論文の内容の要旨

Spin injection and spin transport in superconductors

(超伝導体中におけるスピン注入とスピン輸送)

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Spintronics is an active research field in condensed matter physics, whose aim is to exploit and manipulate the spin degree of freedom. Despite the growing interest in spintronics, however, spin transport in superconductors has not been explored yet especially from the experimental point of view. In this study, we have investigated spin transport in superconductors. We mainly discuss three subjects in the thesis; the spin relaxation time in a superconducting Nb, the inverse spin Hall effect (ISHE) in a superconducting niobium-nitride (NbN) and generation of the spin-triplet supercurrent in the superconductor (S) – ferromagnet (F) – S Josephson junctions.

Spin relaxation for spin currents is an important factor because it determines how long electrons can keep the initial direction of spin angular momentum. The most critical difference in spin transport in superconductors from that in normal metals is that it is mediated by the Bogoliubov quasiparticles, rather than electrons. These Bogoliubov quasiparticles can be regarded as a superposition of electron-like and hole-like excitations, and due to the different energy dispersion than that for electrons, the group velocity of the quasiparticles is smaller than that of electrons. Since spin relaxation occurs after electrons experience many scatterings by phonons and impurities, smaller group velocity brings about longer spin relaxation time. There have been several studies which investigate the spin relaxation time in superconductors, but their results are not conclusive: Due to spurious effects, underestimation or overestimation occurs, and it makes precise evaluation of the spin relaxation time difficult in superconductors.

In our work, we inject spin currents into a superconductor, and investigate the spin relaxation time in the superconducting state. We exclude the spurious effects described above by using the refined device structure, and attempt to estimate the spin relaxation time precisely. To attain this goal, we fabricate the lateral spin valve (LSV) devices. These devices are composed of two ferromagnet wires bridged by a nonmagnet wire. As a ferromagnet, we use permalloy (Py, $Ni_{81}Fe_{19}$), and as a nonmagnet, Cu. In these devices, when a change current passes between one of the two Py wires and the Cu bridge, a spin current is generated in the Cu. This spin current can be nonlocally detected using the other Py wire, and the detected signals are called nonlocal spin valve (NLSV) signals. We choose Nb as a superconductor because it has high critical temperature (T_C = 9.2 K) among metallic superconductors, and also has large spin-orbit interaction (SOI). Large SOI is also good to observe the spin Hall effect (SHE). For materials with large SOI, the spin absorption technique is useful to inject spin currents. When a wire with large SOI is inserted below the Cu bridge in the LSVs, the spin current is partly absorbed into the Cu, because it is energetically favorable for the spin current to enter into the wire with large SOI and relax faster. As a result, the detected NLSV signals in the other Py wire are suppressed. Through this spin absorption technique, we inject spin currents into Nb and investigate the difference in the spin absorption between the normal state and the superconducting state.

We perform the spin absorption experiments both above and below T_C (= 5.5 K in our device). At 10 K, above T_C, the NLSV signals from the LSVs with the Nb middle wire are suppressed compared with those from the LSVs without the Nb middle wire, as in our previous studies. The spin absorption is independent of the magnitude of the charge current we flow between the Py spin injector and the Cu bridge (spin injection current, I). At 370 mK, much lower than T_C, however, the situation becomes drastically different: The spin absorption strongly depends on I, and as I decreases, the NLSV signals increase. These increasing NLSV signals are the signature of the suppressed spin absorption.

To determine the origin of this anomalous behavior in the spin absorption, we measure the resistance close to the Cu/Nb interface (R_I), because the interface is the most sensitive part for spin absorption. Temperature dependence of R_I is first measured. We next fix the bath temperature and modulate I, and simultaneously measure R_I. Then we obtain the same curve for the relation between R_I and T, and R_I and I. This indicates that the effective temperature at the Cu/Nb interface is deviated from that of the bath due to I.

Taking into account these effects, we carry out theoretical calculations. When transport of electrons between the Cu and Nb wire is considered, it is necessary to calculate the density of states (DOS) of Nb. We note that in our LSVs, the Cu/Nb interface is highly transparent owing to the fabrication through the shadow evaporation. For this transparent contact between a superconductor and a normal metal, it is essential to account for the superconducting proximity effect. The DOS of Nb can be calculated with the Usadel equation in this regime. The point to note here is that in the Usadel equation, there is a term which contains the spin relaxation time. Therefore by using the spin relaxation time as a fitting parameter, we can calculate the amount of the absorbed spin current into the Nb wire so as to reproduce the experimental data. We perform the calculation based on this idea, and succeed in reproducing the experimental data of the NLSV signals as a function of I at 370 mK (the bath temperature). From the theoretical fitting, we also obtain the spin relaxation time for each I. The spin relaxation time in the superconducting state is found to increase with decreasing I, and it becomes more than four times larger than that in the normal state when $I < 10$ μA. Considering the effective temperature increase with I, this result is a clear experimental demonstration of the enhanced spin relaxation time in the superconducting state with decreasing temperature, as theoretically predicted.

