, reducing the

free-electron density and producing a downshift in the plasma frequency. Moreover, the fact that small temperature excursions above room temperature produce a significant increase in the diagonal permittivity suggests that the free-electron density or plasma frequency increases with temperature up to about 320 K when the process saturates just as it does below about 160 K. Such behavior is consistent with that of doped semiconductors in which all dopant levels are fully occupied at the lower temperature and exhausted at the higher temperature. Our samples are 3% to 5% Sb rich and it is tempting to consider that this could lead to the creation of states or trapping centers within the minority band gap. Ebert and Shultz[20] have addressed the production of states within the minority band gap for the cases of excess Pt and Mn but not Sb. The experimentally derived density-of-states curve of Kautzky[16] shows a ''tail'' that could be the result of states within the minority band gap. De Groot[21] and others have speculated on the possibility of improving magneto-optic effects in PtMnSb by doping, noting that the observed effects depend critically on the diagonal as well as the off-diagonal elements of the permittivity or conductivity tensor.

There are however several problems with a simple model of thermally activated impurity states. First, it is likely that the presence of states within the gap would adversely affect the minority channel and second, the room temperature optical and magneto-optic behavior of our material is so typical of that produced elsewhere. It is difficult to believe that any doping mechanism alone can reduce the reflectance from that of a typical, poorly conducting metal, to the near dielectric values of 6% to 7% observed at around 100 K. A more sophisticated mechanism for suppressing the majority channel must be sought, discovery of which raises the exciting possibility of achieving very large polar Kerr rotations at room temperature similar to those recorded at around 100 K.

[1] P. G. van Engen, K. H. J. Buschow, R. Jongebreur, and M. Erman, Appl. Phys. Lett. **42**, 202 (1983).

[2] R. A. de Groot, F. M. Mueller, P. G. van Engen, and K. H. J. Buschow, Phys. Rev. Lett. **50**, 2024 (1983).

[3] H. Feil and C. Haas, Phys. Rev. Lett. **58**, 65 (1987).

[4] M. M. Krillova, A. A. Makhnev, E. I. Shreder, V. P. Dyakina, and N. B. Gorina, Phys. Status Solidi **187**, 231 (1995).

[5] P. M. Oppeneer, V. N. Antonov, T. Kraft, H. Eschrig, A. N. Yaresko, and A. Ya. Perlov, Solid State Commun. **94**, 255 (1995).

[6] Long Zhang, Xian-Wu Zou, Hao-Xue Qiao, and Zhun-Zi Jin, J. Appl. Phys. **81**, 4167 (1997).

[7] J. van Ek and J. M. Maclaren, Phys. Rev. B **56**, R2924 (1997).

[8] V. N. Antonov, P. M. Oppeneer, A. N. Yaresko, A. Ya Perlov, and T. Kraft, Phys. Rev. B **56**, 13 012 (1997).

[9] M. J. Otto, R. A. M. van Woerden, P. J. van der Valk, J. Wijngaard, C. F. van Bruggen, C. Haas, and K. H. J. Buschow, J. Phys.: Condens. Matter **1**, 2341 (1989).

[10] J. S. Moodera and D. M. Mootoo, J. Appl. Phys. **76**, 6101 (1994).

[11] G. S. Bains, R. Carey, D. M. Newman, and B. W. J. Thomas, Jpn. J. Appl. Phys. **6**, 6 (1991); J. Magn. Magn. Mater. **104-107**, 1011 (1992); R. Carey, H. Jenniches, D. M. Newman, and B. W. J. Thomas, J. Magn. Soc. Jpn. **17**, 90 (1993); J. Appl. Phys. **75**, 7081 (1994); R. Carey, D. M. Newman, and M. L. Wears, *ibid.* **83**, 6739 (1998); Phys. Rev. B **58**, 14 175 (1998).

[12] J. C. Suits, IEEE Trans. Magn. **8**, 95 (1972).

[13] K. Takanashi, K. Sato, J. Watanabe, Y. Sato, and H. Fujimori, Jpn. J. Appl. Phys., Part 1 **30**, 52 (1991).

[14] J. F. Bobo, P. R. Johnson, M. Kautzky, F. B. Mancoff, E. Tuncel, R. L. White, and B. M. Clemens, J. Appl. Phys. **81**, 4164 (1997).

[15] S. Saito, M. Watanabe, H. Shoji, and M. Takahashi, IEEE Trans. Magn. **33**, 3253 (1997).

[16] M. C. Kautzky, Ph.D. thesis, Stanford University, 1998.

[17] S. Jha, Phys. Rev. **140**, A2020 (1965).

[18] G. D. Di and S. Uchiyama, J. Appl. Phys. **75**, 4270 (1994).

[19] G. Q. Di and S. Uchiyama, Phys. Rev. B **53**, 3327 (1996).

[20] H. Ebert and G. Schutz, J. Appl. Phys. **69**, 4627 (1991).

[21] R. A. de Groot, Physica B **172**, 45 (1991).