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Optical and magneto-optical investigation on electronic structure of ordered ferromagnetic Fe₃Pt

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The optical and magneto-optical properties of ordered Fe₃Pt have been investigated by spectroscopic ellipsometry and magneto-optical Kerr spectroscopy. The diagonal component of the optical conductivity tensor of the compound exhibits a broad absorption peak at about 2 eV, which is shifted by about 0.5 eV to lower energies from the corresponding one in pure bcc Fe. The Kerr angle spectrum of the compound disperses quite similarly in both spectral trend and magnitude to that of pure Fe below 3.5 eV but differently above it. The lower-energy shift of the 2-eV-absorption structure of the compound is interpreted as due to the shift of the minority-spin Fe-*d* states toward E_F through the hybridization with Pt-*d* states. The Kerr effect of the compound is attributable to a large spin-orbit coupling in Pt as well as the well-hybridized spin-polarized bands. © 2001 American Institute of Physics. [DOI: 10.1063/1.1331064]

I. INTRODUCTION

The Fe–Pt alloy system has recently received considerable attention because of its potential applications to magneto-optic recording media. Fe_{*x*}Pt_{1–*x*} alloys in the range $0 < x < 1$ can be both stoichiometric and nonstoichiometric with various degrees of order.^{1,2} For $x \leq 0.25$ ordered and disordered samples have a fcc lattice and a martensitic transformation to a bcc phase occurs for lower Fe compositions. Near $x = 0.5$ an ordered phase of FePt with the CuAu structure can be grown. It exhibits strong tetragonal distortion along the [001] direction, leading to a large magnetic anisotropy and subsequent high magneto-optical Kerr effects (MOKE).³ Also, an ordered phase of Fe₃Pt with the Cu₃Au structure exists near $x = 0.75$.

Magnetically, ordered Fe₃Pt shows ferromagnetism with T_c about 435 K, while for its disordered phase T_c drops to about 260 K.^{4,5} Both phases exhibit anomalies in the thermal-expansion coefficient and spontaneous volume magnetostriction but with different temperature dependencies. Such Invar effects have usually been observed in chemically disordered and magnetically inhomogeneous phases of 3*d* intermetallic alloys such as Fe–Ni and Fe–Pd, giving rise to difficulties in theoretical approach to the effects. Since the discovery of Invar anomalies in ordered and magnetically homogeneous Fe₃Pt, there have been extensive experimental and theoretical investigations on the compound for understanding its electronic and magnetic structures and their relation to the Invar effects.

Theoretical band structure calculations^{2,6} on ordered Fe₃Pt revealed strong hybridization between Fe- and Pt-*d*

states and a resultant unusually high density of states (DOS) at the Fermi level E_F compared to other strong ferromagnets such as Fe, which is thought to provide a necessary condition for a magnetic instability in the compound. Ebert and Akai calculated the absorptive parts of the diagonal and off-diagonal components of the optical conductivity tensor of ordered Fe₃Pt using a spin-polarized linearized muffin-tin orbitals method.⁷ Their calculational results disagree with the present experimental data, presumably because of a different crystal structure, the Fe₃Al structure, they adopted in the calculation. Katayama *et al.* reported MOKE measurements on several kinds of ordered and disordered Fe–Pt alloy films in which annealing leads to substantial reduction in the Kerr rotation angle.¹

In the present work, the optical and magneto-optical properties of ordered Fe₃Pt have been investigated by spectroscopic ellipsometry (SE) and polar MOKE measurements. The diagonal component of the optical conductivity tensor of solids from the SE measurement gives the spectral dependence of electronic interband transitions from occupied to unoccupied states across E_F . MOKE measurements on magnetic solids produce nonzero Kerr rotation and ellipticity due to their different optical response to left- and right-circularly polarized light. Upon reflection from a magnetized surface, normally incident linearly polarized light becomes elliptically polarized with ellipticity ϵ_K and its major axis rotated by the Kerr angle θ_K relative to the polarization axis of the incident beam. The two MOKE parameters are known to arise from the combined effects of net spin polarization and spin-orbit coupling in the magnetic materials. The off-diagonal components of the conductivity tensor can be estimated using the MOKE and SE parameters. The experimental conductivity spectra were compared with theoretical

