Inverse spin Hall effect in Cr: Independence of antiferromagnetic ordering

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Inverse spin Hall effect in Cr: independence of antiferromagnetic ordering

D. Qu[1], S. Y. Huang[2, 1], and C. L. Chien[1*].

[1] Department of Physics and Astronomy, Johns Hopkins University, Baltimore, Maryland, 21218, USA

[2] Department of Physics, National Taiwan University, Taipei, 10617, Taiwan

Abstract:

Chromium is a *3d* spin density wave antiferromagnetic (AF) metal with a Néel temperature of 311 K. By using the thermal spin injection method, we have observed large inverse spin Hall voltage in Cr. With negligible magnetic proximity effect and a large spin Hall angle, Cr is a superior intrinsic pure spin current detector than many *5d* metals. Even more strikingly, the temperature dependent thermal spin injection measurements show that the large spin Hall angle in Cr is independent of the AF ordering and indicating that the inverse spin Hall effect is unrelated to AF ordering in Cr.

The exploration of spintronic phenomena has evolved from manipulating spin-polarized current to that of pure spin current, which may be generated by a few mechanisms, including spin Hall effect (SHE) [1], spin pumping [2], spin Seebeck effect (SSE) [3], and others. On the other hand, the inverse spin Hall effect (ISHE) in a metal with strong spin-orbit coupling can be used to detect pure spin current by converting it to a charge current as described by $\boldsymbol{J_C} \propto \theta_{SH} (\boldsymbol{J_S} \times \boldsymbol{\sigma})$. As the result, a charge current $\boldsymbol{J_C}$ is generated in the direction perpendicular to the spin current $\boldsymbol{J_S}$ with spin index $\boldsymbol{\sigma}$, and θ_{SH} is the spin Hall angle, which quantifies the conversion efficiency between the charge current and the spin current. Since the strength of the spin-orbit coupling scales with Z^4 [4], where Z is the atomic number, high-Z metals mostly *5d* and *4d* metals such as Pt, Au, and W have been extensively explored as pure spin current detectors/generators because of the expected large θ_{SH}. In contrast, *3d* metals, such as Cu, have very small θ_{SH} thus has very low efficiency in converting spin/charge current.

Recently, the prospect of anomalous Hall effect (AHE) in some antiferromagnets (AF) [5] has been suggested theoretically. First principle calculations have shown that IrMn, an antiferromagnet (AF), may acquire a large anomalous Hall conductivity due to its non-collinear AF spin structure and its strong spin-orbit coupling. Although the spin Hall effect in antiferromagnets, such as PtMn, IrMn, and PdMn, have been studied experimentally, the relationship between the spin Hall effect and the antiferromagnetic ordering has not been well established. Since these materials already contain heavy elements with strong spin orbit coupling, it is more difficult to study the mechanism of spin Hall effect in these antiferromagnets [6, 7]. Therefore, the AF metals with small atomic number would be better choices. Very recently, a large inverse spin Hall effect has been observed in *3d* AF Cr by spin pumping experiment [8]. The observed θ_{SH} in Cr is nearly 50 times larger than the theoretical value calculated for Cr [9]. These unexpectedly results lead to the intriguing question of whether the large θ_{SH} in Cr is due to its AF spin structure.

It is interesting to explore the prospect of ISHE in chromium (Cr) for several reasons. Firstly, Cr is a *3d* metal, thus a sizable θ_{SH} would be significant in view of the very small θ_{SH} in other *3d* metals such as Cu. Secondly, Cr metal exhibits an incommensurate spin-density wave ~~(SDW)~~ AF ordering below the Néel temperature (T_N) of 311 K, which allows one to explore pure spin current effects in the paramagnetic as well as the AF states. Thirdly, many *5d* metals except Au on YIG, such as Pt/YIG, Ta/YIG, and W/YIG, show evidences of magnetic proximity effects (MPE) [10, 11], which include the intriguing magnetoresistance (MR) and anomalous Hall effect. It remains to be revealed whether similar MPE also exists in Cr with a smaller value of Z in contact with a ferromagnetic insulator. Finally, theories suggest that AF materials may play key roles in future spintronic phenomena and devices [5]. The ISHE in AF with a low atomic number, such as Cr, is one basic phenomenon for exploration.

