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Colorado State University - Ft. Collins
Sponsored Programs
2002 Campus Delivery
Fort Collins, CO 80523 -2002

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19a. NAME OF RESPONSIBLE PERSON
Mingzhong Wu

19b. TELEPHONE NUMBER
970-491-6312

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INVESTIGATOR(S):

Name: Mingzhong Wu
Email: mingzhong.wu@colostate.edu
Phone Number: 9704916312
Principal: Y

Organization: **Colorado State University - Ft. Collins**
Address: Sponsored Programs, Fort Collins, CO 805232002
Country: USA

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Phone: (970) 491-6312
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Major Goals: Yttrium iron garnet (YIG), with its extremely low magnetic damping, is perhaps the optimum magnetic material for exploration of spin superfluidity. While there have been considerable theoretical studies on spin superfluidity in YIG thin films, so far there has been no experimental work of note. This STIR project aims to use low-damping YIG thin films to demonstrate room-temperature spin superfluidity for the first time. It also aims to explore the spatial characteristics of spin supercurrents. Perpendicularly-polarized spin currents from a lateral spin valve will be pumped into a YIG thin film to excite spin superfluidity. The detection of the spin superfluidity will rely on spin supercurrent-induced resistance changes in a ferromagnetic metal/normal metal bi-layered structure built on the top of the YIG film. The nature of the spin superfluidity state will be confirmed by measuring the spatial characteristics of the spin supercurrent. The results from this project will significantly promote the understanding of the spin superfluidity and spin supercurrent phenomena and will therefore significantly advance the research field of spin superfluidity, a new field that draws inspiration from both the magnetization dynamics and spintronics communities in particular and from the broad condensed matter physics community in general.

Accomplishments: The goal of this STIR project is to demonstrate room-temperature spin super-fluidity (SSF) in yttrium iron garnet (YIG) $Y_3Fe_5O_{12}$ thin films.

To achieve the goal, we need to grow a magnetic metallic thin film with perpendicular magnetic anisotropy (PMA) on a YIG thin film, which will be used to generate a perpendicularly-polarized spin current for the YIG film. We plan to use this perpendicular spin current to induce or excite SSF in the YIG thin film.

Prior to the pandemic-induced shutdown of our labs in March 2020, we focused mainly on the growth of CoFeB and $(Co/Ni)_n$ thin films with PMA on YIG thin films. Unfortunately, we failed to grow films with PMA using this set of materials. The primary reason is that the formation of PMA in these films usually requires the films to be grown on heavy metal thin films, such as Pt. We could consider growing a Pt thin film on the YIG film first and then depositing a CoFeB or $(Co/Ni)_n$ thin film on top, but the Pt film will block spin flow from the PMA film to the YIG film.

In the end of June 2020, work resumed with limited lab access, but our sputtering system experienced technical issues due to the lab shutdown. Right after those issues were fixed, we started to try a different material ? an amorphous ferrimagnetic alloy (CoFe)Tb. After some efforts with optimization of the sputtering conditions, we were able to grow (CoFe)Tb thin films with PMA first on sapphire substrates, and then on $Gd_3Ga_5O_{12}$ substrates. Since YIG materials share the same crystalline structures and almost equal lattice constants with $Gd_3Ga_5O_{12}$, we strongly believe that we can now easily grow (CoFe)Tb PMA films on YIG films.

We will use the PMA (CoFe)Tb film to inject perpendicular spin currents into the YIG film, but we also need to avoid the direct exchange coupling between the two films. Such direct coupling would make the SSF excitation impossible because the PMA film would make the in-plane moments in the YIG film perpendicular. For this consideration, we need to add an ultrathin oxide layer in-between the two films; we will use the spin tunneling through this oxide barrier layer, to transfer spin into the YIG film and thereby excite the SSF. In the last months of this project, we focused on the development of such tunneling structures.

During the closure of our labs, because we were unable to make any experimental efforts, we carried out micromagnetic simulations on the excitation of SSF in YIG thin films. We succeeded in numerically demonstrating SSF in YIG films and studied the effects of the YIG film thickness and the spin polarization. These numerical studies provide important implications for experimental demonstration.

We were not able to demonstrate SSF effects in YIG thin films during the project period. This was largely because of the pandemic-caused constraints and the unexpected technical challenges. However, our work laid the foundation for the future experimental demonstration of the room-temperature SSF effects. The major results of our work are presented in a pdf file uploaded.