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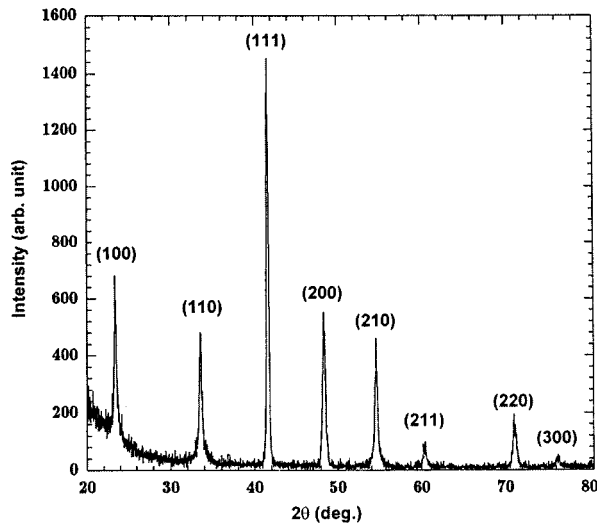


FIG. 1. X-ray diffraction pattern of ordered Fe₃Pt using Al Kα x rays.

spectra evaluated using the spin-polarized band structure of ordered Fe₃Pt, which was calculated by the tight-binding linear-muffin-tin orbitals (TB-LMTO) method.⁸ The electronic and magnetic structures of ordered Fe₃Pt are interpreted through comparison between the experiment and the theory.

II. EXPERIMENT

A polycrystalline Fe₃Pt ingot was prepared by arc melting and subsequent annealing at 600 °C for 24 h. An x-ray diffraction pattern taken with Al Kα radiation of the sample, shown in Fig. 1, indicates that the sample is composed of randomly oriented grains with the well-ordered Cu₃Au structure. Figure 2 shows the result of a magnetization measurement on the sample using a SQUID magnetometer. The magnetization data show that ordered Fe₃Pt is a strong ferromagnet with a saturation magnetic moment of about 9.0 μ_B per formula unit at 7 K. However, a substantial decrease

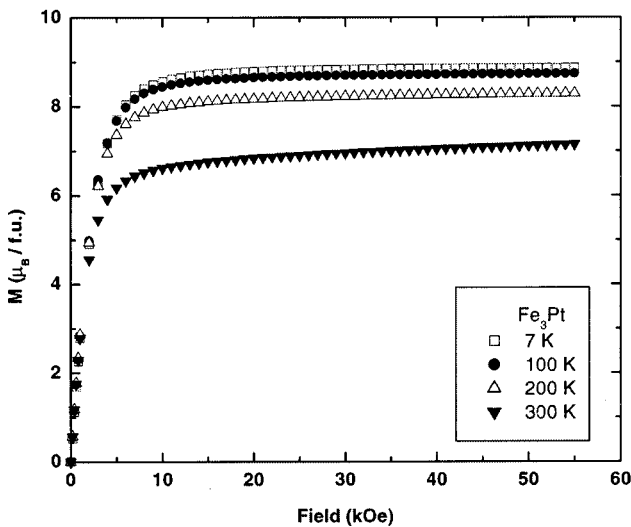


FIG. 2. Magnetization measured on ordered Fe₃Pt.

in the saturation moment between 200 and 300 K is observed, possibly due to a magnetic instability of the compound with temperature.

A planar mirror-like surface was achieved by mechanical polishing with abrasives, the final grade being a paste of 0.05-μm-diameter alumina. SE measurements on the sample were performed at room temperature using an automatic rotating-analyzer ellipsometer in the 1.5–5 eV photon energy region with energy intervals of 0.02 eV. Through the measurement of the complex reflectance ratio ρ (= r_p/r_s) of the p (parallel) and s (perpendicular) field components of the light beam defined with respect to the plane of incidence of the sample the complex dielectric function can be obtained from the equation

$$\epsilon = \sin^2 \phi + \sin^2 \phi \tan^2 \phi \frac{(1 - \rho)^2}{(1 + \rho)^2} \tag{1}$$

by using a two-phase model (air and sample).⁹ The data were taken at an angle of incidence of 68° and a fixed polarizer angle of 45° with respect to the plane of incidence.