In this work, by using the thermal spin injection method, we observed strong ISHE in Cr with a spin Hall angle θ_{SH} comparable to that of W, and larger than those of Ta and Au. Moreover, Cr on YIG does not develop any anomalous Hall signal that appears in Pt, Ta, and W on YIG [12]. We also perform measurements from low temperature to above T_N, showing that the AF order in Cr plays a negligible role in its sizable spin Hall angle. As such, Cr can be an effective pure spin current generator/detector, even better than W and Pt, due to its large spin Hall angle and negligible MPE.

We use polycrystalline Cr thin films of various thicknesses *t* deposited by magnetron sputtering onto thermally oxidized Si or polycrystalline ferromagnetic insulator yttrium iron garnet (YIG) substrates, as shown in Fig.1 (a). The bulk YIG we used is 0.5 mm thick with a rectangular shape of 3 mm by 5 mm. The x-ray diffraction measurement in Fig. 1(b) indicates that the Cr film is polycrystalline with a preferred (110) texture. We patterned the Cr/Si and the Cr/YIG films into a Hall bar structure for the thermal measurement and electrical measurements of resistivity, magnetoresistance, and Hall effect. The width and length of the Hall bar is 0.2 mm and 4.5 mm, respectively.

In the magnetoresistance (MR) measurement an in-plane magnetic field is applied. The resistivity of Cr (6 nm)/YIG is 89 μΩ–cm. As shown in Fig. 2 (a), there is a very small MR in Cr (6 nm)/YIG with an anisotropic MR (AMR)-like behavior, where the field parallel to charge current ($\rho_{\|}$) has a larger resistivity than when field is transverse to the current direction (ρ_T). However, the small MR ratio of 1.0×10^{-5} is about five times smaller than that in W/YIG and one order of magnitude smaller than that in Pt/YIG at the same metal thickness. For Cr/YIG with a thicker Cr, the MR ratio reduces indicating an interfacial origin. The control sample of Cr/Si does not show any measurable MR behavior in the field range of ± 1 kOe.

The unexpected MR in Pt/YIG has recently attracted a great deal of attention due to its unusual characteristics and intriguing origins. Experiments suggest contributions from both spin current and magnetic proximity effect [13]. The MR obtained for Cr/YIG can also be ascribed to these two mechanisms. In the small field region (< 2 kOe), the main contribution to the MR of Cr/YIG is spin current. The intriguing MR in Pt/YIG was first proposed theoretically as the spin Hall MR due to the conversion of spin/charge current within the metallic layer [14], although a more recent theory [15] claims to account for the same MR characteristics but only relied on the interface. At any rate, the magnitude of the MR observed in Cr/YIG is at least an order of magnitude smaller than that in Pt/YIG and barely observable.

In the Hall measurement of both Cr/Si and Cr/YIG with thickness *t* from 6 to 15 nm, we observe only ordinary Hall effect with Hall voltage linearly dependent on the magnetic field in the temperature range of 5 – 200 K and field range of ± 5 T. However, unexpectedly in the thinnest 3 nm Cr sample, in addition to the ordinary Hall effect, we have also observed a clear anomalous Hall effect (AHE) signal for Cr(3 nm)/YIG as shown in Fig. 2 (b). The AHE signal appears above 50 K and remains observable at 300 K. However, we find that the AHE signal also appears in Cr(3 nm)/Si as shown in Fig. 2(c), thus unrelated to the presence of YIG. We note that the unexpected AHE in thin Cr has been reported earlier, but of unknown origin [16]. However, in our case, after capping the 3 nm Cr with a 1.2 nm Si layer, the AHE contribution disappears and only the linear ordinary Hall effect remains, as shown in Fig. 2 (d). Therefore, the AHE in the thinnest unprotected Cr layer comes from surface Cr oxidation resulting in ferromagnetic Cr oxides. It is noteworthy that both Cr/YIG and Cr/Si show no indication of magnetic proximity effect (MPE), in sharp contrast to the *5d* metal on YIG, such as Pt/YIG and W/YIG [12]. Therefore, Cr can be used as an intrinsic pure spin current detector.

