Non-monotonic variation of anomalous Hall conductivity with spin orbit coupling strength

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Abstract

For L1(0) FePt films, the anomalous Hall resistivity is found to be proportional to spontaneous magnetization $M_S$. After the $M_S$ temperature effect is eliminated, $\rho_{xyo}$ can be fitted by $\rho_{xyo} = a_o \rho_{xx} + b_o \rho_{xx}^2$. $a_o$ and $b_o$ change non-monotonically with chemical long range ordering degree $S$. Accordingly, it is indicated that for L1(0) FePt films the spin orbit coupling strength increases monotonically with increasing $S$.

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Anomalous Hall effect (AHE) in ferromagnetic metallic films has been studied extensively because of its intriguing physics [1–16]. It is shown both theoretically and experimentally that the AHE resistivity is fitted by $\rho_{xy} = a\rho_{xx} + b\rho_{xx}^2$ with the longitudinal resistivity $\rho_{xx}$. One has the anomalous Hall conductivity (AHC) $\sigma_{xy} \simeq -a\sigma_{xx} - b$ when $\rho_{xy} \ll \rho_{xx}$, where $a = -\sigma_{sk}/\sigma_{xx}$ with $\sigma_{xx} = 1/\rho_{xx}$, $b = -\sigma_{sj} - \sigma_{int}$, $\sigma_{sk}$ arises from the extrinsic skew scattering at impurity sites, $\sigma_{sj}$ and $\sigma_{int}$ correspond to the extrinsic side-jump scattering and the intrinsic Karplus-Luttinger terms, respectively [2–4]. Very recently, $\sigma_{int}$ has been recalculated by the integration of the Berry curvature $\Omega(\vec{k})$ of the Bloch states in the Brillouin zone [6, 7],

$$\sigma_{int} = -\frac{e^2}{\hbar} \int \frac{d^3k}{(2\pi)^3} \Omega^z(\vec{k})$$

(1)

It is theoretically shown that as caused by the spin orbit coupling (SOC), $\sigma_{sk}$ and $\sigma_{sj}$ exhibit non-monotonic variations with the SOC strength $\Delta_{SOC} = \xi \vec{l} \cdot \vec{s}$ whereas $\sigma_{int}$ demonstrates monotonic variation [7–9]. There is still a lack of experimental evidence because most such experimental studies are focused on 3d transition metallic films [11–15] and $\Delta_{SOC}$ can be tuned only in a small regime. In contrast, L1(0) FePt films, as important magnetic recording media, may be an ideal object for this purpose because $\Delta_{SOC}$ can be tuned by the chemical long range ordering degree $S$, as demonstrated by the enhancement of magneto-optical Kerr effect (MOKE) and spin Hall effect [17, 18]. More importantly, up to date, the SOC mechanism remains unclear although it is of crucial importance for the magnetism of L1(0) FePt films. For example, $\Delta_{SOC}$ in L1(0) FePt films strongly depends on the orbital polarization of Pt atoms but experimental results about the magnitude of the orbital polarization are controversial [19, 20]. Studies of the AHE in L1(0) FePt films as a function of $S$ are helpful to deeply understand the nature of both the AHE and in particular the SOC in L1(0) FePt films.

In this work, we have studied the SOC effect on the AHE by employing L1(0) FePt films, in which the phase transformation and thus $S$ are easily controlled by varying either deposition or post-annealing conditions. $\rho_{xy}$ is found to change in a linear scale of spontaneous magnetization $M_S$ below 300 K. After the thermally driven $M_S$ reduction is considered, $\rho_{xyo}$ can be fitted in a scale of $\rho_{xyo} = a_o\rho_{xx} + b_o\rho_{xx}^2$. It is interesting to find that $a_o$ and $b_o$ both change non-monotonically with $S$. Based on two dimensional electron gas model [8, 9] and other calculations [7], the present AHE experiments indicate the SOC enhancement in L1(0) FePt films.
A series of 10 nm thick L1(0) Fe\textsubscript{50}Pt\textsubscript{50} (=FePt) films with altering \( S \) were grown on MgO(001) substrates by DC magnetron sputtering at different substrate temperatures. The microstructure and the film thickness were identified by x-ray diffraction (XRD) and reflectometry (XRR), respectively. The films were patterned into normal Hall bar and the Hall resistance \( \rho_H \) was measured from 5 K to 300 K. The longitudinal resistance \( \rho_{xx} \) was also measured in the same temperature regime at zero external magnetic field. In experiments, the magnetoresistance of all samples is less than 0.5\%. \( M_S \) was measured as a function of temperature by PPMS. Polar MOKE spectra were measured at room temperature by a home-made Kerr spectrometer [21].