MOKE measurement on the sample was performed at 7 K in an optical cryostat in the 1.5–4.5 eV photon energy region with energy intervals of 0.1 eV. A split-coil superconducting magnet in the cryostat can produce magnetic field up to 7 T. The θ_K and ε_K of the sample were measured under a magnetic field of 1.4 T, at which the magnetic moment of the sample is fully saturated (Fig. 2). In the present polar MOKE measurements, an intensity method employing a photoelastic modulator is used, in which the two magneto-optical parameters can be measured simultaneously with high accuracy.^{10,11} The off-diagonal components of the optical conductivity are related to the magneto-optical parameters by¹²

$$\sigma_{1,xy} = \frac{\omega}{4\pi} (-A\theta_K + B\epsilon_K) \quad \sigma_{2,xy} = -\frac{\omega}{4\pi} (B\theta_K + A\epsilon_K), \tag{2}$$

where A and B are given by

$$A = -k^3 + 3n^2k - k \quad B = -n^3 + 3k^2n + n. \tag{3}$$

The optical constants, n and k, of the sample are obtained by SE at room temperature. The angle of incidence in the present MOKE measurement was less than 4°.

III. THEORETICAL CALCULATIONS

The spin-polarized band structure of ordered Fe₃Pt in the Cu₃Au crystal structure was calculated using the TB-LMTO method based on the atomic-sphere approximation including the effect of the spin-orbit interaction. The exchange-correlation potential was included in the local spin-density approximation (LSDA) with the von Barth–Hedin form. The k-integrated functions were evaluated by the tetrahedron method with 144 k points in the irreducible Brillouin zone. The calculations were carried out using an experimentally determined lattice constant a₀ = 3.735 Å with the muffin-tin radii of Fe and Pt of 1.442 and 1.511 Å, respectively.

The calculated spin-polarized band structure of ferromagnetic Fe₃Pt produced a total magnetic moment of 8.13 μ_B per unit cell, smaller than the experimental value of 9.2

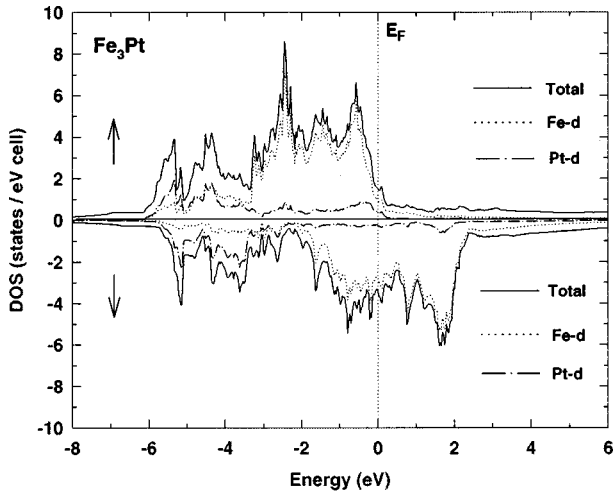


FIG. 3. Spin-resolved density of states of ordered ferromagnetic Fe₃Pt; *d* states from Fe and Pt sites are separately drawn.

μ_B by Ishikawa *et al.*¹³ and the present magnetization measurement result of $9.0 \mu_B$. The magnetic moments of the Fe and Pt sites are evaluated to be 2.60 and $0.33 \mu_B$, respectively, close to the result of Podgorny, 2.54 and $0.21 \mu_B$ from a LMTO calculation,² and rather different from the result of Hasegawa, 2.5 and $0.5 \mu_B$ from an augmented plane-wave method.⁶ The calculational results were reported to be very sensitive to the values of the lattice constant and the approximations made on the shape of the potential.²

The magnetic moment of the Fe site is enhanced compared to that in bcc Fe ($2.22 \mu_B$) while the Pt site also has substantial magnetic moment mostly due to a polarization of Pt-*d* bands through the hybridization with Fe-*d* bands. The nearly filled nature of the majority-spin *d* bands of Fe can explain its enhanced ferromagnetic moment compared to that of pure bcc Fe which has partially filled majority-spin *d* bands.

