論文の内容の要旨

Helicity dependent photocurrent in strong spin-orbit-coupling materials
（強いスピン軌道相互作用を持つ物質におけるヘリシティ依存光電流）

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Spintronics, in which the spin degree of freedom of electrons is used for various functions, is a booming field of research in recent decays. With the new knowledge of the strong spin-orbit coupling (SOC) materials, the research has been focused on injecting, manipulating and detecting spin in these new playgrounds. Recent discovery of topological insulators (TIs) further promotes the developments of spintronics in strong spin-orbit coupling materials. Besides the developments of spintronics in materials aspect, new techniques using optical angular momentum for manipulating spin of electrons are also a great advance in the field. Compared with conventional electrical methods for spin manipulation, optical approaches could provide remote and contact-free spin injection and manipulation. Especially, the combination of optical angular momentum and TIs draws lots of attentions on the helicity dependent photocurrent (HDP), a new way to produce non-reciprocal photocurrent. It is said that HDP is spin-polarized and could be controlled by experimental scheme such as polarization, wavelength, and angle of incidence of illumination light. This special phenomenon not only provides a new possibility for device applications but also opens up a new way for the study of topological surface states (TSSs).

We study the HDP in strong spin-orbit coupling materials in this thesis. We first show that silicon, as a material with weak spin-orbit interaction, exhibits no HDP as expected. Then we investigate thin films of well-studied topological insulator Bi_2Se_3. With our *in-situ* growth/measurement system in ultrahigh vacuum, we have demonstrated that the HDP in Bi_2Se_3 films is strongly related with the TSSs under illumination of light certain wavelength. HDP that contains circular photogalvanic effect (CPGE) and circular photon drag effect (CPDE) mechanisms is observed in this experiment. HDP due to CPGE is generally believed to be related with the asymmetric excitation at spin splitting bands due to selection rules, which would generate a spin-polarized photocurrent as a result. However, HDP due to CPDE is not necessarily generated from exciting electrons in spin splitting bands, because even the excitation in the spin degenerate bands the momentum transfer from photons will lead to an asymmetric distribution of excited carriers in *k* space and results in a photocurrent. In addition, HDP reveals different properties depending on the experimental geometry and illumination wavelength, but not all of them are related with the spin manipulation. With further investigation of the HDP at oblique incidence on thickness dependence of the Bi_2Se_3 film, we found that HDP originated from CPGE changed its sign when the thickness of the Bi_2Se_3 film increased from 4 QL to 7 QL. We believe this observation suggests that HDP from CPGE is strongly related with TSSs of Bi_2Se_3, and the HDP due to CPGE would reverse the flowing direction when TSSs evolving from Rashba surface states to helical Dirac cone surface states. When the TSSs evolves from Rashba states to a helical Dirac cone surface states, the outer subband of the Rashba states is gone. With the same spin injected into this system by a circularly polarized photon, the dominating band of the spin-to-charge conversion changes from the outer Rashba band to inner Rashba band. Therefore this reversing of HDP is inevitable when the HDP originate from TSSs. Moreover, we observed an HDP at the edge of the sample under normal incidence. Normally, CPGE or CPDE mechanism are forbidden by the symmetry of Bi_2Se_3. The HDP we observed at the edge of the sample is due to the inverse spin Hall effect (ISHE) acting on the out-of-plane spin component which is excited by a circularly polarized light at normal incidence. With finite element method simulation, we understand that an electric dipole is formed at the edge due to the charge accumulation by the current of ISHE, and it is this dipole potential that causes the HDP we measured. By shinning the opposite edge of the sample, an electrical dipole with opposite polarity is generated, and the HDP flows in opposite direction.

Next, we have shown the HDP is also observed in Bi(111) films, a trivial but strong SOC material, though the outcome is not as promising as in Bi_2Se_3 films in the aspect of spin manipulation and the intensity of HDP. CPDE rather than CPGE is responsible for the HDP we observed at 635 nm illumination. The high extinction coefficient of bismuth is very high especially at long wavelength. As a result, we almost observe no photocurrent at 1550 nm illumination. This result suggests that though heavy metals also possess strong SOC, the high extinction coefficient will make the light absorption in them very difficult, thus they are not good candidates for the optical spin orientation.

Finally, we examined a series of TIs, $(Bi_xSb_{1-x})_2Te_3$, in which the Fermi level position can be tuned by changing the Bi and Sb compounds ratio. CPGE and CPDE mechanisms are also observed to be responsible for the generation of the HDP. We find that HDP is strongly influenced by the Fermi level position at 1550 nm, however, does not change much at 635 nm. This result further supports the conclusion that HDP could be related with TSSs of the TI at certain wavelength. HDP at normal incidence is also observed in this experiment, but only the bulk insulating sample exhibits such phenomenon. Similar to the case in Bi_2Se_3, we believe that ISHE is the explanation for the HDP at normal incidence. Moreover, we found that HDP due to ISHE is more sensitive to Fermi level position than HDP of CPGE, this is probably due to the fact that out-of-plane spin component only exists at the position far from the Dirac point of TSSs but in-plane spin generally exists in a wide energy range of the helical Dirac cone.

Through the whole study, we conclude that HDP generally exists in materials with strong SOC, but the HDP shows different properties, is originated from different mechanisms, and depends on the specific experimental arrangements. These findings suggest that we need to be careful when concluding the mechanism and origin of the HDP. In addition, TIs show better responds to the circularly polarized light than normal materials in the aspect of HDP which related with the complex spin texture of the materials. We have observed the HDP due to CPGE at all our TI samples at 1550 nm illumination but not on bismuth. Moreover, HDP due to CPGE in TIs could indeed reveal the TSSs feature of TIs. Both the thickness dependence and the Fermi level position dependence experiments suggest that HDP is related with TSSs of TIs, and the variation of such HDP could even reveal the evolution of the TSSs. Besides, out-of-plane spin component can be injected into TIs by optical methods when hexagonal warping effect occurs. With the conclusion that we previous have, HDP of 1550 nm is related with TSSs, and we can conclude that hexagonal warping part of the TSSs could be the origin of the out-of-plane spin. Our study proves several speculations in the field, and our new finding may initiate new applications or researches for spintronics.