Out of plane superconducting Nb/Cu/Ni/Cu/Co triplet spin-valves

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Abstract:

The critical temperature of a triplet superconducting spin valve depends on the effectiveness of the conversion of singlet pairs in the superconductor to triplet pairs which can penetrate a ferromagnet and so drive a strong proximity effect. Here we compare the out-of plane field dependence of the critical temperature in transition metal triplet spin valves with otherwise equivalent samples in which the singlet-triplet converting spin mixer ferromagnet layer is omitted. We report a significant field-dependent difference between the samples which is consistent with a magnetisation orientation dependent spin mixing efficiency in the spin valve. .

Main text

Superconducting spin electronics (superspintronics) is an emerging field that holds great potential for devices with faster performance and greater energy efficiency than current semiconductor architectures.[1, 2] A key potential element for this technology is the superconducting analogue of the spintronic spin valve. In a conventional (singlet) ferromagnet / superconductor/ ferromagnet (F/S/F) superconducting spin valve (SSV) the critical temperature (T_c) is suppressed when the two F layer magnetisations are parallel (P) in comparison with the antiparallel (AP) state.[3-6] The effect occurs because of the addition of the F layer exchange fields in the P state, which disrupts the singlet electron pairing, and their partial cancellation in the AP state.[4]

Recently, a more complete synergy between superconductivity and spintronics has been made possible through the discovery of mechanisms for the generation of odd-frequency spin-triplet Cooper pairs at S/F interfaces.[7-9] A key experimental method for understanding the triplet proximity effect is the measurement of T_c changes in S/F1/N/F2 superconducting triplet spin valves (TSVs) as a function of the misalignment of the magnetisations of F1,2 ferromagnet layers (the spin mixer and drainage layer respectively); N is normal, non-magnetic spacer to decouple the magnetisations of F1 and F2.[10-13] Theories of the triplet

proximity effect suggest that it is maximised, and hence T_c is minimised, when the magnetisation of F1 and F2 are orthogonal, so that triplet pairs generated by the spin mixer layer can penetrate effectively into the drainage layer and so weaken the overall superconductivity.[14, 15]

There are several experimental reports on TSVs which use the standard ferromagnetic metals (Fe, Co, Ni) as the spin mixer and drainage layers.[10-12] Most experiments on such devices control their relative magnetisation directions with the application of field in the plane. However, the largest changes in T_c (ΔT_c), and hence the strongest triplet proximity effects, has been reported for MoGe/Ni/Cu/CrO$_2$ TSVs in which the field is applied out of plane and the relative alignments of the Ni and half-metallic CrO$_2$ magnetisations is determined by the different susceptibilities of the two layers.[13] This result is striking, not only in terms of the magnitude of ΔT_c, but also in its field dependence which continues to increase well beyond the saturation field of both magnetic layers (i.e. when the magnetisation of the two layers is expected to be parallel). In order to explore whether the geometry of the experiment, or the half-metallic nature of the CrO$_2$ was the controlling factor in the magnitude and field-dependence of ΔT_c the experiments reported in this paper aimed to repeat the study but with CrO$_2$ replaced by Co.

One of the problems with the out of plane geometry is that substantial fields need to be applied normal to the S layer, which leads to Abrikosov vortex nucleation and a substantial T_c suppression and so it is important to understand the behaviour of control samples in which the spin mixer layer is absent. We have therefore performed $T_c(H)$ measurements on Nb(20)/Cu(5)/Ni(2)/Cu(5)/Co(50) TSVs (layer thicknesses in nm) and otherwise identical control samples for which the Cu(5)/Ni(2) bilayer was omitted. The Cu layers are included to magnetically decouple the F1/F2 layers and reduce the magnetic dead layer which forms at a Nb/Ni interface.[16] The Nb layer is chosen to be thick enough to exhibit superconductivity at reasonable temperatures. The thickness of the Ni F1 layer was comparable to the singlet coherence length[16] and similar to that used in Singh *et al*.[13] By changing the relative magnetisation directions of F1 and F2, the triplet Cooper pair potential is varied accordingly and hence the amount of singlet pairs that converted to triplet is changed.

