Magnetooptical Spectra in Pt/Co and Pt/Fe Multilayers

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Magnetooptical (MO) spectra were measured between 1.2 and 5.9 eV in Pt/Co and Pt/Fe multilayers and corresponding Pt-Co and Pt-Fe alloys produced by DC magnetron sputtering. Real and imaginary parts of off-diagonal conductivity-tensor elements were calculated using optical constants n and k, deduced from the reflectivity spectra measured with synchrotron radiation. The off-diagonal conductivities thus obtained suggest that magnetooptical spectra of multilayers are caused by those of corresponding alloys formed at the interface. To confirm this assumption, a simulation of MO spectra was performed by means of the virtual optical constant method using diagonal and off-diagonal conductivity of the alloy layer determined by the experiment. The results of calculation provide evidence for the presence of an alloy layer at the interface between Pt and transition metal.

KEYWORDS: magnetooptical Kerr rotation, magnetooptical Kerr ellipticity, Pt/Co multilayer, Pt/Fe multilayer, reflectivity, conductivity tensor, virtual optical constant method

§1. Introduction

Recently, multilayers consisting of Pt and transition metal have been intensively studied as new materials for high-density magnetooptical (MO) recording media suited for short-wavelength laser light.¹⁾ Hashimoto et al. already succeeded in recording and reading small-size bits with considerable carrier-to-noise ratio using a 488 nm line of Ar⁺ laser.²⁾ Zeper et al. reported MO spectra in Pt/Co multilayers between 1 and 5 eV.³ Moog et al.⁴ simulated a MO spectrum in multilayers using the new matrix formalism developed by Zak et al.5) and found that the observed MO spectra in the high photon energies cannot be explained solely by the phenomenological multilayer optics unless a strong MO contribution from the Pt atom is assumed. We have also been working on MO studies in Pt/Co multilayers and alloys and have pointed out that MO effects of Pt/Co multilayers are caused by the presence of alloys at the interfaces between layers.6)

In order to elucidate the origin of the large MO effect in multilayers, we measured both the Kerr rotation and ellipticity spectra for photon energies between 1.2 and 5.9 eV in multilayers with several thickness ratios as well as in an alloy film of corresponding composition by means of the polarization modulation technique using a piezobirefringent modulator.⁷⁾

We also measured reflectivity spectra between 0.5 and 25 eV, from which optical constants were calculated using the Kramers-Kronig relationship. Diagonal and offdiagonal elements of effective optical conductivity tensor were evaluated from these spectra. We performed simulation of MO spectra assuming the existence of an alloy layer at the interface. For this purpose we employed a simple virtual optical constant method, which proved to be a powerful tool for explaining the enhancement of the MO effect in Fe/Cu multilayers.⁸⁾

§2. Experimental

Pt/Co and Pt/Fe multilayers as well as corresponding alloy films were prepared using the DC magnetron sputtering technique on glass substrates. Formation of multilayer structure was confirmed using the small-angle X-ray diffraction technique. Layer thickness and composition of films are given in Table I. The first and the second column represent the designed thickness, ratio (or composition ratio) and the designed thickness, respectively. The third, the fourth and the fifth column are the period determined by X-rays, the composition determined by fluorescent X-ray analysis and the realized thickness, respectively. In the last column are listed the thicknesses of Pt and transition metal layers estimated from the composition, where the presence of an alloy layer at the interface is not taken into consideration.

MO Kerr spectra were measured by means of the polarization modulation technique using a piezobirefringent modulator. The apparatus is essentially the same as that described in ref. 7, with some improvements to ensure short wavelength measurements. We used a 150 W Xe-lamp made of fused silica as a light source, a double monochromator of 25-cm focal length with a 1200 groove/mm grating blazed at 200 nm. A CaF₂-type piezobirefringent modulator (Hinds International Type PEM CF3) was employed to modulate optical retardation. The polarizers were Rochon prisms made of MgF₂.

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 Table I.
 Layer thickness (designed), total film thickness (designed), period (measured), composition (analyzed), total thickness (measured) and layer thickness (estimated from composition) of multilayers. Parameters for alloy films are also listed.