We next investigate the SHE in a superconductor. In place of Nb used in the above study, we use niobium-nitride (NbN) in the present case owing to higher T_C. The device is composed of a Py wire and a NbN wire bridged by a Cu wire. Using the spin absorption technique, we can inject pure spin currents into the NbN wire. The injected spin currents are converted into charge currents through the ISHE, which can be detected as a voltage difference between the two edges of the NbN wire. The detected voltage depends on the orientation of the spin polarization of the injected spin currents, which follows the direction of the magnetization of the Py spin injector. Thus during the measurements we apply the inplane magnetic field to control the magnetization of the Py.

We perform the ISHE measurements both at 20 K (> T_C = 10 K) and 3 K (< T_C). At 20 K, we observe typical inverse spin Hall signals (ΔR_{ISHE}), and ΔR_{ISHE} do not depend on the magnitude of the spin injection current (I).

We next measure the ISHE at 3 K, then ΔR_{ISHE} first decreases with decreasing I, and then for I < 100 μA they increase dramatically. With I = 0.01 μA, the signal is more than 2000 times larger than that in the normal state.

To confirm that the observed signals derive from the ISHE, we measure the angular dependence of the signals on the angle θ between the external magnetic field and the longitudinal axis of the Py spin injector. Then the angular dependence shows the sinusoidal relation to θ, a signature of the ISHE.

We also investigate how superconductivity of NbN plays a role for this enormous ISHE. As noted above, the unique feature of spin transport in superconductors is that it is mediated by the Bogoliubov quasiparticles. These quasiparticles are composed of a combination of electron-like and hole-like excitations. At equilibrium, the number of quasiparticles in the electron-like branch and hole-like branch is balanced. When the ISHE occurs in superconductors, this balance between the two branches is broken, and the charge imbalance (CI) occurs. This charge CI effect is a nonequilibrium phenomenon, and has to relax in a certain time or length. The ISHE can be detected through the CI effect in the superconducting state, and to obtain the signals one has to place the voltage probes with in the length (CI length) from the region where the ISHE occurs. In the ISHE in NbN, due to large SOI of NbN thus the small spin diffusion length, the ISHE arises just below the Cu/NbN interface in the NbN wire. Therefore if superconductivity plays a role for the enormous ISHE, there should be a distance dependence of the detected signals between the Cu/NbN junction and the voltage probe (d). To confirm this scenario, we prepare two devices with different d, d = 0.4 μm and 10 μm. As a CI length, we use 4 μA from the value for Al as a reference. We measure ΔR_{ISHE} for the two devices at 3 K and 20 K. At 20 K, they show almost the same magnitude of ΔR_{ISHE}. However, at 3 K, while the device with d = 0.4 μm shows very large signals, signals from that with d

= 10 μm are strongly suppressed. Based on these results, we can conclude that the signals are detected via the CI effect and superconductivity of NbN should play an important role for the enormous ISHE.

We next carry out calculations to analyze the experimental data. ΔR_{ISHE} is proportional to the longitudinal resistivity ρ_{xx} and its quadratic, ρ_{xx}^2. In superconductors, ρ_{xx} has to be replaced by ρ_{qp}, the resistivity of quasiparticles. Due to the superconducting gap, ρ_{qp} is written as $\rho_{xx}/2f_0$ (Δ), where f_0 (Δ) is the Fermi distribution function at the superconducting gap Δ. Since ρ_{qp} is increasing with decreasing temperature, ΔR_{ISHE} can also be enhanced. Based on this idea, we perform numerical calculations, and obtain increasing ΔR_{ISHE} with decreasing temperature (T). We note here that in our experiments, is increasing with I, not T. To investigate the relation between I and T, we measure the resistance close to the Cu/NbN interface (R_{I}) in the same way as that described above. Then we obtain a good agreement between the relation R_{I} vs temperature and R_{I} vs I. By comparing these two relations, we can relate the effective temperature at the Cu/NbN interface to I. However, direct substitution of this relation into the equation between ΔR_{ISHE} and T does not reproduce the enormous enhancement of ΔR_{ISHE} with decreasing I. Based on the obtained relation between I and T through R_{I}, we assume that the effective temperature at the Cu/NbN interface is proportional to the square-root of I. By using this relation we can reproduce the experimental data fairly well.

The final subject is generation of the spin-triplet supercurrents in SFS Josephson junctions. Spin-triplet supercurrents are attractive in terms of spintronics because they can carry spin angular momentum truly without dissipation. In recent years, there have been reports on the observation of spin-triplet supercurrents in SFS Josephson junctions by using strong ferromagnets as Fs. However, no studies have directly linked spin-triplet supercurrents to spintronics.

To explore the potentiality of spin-triplet supercurrents for spintronics, we fabricate the SFS Josephson junction composed of the Co ferromagnetic wire and two tungsten (W) wires deposited by the Focused-Ion-Beam (FIB) system. W deposited by FIB systems is known to show T_{C} much higher than that of bulk W. We first measure superconductivity of W itself, and observe T_{C} = 9.5 K for our devices. We next measure the superconducting transition of the W-Co-W Josephson junction, where W-W distance (d) is 600 nm. Then we obtain the zero resistance through the junction at 7 K, even though d is much larger than the coherence length of the ferromagnet based on spin-singlet supercurrents. We also confirm that ferromagnetism of the Co wire is sustained by measuring the anisotropic magnetoresistance at room temperature. These facts support the observation of spin-triplet supercurrents in our device, which might be induced by the spin active interface due to the strong SOI of W wires.