We use longitudinal spin Seebeck effect (LSSE), an established method for generating pure spin current via the vertical temperature gradient, to inject spin current from YIG into the adjacent metal layer. The spin current is then converted into a charge current by the ISHE and we measure the ISHE voltage as illustrated in Fig. 1(a). The vertical temperature gradient is accomplished by placing the sample between two Cu blocks maintained at two different uniform temperatures. Most of the temperature drop occurs in YIG, because its thickness is five orders of magnitude larger than that of the metal thin film.

As shown in Fig. 3 (a), the sign of the ISHE voltage in Cr/YIG is opposite to those in Pt/YIG and Au/YIG [12], thus instantly reveals that the sign of θ_{SH} in Cr is opposite to those of Pt and Au. The opposite sign of θ_{SH} is due to the less than half filled *3d* shell in Cr. The ISHE voltage is antisymmetric in field and is proportional to the temperature gradient. For 6 nm Cr on YIG, a sizable thermal voltage of - 4.7 μV is observed from the positive saturation field to the negative saturation field, measured from a wire length of 4 mm, under a temperature gradient of 20 K/mm as shown in Fig. 3 (a). In the control sample of Cr (30 nm)/Si, there is no discernable voltage under the same temperature gradient because the lack of spin current. This is also different from the sizable anomalous Nernst effect (ANE) observed in the ferromagnetic metal Py/Si, because the absence of net magnetization in AF metal Cr [17]. These results indicate that the spin current generated from YIG is converted to a charge current in Cr due to ISHE alone, and Cr itself does not generate any measurable thermal voltage at the transverse direction under a perpendicular temperature gradient. Therefore, the thermal voltage observed in Cr/YIG can be attributed solely to the ISHE in Cr.

We recently demonstrated a method for the determination of the relative spin Hall angle and the spin diffusion length of a material by thermal spin injection [12]. It entails measuring the electrical resistivity and thermal voltage of a series of films of different thicknesses and self-consistently determining the spin current conversion to charge accumulation. For all metal films, the resistivity is constant in thick layers but increases with decreasing thickness at small thicknesses due to surface scattering. Therefore, the thickness dependence of resistivity must be established experimentally and measured in each case. The results of the Cr films are shown in Fig. 3(b). The resistivity for very thick films is about 25 μΩ-cm, but its value rises sharply to a few hundreds of μΩ-cm for the thinnest films. The measured thermal voltage decreases with increasing Cr thickness as shown in Fig. 3(c) due to the decay of spin current. The thermal voltage ΔV_{th}, the Cr film thickness t, and resistivity $\rho(t)$ can be described by the following relation

$$\Delta V_{th} = 2[CL\,\nabla T]\,[\rho(t)\;\theta]\,[(\lambda_{SF}/t)\tanh(t/2\lambda_{SF})] \qquad (1)$$

where C is spin injection efficiency related to the magnetic properties of YIG and the spin mixing conductance at the interface, L is the length of the patterned Cr film, ∇T is temperature gradient, θ_{SH} is the spin Hall angle, and λ_{SF} is the spin diffusion length. In our measurements, ∇T and L have been fixed, whereas t, $\rho(t)$, and ΔV_{th} have been measured for every film. Assuming a constant C, by fitting the values of $\Delta V_{th}/\rho(t)$ to Eq.(1) we can extract the spin Hall angle of Cr relative to that of Au and a spin diffusion length for Cr. Our results can be described by Eq. (1) very well as the sold line in Fig. 3(d). We obtain the relative spin Hall angle of -1.38 for Cr (relative to Au), which is comparable to that of W, and larger than that of Ta, assuming the same interface spin current transport efficiency. A comparison of the spin Hall angle and spin diffusion length of Cr with those of the other *5d* metals is shown in Table 1. The spin diffusion length of Cr is 2.1 nm, comparable to the values in the literature [18].