Designed values		Measured values			Estimated ratio
Pt/TM (Å) (Å)	Thickness (Å) -	Period (Å)	Composition (Pt at.%)	Thickness (Å)	
Pt(10)/Co(5)	2000	14.0	60.3	1840	Pt(9.5)/Co(4.5)
Pt(18)/Co(5)	500	21.4	73.98	534	Pt(17.0)/Co(4.4)
Pt ₆₀ Co ₄₀	2000		60.2	1915	-
Pt(10)/Fe(5)	2000	14.6	66.98	2007	Pt(10.5)/Fe(4.1)
Pt(20)/Fe(5)	2000	24.3	81.15	2155	Pt(20.6)/Fe(3.7)
Pt ₆₁ Fe ₃₉	2000		58.3	1760	_

Ellipsoidal mirrors were used for focussing optics to avoid chromatical aberration. The signal was detected by a photomultiplier with an InGaAs target and amplified by two lock-in amplifiers. This system covers the wavelength region between 210 and 1000 nm. The details of the apparatus will appear in a later publication.

Reflectivity spectra were measured using a Hitachi U-3410 spectrophotometer between 190 and 2600 nm. The VUV-reflectivity spectra were measured for the first time between 50 and 500 nm using a Seya-Namioka-type spectrometer at the beam line BL-1 in the Synchrotron Radiation Laboratory of ISSP, the University of Tokyo. Both spectra were combined to cover photon energies from 0.5 to 25 eV, with the connecting wavelength around 300 nm, where the continuity of the energy-derivative was observed. Optical constants n and k were deduced from the reflectivity by Kramers-Kronig analysis with appropriate extrapolation parameters determined to reproduce optical constants obtained by spectroscopic ellipsometry.

§3. Results

3.1 Spectra of Pt/Co multilayers and the corresponding alloy

In Fig. 1 are shown spectra of polar MO Kerr rotation and ellipticity in Pt(18)/Co(5), Pt(10)/Co(5) multilayers and the Pt₆₀-Co₄₀ alloy. Kerr rotation and Kerr ellipticity are shown by curves with closed symbols and open symbols, respectively. In this paper the positive sense of the



Fig. 1. Magnetooptical Kerr spectra in Pt(10)/Co(5) and Pt(18)/Co(5) multilayers and $Pt_{60}Co_{40}$ alloy. In this figure closed symbols denote Kerr rotation and open symbols Kerr ellipticity.

Kerr rotation is defined as a clockwise rotation of the linear polarization viewed by an observer confronting the reflected beam, with the magnetic-field direction pointing to the destination of incident light, while the sense of the Kerr ellipticity is determined so that it is consistent with that obtained using the Kramers-Kronig relationship⁹⁾ of the Kerr rotation spectrum.*

Kerr rotation spectra of $Pt_{60}Co_{40}$ and Pt(x)/Co(5)(x=10 and 18) films share a common feature, with a peak at around 3.9 eV and a crossing point of the abscissa around 5.8 eV. Nevertheless, considerable difference is observed in the magnitude of the maximum rotation as well as in the low-energy structure which is observed in the alloy but is lost in multilayers. The overall features of Kerr ellipticity spectra in the alloy and the multilayers are also quite similar to each other. However, the energy position of zero-crossing ellipticity differs from sample to sample.

The reflectivity spectra of the $Pt_{60}Co_{40}$ alloy and the Pt/Co multilayers are shown in Fig. 2. Reflectivity in these films decreases up to 8.5 eV where it takes a minimum and increases again towards higher energies.

Figure 3 shows real and imaginary parts of the diagonal element of the conductivity tensor only below 6 eV. Conductivity spectra for the 0.5-25 eV region offer



Fig. 2. Reflectivity spectra in Pt/Co multilayers and Pt-Co alloy between 0.5 and 25 eV.

^{*}The sense of the rotation was mistaken in our previous paper⁶⁾ and should be reversed, while the sense of the ellipticity should remain unchanged.