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PARTICIPANTS:

Participant Type: PD/PI
Participant: Mingzhong Wu
Person Months Worked: 1.00 **Funding Support:**
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Participant Type: Postdoctoral (scholar, fellow or other postdoctoral position)
Participant: Chuanpu Liu
Person Months Worked: 12.00 **Funding Support:**
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Partners

,

I certify that the information in the report is complete and accurate:
Signature: Mingzhong Wu
Signature Date: 4/22/21 10:24PM

1. Summary of major efforts

The goal of this STIR project is to demonstrate room-temperature spin super-fluidity (SSF) in yttrium iron garnet (YIG) $Y_3Fe_5O_{12}$ thin films.

To achieve the goal, we need to grow a magnetic metallic thin film with perpendicular magnetic anisotropy (PMA) on a YIG thin film, which will be used to generate a perpendicularly-polarized spin current for the YIG film. We plan to use this perpendicular spin current to induce or excite SSF in the YIG thin film.

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In the end of June 2020, work resumed with limited lab access, but our sputtering system experienced technical issues due to the lab shutdown. Right after those issues were fixed, we started to try a different material — an amorphous ferrimagnetic alloy (CoFe)Tb. After some efforts with optimization of the sputtering conditions, we were able to grow (CoFe)Tb thin films with PMA first on sapphire substrates, and then on $Gd_3Ga_5O_{12}$ substrates. Since YIG materials share the same crystalline structures and almost equal lattice constants with $Gd_3Ga_5O_{12}$, we strongly believe that we can now easily grow (CoFe)Tb PMA films on YIG films.

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We were not able to demonstrate SSF effects in YIG thin films during the project period. This was largely because of the pandemic-caused constraints and the unexpected technical challenges. However, our work laid the foundation for the future experimental demonstration of the room-temperature SSF effects. The major results of our work are presented below.

2. Growth of (CoFe)Tb thin films with PMA

We succeeded in using DC sputtering to grow (CoFe)Tb thin films with PMA and square-like magnetic hysteresis loops. Such thin films will be used as spin polarizers to produce for YIG films a spin current that is polarized perpendicularly, or out-of-plane. Note that (CoFe)Tb is an amorphous ferrimagnetic alloy in

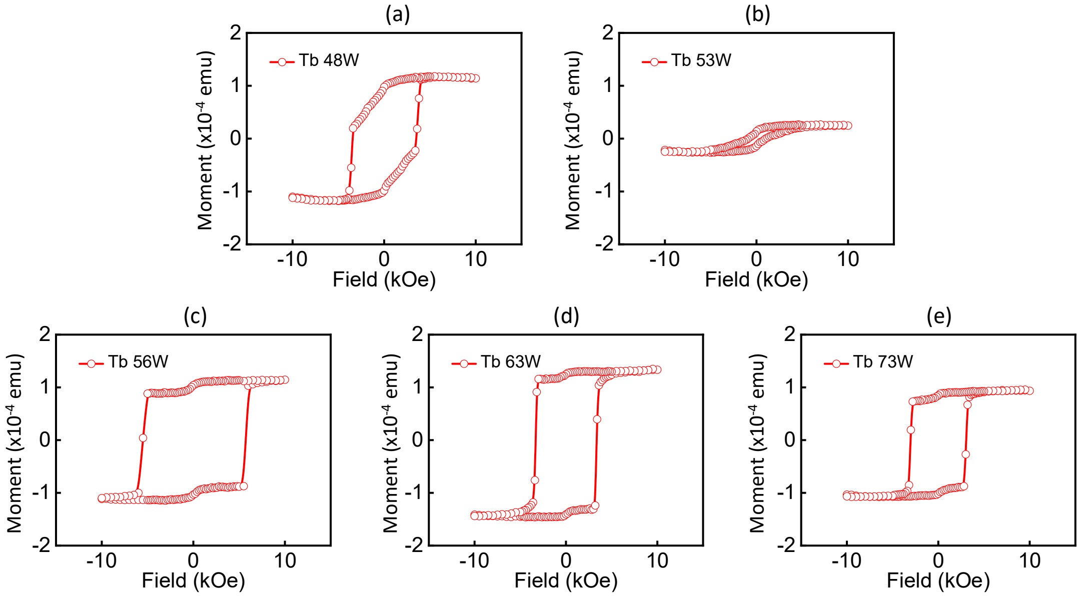

Fig. 1. Magnetic hysteresis loops measured on (CoFe)Tb thin films grown on sapphire substrates by co-sputtering. The power of the CoFe sputtering was 80 W for all the films, while the power of the Tb sputtering was different for different films, as indicated. The loops were measured by a vibrating sample magnetometer under a perpendicular magnetic field.

which the magnetic moment of the transition metal element sub-lattice (CoFe) is antiparallel to that of the rare earth sub-lattice (Tb). This material has been previously used as a spin polarizer in magnetic tunnel junction devices.