Deposited at ambient temperature, the FePt film is of disordered fcc structure with (111) preferred orientation. At high substrate temperatures, peaks begin to appear near 24 degrees and 48 degrees corresponding to L1(0) phase (001) and (002) orientations, as shown in Fig. 1(a). It is found that when the substrate temperature is increased, \( S \) as calculated from the intensities of (001) and (002) peaks [22] increases from 0 to 0.86 and the lattice constant along the \( c \) axis decreases from 0.382 nm to 0.374 nm. Therefore, the long range chemical ordering and the lattice distortion happened simultaneously. Due to its crucial importance for calculations of \( \rho_{xy} \) and \( \rho_{xx} \), the film thickness was measured by XRR at low angles, as shown in Fig. 1(b), and found to be 10 ± 0.5 nm.

Figure 1(c) shows typical Hall loops at 5 K. For \( S = 0 \), the Hall loop is slanted with hard axis along the film normal direction. For large \( S \), the loop becomes squared with large coercivity. Apparently, the perpendicular magnetic anisotropy is established in L1(0) FePt films. For the Hall loop of ferromagnetic films, Hall resistivity \( \rho_H = R_O H + 4\pi M(H)R_S \), where \( R_O \) and \( R_S \) are coefficients of ordinary and anomalous Hall effects, respectively. By extrapolating the saturation curve of \( \rho_H \) versus \( H \), the AHE resistivity \( \rho_{xy} \) is achieved and found to decrease for large \( S \) [23]. As shown in Fig. 1(d), for \( S = 0 \) and 0.86 the normalized spontaneous magnetization decreases by about 15\% with increasing temperature from 5 K to 300 K. Apparently, the \( M_S \) reduction cannot be ignored because the Curie temperature of 700-750 K is not sufficiently high [24]. Moreover, for \( S = 0 \) and 0.86, \( M_S \) changes in a linear scale of \( T^2 \), hinting either the excitation of interacting spin waves or long-wavelength, low-frequency fluctuations [13, 25]. For all samples, one has \( M_S = M_0 f(T) \), where \( M_0 \) and \( f(T) \) are the spontaneous magnetization at zero temperature and the temperature dependent factor, respectively.
Figures 2(a) and 2(b) show that $\rho_{xx}$ and $\rho_{xy}$ both increase with temperature but decrease with $S$. $\rho_{xx}$ approaches the residual resistance $\rho_o$ near zero temperature and the latter becomes small for high $S$ possibly due to both improvement of the crystalline quality and reduction of the density of static defects at elevated substrate temperatures. Figures 2(c) and 2(d) show typical curves of $\rho_{xy}/\rho_{xx}$ versus $\rho_{xx}$ as a function of temperature. For all samples, the curves of $\rho_{xy}/\rho_{xx}$ versus $\rho_{xx}$ have downward curvatures and cannot be fitted with the formula $\rho_{xy}/\rho_{xx} = a + b\rho_{xx}$. In order to reveal the relationship between $\rho_{xy}$ and $\rho_{xx}$, they are often measured as a function of temperature. At the same time, $M_S$ generally decreases with increasing temperature for ferromagnetic materials with low Curie temperature. If $\rho_{xy}$ is proportional to $M_S$, one has the following equation.

$$\rho_{xy} = \rho_{xy0} f(T)$$

(2)

As such, the $f(T)$ independent AHE resistivity $\rho_{xy0}$ can be fitted by $\rho_{xy0}/\rho_{xx} = a_o + b_o\rho_{xx}$. As shown in Figs. 2(c) and 2(d), $\rho_{xy0}/\rho_{xx}$ can be fitted by a linear function of $\rho_{xx}$. Such salient linear dependence indicates that for FePt films $\rho_{xy}$ is proportional to $M_S$ in the sampling temperature region. Similar phenomena have been observed in Ni alloys, Heuslers(CoMnSb, NiMnSb, and Co$_2$CrAl), Si-based magnetic semiconductor, and other ferromagnetic compounds [13, 26–31]. It has been pointed out that the skew scattering contribution has linear dependence on $M_S$ [8, 32]. As a result, one has $a = a_o f(T)$ and $b = b_o f(T)$, where $a_o$ and $b_o$ are $f(T)$ independent. The intrinsic $\sigma_{\text{int}}$ is proved to have linear dependence on $M_S$ by both the Karplus-Luttinger model and the integration of the Berry curvature [2, 13], so does $\sigma_{sj}$. Therefore, one has $\sigma_{xy0} \approx -a_o \sigma_{xx} - b_o$, where $a_o = -\sigma_{sko}/\sigma_{xx}$; $b_o = -\sigma_{sjo} - \sigma_{\text{into}}$, and $\sigma_{xy0(\text{sko, sjo or into})} = \sigma_{xy(\text{sk, sj or int})}/f(T)$. In the following, we will become concerned about corresponding $f(T)$ independent physical quantities.