Fig. 3. Diagonal element of conductivity tensor in Pt/Co multilayers and Pt-Co alloy between 1.2 and 5.9 eV. Closed symbols denote real part and open symbols imaginary part.

plenty of information on the electronic structures and will be discussed elsewhere.

Following the procedures described in ref. 10, real and imaginary parts of the off-diagonal element σ_{xy} of the conductivity tensor were deduced from the MO Kerr spectra of Fig. 2 using the optical constants *n* and *k* obtained from the Kramers-Kronig analysis of reflectivity spectra, and are illustrated in Fig. 4.*

The curves of σ_{xy} thus deduced for the three samples show unexpectedly good agreement among each other, despite the considerable difference in the raw MO spectra of Fig. 2. This fact seems to suggest that the MO effect in the Pt/Co multilayers is essentially the same as that in the alloy.

3.2 Spectra of Pt/Fe multilayers and the corresponding alloy

Similar experiments were carried out in the Pt(10) /Fe(5) and Pt(20)/Fe(5) multilayers and the $Pt_{61}Fe_{39}$ alloy film. The alloy films of different compositions were also investigated. However, for simplicity only the results in the alloy with the above composition are shown in this paper.

Figure 5 shows polar MO Kerr spectra in Pt(10)/Fe(5)and Pt(20)/Fe(5) multilayers and $Pt_{61}Fe_{39}$ alloys. MO Kerr rotation spectra of the multilayers show a peak around 4.2 eV, which is about 0.3 eV higher than the peak position of Pt/Co multilayers.

The reflectivity spectra of these films are shown in Fig. 6. They show a decrease up to 9 eV, followed by a rather flat part extending to 25 eV. Real and imaginary parts of the diagonal element of the conductivity tensor were deduced by Kramers-Kronig analysis and are shown for photon energies between 1.2 and 5.9 eV in Fig. 7.

Real and imaginary parts of the off-diagonal element of the conductivity tensor were calculated from the MO Kerr spectra of Fig. 5 using the optical constants n and k



Fig. 4. Off-diagonal element of conductivity tensor in Pt/Co multilayers and Pt-Co alloy between 1.2 and 5.9 eV. Closed symbols represent real part and open symbols imaginary part.



Fig. 5. Magnetooptical Kerr spectra in Pt(10)/Fe(5) and Pt(20)/Fe(5) multilayers and $Pt_{61}Fe_{39}$ alloy. In this figure closed symbols denote Kerr rotation and open symbols Kerr ellipticity.



Fig. 6. Reflectivity spectra in Pt/Fe multilayers and Pt-Fe alloy between 0.5 and 25 eV.

obtained from the Kramers-Kronig analysis of reflectivity spectra and are illustrated in Fig. 8. In contrast to the case of Pt/Co the off-diagonal terms of the three films investigated do not show coincidence among each other.

^{*}The curves of σ_{xy} differ substantially from those reported in ref. 6 due to the wrong sign convention of Kerr rotation as described in the preceding footnote.



Fig. 7. Diagonal element of conductivity tensor in Pt/Fe multilayers and Pt-Fe alloy between 1.2 and 5.9 eV. Closed symbols denote real part and open symbols imaginary part.



Fig. 8. Off-diagonal element of conductivity tensor in Pt/Fe multilayers and Pt-Fe alloy between 1.2 and 5.9 eV. Closed symbols represent real part and open symbols imaginary part.

§4. Discussion

From the above experimental results, we have found that the off-diagonal term of the conductivity tensor in Pt/Co multilayers and that of the $Pt_{60}Co_{40}$ alloy film show surprising coincidence with each other, suggesting the presence of interfacial alloy layers in multilayers. In reality, the concentration of Pt(Co) atoms in the Co(Pt) layer shows a gradual decrease with increasing distance from the interface. Small-angle X-ray diffraction can provide the thickness of the intermixed (alloy) layer by analyzing the ratio of the main peak to the satellite peak intensity. From our preliminary experiment, the thickness of the alloy in the Pt/Co multilayer was estimated as 2–3 atomic layers.

