論文の内容の要旨

Optical spin-charge interconversion at non-magnetic metal/oxide interfaces

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Spintronics is based on two physical degrees of freedom: spins and charges of electrons or holes. The field of spintronics was born in 1988 with the discovery of the giant magneto-resistance by Albert Fert and Peter Grunberg, who were awarded by the Nobel prize of physics in 2007. Since then, researchers used this discovery to make significant improvements in data storage. These advancements led to the development of a new era of magnetic drives. Recently, spin conversion, a generic term for all conversion phenomena based on the principle of angular momentum conversion, is rising big interests in the electronic industry due to its possibility to lead to the development of new experimental methods as well as the implementation of novel spin conversion mechanisms. The further development of spin-conversion functionalities relies on the microscopic understanding of the interaction among quasiparticles such as electrons, spins, magnons, phonons, and photons. Recently, new types of spin-charge interconversion in non-magnetic metal/Bi_2O_3 interfaces were experimentally demonstrated where Rashba spin orbit coupling are presented. This Rashba-type spin-orbit interaction leads to the generation of magnetization from an applied electric field (direct Edelstein effect) and an electric current from a magnetic field or the magnetization (inverse Edelstein effect). This effect has some similarities to spin Hall effect and inverse spin Hall effect that also induce some spin-charge interconversion also due to spin-orbit interaction. Especially, in semiconductor/metal systems such as GaAs/Pt, it has been shown that by injecting circularly polarized light coupled with electron spins in semiconductors and it will produce some transverse current through inverse spin Hall effect. Similarly, one can wonder if interactions between polarized light and Rashba-type metal/oxide system are possible as well. Relatively few studies have been done related to the interaction electron and polarized light in the presence of spatial inversion asymmetry.

In this work, we are focusing on investigating responses of chirality and polarization to expand the functionality of the spintronics devices. First, we investigate the creation of spin accumulation at the interface of metal/oxide system by direct Edelstein effect. By injecting AC charge current and a magneto-optical Kerr effect system, we will detect spin accumulation and give a proof of that Cu(Ag)/Bi_2O_3 have a strong Rashba effect at the interface. Then, we will investigate the possibility of the creation of charge current by injection of polarized light on Cu/ Bi_2O_3 interface by inverse Edelstein effect. Above the band gap of Bi_2O_3, spin to charge conversion is expected, similar to Pt/GaAs interface. But more surprisingly, we also observe spin to charge conversion below the band gap of Bi_2O_3.

Spin accumulation induced by spin Hall effect was first directly observed when passing non-polarized electrical current in bulk GaAs and strained InGaAs by using a magneto-optical Kerr effect system. The detected spin polarization was found to be opposite at opposite edge of the samples. Analogously to spin Hall effect, direct Edelstein effect is a physical phenomenon that permits the generation of non-equilibrium spin polarization from electrical charge current, which in turn leads to the build-up of spin accumulation.

We show the observation of the spin accumulation in non-magnetic metal/bismuth oxide interface by using time-resolved transverse magneto-optical Kerr effect (TR-TMOKE) at a laser wavelength of 408 nm. Generally, TMOKE should result in a change in intensity rather than a change in Kerr rotation, when working with pure p-polarized light. However, in our TR-TMOKE setup, signal significantly enhanced by utilizing mixture of s- and p-polarized light. We apply modulated AC voltage along the interface, which aligns spins perpendicularly to the electrical charge flow. We show that TR-TMOKE signals for Cu/Bi_2O_3 and Ag/Bi_2O_3 interfaces coincide with the excitation AC sinusoidal voltage. Correlation between the AC excitation voltage and the TR-TMOKE signal directly assures the presence of spin accumulation induced by direct Edelstein effect in our devices. More interestingly, the MOKE signals from these two interfaces show opposite phases, indicating opposite spin-momentum locking configuration. This opposite spin-momentum locking has also been confirmed by spin pumping experiments in the same interfaces done by our group. We also describe the relation between the amplitude of our TR-TMOKE signals and the expected spin accumulation. The amplitudes of Cu/Bi_2O_3 and Ag/Bi_2O_3 signals are giving a ratio of 1.45 and are in good agreement with the ratio of their estimated spin accumulation equal to 1.69. Also, we explain the difference of the optical detections of spin accumulation generated by spin Hall effect and Edelstein effect. Intrinsically, spin accumulation generated by bulk spin Hall effect and direct Edelstein effect at interfaces have a different distribution of spin orientations. Spin polarization via bulk spin Hall effect is oriented perpendicular to the charge current on all the four planes which are transverse to the charge current. In contrast, spin accumulation via Edelstein effect is in-plane at interfaces, homogeneous and perpendicular to the charge current direction. In the experiments, the spin accumulation is generated at the Cu/Bi_2O_3 and Ag/Bi_2O_3 interfaces, and it is possible for these spins to diffuse into the metal. If the spin diffusion length is larger than the thickness of nonmagnetic metal layers, the quantity of detected spin accumulation will be increased with the penetration depth of the laser. We evaluate a possible enhancement of the signal by a factor of 9.3 in our sample. In the end, we show for the first time detection of spin accumulation at the interface of non-magnetic metal/oxide and give an estimation of the detected spin accumulation by our system of 130.2 μeV for Cu/ Bi_2O_3 and 220.4 μeV for Ag/ Bi_2O_3.