Figure 1 presents the magnetic moment vs. field hysteresis responses measured on five (CoFe)Tb thin film samples by a vibrating sample magnetometer (VSM) under a perpendicular magnetic field. The films were all grown on (0001)-oriented sapphire (Al_2O_3) substrates, via co-sputtering of a $Co_{35}Fe_{65}$ target (99.99% purity) and a Tb target (99.9% purity) at room temperature. The base pressure of the sputtering chamber was 1.5×10^{-7} Torr. The power of the $Co_{35}Fe_{65}$ sputtering (DC) was fixed at 80 W, while the power of the Tb sputtering (AC) was varied as indicated in Fig. 1. The film thickness has not been precisely characterized, but is estimated to be about 15 nm on the basis of the growth rate and the sputtering time (5 minutes).

The data in Fig. 1 show two major results. First, the "Tb 53W" sample exhibits the smallest magnetic moment, as shown in Fig. 1(b). This result seems to indicate that in the samples prepared with the Tb sputtering power higher than 53 W, the magnetic moment of the Tb sub-lattice is dominant, while in the samples prepared with the Tb sputtering power lower than 53 W, the magnetic moment of the Co sub-lattice is dominant. We will confirm this conclusion via magneto-optical Kerr effect measurements in the future. Second, the strength of the PMA strongly depends on the power of the Tb sputtering. In terms of maximizing the PMA strength, the optimal sputtering power is about 56 W.

Figure 2 presents a VSM hysteresis loop measured on a (CoFe)Tb thin film grown on a (111)-oriented single-crystal $Gd_3Ga_5O_{12}$ substrate. The sputtering parameters for this film were the same as for the film cited in Fig. 1(e). One can see that the loop is nearly square, and the coercivity is as high as about 2400 Oe. This indicates that this film can be used as the spin polarizer needed for the SSF demonstration. We believe that we can easily grow such PMA films on YIG films in the future since YIG and $Gd_3Ga_5O_{12}$ share the same crystalline structures, almost equal lattice constants, and very similar chemical properties.

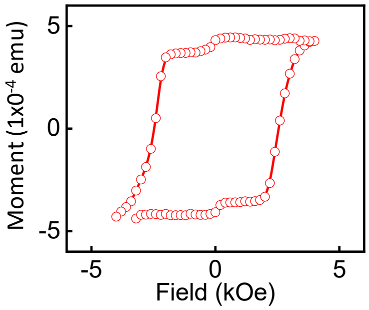

Fig. 2. A magnetic hysteresis loop measured on a (CoFe)Tb thin film grown on a single-crystal $Gd_3Ga_5O_{12}$ substrates by co-sputtering. The loop was measured by a vibrating sample magnetometer under a perpendicular magnetic field.

3. Development of Al_2O_3-based tunneling structures

In order to avoid the direct exchange coupling between the YIG film and the PMA metallic film and at the same time enable the uniform transfer of spin from the PMA film to the YIG film, it is necessary to grow a magnetic tunneling structure on top of the YIG film. We explored and succeeded in the fabrication of such tunneling structures.

Figure 3 shows the results on a NiFe/Al_2O_3/CoFe tunneling structure. The fabrication of this structure involves the following major steps: (1) fabrication of a NiFe thin film strip that has a thickness of about 5 nm and a width of 4 μm, (2) fabrication of a "20 μm by 20 μm" Al square element on the top of the NiFe strip, (3) natural oxidation of the Al element at room temperature in the atmosphere for 24 hours, to form a Al_2O_3 barrier layer, and (4) fabrication of a CoFe strip on top that has a thickness of about 5 nm and a width of about 4 μm. Two notes should be made. First, the bottom NiFe strip and the top CoFe strip are perpendicular to each other, as shown in Fig. 3(a). This geometry eases the tunneling resistance measurements. Second, Al_2O_3 was chosen as the barrier because it is a common tunneling barrier material.