In the present work, we simulated MO spectra assuming the existence of the PtCo alloy layer with an average composition at the interface: i.e., the multilayer can be regarded as having a $[Pt/PtCo/Co/PtCo]_n$ structure as illustrated in Fig. 9. As the PtCo at the interface, we assume the ordered alloy of Pt₆₀Co₄₀, taking into account



Fig. 9. A structure assumed for the calculation of MO spectra of the Pt/Co multilayer with an interfacial alloy.

the similarity of the σ_{xy} spectrum of this alloy to that of the Pt(10)/Co(5) multilayer. We performed a simulation of MO spectra of the structure using the virtual optical constant method, which proved to be powerful for explaining the experimental MO spectrum in Fe/Cu multilayers.⁸⁾ In the present calculation, we employed optical constants and off-diagonal elements of Pt₆₀Co₄₀ alloy determined in the present study, as well as the reported values of optical constants of Pt¹¹ and Co¹² and off-diagonal element of dielectric permeability tensor in Co.¹³⁾

We performed the calculation varying the thickness x of PtCo alloy formed at the interface. As stated in the preceding section, the thickness of each layer listed in the last column of Table I was determined from the analyzed composition without assuming interfacial mixing. If an interfacial alloy layer of thickness x is assumed, thicknesses of Pt and Co layers remaining unalloyed should be readjusted so that the total composition for one period remains unchanged. For this purpose, the density of PtCo alloy is necessary, which was estimated assuming that the crystal structure is face-centered tetragonal with lattice constants of a=2.682 Å and c=3.675 Å and that two formula units are present in a unit cell.¹⁴

The simulated Kerr rotation spectra for several values of alloy thickness x are given in Fig. 10. As shown by



Fig. 10. Simulated Kerr rotation spectra in Pt/Co multilayers assuming the presence of an alloy layer with thickness x at the interface.

open circles, the calculation without the formation of the alloy layer (x=0) failed to explain not only the overall spectral feature but also the magnitude of peak rotation in the Pt/Co multilayer. The Kerr rotation are too small to reproduce the observed spectra. This result is consistent with the simulation⁴⁾ using the matrix formalism reported in ref. 5.

Spectral shape as well as peak value of Kerr rotation depends strongly on the thickness of the alloy layer. Thickness x of the alloy layer reaches a maximum when whole Co atoms in the layer undergo alloying. In our Pt(10)/Co(5) multilayer, the maximum value of x is 6.98 Å, for which the spectrum is nearly identical to that of the alloy. The best fit to the experimental curve (dotted line) is obtained for x=6.5 Å. This means a total thickness as great as 13 Å undergoes alloying in one period of 14 Å. A simulation in the Pt(18)/Co(5) multilayer was also performed, in which the alloy layer thickness giving the closest fit to the experiment was determined as 6.7 Å. It is thus found that the thickness of the alloy layer is not dependent on the Pt layer thickness.

Similar calculations were carried out in Pt(10)/Fe(5) multilayers. The results are shown in Fig. 11. In this case lattice constants (a=3.905 Å, c=3.735 Å) were quoted from Buschow's paper.¹⁵⁾ The trend in which the spectral shape varies with the interface layer thickness is quite similar to the case of Pt(10)/Co(5) multilayers shown in



Fig. 11. Simulated Kerr rotation spectra in Pt/Fe multilayers assuming the presence of an alloy layer with thickness x at the interface.

Fig. 10. The overall features of the experimental curve are very close to the simulated spectrum with the maximum alloy thickness 6.4 Å. For the Pt(20)/Fe(5) multilayer, the best agreement was obtained when x equals 5.5 Å. Further details of the simulation will be described elsewhere.

§5. Conclusions

MO spectra and reflectivity spectra were measured in multilayers of Pt/Co and Pt/Fe and alloy films. Simulation by the virtual optical constant method revealed that spectral behaviors of multilayers can be explained by formation of an alloy layer with the thickness of approximately 6.5 Å at the interface between the Pt layer and the transition metal layer comprising the multilayers.

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