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Direct optical detection of current induced spin accumulation in metals by magnetization-induced second harmonic generation

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Strong spin-orbit coupling in non-magnetic heavy metals has been shown to lead to large spin currents flowing transverse to a charge current in such a metal wire. This in turn leads to the buildup of a net spin accumulation at the lateral surfaces of the wire. Spin-orbit torque effects enable the use of the accumulated spins to exert useful magnetic torques on adjacent magnetic layers in spintronic devices. We report the direct detection of spin accumulation at the free surface of nonmagnetic metal films using magnetization-induced optical surface second harmonic generation. The technique is applied to probe the current induced surface spin accumulation in various heavy metals such as Pt, β-Ta, and Au with high sensitivity. The sensitivity of the technique enables us to measure the time dynamics on a sub-ns time scale of the spin accumulation arising from a short current pulse. The ability of optical surface second harmonic generation to probe interfaces suggests that this technique will also be useful for studying the dynamics of spin accumulation and transport across interfaces between non-magnetic and ferromagnetic materials, where spin-orbit torque effects are of considerable interest.
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The utilization of the spin degree of freedom of electrons has led to the success of spintronics in high density magnetic memory devices.[1] Spin polarized currents[2,3] are being increasingly used to apply magnetic spin transfer torques for magnetization switching in magnetic devices as they operate at switching currents orders of magnitude lower than traditional Oersted field switching.[4–6] The spin-orbit torque effect, typically identified as the spin-Hall effect,[7–10] which is the generation of a spin current transverse to an applied charge current as a result of spin-orbit coupling, has emerged as a promising new low current switching technique, particularly with the demonstration of giant spin-Hall effect in the highly resistive β phase of Ta.[10] The effect leads to the accumulation of spins of opposite orientation at opposite faces of the spin-orbit material, transverse to the charge current direction.[11]

The transverse spin accumulation that arises from a charge current is usually quantified by the spin-Hall angle[10] $\theta_{sH} = J_s/J_c$, where J_c is the charge current and J_s is the transverse spin current density. Large spin-Hall angles have been measured in heavy metals such as Pt,[12] β-Ta,[10] and β-W.[13] Although optical techniques have been used to detect and measure the current induced spin accumulation in semiconductors such as AlGaAs,[14] GaAs,[11,15] and strained InGaAs,[11] until recently the effect in metals was studied exclusively by electronic methods[16] or by the study of its interaction with a magnet.[10,12,17] Direct detection of the current induced spin accumulation in non-magnetic metals was recently reported by van 't Erve *et al.*[18] with magneto-optical Kerr effect (MOKE), and with spin polarized positron beams by Zhang *et al.*[19] A direct optical probe of the effect in the non-magnetic metals is attractive because it allows separation of the effect of the spin-current in the non-magnetic metal from the spin-transport across the non-magnetic/magnetic interface.

Here, we propose an alternate technique to directly probe the current induced spin accumulation at the surface optically through magnetization-induced second harmonic generation (MSHG).[20] Second order nonlinear optical effects are observable only in systems where both space-inversion and time reversal symmetries are broken, and hence, the MSHG signal is only sensitive to the magnetization at surfaces and interfaces. This is ideal for our measurements as the current induced spin polarization accumulates in a very thin layer at the surfaces of thin films. Although spin accumulation has already been optically probed with MOKE,[18] the signal to noise ratio (SNR) was extremely low for the W films they measured (SNR $\sim$ 5:1) and insufficient for Pt films (SNR $\sim$ 2:1). Moreover, the MSHG sensitivity at surfaces and interfaces has been shown to be up to 2 orders of magnitude higher than with traditional MOKE.[21] Also, the reflected second harmonic intensity is proportional to the square of the incident intensity.[20,22] As a consequence of this, the effective optical skin depth in metals for MSHG is half the skin depth for a linear magneto-optic technique such as MOKE. Therefore, the signal contribution from the bottom interface of the metal film is significantly reduced compared to the case of MOKE, which allows for the study of thinner films. We report here the detection of the current induced spin accumulation in thin films of Pt, β-Ta, and Au through MSHG. In our experiments, we utilize a pulsed laser that enables an easy and direct observation of time-dependent dynamics of the spin accumulation that follows the applied current pulse, which is reported here for Pt films. The MSHG technique presented here can also be used complementary to MOKE to gain further insights into the spin accumulation mechanism.[20]

