Spintronic devices using thin films of heavy metals: understanding the role of strong spin-orbit coupling and proximity-induced magnetism

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Abstract

Very little is known about the electronic properties of the heavy actinide as thin films mixed into non-magnetic and magnetic materials. The availability of thin film growth systems with these materials is very limited: here we use an actinide-dedicated DC magnetron sputtering system at the University of Bristol to investigate their possible application in spintronic applications.

1. Research Objectives

Current spintronics research is focussed on heavy metals as a source of highly spin polarized currents. They can give rise to large spin Hall effects which can be used for magnetization switching. Initial investigations of the transport properties of dilute-U/Cu, were begun to compare and contrast with previous studies of the spin-orbit coupling of Bi and Ir atoms in Cu, as shown in Fig. 1 below [1]. A key question in the community is what materials are most suitable to provide the largest signals. Actinide materials are of interest in the question, since in the most naive picture the spin-orbit coupling strength scales as a function of the atomic number to the fourth power. The objectives of this project are to continue this work and include Th as an additional type of impurity dopant as well as U. We will synthesize thin films of copper with dilute uranium or thorium content, investigating several different Th concentrations (typically in the range of a few %).

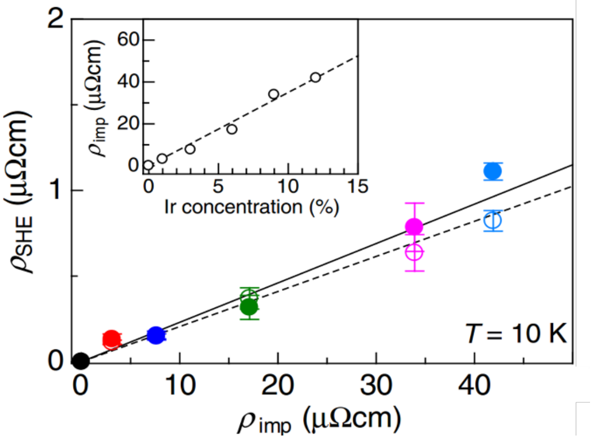

Fig. 1: Spin Hall resistivity in Cu as a function of induced resistivity due to Ir impurities. Taken from Ref. [1].

The behavior of actinide materials in proximity to ferromagnetic materials should also be understood, both to allow integration in more complex spintronic devices, and also for fundamental studies. A key point is that previous x-ray magnetic circular dichroism studies of uranium/ferromagnet thin film superlattices have indicated an induced moment in the U, which is especially big when the ferromagnet is iron, approaching 0.1 Bohr magnetons per atom just at the interface [2, 3], also see Fig. 2.

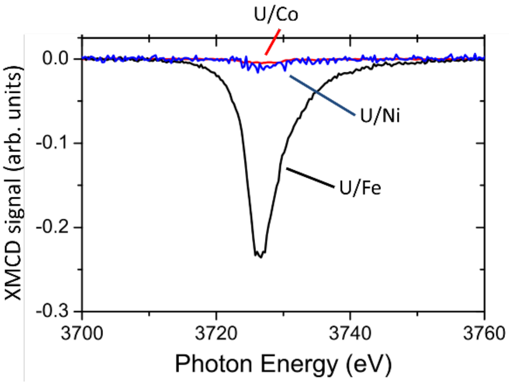

Fig. 2: XMCD data of U/Ferromagnet superlattices for three different elemental ferromagnets. Taken from Ref. [3].

2. Research Contents

The main results of this work have not been finalized due a delay in the transfer of the funds to Bristol. The contents of the research is focused on actinide materials in proximity to non-magnetic and ferromagnetic materials. For the former, we will continue the initial investigation of the transport properties of dilute-U/Cu, to compare with previous studies of the spin-orbit coupling of Bi and Ir atoms in Cu. These measurements are of interest since naively the spin-orbit coupling strength scales as a function of the atomic number to the fourth power. For proximity effects with ferromagnetic materials, previous x-ray magnetic circular dichroism studies of uranium/ferromagnet superlattices have indicated an induced moment in the U, which is especially big when the ferromagnet is iron. We aim to understand the strength of the spin-orbit interaction in actinide thin films, including the importance of the induced moment, using magnetization, magnetoresistance, and ferromagnetic resonance measurements in polycrystalline layers. We can compare to the literature on Pt, where the induced moment has also been discussed. We will contrast the behavior of different ferromagnets also with Th, whose electronic structure is less dominated by the 5f electrons: a far weaker induced moment is expected.