Figures 3(a) and 3(b) show that $a_o$ and $b_o$ change non-monotonically with $S$. For $S = 0$ and 0.86, $a_o$ approaches zero and has a minimal value at intermediate $S$. $b_o$ is equal to 700 (Ωcm)$^{-1}$ for $S = 0$ and increases with a maximal value of 900 (Ωcm)$^{-1}$, and finally approaches a saturation value of 600 (Ωcm)$^{-1}$ for $S = 0.86$. Opposite signs of $a_o$ and $b_o$ indicate that the skew scattering has contribution to the AHC in an opposite way to those of the side-jump and the Karplus-Luttinger terms, as observed in bcc Fe films [15]. Very recently, a new approach has been proposed to fit the data by Tian et al [15], in which $\rho_o$ (induced by impurity) and $\rho_{xxT}$ (contributed by phonon) are considered
to have different contributions in the skew scattering term. For L1(0) FePt films, one has
\[ \rho_{xyo} = a'\rho_o + a''\rho_{XXT} + b'\rho_{xx}^2. \] As shown in Fig. 3(b), the values of \( b_o \) and \( b'_o \) are close to each other for high \( S \). This is possibly because when \( \rho_o \) becomes small, the difference between the new and the conventional approaches becomes negligible as observed in Fe films [15]. More importantly, \( b'_o \) also exhibits non-monotonic variation with \( S \) similarly to \( b_o \). Finally, the AHE of L1(0) FePt films with \( S = 0.8 \) has very recently been studied [16], in which values of \( a \) and \( b \) are dramatically different from the present results of \( a_o \) and \( b_o \), which is likely because all data in Ref.[16] were analyzed from \( \rho_{xy} \) instead of \( f(T) \) independent \( \rho_{xyo} \).

It is easy to understand the non-monotonic variations of \( a_o \) and \( b_o \) according to theoretical models about the AHC [7–9] under the assumption that \( \Delta_{SOC} \) increases monotonically with \( S \), i.e., the SOC constant \( \xi \) of Pt and Fe atoms is *equivalently* enhanced during the phase transformation. Here, the effect of the exchange split energy on the AHC can be neglected although the AHC arises from the interplay between the SOC and the exchange split. This is because the effective spin magnetic moment does not change much during phase transformation and thus the exchange splitting energy between spin-up and spin-down bands is expected to change little with \( S \) [19, 20]. Firstly, Sinitsyn *et al* have studied the AHC dependence on \( \xi \) in the two-dimensional Dirac model system by a modified semiclassical transport approach [8]. According to Eq.72 in this literature, \( \sigma_{sko} \) has a minimum at \( \Delta_{gap}/vk_F \simeq 0.5 \) with \( \Delta_{gap} \propto \xi^2 \) and \( v \) being the model parameter [16, 33]. Alternatively, in the framework of quantum transport theory, Wölle and Muttalib have studied the AHE in (quasi) two-dimensional disordered metallic band ferromagnet [9]. According to Eq.12 in Ref.[9], \( \sigma_{sko} \) achieves a minimum at \( \xi/\xi_o \simeq 0.8 \) with \( \xi_o \) being the bare one. Since \( \sigma_{xx} \) is independent of \( \xi \), the non-monotonic variation of \( a_o \) in Fig. 3(a) is easily understood.

Secondly, as the sum of \( \sigma_{sjo} \) and \( \sigma_{into} \), the non-monotonic variation of \( b_o \) in Fig. 3(b) further confirms above assumption. On one hand, \( \sigma_{into} \), which exists in perfect crystals, is theoretically predicted to increase linearly at small \( \xi \) and to reach saturation for large \( \xi \) as shown by Fig.4 in Ref.[7] and Eq.58 in Ref.[8]. It is indirectly confirmed by the Kerr rotation and the ellipticity enhancement in L1(0) FePt films, compared with the disordered FePt films, as shown in Fig. 4 [17]. By utilizing sum rules for the optical constants [34], the intrinsic AHC (at the circle frequency \( \omega = 0 \)) is suggested to be enhanced because the intrinsic AHC has the same origin (both spin polarization and SOC) as the
MOKE [7]. On the other hand, since $\sigma_{sjo}$ obeys the $\xi$ dependence similar to that of $a_o$ as shown by Eq.76 in Ref.[8] and Eq.17 in Ref.[9], it also changes non-monotonically with $S$. Furthermore, it should be pointed out that the impurity state can be excluded for the non-monotonic variation trends of $a_o$ and $b_o$. Since $\sigma_{sko}$ and $\sigma_{xx}$ are both inversely proportional to the impurity concentration, as the ratio $\sigma_{sko}/\sigma_{xx}$, $a_o$ is independent of the impurity concentration. $\sigma_{sjo}$ is also independent of the impurity concentration albeit it arises from the interplay of the impurity scattering and the SOC [10]. As a consequence of the SOC in ideal crystals, $\sigma_{into}$ is not related to the impurity state at all. According to the theoretical prediction by Yao, Sinitsyn, and Wölffe et al [7–9], the present experimental results indicate that the $\Delta_{SOC}$ increases with increasing $S$.