It has been known that *3d* metals with low Z should have very small spin Hall angles comparing to those of the *5d* metals. For example, the spin Hall angle in Cu is only 0.0032, which is nearly 20 times less than the *5d* metal Au, and 2 times smaller than the *4d* metal Ag [19]. However, the determined spin Hall angle in the *3d* metal Cr by LSSE is large and comparable to the *5d* metal W.

Due to the unusual spin density wave antiferromagnetic ordering in Cr, it is important to investigate whether the large spin Hall angle in Cr is in anyway related to its AF order. Thus, we performed measurements from 30 K to 345 K with Cr in the AF state to above T_N of 311 K of bulk Cr. We measured the thermal voltage of a 10-nm thick Cr film with 1.2 nm Si capping layer from 30 K to 345 K in a cryostation. For bulk Cr, T_N is 311 K but reduces to 300 K for the 80-nm Cr film on YIG substrate from resistance measurements. For thinner Cr layers, T_N is even lower. Thus, the temperature range of 30 – 345 K should cover T_N of all Cr samples, bulk or thin films. A Cernox thermometer reads the temperature of the sample on the cool side and the resistivity of the Cr film is measured on the hot side. By adjusting the heater power we keep the temperature gradient on the Cr/YIG sample close to 20 K/mm. According to Eq. (1), the ISHE voltage depends on temperature gradient and resistivity of the film. Therefore, we plot $V/(R\Delta T)$ vs. hot side temperature in Fig. 4 (a) to capture the intrinsic temperature dependent behavior of the Cr/YIG system.

From 345 K to 100 K, the V/(RΔT) of Cr/YIG steadily increases before sharply decreasing at lower temperatures. This strong temperature dependence may be related to the AF ordering in Cr. However, in the control experiment with 5-nm Pt film on YIG, the values of V/(RΔT) show virtually the same dependence as shown in Fig. 4 (b). Therefore, the temperature dependence of the ISHE voltages in both Cr/YIG and Pt/YIG is unrelated to the AF ordering in Cr but due to the thermal injection mechanism and the physical properties of YIG. Thermal injection mechanism using ferromagnetic insulator relies on thermally excited magnons, which freeze at very low temperature. Thus the ISHE voltage reduces towards zero as 0 K is approached as experimentally observed. The maximum ISHE voltage at 100 K and decreasing at higher temperatures are likely due to the change of the magnon properties in YIG. Recently, numerical calculations of the YIG magnon thermal conductivity show that the temperature dependence of YIG magnon thermal conductivity is non-monotonic with a maximum around 70 K [20]. At low temperatures only low-energy magnons with long wavelength are excited while at high temperatures the lifetime of high-energy magnons with short wavelength decreases significantly. The same temperature dependences for Pt/YIG and Cr/YIG demonstrate that the AF ordering in Cr has no apparent effect on the ISHE and the spin Hall angle of Cr.

We are aware of the very small theoretical spin Hall angle (-0.15%) for Cr assumes Cr to have a hexagonal structure and not bcc [9]. The very small calculated value is also incompatible with the experimental value we observed by DC based thermal spin injection, nor the large value of (-5%) obtained by Wang *et al.*, using spin pumping [8]. The large spin Hall angle in Cr remains to be accounted for theoretically but the AF order in Cr appears to be irrelevant.

In conclusion, we report a large ISHE in the *3d* metal Cr, with a very large spin Hall angle and a short spin diffusion length comparable to those of W. More importantly, we demonstrate that the large ISHE in Cr is unrelated to its spin density wave AF ordering. With negligible small magnetic proximity effect, Cr with large spin Hall angle is a superior spin current generator/detector better than many *5d* metals, including Pt and W, in pure spin current phenomena and devices.