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Sputtered Pt and β-Ta films, and evaporated Au films of different thicknesses on a thermally oxidized Si substrate patterned into $50\,\mu m \times 50\,\mu m$ squares and contacted with 80 nm thick evaporated gold contacts served as our samples (see Fig. 1 inset). The laser used was an 800 nm wavelength, regeneratively amplified Ti:sapphire laser which outputs $\sim$70 fs pulses, each of energy up to $\sim$1 μJ, at 252 kHz repetition rate. A polarizer was used to render the incident beam p-polarized. The laser beam was incident at an angle of 45° on the sample surface, focused to a $\sim$30 μm spot. The beam was attenuated to produce a maximum incident fluence at the sample estimated to be approximately 300 μJ/cm^2. The reflected light was filtered such that only the 400 nm second harmonic is passed, and the dominant 800 nm fundamental was removed. This was then passed through another p-polarized polarizer to a photo multiplier tube. The output was amplified, and the second harmonic intensity was measured by a photon counter. The schematic of our setup is shown in Fig. 1. Alternate positive and negative current pulses, each 200 ns in duration with amplitude in the range of $\sim$150 mA, separated by the laser pulse period, were applied and synchronized with the arrival of laser pulses onto the sample. For the described configuration of the polarizers, the measured magnetic asymmetry signal from MSHG is sensitive to the component of magnetization M_y transverse to the plane of incidence.[20,22] This is indeed the expected polarization direction of accumulated spins as the current in the sample is in the plane of incidence as shown in the schematic in Fig. 1. The intensity of the reflected second harmonic in a 10 nm Pt film was confirmed to vary quadratically with the incident fluence up to $\sim$500 μJ/cm^2 (see Fig. S1[23] in supplementary material).

We measured the magnetic asymmetry or contrast A defined in Eq. (1),[20] where $I^{2\omega}(M)$ is the intensity of the second harmonic for a surface magnetization of M. In the case of current induced spin accumulation, the magnetization ($\pm M$) at the surface of the film is related to the spin accumulation, which is proportional to the applied current ($\pm J$).[24]

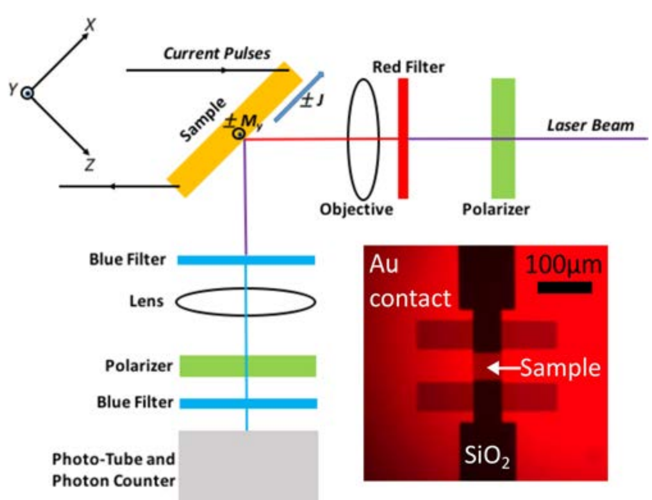

FIG. 1. Schematic of the magnetization-induced second harmonic generation set-up. The current J is applied along x and the magnetization M due to spin accumulation is along y, perpendicular to the plane of incidence. The current pulses and photon counter are synchronized with the laser pulse with a delay generator. Inset: Microscope image of the sample.

$$A = \frac{I^{2\omega}(+M) - I^{2\omega}(-M)}{I^{2\omega}(+M) + I^{2\omega}(-M)} = \frac{I^{2\omega}(+J) - I^{2\omega}(-J)}{I^{2\omega}(+J) + I^{2\omega}(-J)}. \quad (1)$$

The asymmetry for a ferromagnetic in-plane magnetized CoFeB film was found to be $\sim$80% with an external field of $\pm$40 mT and no current in the sample.

We utilize the asymmetry normalized with the applied charge current density (A/J_C) as a measure of the spin accumulation because the spin accumulation scales with J_C. For a 20 nm thick Pt sample, an asymmetry A of 6.1% was observed for a current density of $J_C = 1.5 \times 10^7$ A/cm^2 with 10 $V_{p\text{-}p}$ driving voltage. The sample stage was rotated and the asymmetry A was plotted as a function of θ, the angle between the current direction, and the plane of incidence, as shown in Fig. 2(a). As can be seen, the asymmetry varies as $\cos\theta$. The magnetic asymmetry A is zero when the current is perpendicular to the plane of incidence and changes sign if the sample is rotated further. The setup being sensitive to M_y proves that the magnetization component we measure is perpendicular to the current, and that its direction changes with that of the current. Furthermore, Fig. 2(b) shows that A scales linearly with the amplitude of the current pulses applied through the film, as is expected. It is important to ensure that the measured signal