Theoretical calculations are encouraged to address observed features of the AHC in L1(0) FePt films with varying $S$. Here, well defined crystalline structure in L1(0) FePt films favors direct comparison between experiments and calculations. Although $\sigma_{int}$ is proportional to $M_S$ in the temperature regime from 0 K to 300 K, the SOC effect on the AHC cannot be taken into account by perturbation approach because $a_o$ and $b_o$ vary non-monotonically with $\xi$ as discussed above. Since $M_S$ is shown to obey linear dependence on $T^2$, the $M_S$ reduction is proposed to be caused by long-wavelength, low frequency fluctuation of spin orientation at finite temperatures by Zeng et al [13]. Accordingly, this difficulty in theory is overcome. For L1(0) FePt films with small $S$, $\sigma_{sj}$ also shows the linear dependence on $M_S$, however, different from observations of Mn$_5$Ge$_3$ where $\sigma_{sj}$ is neglected. Furthermore, the increase of $\Delta_{SOC}$ with $S$ indicates the enhancement of orbital polarization of Pt atoms due to chemical long range ordering because the SOC at Pt sites plays a dominant role in the magnetism of L1(0) FePt alloys [35, 36], although the orbital polarization of Fe atoms is observed to be enhanced by about 300% [19, 20]. In calculations, the lattice distortion and the chemical ordering should be taken into account because the former and the latter ones have great impact on the orbital polarization in the magnitude and the anisotropic distribution, respectively.

In conclusion, it is likely the first time to have studied the AHC dependence on $\Delta_{SOC}$ in experiments, by employing L1(0) FePt films. As the ratio $\rho_{xy}/f(T)$, $\rho_{xyo}$ can be parameterized by $\rho_{xyo} = a_o\rho_{xx} + b_o\rho^2_{xx}$. Accordingly, $\rho_{xy}$, $\sigma_{xy}$, $\sigma_{sk}$, $\sigma_{int}$, and in particular $\sigma_{sj}$ are all proportional to $M_S$ as a function of temperature in the regime of 0-300 K. It is interesting to find that $a_o$ and $b_o$ change non-monotonically with $S$. Accordingly, $\Delta_{SOC}$ is
verified to be enhanced in L1(0) FePt films and to increase monotonically with $S$. The present state of the art results will also be helpful to study electronic structure of L1(0) FePt films.

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FIGURE CAPTIONS

Figure 1 Typical XRD spectra at high angles (a) and XRR at low angles (b), Hall loops at 5 K(c), and temperature dependence of the normalized spontaneous magnetization, i.e., \( f(T) \)(d). In (d), the lines refer to linear fit results.

Figure 2 \( \rho_{xx} \)(a) and \( \rho_{xy} \)(b) versus temperature for FePt films with various \( S \), the ratio (\( \alpha \)) of \( \rho_{xy}/\rho_{xx} \)(black squares) and \( \rho_{xyo}/\rho_{xx} \)(red circles) versus \( \rho_{xx} \) for \( S = 0.86 \) (c) and 0 (d). Here, symbols in (a)-(d) refer to measured results, solid lines for \( \rho_{xyo}/\rho_{xx} \) curves in (c) and (d) to fitted results by the linear function, and other lines in (a)-(d) serve a guide to the eye.

Figure 3 \( a_0 \)(a), \( b_0 \) and \( b'_0 \)(b) as a function of \( S \), which are fitted from the curves of \( \rho_{xyo}/\rho_{xx} \) versus \( \rho_{xx} \).

Figure 4 Polar Kerr rotation \( \theta_K \) (a) and ellipticity \( \epsilon_K \) (b) spectra of FePt films with \( S = 0 \) and 0.86. During measurements, the samples were in the saturation state. Lines serve a guide to the eye.
FIG. 1:
FIG. 2:
\( b_0 \) and \( b'_0 \) (10^3 \text{ cm}^{-1})

FIG. 3:
FIG. 4: