Direct optical observation of spin accumulation at nonmagnetic metal/oxide interface

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We report the direct observation of uniform in-plane spin accumulation at room temperature by magneto optical Kerr effect, at the interface formed between nonmagnetic metal (Cu, Ag) and oxide (Bi$_2$O$_3$). Recent reports show spin to charge conversion at these interfaces suggesting the presence of Rashba like spin orbit coupling (SOC). The formation of spin accumulation is the result of current induced spin polarization at our interfaces (direct Rashba–Edelstein effect), without external magnetic field or proximity to ferromagnetic materials. We observe opposite orientation of spin accumulation at Cu/Bi$_2$O$_3$ and Ag/Bi$_2$O$_3$ interfaces reflecting their opposite sign of Rashba SOC (Rashba parameter). Moreover, estimation of spin accumulation from values of Rashba parameters obtained by independent spin pumping measurements, agrees well with the difference in amplitude of our normalized Kerr signals for Cu/Bi$_2$O$_3$ and Ag/Bi$_2$O$_3$ interfaces. Uniform in-plane spin accumulation due to Rashba-Edelstein effect can be applied for spin filter devices and efficient driving force for magnetization switching. Published by AIP Publishing.

Since the discovery of Rashba effect in 1959, there has been a growing number of theoretical studies and experimental evidence of innovative concepts, which in great part led to the formation of the Spintronics field. The recent discovery of new topological class of materials, the control of spin orientation of moving electrons, and spin to charge conversion, magnified enormously the interest in Rashba type systems, which nowadays goes beyond semiconductors.1–3 Generation of spin accumulation from non-spin-polarized electrical current is particularly attractive for the development of new device concepts, which can be manipulated by electric fields.4 Spin accumulation induced by spin Hall effect was first directly observed when passing nonpolarized electrical current in bulk GaAs and strained InGaAs without external magnetic fields.5 The detected spin polarization was found to be opposite at opposite edge of the samples. Immediately after that, spin Hall effect was confirmed by direct electronic measurements.6 Analogously, Rashba–Edelstein effect describes how moving electrons in an electric field experience a momentum dependent magnetic field, which couples to their spin angular momentum. This physical phenomenon permits the generation of non-equilibrium spin polarization from electrical charge current, which in turn leads to the build-up of spin accumulation.1–3

Spin accumulation at a Rashba interface is the result of inversion asymmetry in two-dimensional electron systems (2DES). Bychkov and Rashba proposed that the lack of inversion symmetry creates an interfacial out of plane electric field $E = E_z$, which results in an additional spin-orbit coupling (SOC) in 2DES.7 The Hamiltonian of this Rashba SOC is $H_R = z_R(k \times z) \cdot \sigma$, where $z_R$ is the Rashba parameter, $z$ is the unit vector perpendicular to the interface and $\sigma$ is the Pauli spin vector. Rashba SOC leads to spin splitting of the electron bands depending on the electron wavevector $k$. In equilibrium, the number of occupied states for spin up and spin down is equal, however, when electrical charge current flows at the Rashba interface (i.e., along x-axis), electrons experience an effective in-plane magnetic field, $B_{R}$, perpendicular to their direction of flow, creating a net out of equilibrium spin polarization. This effect is also known as the direct Rashba Edelstein effect (DREE).1 Figure 1(a) shows a schematic representation of the equilibrium spin polarized configuration of a Rashba-type interface in energy dispersion as a function of in-plane momentum $k$, and Fig. 1(b) its Fermi contour representation. Figure 1(c) shows the non-equilibrium spin polarization state when passing electrical charge current. Out of equilibrium, the Fermi contours are displaced in $k_{\perp}$ by an amount $\delta k_{\perp} = -e \rho_{NM} J_{inj} \tau (1 + \eta)/\hbar$, with $\eta = m^*_e z_R/(\hbar^2 k_F)$, where $\rho_{NM}$ is the resistivity of the nonmagnetic layer, $J_{inj}$ is the density of the injected current at the interface, $\tau$ is the momentum relaxation time of the 2DES, $m^*_e$ is the effective mass of the electron, and $z_R$ is the Rashba parameter which reflects the strength of SOC. Figure 1(d) depicts schematics of the spin accumulation when electrons flow along x-axis at the interface between a nonmagnetic metal (NM) and Bi$_2$O$_3$ with Rashba SOC.