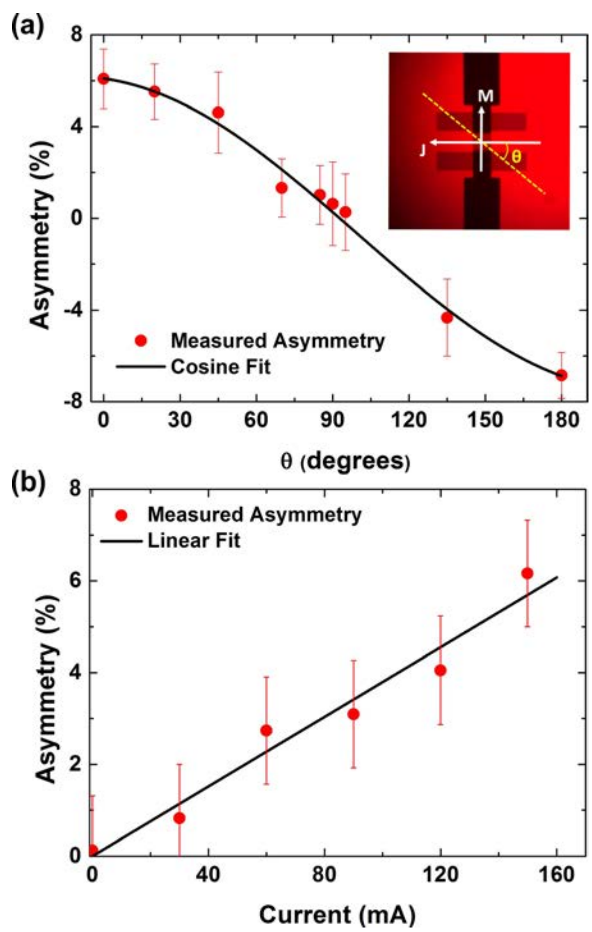

FIG. 2. (a) Magnetic asymmetry as a function of the angle θ between the direction of current and the plane of incidence (yellow dotted line in inset). The MSHG signal in the reported configuration is sensitive to the component of magnetization transverse to the plane of incidence. The cosine dependence of A on θ indicates that the magnetization is perpendicular to the direction of current. (b) Magnetic asymmetry as a function of the amplitude of the current pulses in the 20 nm Pt sample.

does not arise from spin polarization due to the Oersted magnetic field generated by the charge current. The amplitude of the current pulse through the film being 150 mA, the Oersted field at the surface of the film is calculated to be approximately 1.9 mT. However, the asymmetry remained unchanged when a larger transverse magnetic field of 40 mT was applied along *y* using an external magnet, in conjunction with the current. Thus we rule out the possibility of the MSHG signal arising from the Oersted field of the current, and conclude that the asymmetry is a measure of the spin-orbit induced spin accumulation in our sample.

Table I summarizes the measured normalized asymmetry for different samples with current in the plane of incidence. All the measurements were done with the current direction in the plane of incidence, with a 10 $V_{p\text{-}p}$ driving voltage. The asymmetry measured was then divided by the corresponding current density in order to obtain the normalized asymmetry reported in the table. The asymmetry for all the samples with zero current was found to be zero within the shot noise level. The high resistivity 30 nm β-Ta film was sputtered and capped with a 1 nm sputtered layer of Ti to prevent the oxidation of the top surface of Ta since the MSHG signal is extremely sensitive to surface smoothness and oxidation.[20] We believe that the asymmetry in the MSHG signal arises at the Ta/Ti interface. The asymmetry in the Ta/Ti sample is negative and opposite in sign to that of Pt as is expected from previous spin-Hall effect experiments.[10,12] The evaporated pure Au films of 10 nm and 20 nm thickness show a small normalized asymmetry with the same sign as Pt and with a magnitude ~20% of that of Pt. This result is consistent with the previously reported non-negligible spin-Hall angle for Au,[25] and the ratio with that of Pt is close to the one reported by the Hoffman group.[26,27] Au and Pt films do not need a capping layer as they are not prone to oxidation. The asymmetry in the sputtered 10 nm Cu film capped with 1 nm evaporated Al is consistent with zero within the limits of the experimental noise. The magnitude of the asymmetry in Cu is consistent with the data of Zhang *et al.*[19]