Moving on, we show the possibility of the creation of charge current by circularly using the same devices. We start with a measurement of the optical absorption of our Cu/ Bi_2O_3 device by UV-Vis-NIR absorption spectroscopy, in the range from 0.9 eV to 6.2 eV. We find out that the absorption spectra displays two interesting optical transitions, one at 3.13 eV and the other at 2.12 eV. The optical transition at 3.13eV corresponds to the band gap of Bi_2O_3 and the transition around 2.12 eV corresponds to the plasmonic absorption of copper. To characterize deeply these two optical transitions, we use continuous wave laser at different energies (1.15 eV, 1.96 eV, 3.05 eV). We vary the incidence angle θ, and the photon polarization by using a linear polarizer and a quarter wave plate mounted on a mechanical rotator. The photovoltage is detected perpendicularly to the incidence beam by a lock-in amplifier synced with a mechanical chopper. We show that light-polarization information can be converted into an electric signal by combining the optical selection rules and the inverse Edelstein effect. By using a laser with a power of 1mW and an energy of 3.05 eV, which is close to the band gap of Bi_2O_3, we measure the incidence angle dependence of the detected circular photovoltage and it shows a maximum value at an incidence angle $\theta = 50°$ with a value of 3.1 μV. We explain this result by analogy of previous experiments in Pt/GaAs that shows inverse spin Hall effect. Indeed, in a semiconductor, the optical selection rules for interband transitions induce spin-polarized electrons in the conduction band via the absorption of circularly polarized light. This process transfers light circular polarization into electron-spin polarization. Then, depending on the spin momentum locking at the interface, the spin polarized electron diffuses transversally in a direction.

We also measure the circular photovoltage depending on the incidence angle dependence by using a laser at 1.96 eV energy. More surprisingly, it shows that this dependence of photovoltage is different from the behavior at 3.05 eV energy. The voltage behavior created by the circularly polarized light for a laser power of 1.5 mW looks linear and can reach 15 μV which is five times bigger than at 3.05 eV. Using the previous scenario, it would be expected that no circular photovoltage could be detected because the energy of the laser is below the band gap of Bi_2O_3. One hypothesis is to attribute this circular photovoltage to plasmonic like absorption of the heterostructure. Photovoltaic devices based on plasmon induced hot electrons at metal/oxide nanoparticles have shown large conversion efficiencies. We compare this circular photovoltage with the response of a Cu(111) layer to a circularly polarized light at 1.96 eV, where the optical absorption of plasmon surface states is expected. However, we do not observe significant transverse voltage related to circularly polarized light. Also, we measure the sample with a laser at 1.15 eV energy. Neither do we observe transverse voltage. This suggests that a combination of plasmon-induced hot electrons and inverse Edelstein effect as the origin of our polarised photovoltage is at the Cu/Bi_2O_3 interface.

One hypothesis to explain the difference of oblique incidence dependences at 1.96 eV and 3.05 eV can be that two main factors are contributing to the signal: the degree of circular polarisation and the angle dependence between spin polarization vector (σ_s) and the spin current (J_s) such as $V_c \propto \sigma_s \times J_s$. Above the band gap, the degree of circular polarization depends on the absorption coefficients for s-polarised component and p-polarised component, which are unaffected at normal incidence (θ =0°) and decrease significantly at higher angles. J_s is negligible at normal incidence (θ =0°) and gradually increases towards the maximum at grazing angles. As a result, for an excitation energy above the Bi_2O_3 band gap, a minimum of V_C is located at θ=0° and a maximum is at θ=50°. However, below the band gap of Bi_2O_3, the photons are not absorbed in the

Bi_2O_3. Meanwhile, their degree of circular polarization is negligibly affected by the incidence angle, just only showing that oblique incidence dependence on $\sigma_s \times J_s$, possibly explaining the linear dependence.

To summarize, we studied the effects of optical spin-charge interconversion at the non-magnetic metal/oxide interfaces. Especially, we are able to characterize the spin accumulation at Cu/Bi_2O_3 and Ag/Bi_2O_3 interfaces and show that these interfaces have opposite spin momentum configuration. Also, we illustrate that circularly polarized light with an energy near the band gap of Bi_2O_3 can be converted into charge current in Cu/Bi_2O_3 interface. More surprisingly, we reveal the existence of helicity dependent photovoltage at energy range close to the plasmon resonance of copper. Also, the result from optical spectroscopy shows plasmonic like absorption at Cu/Bi_2O_3 interface. Based on these results, we hypothesize a new mechanism for photovoltage generation that relies on plasmon resonance and Rashba interfaces. To sum up, we demonstrate that plasmonic energy conversion holds the promise for efficient mechanism of electron-hole separation in photovoltaic devices at low costs. Moreover, the study reflects the relevance of selecting appropriate engineering of heterojunctions. Furthermore, considering the increasing interest of systems with spatial inversion asymmetry, we expect that the presented work would motivate further studies on advancing conversion efficiencies and further understanding towards spintronics in photovoltaics.