Recent reports show spin to charge conversion at the Cu/Bi$_2$O$_3$ interface by spin pumping8 and spin Seebeck effect.9 Spin to charge conversion was also reported at the Ag/Bi interface10 suggesting the existence of Rashba SOC at Ag/Bi$_2$O$_3$ interface. From these previous reports, we can indirectly infer the presence of spin accumulation on these non-magnetic metal/oxide interfaces. We investigate the in-plane interface

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spin accumulation at the Cu/Bi$_2$O$_3$ and Ag/Bi$_2$O$_3$ by time-resolved transverse magneto-optical Kerr effect (TR-TMOKE) at a laser wavelength of 408 nm. Our samples consist of a stacking bilayer of 20 nm thickness of nonmagnetic metal (Cu, Ag) and 10 nm of Bi$_2$O$_3$. X-ray diffractometer spectroscopy at shallow angles reveals preferential Cu (111) and Ag (111) orientation at the interface, and amorphous Bi$_2$O$_3$ (see supplementary material). We apply modulated AC voltage along the interface, which produces spin accumulation oriented perpendicular to the electrical charge flow. Correlation between the AC excitation voltage and the TR-TMOKE signal directly assures the presence of spin accumulation induced by DREE in our devices. Generally, TMOKE should result in a change in intensity rather than a change in Kerr rotation, when working with pure p-polarized light ($\eta = 90^\circ$). However, when working with mixed s- and p-polarized light ($\eta = 45^\circ$), TMOKE signal is significantly enhanced and leads to appearance of a Kerr rotation component $\Delta \phi$, as previously reported.\textsuperscript{11–13} In our TR-TMOKE setup we polarized our incidence light at $\eta = 45^\circ$ mixing the s- and p-polarizations.\textsuperscript{14} As the Kerr ellipticity of the Rashba interface is stronger than Kerr rotation due to strong dichroism effect,\textsuperscript{15–17} we used a quarter wave plate (QWP) as commonly used for Kerr ellipticity when characterizing Nickel films.

Figures 2(a) and 2(b) show the TR-TMOKE signal for Cu/Bi$_2$O$_3$ and Ag/Bi$_2$O$_3$ interfaces, respectively. Fitting (solid lines) show signal periodicity of 10 ns (100 MHz) in time (frequency) for both Cu/Bi$_2$O$_3$ and Ag/Bi$_2$O$_3$, which coincide with the excitation AC sinusoidal voltage at a frequency of 100 MHz passing through the interface. From Figs. 2(a) and 2(b), it is possible to observe opposite phase between TR-TMOKE signals of Cu/Bi$_2$O$_3$ and Ag/Bi$_2$O$_3$, indicating opposite spin momentum locking configuration. The spin accumulation corresponds to the initial splitting of the spin sub-bands (spin locking) of the free energy dispersion, $E_{\pm}(k) = E_0 + (\hbar^2 k^2/2 m_0^*) \pm |s_R||k|$. The orientation of the spin polarization on each spin sub-band is defined by $P_{\pm}(k) = \pm \frac{s_R(-k_x,k_z,0)}{|k|}$. Both, the orientation of the initial spin configuration and the corresponding spin accumulation directly dependent on the Rashba parameter $z_R$ and the effective mass $m_0^*$, which determines the splitting order of inner and outer energy branches $(E_+, E_-)$.\textsuperscript{18}

The estimated $z_R$ for Cu/Bi$_2$O$_3$ and Ag/Bi$_2$O$_3$ from independent spin pumping experiments show $z_R = -0.25 \pm 0.03$ eV A and $z_R = +0.18 \pm 0.04$ eV A,\textsuperscript{19} respectively, while the effective mass is assumed negative for both systems.\textsuperscript{10,18} According to their sign of $z_R$, the expected orientation of spin states in the Fermi contour representation are depicted in Figs. 2(c) and 2(d) for Cu/Bi$_2$O$_3$ and Ag/Bi$_2$O$_3$ interfaces, respectively.

Spatially uniform spin accumulation is also expected to originate from flow of electric current on Rashba type interfaces.\textsuperscript{1,3} By scanning the position of our laser spot along the Ag/Bi$_2$O$_3$ interface, we examine the relative change on amplitude of TR-TMOKE signal. Figure 3(a) shows the comparison of the obtained TR-TMOKE signal for five different laser spot positions along the interface parallel to the electrical charge flow, as sketched on Fig. 3(b). The full area of our sample is $200 \mu m \times 600 \mu m$ and the nominal laser spot diameter is about $3.55 \mu m$. The amplitude of the TR-TMOKE signal is constant along the Ag/Bi$_2$O$_3$ interface reflecting spatially homogenous spin accumulation, and inferring negligible influence of heat gradients in our measurements.