The photon count rates were of the order of ~100/min for the Pt and Ta films, and ~1000/min for the Au and Cu films. The measurements were integrated over a time period of 20 min to obtain adequate signal/noise ratio. The standard error bars obtained for different samples, measured over twenty repeated experiments of 1 min integration time each, are consistent with the photon counting shot noise (see Fig. S2 in supplementary material[23] for photon counting statistics of the 10 nm Pt film). In order to interpret the increase in asymmetry for the Au and Pt samples when decreasing the film thickness, the non-linear susceptibility tensor elements, $\chi^{(2)}$, are needed. Top and bottom interfaces are expected to have complex susceptibilities of different amplitude and phase. We start with the assumption that $\chi^{(2)}$[air/Pt] and $\chi^{(2)}$[Pt/SiO_2] will have similar order of magnitude and will be phase shifted by 180° due to opposite symmetries of the metal/insulator interfaces.[28,29] Under this assumption, for a spin accumulation of opposite orientation on both interfaces as expected for spin-Hall effect, the SHG fields from the top and bottom interfaces should add constructively. This then implies the observed increase in the absolute value of the asymmetry in thinner films as we are more sensitive to the bottom interface as a result of decreased absorption through the film. Quantitative modeling of the effects of multiple reflections of both the fundamental and second harmonic fields in the multilayer structure is currently in progress and will be published separately. The competition between the spin-Hall and the Rashba effects in contributing to the surface magnetization[30] may also play a role in determining the thickness dependence of the normalized magnetic asymmetry. The ability of our technique to directly measure the spin accumulation at the normal metal surface independently of effective torques on a ferromagnetic layer in contact with the normal metal[30] will be of great value in helping to identify the relative roles of spin-Hall and Rashba effects in various device structures. This will be the subject of future work.

The experiments described above confirm that MSHG is an effective technique to detect spin accumulation with an improved signal to noise ratio compared with MOKE.[18] We then studied the time dynamics of the spin accumulation in the 10 nm Pt sample using a unipolar 2 ns wide current pulse. The voltage pulse waveform was measured across the sample contacts with an RF active probe, and the current waveform is estimated based on the measured resistance of the sample. The relative time of arrival of the laser and current pulses at the sample were varied using an electronic delay generator, and at each delay time, the magnetic asymmetry was measured. The results are shown in Fig. 3. Within the observed noise level, the spin accumulation pulse is seen to follow the current pulse on the time scale of ~200 ps. Similar results were observed in the 20 nm Pt film. We can therefore conclude that the MSHG is suitable for time-

TABLE I. The magnetic asymmetry *A* normalized with charge current density J_C for different metallic films with current direction along the plane of incidence and 10 $V_{p\text{-}p}$ driving voltage.

Sample	Asymmetry/current density [% $cm^{-2}/(10^7$ A)]
Pt (10 nm)	5.12 (±0.51)
Pt (20 nm)	4.05 (±0.68)
β-Ta (30 nm)/Ti (1 nm)	−5.78 (±2.02)
Au (10 nm)	1.10 (±0.04)
Au (20 nm)	0.50 (±0.04)
Cu (10 nm)/Al (1 nm)	0.11 (±0.16)

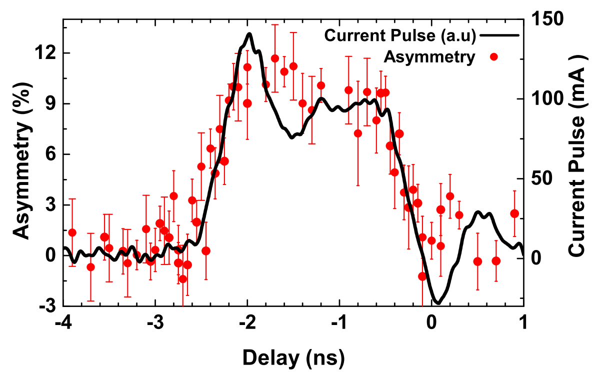

FIG. 3. Spin accumulation at the surface of the 10 nm Pt sample as a function of the delay of the laser pulse relative to the 2 ns current pulse. The "zero" time delay is arbitrary. The current magnitude was estimated from the voltage signal across the sample contacts and separate measurement of the sample resistance.

dependent studies of the current induced spin accumulation. Experiments with higher time resolution are in progress.

To summarize, we have shown that the magnetization-induced second harmonic generation is a promising technique to detect and probe the current induced spin accumulation at the surface of metallic thin films. We have verified that the spin accumulation is perpendicular to the direction of flow of charge current and that the measured magnetic asymmetry signal due the spin accumulation scales with the applied charge current density. The surface magnetization due to spin accumulation was detected in the metals Pt, β-Ta and Au. The sign of the asymmetry in β-Ta and Pt are opposite to each other, and the sign in Au is the same as in Pt. The magnitude of the asymmetry normalized to charge current density is negligible in Cu, and is small in Au, as is expected. We have also studied and reported the time dynamics of the accumulated spins in 10 nm and 20 nm Pt films and find that the spin accumulation closely follows the charge current on a 200 ps time scale. The surface/interface sensitivity of MSHG, in addition to the simplicity and the ability to detect spins directly independent of spin transport into an adjacent magnetic layer, makes it a strong candidate to study spin accumulation at metallic surfaces and interfaces. It also offers the promise of performing detailed time-dependent dynamic studies of spin accumulation.

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