Spectroscopic evidence of high transport spin-polarization in ferromagnetic CuFeSb

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Abstract

We have measured the transport spin polarization in the itinerant ferromagnet CuFeSb by spin-resolved Andreev reflection spectroscopy. From the analysis of the Andreev reflection spectra within a modified Blonder-Tinkham-Klapwijk (BTK) formalism that includes Fermi-level spin polarization and the spectral broadening due to finite quasiparticle lifetime, we found that the intrinsic transport spin polarization in CuFeSb is approximately 50%. This is significantly different from the half-metallic behaviour of CuFeSb as expected from band-structure calculation. We attribute this difference to possible difference in the Fermi velocities in the spin up and the spin down bands of CuFeSb.

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It is believed that the exotic superconductivity in the iron-based pnictide and chalcogenide superconductors originates from a magnetically driven pairing mechanism where the superconducting order is thought to be coupled with spin fluctuations\cite{1-5}. Therefore, in order to understand the nature of coupling between the superconducting order and spin fluctuations, it is imperative to understand the nature of magnetism and the spin fluctuation in the parent compounds from which the superconducting states are derived through doping. Usually the parent compounds of the pnictide and the chalcogenide superconductors are known to be antiferromagnetic where the antiferromagnetism is thought to be promoted by spin density waves (SDW) associated with Fermi surface nesting\cite{6-13}. More recently it has been shown that CuFeSb, which is isostructural to the iron-based layered superconductors e.g., Li(Na)FeAs\cite{14}, has a ferromagnetic ground state\cite{15}. A close relative of this compound CuFeAs stabilizes in an antiferromagnetic ground state with a Neel temperature of 9 K\cite{16}. The ferromagnetic order in CuFeSb is thought to originate from the large height of Sb from the Fe plane. This fact also supports the hypothesis that the competing magnetic interactions in ferropnictide superconductors is decided by the anion height i.e., there is a gradual change in the magnetic properties from superconductivity to antiferromagnetism to ferromagnetism on moving in the increasing order of anion height from LiFeAs to CuFeAs to CuFeSb \cite{15-17}. Until now CuFeSb has been known to be the only material in the FeAs or FeSb family that shows a ferromagnetic ground state. Therefore it is most important to understand the Fermi surface properties of this unique system by spectroscopic measurements – particularly, the nature of the Fermi surface spin polarization and the degree of spin fluctuations.

In this paper we have employed spin-resolved Andreev reflection spectroscopy using conventional superconducting tips to measure the spin polarization at the Fermi level of CuFeSb \cite{18}. From the analysis of the Andreev reflection data between the superconductor and the ferromagnet, we found the evidence of a high degree of transport spin polarization approaching 50% in CuFeSb. In addition to the insight regarding the magnetic interactions in the ferropnictide superconductors that might emerge from this result, it should be noted that the ferromagnets with more than 50% transport spin polarization are potential candidates as spin source in spintronic devices.

The transport through a ballistic point-contact between a normal metal and a superconductor is dominated by Andreev reflection that involves the reflection of a spin up (down)
electron as a spin down (up) hole from the interface\cite{19}. The Andreev reflection spectra ($dI/dV$ vs. $V$) are traditionally analyzed by the theory developed by Blonder, Tinkham and Klapwijk (BTK)\cite{20}. This theory assumes a delta-function potential barrier whose strength is characterized by a dimensionless parameter $Z$ which is proportional to the strength of the barrier. Ideally, for elemental superconductors where the quasiparticle life-time is very large, the Andreev reflection spectra can be fitted using two fitting parameters, $Z$ and $\Delta$, the superconducting energy gap. For all non-zero values of $Z$, the $dI/dV$ spectra show a double-peak structure symmetric about $V = 0$. However, in the system where the life-time of the quasiparticles is finite due to inelastic processes at the interface, the Andreev reflection spectrum undergoes broadening. This broadening is accounted for in the modified BTK theory where a complex component $i\Gamma$ is artificially added to the quasiparticle energy ($E$)\cite{21}. In such cases the spectra are analyzed using three fitting parameters, $Z$, $\Delta$ and $\Gamma$.