We now discuss about the relative amplitude of our TR-TMOKE signals and the expected spin accumulation. The non-equilibrium 3D density of spins at the interface is related to the electrical charge current density through the Rashba parameter $z_R$, as follows: $\langle \delta S \rangle = m_0^* z_R I_{ex}/e \hbar \nu$,\textsuperscript{20} where $\hbar \nu$ is the Fermi energy, which are taken from the nonmagnetic metal values ($\hbar \nu = 7$ eV for Cu and $\hbar \nu = 5.5$ eV for Ag), and $m_0^*$ is the effective mass, which is approximated to the mass of a free electron at our interfaces ($m_0 = 9.11 \times 10^{-31}$ kg). For our estimation, we assign an interface thickness of one atomic lattice.
of typical thickness $t = 0.4 \text{ nm}$\textsuperscript{8,10}. The interfacial electrical current density $J_{cx}$ in our measurements in Figs. 2(a) and 2(b) are $1.87 \times 10^{12} \text{ A/m}^2$ and $2.16 \times 10^{12} \text{ A/m}^2$, giving a total $\langle \delta S \rangle$ of $14.0 \mu \text{eV} (3.61 \times 10^{23} \text{ m}^{-3})$ and $23.7 \mu \text{eV} (3.82 \times 10^{23} \text{ m}^{-3})$ for Cu/Bi$_2$O$_3$ ($\alpha_R = -0.25 \pm 0.03 \text{ eVÅ}$) and Ag/Bi$_2$O$_3$ ($\alpha_R = +0.18 \pm 0.04 \text{ eVÅ}$) interfaces, respectively. Assuming that the spin accumulation created at the interface diffuses in the bulk, and the spin diffusion lengths at room temperature for Cu\textsuperscript{21} and Ag\textsuperscript{22} are much larger than our layer thickness (20 nm), the detected spin accumulation would depend mainly in the penetration depth of our laser into the metal layers, where the extinction ratio $\kappa$ for both metals (Cu, Ag) is very similar at our laser wavelength (408 nm)\textsuperscript{23}. Within this scenario, even if taking a conservative small penetration depth ($< 10 \text{ nm}$), our spin accumulations are in good approximation (order of magnitude) with the spin accumulation in previous reports\textsuperscript{24,28}. Nevertheless, systematic studies of the mechanism of spin diffusion from Rashba interface into bulk, by other techniques such as spin Hall magnetoresistance (SMR)\textsuperscript{25} is necessary for completing description of the optically detected spin accumulation.

We compare the ratio between TR-TMOKE signal amplitudes for Cu/Bi$_2$O$_3$ and Ag/Bi$_2$O$_3$ and the ratio of their total spin accumulations. The amplitudes of our signals have been normalized by the corresponding reflectivity for Cu/Bi$_2$O$_3$ and Ag/Bi$_2$O$_3$ at our laser wavelength (408 nm)\textsuperscript{23}. From our fittings in Fig. 2, we obtain peak amplitudes of 0.077 and 0.112 for Cu/Bi$_2$O$_3$ and Ag/Bi$_2$O$_3$, giving a ratio of 1.45, which is in good agreement with the ratio of their estimated spin accumulation equal to 1.69. For our estimations, we assume negligible spin Hall effect at our samples, as it is been tested previously by spin pumping measurements for Py/Cu and Py/Bi$_2$O$_3$ bilayers\textsuperscript{8}.

In summary, we observed the homogeneous in-plane current induced spin accumulation at room temperature due to Rashba-like SOC at nonmagnetic metal/oxide interface by TR-TMOKE. Opposite phase of TR-TMOKE signal for Cu/Bi$_2$O$_3$ and Ag/Bi$_2$O$_3$ interfaces corresponds to their intrinsic opposite spin configurations, reflecting their sign of Rashba parameter $\alpha_R$, independently determined by spin pumping measurements. Comparison of the ratio of our TR-TMOKE signals and estimation of spin accumulation $\langle \delta S \rangle$ for Cu/
Bi$_2$O$_3$ and Ag/Bi$_2$O$_3$ show good agreement, proving the values of their Rashba parameter $\zeta_R$ by characterization method without external magnetic field or proximity to ferromagnetic layers. Our results also show the feasibility of characterizing the spin accumulation by Kerr effect magnetometry.\textsuperscript{11,24–30} We conclude that our observation of spin accumulation is the result of the efficient generation of spin accumulation by DREE at our interfaces,\textsuperscript{8,10,20} and the high sensitivity of our experimental setup, which combines sine current modulation and lock-in detection techniques, strongly minimizing the experimental setup, and independent spin pumping data and estimation of spin pumping.

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See supplementary material for further details on sample fabrication, X-ray spectroscopy characterization, experimental setup, and independent spin pumping data and estimation of spin accumulation.

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19Spin pumping experiments similar to those performed in Refs. 8 and 10 show $\zeta_R = -0.25^{\pm}0.03$ eVÅ and $\zeta_R = +0.18^{\pm}0.04$ eVÅ, for Cu/Bi$_2$O$_3$ and Ag/Bi$_2$O$_3$, from independent spin pumping experiments, see supplementary material. Detail explanation of these results will be reported elsewhere.