The polycrystalline heterostructures were grown by dc-magnetron sputtering on unheated Si/SiO$_2$ substrates at a pressure below 10^{-8} mbar. Electrical transport measurements on these samples were performed using a four-probe geometry in a Quantum Design Physical Properties Measurement System. The magnetic measurements were performed at 300 K and 10 K using a Princeton vibrating sample magnetometer and Quantum Design MPMS respectively.

The magnetisation M versus applied field H of a control sample stack at 10 K is shown in Fig. 1. The magnetisation of the Co (which dominates the signal) shows an in-plane saturation field (H_S) of about 0.02 T which is much smaller than the out of plane H_S of about 2 T. Epitaxial (0001) Co films show a distinct transition from in-plane to out of plane anisotropy as the thickness increases above 40 nm which is driven by the magnetocrystalline anisotropy in this direction overcoming the shape anisotropy which typically dominates in thin films.[17] Polycrystalline films show a similar transition at a larger thickness reflecting the reduced net magnetocrystalline anisotropy.[18, 19] It is clear that our Co films are below this thickness threshold: the in-plane $M(H)$ measurements show easy-axis behaviour with low coercivity and magnetic force microscopy shows no evidence for the stripe domains typically seen for out of plane anisotropy. Nevertheless the out-of-plane $M(H)$ curve shows a significant remanence at zero field, indicative of a weak out-of plane anisotropy which probably originates from the (0001) growth texture of the film.[19, 20] The $M(H)$ of Ni shows less anisotropy and is saturated in both directions at about 0.5 T. The saturation magnetisation M_S is 4×10^5 A/m , which is comparable to other reports.[13]

Resistance vs temperature ($R(T)$) measurements for the TSV and control samples are shown in Fig. 2(a,b) for two different fields. We define T_c as the temperature at which the resistance has decreased to 50% of the normal state value. In Fig. 2(a) one can see that, at zero field, T_c of the TSV is around 1 K lower than the control sample which is probably due to the greater thickness of non-superconducting metals in the TSV than the control sample. By applying a field of 0.5 T in-plane and out-of-plane, as shown in Fig. 2(b), the transition T_c out of plane is about 0.8 K lower than T_c with an in-plane field.

The variation of T_c as a function of applied field H in both samples is plotted in Fig. 2(c). The anisotropic field dependence of T_c in the superconducting thin films is well understood in

terms of Ginzburg-Landau theory (see for example Krasnosvobodtsev *et al.*[21]). In perpendicular field $T_c(H_\perp)$ can be shown to be given by

$$T_c = T_{c0} - \frac{2\pi\xi(0)^2 H_\perp}{\Phi_0}$$

where $\xi(0)$ is the zero-temperature Ginzburg-Landau coherence length and Φ_0 is the flux quantum. In contrast, in a parallel magnetic field, a thin film exists in a vortex-free state which greatly increases the upper critical field (H_{c2}) and a field dependence of T_c which tends to $T_{c0} - T_c \propto H_\parallel^2$ at high fields. As shown in Fig 2(c), $T_c(H)$ in both samples and field directions is broadly consistent with these models.

However, $T_c(H_\perp)$ of the trilayer spin valve is not strictly linear at low magnetic field, as shown in Fig. 2(d), in which the normalised T_c of both samples is plotted. While the control sample is essentially linear with field, the T_c of trilayer sample significantly deviates from this. To make this deviation clearer, in Fig. 3 we plot the difference between the normalised $T_c(H_\perp)$ for the two samples (i.e. $\Delta T_c/T_{c0}$ for this field orientation). In contrast, the in-plane samples over the same field range show essentially identical normalised $T_c(H)$ behaviour (see Fig. 2(d)).

The hypothesis is that the T_c deviations highlighted in Fig. 3 originate from a triplet proximity effect which is dependent on the relative alignment of the Co and Ni layers when the field is applied out of plane. In-plane, the magnetisation curves show that both F layers are essentially parallel throughout the field range explored and so the spin-one triplet proximity effect should be eliminated.