When the metal in the metal-superconductor point-contact is a ferromagnet, the Fermi level is expected to be spin polarized\cite{13}. This means, the density of states of the up-spin electrons ($N_\uparrow$) is not equal to the density of states of the spin down electrons ($N_\downarrow$) at the Fermi level. Therefore, $|N_\uparrow - N_\downarrow|$ electrons encountering the interface cannot undergo Andreev reflection because they do not find accessible states in the opposite spin band. Therefore, in a point-contact between a ferromagnetic metal and a conventional superconductor, Andreev reflection is suppressed. By measuring the degree of this suppression the spin-polarization of the Fermi surface is measured\cite{22–24}. In order to extract the absolute value of the Fermi level spin polarization, first the BTK current is calculated for zero spin polarization ($I_{BTKu}$) and 100% spin polarization ($I_{BTKp}$) respectively. Then the current for an intermediate spin polarization $P_t$ is calculated by interpolation between ($I_{BTKu}$) and ($I_{BTKp}$) following the relation $I_{total} = I_{BTKu}(1 - P_t) + P_tI_{BTKp}$. The derivative of $I_{total}$ with respect to $V$ gives the modified Andreev reflection spectrum with finite spin polarization in the metal. This model is used to analyze the spin-polarized Andreev reflection spectra obtained between a ferromagnetic metal and a superconductor by using four fitting parameters $Z$, $\Delta$ and $\Gamma$ and $P_t$. It should be noted that for the measurement of the spin polarization of ferromagnets usually standard conventional superconducting probes are used for which the value of $\Delta$ is known. In addition, in order to have superconductivity, $\Gamma$ cannot be arbitrarily large with respect to $\Delta$. Therefore, effectively only two parameters, $Z$ and $P_t$, are tuned freely during the analysis of spin-polarized Andreev reflection spectra. For our analysis the
FIG. 1: Point-contact Andreev reflection spectra between superconducting Nb tip and CuFeSb. The red dotted lines show the raw experimental data and the solid black lines show the theoretical fit (see text). All the spectra are normalized with respect to the high-bias conductance. \((dI/dV)_{N}\) indicates normalized differential conductance. The measurement temperature and the fitting parameters are listed in the respective panels.

Theoretical spectra were generated by a code written in python.

All the measurements reported in this paper were performed on a polycrystalline pellet of CuFeSb. CuFeSb shows a ferromagnetic transition around 380 K\cite{15}. The Andreev reflection spectroscopic measurements were performed by measuring the transport characteristics of several ballistic point-contacts between CuFeSb and the elemental superconductors niobium (Nb) and lead (Pb) respectively using a home-built point-contact spectroscopy probe in a liquid helium cryostat. First the polycrystalline pellet of CuFeSb was polished and
mounted on a copper disc which is used as the sample holder in the home-built probe. A calibrated cernox thermometer and a heater were attached to the same copper disc for precise measurement of the temperature and for controlling the sample temperature during the Andreev reflection spectroscopic measurements. Two 100 µm thick gold wires were mounted on the sample for transport measurements. The superconducting tips were fabricated from 250 µm diameter wires of Nb and Pb respectively. The tips were mounted on a teflon piece connected to the head of a 100 threads per inch differential screw. Two more 100 µm thick gold wires were mounted on the tip. The probe was then mounted inside the static variable temperature insert (VTI) of a liquid-helium cryostat. The static VTI was surrounded by a dynamic VTI with a micro-capillary that allowed us to perform measurements down to 1.4 K.

The ballistic point-contacts between the sample and the tips were fabricated and controlled by moving the tip up and down by rotating the differential screw manually. For electrical measurements a lock-in modulation technique was employed where a dc current was coupled with a small current drawn from the sinusoidal output of a digital lock-in amplifier (Stanford Research Systems, model: SR-830) through a passive current source. The modulated current was sent through the point-contacts. The dc component of the voltage drop across the point-contacts was measured by a digital voltmeter (Keithley Model: 2000) and the ac component was measured by the same lock-in amplifier. The data acquisition was done by using a lab-view programme developed in house. The ac voltage drop measured in the lock-in is proportional to differential change in voltage (dV). dI/dV plotted against the dc voltage drop V across a point-contact gives the point-contact Andreev reflection spectrum. dI is proportional to the excitation current set during the experiment.

In Figure 1 (a,b,c) we show three representative Andreev reflection spectra between a Nb-tip and CuFeSb. The spectra clearly show the double-peak structure symmetric about V = 0, which is the hallmark of Andreev reflection. For low values of Z, these peaks appear close to the energy gap of the superconductor. The solid lines show the theoretical fits as per the model described above. The superconducting energy gap of niobium is found to be approximately between 1 meV and 1.5 meV for all the point-contacts that we have analyzed, indicating the proximity of the ferromagnet does not suppress the superconductivity of the point-contacts significantly. The value of Γ remained zero for all the spectra, which means the broadening due to finite quasiparticle lifetime is absent at the point-contact. This
FIG. 2: Point-contact Andreev reflection spectra between superconducting Pb tip and CuFeSb. The red dotted lines show the raw experimental data and the solid black lines show the theoretical fit (see text). All the spectra are normalized with respect to the high-bias conductance. \((dI/dV)_N\) indicates normalized differential conductance. The measurement temperature and the fitting parameters are listed in the respective panels.

fact also indicates that the spin-fluctuation in the system is not significant as strong spin-fluctuations is also known to give rise to large \(\Gamma\)\textsuperscript{[25]}. Therefore, the theoretical fits are obtained by essentially tuning two parameters namely \(Z\) and \(P_t\), this makes the fit accurate and the fitting parameters unique. It is found that the raw data deviate slightly from the fit at certain points (notice the dip structures in \(dI/dV\)). Such deviation is known to originate from the critical current of the superconductor when a small part of the Maxwell’s resistance is also measured along with the Sharvin resistance in the point-contacts close to
the ballistic regime. The experiments were repeated with superconducting Pb-tips to confirm the reproducibility (Figure 2). All the spectra measured with Pb tips were found to be considerably broader than the theoretically generated spectra. This can be attributed to the low-energy phonon modes of Pb that couple strongly with the quasiparticles and might modify the point-contact spectra. However, the low-energy part of the spectra clearly show the signature of Andreev reflection (the double-peak structure symmetric about $V = 0$). This part of the spectra were fitted nicely and the relevant parameters were extracted by fitting the low-bias portion of the spectra. Again, in such fittings, the value of $\Gamma$ remained almost zero and the superconducting energy gap was found to be approximately 1$meV$ which is same as the superconducting energy gap of bulk Pb.

The dependence of $P_t$ on $Z$ for both Nb and Pb based point-contacts is shown in Figure 3. For most of the Nb/CuFeSb point-contacts, the value of $Z$ was found to be small ($< 0.2$) For such point-contacts, the maximum measured value of $P_t$ is found to be 52%. For the Nb/CuFeSb $P_t$ did not change noticeably with $Z$. It should be noted that within the BTK formalism, no correlation between $P_t$ and $Z$ is expected. For the Pb/CuFeSb point-contacts, however, $P_t$ shows small dependence on $Z$ and the dependence is linear – the measured value of $P_t$ decreases with increasing strength of the barrier (represented by $Z$). Although this dependence is not understood within the BTK formalism, for a vast majority of ferromagnetic point-contacts such dependence was found in the past. In such cases the dependence is attributed to spin depolarization at a magnetically disordered scattering barrier formed at the interface. In such cases, the conventional way of finding the intrinsic transport spin polarization is to extrapolate the $P_t$ vs. $Z$ curve to $Z = 0$. By doing this extrapolation, the intrinsic $P_t$ is found to be approximately 47% which is nearly equal to the value measured with the Nb tip. Therefore, it is rational to conclude that the degree of spin polarization at the Fermi level of CuFeSb is approximately 50%.

It should be noted that the accurate value of the transport spin polarization can be obtained only from the point-contacts that are in the ballistic or diffusive limit of transport. For our analysis, we have carefully chosen only the spectra that show the double-peak structure symmetric about $V = 0$, which is a clear signature of Andreev reflection in the ballistic or diffusive limit. One signature of a non-ballistic point-contact is sharp conductance dips symmetric about $V = 0$ associated with the critical current of the point-contacts. We have ignored all the spectra showing sharp conductance dips. Some of the spectra that
FIG. 3: Dependence of transport spin-polarization on $Z$ for point-contacts on CuFeSb with Nb and Pb tips. The red circles show the data points for Nb tips and the blue stars show the data points for Pb tips. The solid lines show the respective linear fits: Red line for Nb tips and blue line for Pb tips. Though $P_t$ shows different dependence on $Z$, the intrinsic value of $P_t$ extracted for $Z = 0$ is identical for both Nb and Pb tips.

we have presented here show very small conductance dips indicating that the point-contacts were close to the ballistic limit containing negligibly small contribution of thermal resistance.

The experimentally obtained value of the intrinsic spin-polarization at the Fermi level is significantly different from the spin-polarization estimated by local spin density calculations earlier performed by Sargolzaei.\cite{27} According to this calculation, the Fermi-level is almost 100% spin polarized that makes CuFeSb a half-metallic ferromagnet. This discrepancy between the experimental results and theoretical prediction may be explained as follows.

In spin-resolved Andreev reflection spectroscopy the difference in up-spin current ($J^\uparrow$) and down spin current ($J^\downarrow$) is measured. Therefore, the relevant definition of spin polarization in such measurements should be the ”transport spin polarization” $P_t = \frac{|J^\uparrow - J^\downarrow|}{J^\uparrow + J^\downarrow}$ \cite{13, 23}. Now, in the ballistic limit of the point-contact, $J^\uparrow = < N^\uparrow v^\uparrow F >$ and $J^\downarrow = < N^\downarrow v^\downarrow F >$, where $v^\uparrow F$ and $v^\downarrow F$ are the Fermi velocities of the spin up electrons and the spin down electrons respectively and the symbol $<>$ indicates an averaging over the respective Fermi surfaces\cite{13, 28}. Even when the point-contacts are not in the pure ballistic limit but in the diffusive regime of transport where only elastic scattering processes at the contact region are allowed, the definition of transport spin polarization has a dependence on the fermi velocities. In the
diffusive regime, $J_\uparrow = \langle N_\uparrow v_{F\uparrow}^2 \rangle$ and $J_\downarrow = \langle N_\downarrow v_{F\downarrow}^2 \rangle$. Therefore, even though the spin polarization as per regular definition ($P = \frac{|N_\uparrow - N_\downarrow|}{N_\uparrow + N_\downarrow}$) is high, it is possible that the lower value of the measured transport spin polarization ($P_t$) is due to a significant difference in $v_{F\uparrow}$ and $v_{F\downarrow}$. The possibility of this difference should be explored theoretically.

In conclusion, we have measured the transport spin polarization of ferromagnetic CuFeSb. From the analysis of the Andreev reflection spectra we obtain a spin polarization of approximately 50% which is significantly lower than the expected 100% Fermi surface spin polarization as per band structure calculations. The lower degree of measured transport spin polarization can be attributed to a possible difference in the Fermi velocities of the spin up and the spin down electrons.

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