We will synthesize thin films of Cu, Co, Ni and Fe with dilute U or Th, investigating different concentrations (in the range of a few %). These will be grown in an actinide-dedicated system at the University of Bristol. The transport properties of these samples, including anomalous Hall effect and spin Hall effect, will be measured as a function of temperature and applied field. We aim to compare our experiments to theoretical results using relativistic bandstructure calculations correctly describing the dilute impurity system. The combination with transport calculations performed in Bristol will allow for a detailed understanding of the underlying mechanisms. If time permits we will also exam the ferromagnets Gd (a localized moment system) and YIG (an insulating material). However preliminary calibration and alloy thin films samples have been grown for studies of magnetic anisotropy.

3. Research results

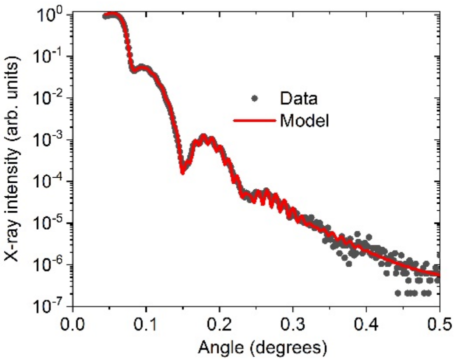

Fig. 3: XRR calibration of a Nb/U bilayer grown on glass at room temperature.

Preliminary samples of U:Fe alloys have been investigated. A key difference between the U:Cu alloys and the U:Fe samples is the sputtering rate of the magnetic elements is significantly smaller than the Cu. The samples are grown by co-sputtering – i.e. the U and Fe are deposited simultaneously. In order to achieve the lowest U concentrations a stable plasma at the lowest power must be utilized. The Fe and U rates were separately calibrated via x-ray reflectivity (XRR) on bilayers of Nb/Fe and Nb/U on glass substrates. Here the Nb layer is to prevent oxidation of the Fe or U. An example XRR plot and fit is shown in Fig. 3. However for the parameter space available we were unable to achieve U concentrations below 20%, which makes it difficult to model the scattering as from isolated U atoms in the magnetic matrix.

In order to achieve lower U concentrations we have fabricated and are currently testing a mask which is placed in front of the U sputter gun to significantly reduce the rate of deposition of U at the substrate position and enable the growth of U concentrations down to 1%.

Despite this setback we have begun preliminary measurements of the effect of U incorporation into the Fe thin film on the magnetic properties, as well as initial theoretical modeling of U and Th impurities in Cu using relativistic bandstructure calculations. The theoretical method to quantitatively predict the Spin Hall angles in dilute alloys is based on a relativistic Greens function method ideally suited for the treatment of impurity systems avoiding artificial boundary conditions. In Fig. 4 we give a visualization of the asymmetric scattering induced by the spin-orbit coupling coming from the heavy impurities U and Th in Cu. Preliminary calculations point to large Hall angles of 7.5% (Cu(U)) and 2.9% (Cu(Th)). Further optimization of the theoretical work will be undertaken once a systematic set of experimental measurements of the longitudinal resistivity versus U concentration have been completed.