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*clchien@jhu.edu

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	Pt	**Au**	**Ta**	**W**	**Cr**
θ / θ_{Au}	**4.33**	**1**	**-0.46**	**-1.43**	**-1.38**
λ_{SF} (nm)	**2.5**	**9.5**	**1.7**	**1.5**	**2.1**

Table 1. Spin Hall angles and spin diffusion length of several metals relative to Au obtained from a self-consistent measurement. The spin mixing conductance of different metals on YIG is assumed to be the same for a simple intuitive comparison.

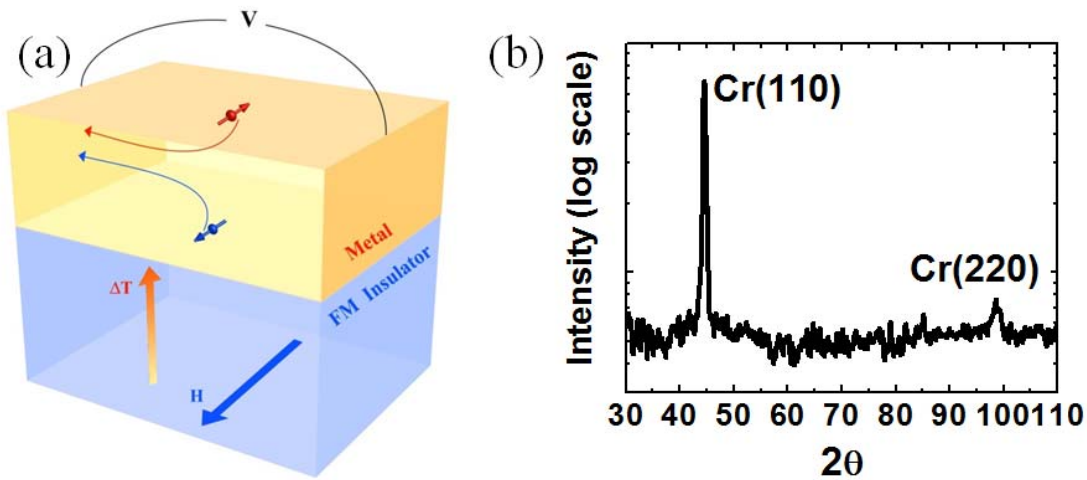

Fig. 1. (Color online) (a) Schematic diagram of longitudinal spin Seebeck effect in ferromagnetic insulator YIG and the inverse spin Hall effect in the adjacent metal layer. (b) X-ray diffraction of 200 nm thick Cr film on Si substrate with (110) texture.