Figure 3 shows the spin-projected DOS of ordered Fe₃Pt. The *d*-band contributions from Fe and Pt sites are also displayed separately. It is seen that the upper part of the valence bands is dominated by Fe-derived *d* states due to their lower binding energies than Pt-derived *d* states. Although the Fe and Pt subbands are well separated for both spins, hybridization between them is strong enough to have considerable Pt-*d* DOS near E_F . The minority-spin Fe-*d* bands are seen to disperse across E_F and have the largest contribution to the total DOS near E_F , which is larger by about a factor of 2 than that of pure bcc Fe.¹⁴ The calculated total DOS at E_F , $N(E_F)$, for the compound is 4.83 states/eV cell. From the experimental electronic specific heat, one can estimate $N(E_F)$. The coefficient γ of the electronic specific heat is given by

$$\gamma = \frac{\pi^2}{3} N(E_F) k_B^2 (1 + \lambda + \mu), \quad (4)$$

where k_B is Boltzmann's constant and λ and μ the mass enhancement factors due to electron–electron and electron–phonon interactions, respectively. The theoretical value of γ

for ferromagnetic Fe₃Pt (without λ and μ) is 9.57 mJ/mol K² from the present calculation. It is in good agreement with the experimental result of 9.6 mJ/mol K² by Sumiyama *et al.*¹⁵

The interband contribution to the diagonal and off-diagonal parts of the optical conductivity tensor was calculated using Kubo's linear-response theory expressed as

$$\tilde{\sigma}_{xx}(\omega) = \frac{ie^2}{m^2 h \Omega} \sum_{\mathbf{k}} \sum_{n,l} \left(\frac{|\pi_{nl}^+|^2 + |\pi_{nl}^-|^2}{2\omega_{nl}} \right) \times \left(\frac{1}{\omega - \omega_{nl} + i\epsilon} + \frac{1}{\omega + \omega_{nl} + i\epsilon} \right), \quad (5)$$

and

$$\tilde{\sigma}_{xy}(\omega) = \frac{e^2}{m^2 h \Omega} \sum_{\mathbf{k}} \sum_{n,l} \left(\frac{|\pi_{nl}^+|^2 - |\pi_{nl}^-|^2}{2\omega_{nl}} \right) \times \left(\frac{1}{\omega - \omega_{nl} + i\epsilon} - \frac{1}{\omega + \omega_{nl} + i\epsilon} \right), \quad (6)$$

where n and l represent the unoccupied and occupied energy band states at wave vector \mathbf{k} , respectively. The momentum operator is replaced by

$$\boldsymbol{\pi} = \mathbf{p} + \frac{\hbar}{4mc^2} \boldsymbol{\sigma} \times \nabla V, \quad (7)$$

where $\boldsymbol{\sigma} \times \nabla V$ represents the spin-orbit contribution. Then, the matrix elements are expressed by

$$\pi_{nl}^{\pm} = \frac{1}{\sqrt{2}} \langle nk | \mp \pi_x + i \pi_y | lk \rangle, \quad (8)$$

where π_{nl}^{\pm} represent matrix elements for the left- and right-circularly polarized components of the momentum operator, respectively. The theoretical form of σ_{xy} indicates a possibility of extensive cancellation of terms with opposite signs so that $|\sigma_{xy}|$ is typically of the order of 1% of $|\sigma_{xx}|$. Thus σ_{xy} can be a more sensitive test of the electronic structure than σ_{xx} .

IV. RESULTS AND DISCUSSION

In Fig. 4, the real (absorptive) and imaginary (dispersive) parts of the diagonal component ($\sigma_{xx} = \sigma_{1xx} + i\sigma_{2xx}$) of optical conductivity tensor of ordered Fe₃Pt from the present SE measurement are exhibited. A strong interband absorption structure exists around 2 eV in the spectra. Also, the experimental σ_{1xx} spectrum is compared with theoretical spectrum calculated from the spin-polarized band structure of ordered Fe₃Pt in the figure. For a direct comparison with the experimental spectra the calculated spectra were convoluted with a constant Lorentzian broadening factor of 0.5 eV to take into account the lifetime broadening of the excited quasiparticle state. Although the theoretical σ_{1xx} spectrum shows a substantial discrepancy with the experimental spectrum in the absorption strength, it reproduces well the absorption peak near 2 eV as well as the spectral trend in the whole energy range. Due to the high surface sensitivity of ellipsometry, measured conductivities frequently fall below calculated values for rough or contaminated surfaces. So,

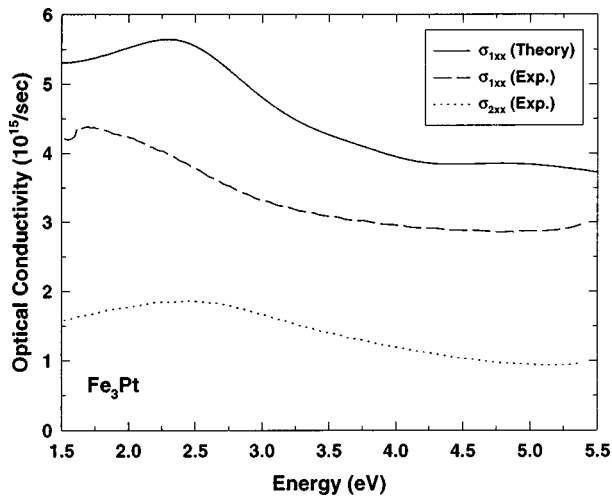


FIG. 4. Real and imaginary parts of diagonal component of optical conductivity tensor (σ_{xx}) of ordered Fe₃Pt. Real part of the conductivity is compared with calculated σ_{xx} .

such a discrepancy can be partly due to surface roughness or an oxide overlayer. The 2 eV absorption in the compound is interpreted as mainly due to direct interband transitions involving Fe-like *d* character in the minority-spin bands distributed across E_F , that is, between occupied *d* and unoccupied *p* ($d \rightarrow p$) and occupied *p* and unoccupied *d* ($p \rightarrow d$). In pure bcc Fe, such absorption structure is located around 2.5 eV.¹⁶ The lower-energy shift of the 2-eV-absorption structure of the compound compared to that of pure Fe is interpreted as due to the shift of the Fe-*d* states toward E_F through the hybridization with Pt-*d* states.

Figure 5 shows the measured θ_K and ϵ_K of ordered Fe₃Pt. The θ_K spectrum exhibits a broad peak at about 3.5 eV, whereas the ϵ_K spectrum shows a large slope in the same energy region, consistent with the Kramers–Kronig relation. θ_K remains negative in the whole energy range, dropping towards lower and higher energies, reaching -0.5° at 1.5 and 4.5 eV. Also, the θ_K spectrum of the compound is simi-

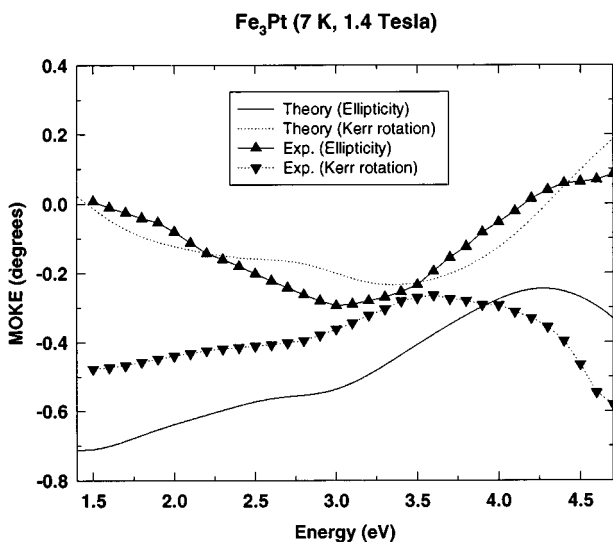


FIG. 5. Kerr rotation (θ_K) and ellipticity (ϵ_K) data for ordered Fe₃Pt. Experimental data are compared with result of theoretical calculation.

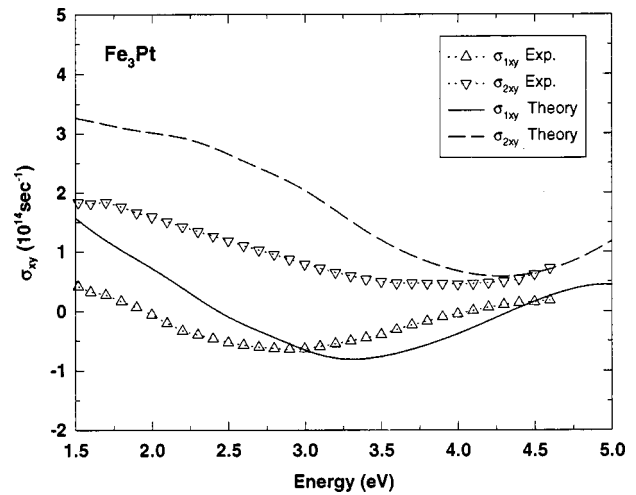


FIG. 6. Real and imaginary parts of off-diagonal component of optical conductivity tensor of ordered Fe₃Pt. Experimental data are compared with result of theoretical calculation.

lar to that of pure bcc Fe^{1,17} below 3.5 eV, in magnitude as well as in spectral trend. Above 3.5 eV, the θ_K of Fe₃Pt goes down to more negative values while that of bcc Fe remains nearly constant at about -0.3° . Although it is now clear that the MOKE is caused by the interplay of net spin polarization and spin-orbit coupling, the origin of the spectral feature of θ_K and ϵ_K has not been understood in detail. Comparison of the experimental θ_K of FePt with FePd¹⁸ showed that the two spectra vary similarly in the whole energy region (1.5–4.5 eV) but the magnitude of θ_K of FePt is larger than that of FePd at higher energies, which is interpreted as due to a larger spin-orbit coupling in Pt than in Pd. Also, by numerically manipulating the spin-orbit coupling strength the magnitude of θ_K was reported to increase at the higher energies for these compounds.¹⁸ Therefore, the Kerr effect of Fe₃Pt is attributable to a large spin-orbit coupling in Pt as well as the well-hybridized spin-polarized energy bands of the compound.

The experimental MOKE spectra are compared with the result of theoretical calculations in Fig. 5. The overall trends of the theoretical spectra reproduce the observed θ_K and ϵ_K reasonably well, except that the peak in θ_K in the theoretical spectrum shifts to higher energies by about 0.5 eV from that in the experimental spectrum and the minimum in ϵ_K in the theoretical spectrum shifts to higher energies by about 0.5 eV from that in the experimental spectrum. Such discrepancy is interpreted as due to errors in using the wave functions and energies of the ground state obtained from LSDA for describing the excited state of transition-metal compounds with localized *d* electrons, i.e., neglecting self-energy corrections. In ferromagnetic Fe, Co, and Ni, the theoretical MOKE structures are usually displaced toward higher energies with respect to the experimental spectra.¹⁹ The shift is the highest for Ni and the smallest for Fe. The discrepancy becomes smaller by increasing the lattice constant because a larger lattice constant leads to a narrower band width.

In Fig. 6, the real (σ_{1xy}) and imaginary (σ_{2xy}) parts of the experimental off-diagonal conductivities of ordered Fe₃Pt evaluated using the measured MOKE and SE spectra are

shown. It is seen that the spectral trend of σ_{2xy} is quite similar to that of σ_{1xx} , exhibiting a broad absorption peak at about 2 eV. Also, the experimental σ_{1xy} and σ_{2xy} spectra are compared with the theoretical spectra in Fig. 6. For absorptive σ_{2xy} , overall experimental features are reasonably well reproduced in the theoretical spectrum except for the magnitude. That is, theory predicts larger values than are experimentally observed, as in the case of absorptive σ_{1xx} in the diagonal component. The spectral trend of dispersive σ_{1xy} looks similar to the ellipticity ϵ_K . Also, for the minimum position in the σ_{1xy} spectrum around 3 eV the theoretical minimum is located at higher energies by about 0.5 eV than the experimental minimum, similar to the case of ϵ_K .

V. SUMMARY AND CONCLUSION

The optical and magneto-optical properties of ordered Fe₃Pt were investigated by SE and MOKE and the results were compared with theoretical calculations based on spin-polarized band structure of the compound. The absorptive parts of the diagonal and off-diagonal components of the measured optical conductivity tensor exhibit broad absorption peaks at about 2 eV. It is interpreted as due to the shift of the minority-spin Fe-*d* states toward E_F through the hybridization with Pt-*d* states. The measured θ_K of the compound varies quite similar to that of pure bcc Fe in both spectral trend and magnitude below 3.5 eV. At higher energies θ_K of the compound becomes more negative while that of pure Fe remains constant. It is attributable to a large spin-orbit coupling in Pt as well as the well-hybridized spin-polarized energy bands of the compound.

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