In order to confirm this explanation, we start from a simple model for the triplet proximity effect. Houzet and Buzdin[14] show that, for limits which are relevant to our device structure, the spin mixing efficiency $P \propto \sin(|\theta_{F1} - \theta_{F2}|)$. Our unpatterned samples are certainly not single domain but using a simple Stoner model for the magnetisation we can estimate the angles of the magnetisation of each layer – i.e. $\sin(\theta(H)) = M(H)/M_s$.

In Fig. 3 we plot $P(H)$ calculated from the $M(H)$ plots in Fig. 1. It is evident that this qualitatively explains the observed $\Delta T_c(H)$ dependence. The inset diagrams illustrate the magnetic configurations from which this is derived: at zero field, the Co magnetisation

shows a significant remanence (but without forming stripe domains) and so is misaligned with the Ni film which is aligned with the plane. With increasing field, the much lower susceptibility of the Ni means that the magnetisations of the two F layers first becomes parallel and hence P = 0 at ~30 mT: The minimum triplet proximity effect occurs as the system approaches the alignment field and so consequently the minimum T_c suppression is observed at this point. As the field is increased, the magnetisation angles diverge as the Ni layer rapidly tends to out of plane alignment with the maximum, and hence the maximum value of P, appearing when the Ni reaches out of plane alignment while the Co remains at a low angle to the film plane. Finally, the Co layer more slowly approaches saturation and so the P and the triplet proximity effect gradually decreases again.

The maximum value of $\Delta T_c/T_{c0}$ is ~ 0.015, which translates to ~ 100 mK given that T_{c0} ~ 6K which is comparable to previous in-plane rotation measurements of triplet spin valves.[11] It is however significantly larger than previous out of plane measurements using transition metal ferromagnets,[12] probably because our experiments used a Ni mixer layer rather than the CuNi alloy by Zdravkov *et al.*.[12] This is consistent with previous work in which using Ni rather than CuNi mixer layers has been shown to lead to much larger critical currents in triplet Josephson junctions.[22] The value is clearly much lower than that (800 mK) obtained with CrO_2 by Singh *et al.*;[13] nevertheless, it is clear that the field dependence of the effect reported here is very different to that seen in CrO_2 TSVs: in the latter case, the peak value occurs at ~2 T, well above the saturation field for both magnetic layers.

In conclusion, we have shown that significant triplet proximity effects can be achieved in an out of plane geometry using transition metal ferromagnets. Although our devices required substantial fields (compared to fields required for a standard in-plane spin valves[5, 23]) to generate the maximum effects, even low fields of a few tens of mT generated measurable T_c changes (i.e. significantly below those required for CrO_2 devices).[13] With careful optimisation of the anisotropies of the F layers it should be possible to create devices in which much smaller fields (for example a few mT [24]) of magnitudes which could be generated by control lines, or even spin transfer torque effects could achieve T_c control. This would both increase the potential for practical device development and reduce the competing field-induced T_c suppression.

Acknowledgements

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FIG 1. The magnetisation *M* versus applied field *H* at 10 K for (a) Nb(20)/Cu(5)/Co(50) and (b) for Cu(5)/Ni(2)/Cu(5) samples (layer thicknesses in nm). In-plane measurements blue triangles; out of plane measurements red circles.

FIG 2. Resistivity of TSV and control samples: (a) resistive transitions at zero field; (b) 0.5T applied in-plane (IP) and out of plane (OOP) (c) superconducting transition temperature (T_c) as a function of field; (d) normalised T_c as a function of field. The key in (c) applies to all panels.

FIG. 3. (Data points) the difference of the normalised critical temperature t_c (= T_c/T_{c0}) between the triplet spin valve and control sample from Fig. 2(d) as a function of out-of-plane field. The solid line shows the spin mixing efficiency P calculated from the magnetisation of the two ferromagnetic layers as discussed in the text. The arrows illustrate schematically the angle of the magnetisations of the Ni (thin, red) and the Co (thick, blue). The inset shows the dependence of the sine of the magnetisation angle of the Ni and Co layers versus out of plane field derived from fits to the data of Fig. 1.