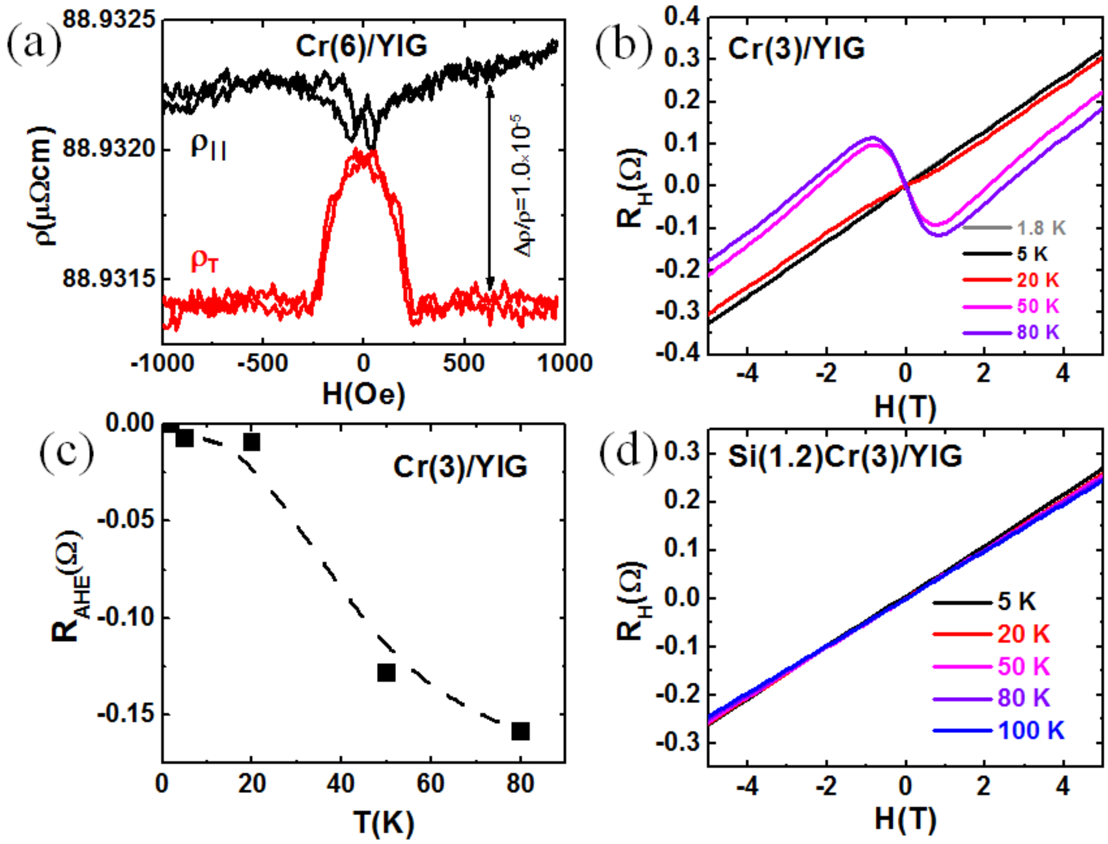

Fig. 2. (Color online) (a) Magnetoresistance of 6 nm thick Cr film on YIG substrate. The MR ratio is 1.0×10^{-5}. (b) Hall resistance of 3 nm thick Cr film on YIG. (c) Anomalous Hall resistance of 3 nm thick Cr film on YIG. (Dashed line is guide to the eye) (d) Ordinary Hall effect of 3 nm thick Cr capped by 1.2 nm thick Si on YIG.