Figure 3(b) shows the tunneling data measured with the configuration given in Fig. 3(a). The high voltages at low fields correspond to the high-resistance state in the structure, in which the magnetization vectors in the bottom NiFe and top CoFe strips are along their length directions and are therefore perpendicular to each other. The low voltages at relatively high fields correspond to the low-resistance state for which the magnetization vectors in both the strips are along the external field direction and are therefore parallel to each other. These results clearly demonstrate the spin tunneling effect in the Al_2O_3-based

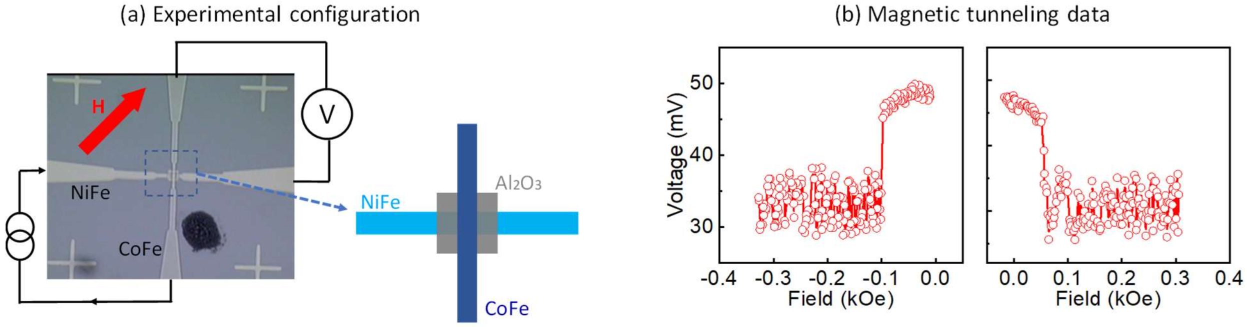

Fig. 3. Magnetic tunneling effects in a NiFe/Al_2O_3/CoFe layered structure. The measurements were performed in an in-plane magnetic field, as indicated by the red arrow in (a), with a DC current of 20 μA.

tunnel structure. This work provides useful implications for the future development of YIG/Al/Al_2O_3/(CoFe)Tb structures. We expect that perpendicularly-polarized spin in the top (CoFe)Tb layer can tunnel through the Al_2O_3 barrier and then reach the bottom YIG film to excite the SSF.

4. Numerical simulations of SSF in YIG films

We carried out OOMMF simulations on the excitation of the SSF in YIG thin film strips with a length of 10 μm, a width of 2 μm, and a thickness in the 3-10 nm range, with a configuration illustrated in Fig. 4. The mesh was set to 5 nm × 10 nm × (3-5) nm. The saturation magnetization of the YIG was set to 139.26 kA/m, and the exchange constant A was 3×10^{-12} J/m [Nature Communications 9, 738 (2018)]. The damping constant of the YIG film was set to 0.002, which is typical for YIG films thinner than 10 nm. The magnetization in the YIG film was excited by injecting a spin current with perpendicular polarization to the middle of the YIG film strip, as shown in Fig. 4. The spin current is injected by applying an electric current with designated spin polarization. The injection region has a width of 0.1 μm and a length of 2 μm. A time step of 25 fs was used in each stage, and 500 stages were taken in the simulations.

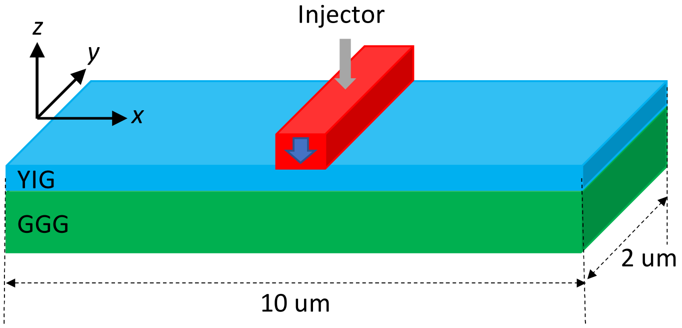

Fig. 4. Configuration for micromagnetic simulations of SSF in an yttrium iron garnet (YIG) thin film strip on a $Gd_3Ga_5O_{12}$ substrate.