Experimentally, specifically so far we have looked at the in-plane magnetic anisotropy at room temperature using a vibrating sample magnetometer. Some typical *M(H)* data are shown in Fig. 5: here we compare two samples with the same nominal total thickness of 20 nm. The pure Fe sample shows a clear angular variation of the magnetic anisotropy. The in-plane magnetization is expected for a film of this thickness, and the magnetic anisotropy is likely the well-known induced anisotropy due to stray fields in the sputtering chamber during growth. In contrast with a significant concentration of U there are two main observations: the magnetization is significantly suppressed leading to far more noisy *M(H)* data, and the angular variation in the anisotropy is far weaker than for the pure Fe sample. This may be associated with the strong spin-orbit scattering of U [4]. To confirm this issue we need to grow lower concentrations fo U, as discussed above, and also compare the results to alloys with Pt and other lighter elements. We also undertook preliminary anisotropic magnetoresistance and anomalous Hall effect measurements on these samples after patterning into Hall bars using optical lithography and acid etching, but the signals were relatively weak in the U-alloy samples.

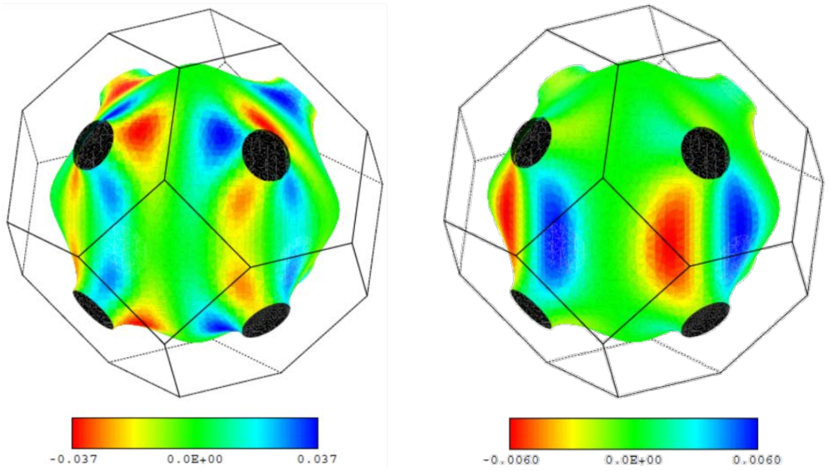

Fig. 4: Visualization of the skew scattering process induced by heavy impurities in Cu. On the Fermi surface of Cu the colour code represents the difference in scattering times for spin-up vs spin- down electrons. The breaking of the four-fold rotational symmetry is a direct measure of asymmetric scattering for spin-up and spin-down electrons via the skew scattering at the heavy impurities of U (left) and Th (right) in Cu.

In the future we also plan to examine ferromagnetic resonance of these systems and anomalous Hall effect data as well. We have been unable so far to acquire a Th target, but we hope in the coming months that we can also obtain Th to compare the results with U as well as Pt.

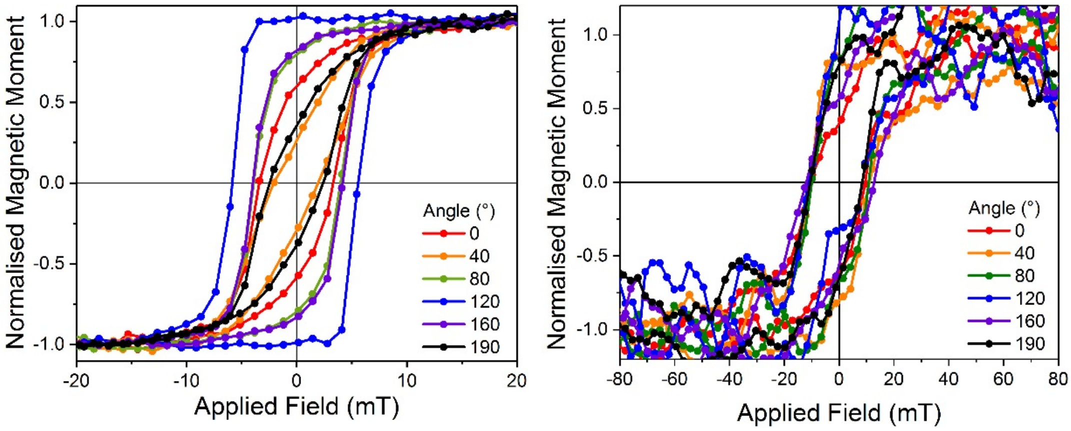

Fig. 5: *M(H)* data versus in-plane angle of pure Fe (left) and a U(20 atomic %):Fe alloy (right) at room temperature. Film thickness is 20 nm in both cases.