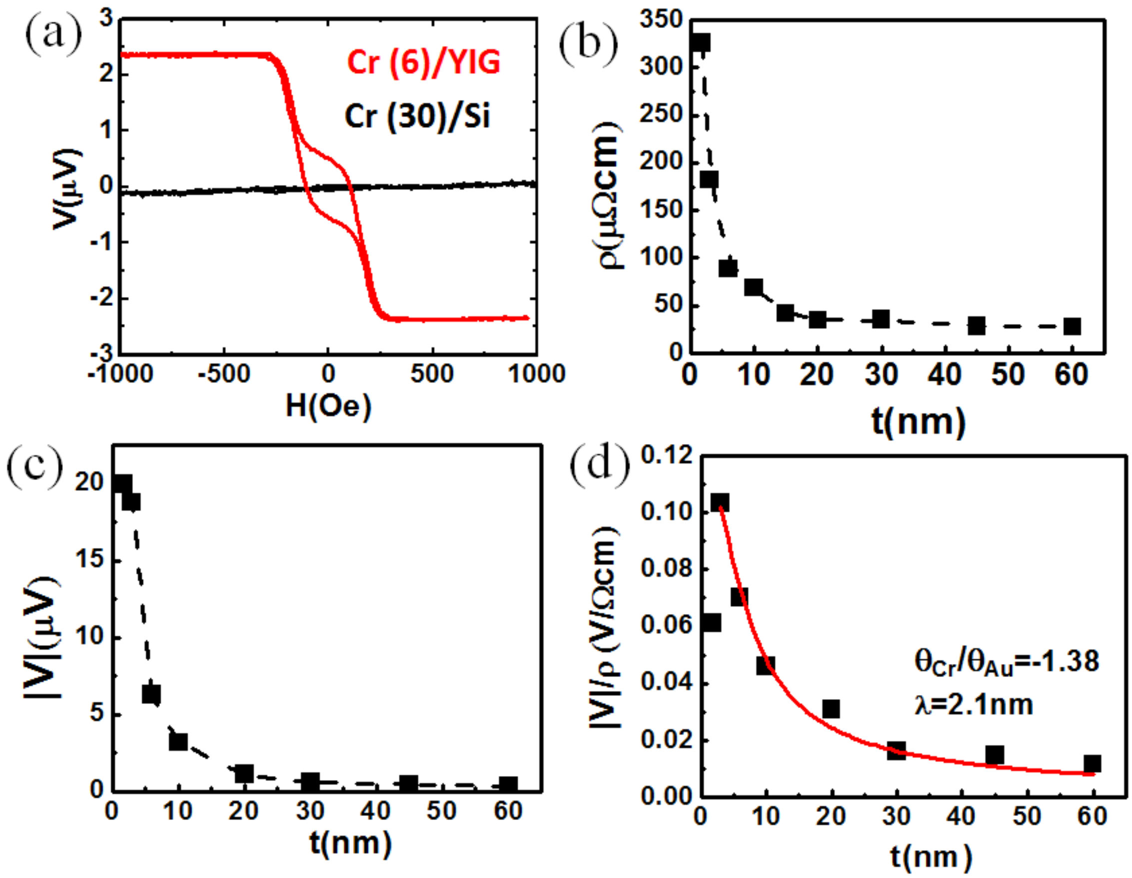

Fig. 3. (Color online) (a) Thermal voltage obtained in 6 nm thick Cr film on YIG and 30 nm thick Cr on Si with a temperature gradient of 20 K/mm. (b) Thickness dependent resistivity of Cr thin films on YIG. (c) Thickness dependent thermal voltage of Cr thin films on YIG. (Dashed lines are guides to the eye) (d) Voltage over resistivity with thickness for the analysis of relative spin Hall angle and spin diffusion length in Cr. Red line is fitting from Eq. 1.

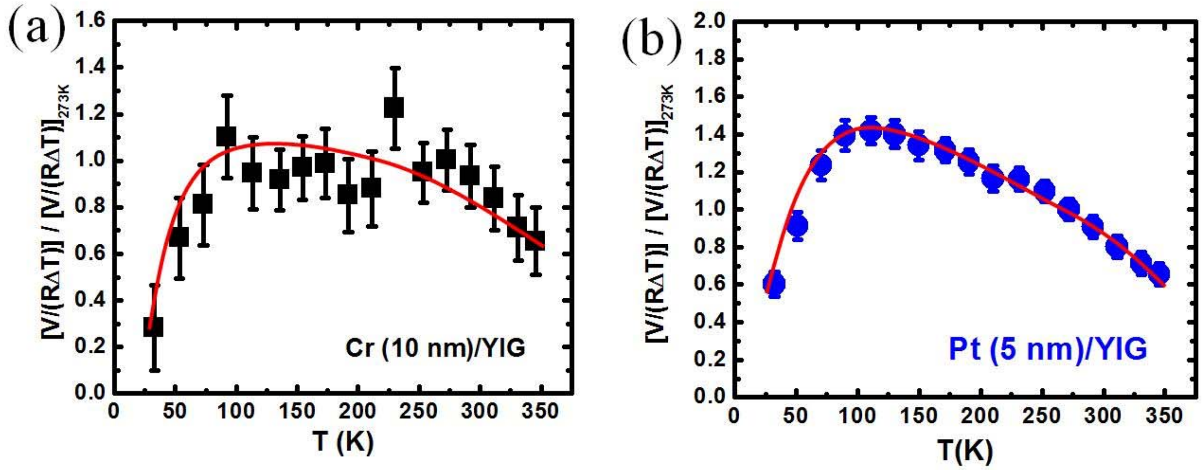

Fig. 4. (Color online) Temperature dependence of thermal voltage divided by film resistance and temperature difference [V/(RΔT)] of the (a) Cr(10 nm)/YIG and (b) Pt(5 nm)/YIG from 30 K to 345 K (metal side temperature). The [V/(RΔT)] values of Cr and Pt at 273 K are used as reference data for each plot. Solid lines are guides to the eyes.