Figure 5 shows the effects of the YIG film thickness on the excitation of the SSF. In each panel, the color map shows the top view of the spin states in the YIG strip after a simulation time of 12.5 ns, with the color denoting the *y* component of the magnetization. The simulation used a charge current density of 1×10^{11} A/m^2 and a spin polarization rate of 0.4 [Nature Materials 3, 877 (2004)].

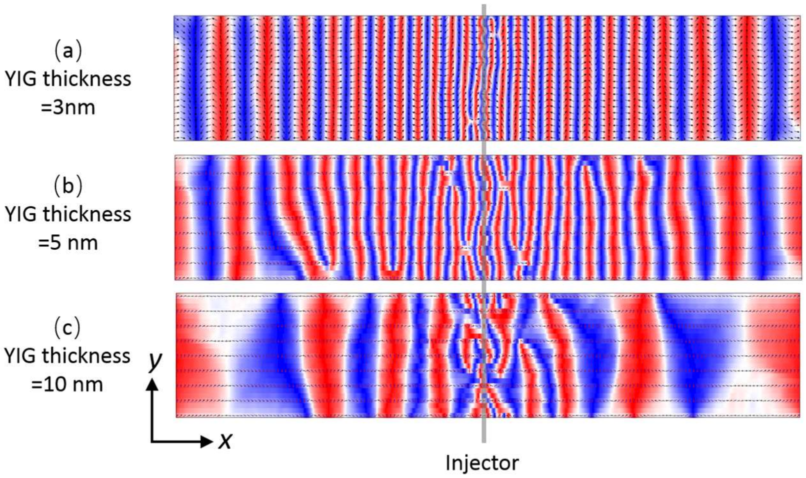

Fig. 5. Simulation results showing the effects of the YIG film thickness on the excitation of the SSF. Each map shows the top view of spin states in the YIG film strip, with the color denoting the *y* component of the magnetization vector.

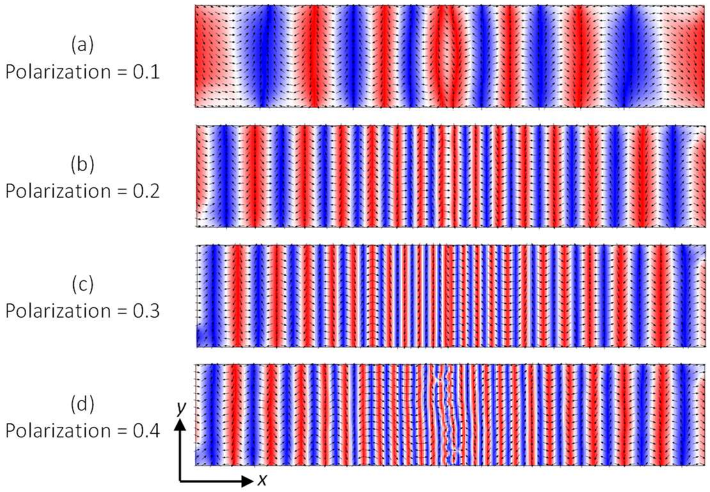

Fig. 6. Simulation results showing the effects of the spin polarization rate of the spin current injected to the YIG film strip. Each map shows the top view of spin states in the YIG film strip, with the color denoting the *y* component of the magnetization vector.

One can see from Fig. 5(a) that phase-resolved patterns are clearly formed along the *x* direction, and the phase gradient decreases as one goes away from the injection area to the two ends of the YIG film strip. The observed patterns indicate the presence of the SSF in the YIG film, while the decrease in the phase gradient reflects the decay of the SSF amplitude. A comparison of the three maps in Fig. 5 indicates that, for a given spin current, the thinner the YIG film is, the stronger and the more coherent the SSF is. With the same spin injection, no SSF was observed in the 15-nm-thick and 20-nm-thick YIG films (not shown in Fig. 5). This threshold behavior is consistent with T. Schneider's work reported in arXiv:1811.09369.

Figure 6 presents simulation results that show the effects of varying the spin polarization of the injected current. The maps were all obtained for a 3-nm-thick YIG film strip and a charge current density of 1×10^{11} A/m^2 after a simulation time of 12.5 ns. A comparison of the maps indicates that the stronger the spin polarization is, the stronger the SSF is.