4. Spin-Hall effect measurement by FMR

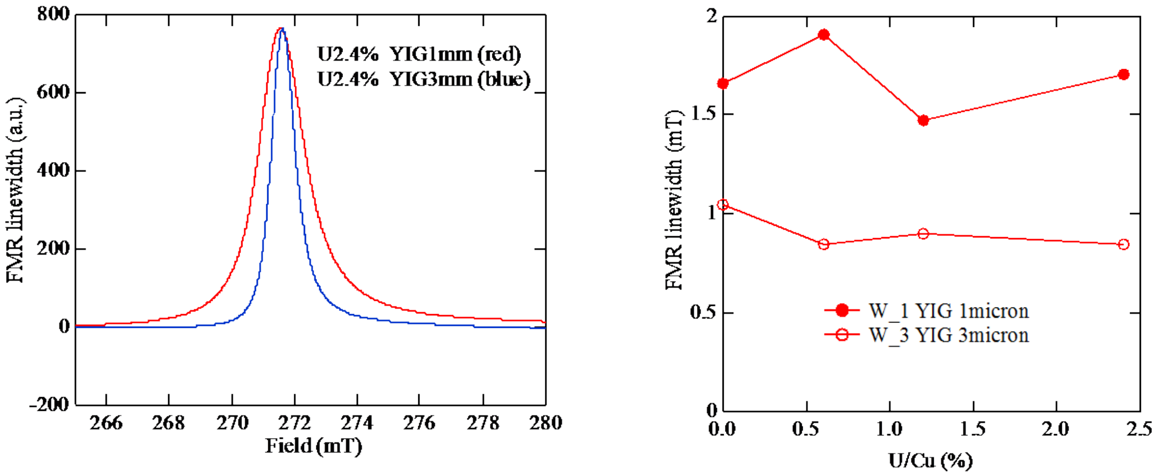

Fig. 6: Typical FMR spectra around 4kOe (left). The spectra can be well fitted with the Lorentzian functions. The linewidth is defined as full width at half maximum (FWHM) of fitted Lorentzian function. U concentration dependence of FMR linewidth (right). No clear dependence is observed in the present samples.

We have tried to detect the spin-Hall effect using Ferromagnetic resonance (FMR) method in diluted-U/Cu thin films prepared by magneto-sputtering method. At first, YIG ferromagnet (1 and 3 μm) thin film is developed on substrate, then 0%-2.4%U/Cu thin films are developed on the YIG thin film. If the spin-Hall effect appears due to the diluted U atoms, FMR linewidth increases with increasing the U concentration. Figure 6 shows typical FMR spectra and U-concentration dependence of FMR linewidth obtained by LS fitting based on the Lorentzian function. Unfortunately, a significant concentration-dependence of linewidth has not been observed in the present samples. Although the reason for the absence is not clear at the present, it seems that a polarized spin injection from YIG to dilute-U/Cu thin film does not work well in the present case, due to a mismatching at interface between YIG and dilute-U/Cu.

It might be possible that Permalloy (Ni/Fe) ferromagnet thin film is better than YIG for dilute-U/Cu thin film, or the thickness of YIG was too large. Indeed, the spin-Hall effect is detected in pure U-pemalloy film by FMR in previous study. We are planning to prepare different thin films with Permalloy and thinner YIG.

Conclusion

In conclusion, although the project has been delayed we have made significant initial progress in growing and characterizing the magnetic properties of U:Fe alloys. A clean change in magnetic anisotropy has been observed for relatively high concentrations of U doped in Fe. Efforts are currently underway to reduce the sputtering rate of the U to fully explore the most relevant low density limit in magnetic and non-magnetic systems.

5. References

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