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Conduction of spin currents through insulating oxides

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Abstract

Spintronics is a field of electronics based on using the electron spin instead of its charge. The recent advance in the manipulation of pure spin currents, i.e. angular momentum transfer not associated to conventional charge currents, has opened new opportunities to build spin based devices with low energy consumption [1]. It has also allowed to integrate ferromagnetic insulators in spintronic devices, either as spin sources [2–6] or spin conductors [2, 7, 8] using their magnetic excitations to propagate a spin signal. Antiferromagnetic insulators belong to another class of materials that can also sustain magnetic excitations, even with a higher group velocity [9]. Hence, they have potential as angular momentum conductors, possibly making faster spin devices. At the opposite end, angular momentum insulators are also required in spintronic circuits. The present letter underlines some essential features relevant for spin current conduction, based on measurements of angular momentum transmission in antiferromagnetic NiO and in the non-magnetic light element insulator SiO_2.

Spin conductors are materials through which a spin current can flow. These are conventionally normal metals where conduction electrons provide the necessary mobile spinned entities. The added properties of light mass and low resistivity have made Al one of the best spin conductors so far [10, 11], with spin diffusion lengths as large as one micron. Another alternative to propagate a spin current is to use spin excitations in magnetically ordered materials. Magnons are the elementary excitations in ferromagnets (FM), each reducing the total spin by one unit of $\hbar$, and hence the magnetic moment by $g\mu_B$ [9] where μ_B is the Bohr magneton and g the Landé factor. They can propagate over distances in the millimetre range in the insulating ferrimagnet Yttrium Iron Garnet (YIG), which has unsurpassed low damping [2–7]. Therefore, magnetic insulators represent an attractive medium for propagating spin information over long distances. However, these spin excitations have a direction imposed by that of the magnetization, which can also potentially couple to that of other magnets in the circuit through direct contact or stray fields. Non magnetic elements are not restricted by these limitations. Interestingly, elementary magnetic excitations also exist in antiferromagnets (AF), materials with two magnetic sublattices oppositely oriented, resulting in a global zero moment [9]. Antiferromagnetic magnons, like their ferromagnetic counterparts, correspond to the reversal of a single spin in an AF crystal. Thermally generated AF magnons carry random spin directions resulting in zero global angular momentum, but each of these entities can transport spin information on significant distances and faster than in ferromagnets. We intend here to demonstrate that angular momentum can be transmitted through an AF insulator inserted between a spin source and a spin sensor.

Spin sources have conventionally relied on extracting spin polarized conduction electrons from ferromagnetic metals. This can be achieved by driving a charge current through the ferromagnet or applying thermal gradients (Peltier effect) [12]. A non-local geometry can also be designed so as to extract a pure spin current 'unpolluted' by charge currents [10, 13]. A contactless means of emitting angular momentum can also be achieved using a FM undergoing ferromagnetic resonance (FMR). Damping processes generate spin currents at the interface with a normal metal, via a phenomenon known as spin pumping [14]. This recent discovery has far reaching implications as magnetic insulators can now be used as active elements in spintronic devices. In particular, Yttrium Iron Garnet is the material of choice [2–7] and the one we selected as our radio-frequency driven spin source.

At the opposite end of the circuit is the spin-sensor, relying on the conversion of a spin

current into a charge current, through a mechanism known as the inverse spin Hall effect (ISHE)[10, 11]. The effect is based on spin-orbit coupling, as originally described for the direct spin Hall effect [15, 16], and it enables spin current sensing, including those emitted from a ferromagnet undergoing FMR [2, 17]. The best ISHE materials are heavy element metals, for which spin-orbit effects are enhanced. Pt is the most commonly used material, and the one we chose here, but others like Ta or W are also promising candidates [5, 18–20].

The device we design is thus a simple tri-layer with spin source and sensor separated by the material through which spin conduction is to be measured. We propose here to study nickel oxide, a well known antiferromagnetic insulator with a Néel point well above room-temperature [21]. A series of tri-layers YIG|NiO|Pt were grown on (111) GGG substrates, each with a single crystalline 200 nm thick film of YIG (deposited by Liquid phase Epitaxy) and a 5 nm thick Pt layer. The NiO layers have thicknesses ranging from zero to 30 nm and were deposited under a 300 Oe magnetic field. Importantly, magnetometry measurements evidence the absence of exchange bias in our trilayers, whose coercivity is around 0.5 Oe, totally unchanged from that of the bare YIG film. This demonstrates that NiO is magnetically decoupled from the YIG, unlike what is systematically found for the NiO|Ni system [22]. The samples are then mounted on a 500 μm wide, 2 μm thick Au transmission line cell used for microwave generation up to 20 GHz. The long axis of the 5 mm $\times$ 1 mm samples is mounted parallel to the excitation field h_{rf} as indicated in the inset of Fig.1. The inverse spin Hall voltage V_{ISH} in Pt is measured by a lock-in technique (with the microwave power turned on and off at a frequency of a few kHz) with electrical connections through gold leads on each side of the area of excitation. When in ferromagnetic resonance, YIG emits a spin current at the interface with NiO, itself in contact on the other side with the Pt layer converting spin into charge.

The ISHE generated in the reference YIG|Pt sample is shown in Fig. 1-a, evidencing the significant spin current emitted when the YIG enters resonance, as already reported [5]. Measured Hall voltages are odd in field and can reach here 27 μV at 3.85 GHz for an output power of 10 dBm (i.e. a radio-frequency field of 0.2 Oe). The effect can be understood using three key parameters: the spin mixing interface conductance $G_{\uparrow\downarrow} \simeq 10^{14}\ \Omega^{-1}\cdot\mathrm{m}^{-2}$, the spin Hall angle $\theta_{SH} \simeq 0.05$ and the spin diffusion length $\lambda_{sd}^{\mathrm{Pt}} = 2$ nm. These are representative of the physics at the heart of the effect. The dynamically generated spin current is emitted through the interface which acts as a semi-transparent medium described by the 'conduc-

tance' $G_{\uparrow\downarrow}$. The exact nature of this parameter is at present not completely clear, but experimentally, metallic interfaces have a much better transparency than insulator/metallic ones [23]. This probably reflects the importance of a common vector of spin transport, being the conduction electrons in this specific case. The second important quantity is λ_{sd}, the distance on which spin memory is preserved in the normal metal, set by the by the spin-flip scattering rate. It is rather short in Pt where spin currents are efficiently converted into charge currents by the inverse spin Hall effect. The figure of merit of this process is the spin Hall angle θ_{SH}, determined by the difference in average scattering angle between spin up and spin down electrons.

Once the angular momentum source and detector tested using the YIG|Pt structure, the effect of inserting the NiO layer can be studied. For NiO thicknesses ranging from 2 nm to 15 nm a ISHE voltage can be measured in the Pt layer as shown in Fig. 2-a. We argue that this is the conceptual demonstration that antiferromagnets can indeed conduct angular momentum. The amplitude of the ISHE signal is however much lower than in the reference YIG|Pt sample (compare Fig. 1-a and 1-b), thus evidencing the low transparency of the YIG|NiO interface and/or the imperfect transmission of NiO. It is therefore interesting to study the evolution of the angular momentum transfer as a function of NiO thickness, as shown in Fig. 2. The spin current signal is found to decrease in an exponential fashion leading to an estimated diffusion length around 2 nm. This is extremely small compared to what one could expect if magnons were propagating through the antiferromagnet. Instead, this corresponds more to the actual size of these entities, hinting to a non propagative angular momentum transport. We would like to point out here that this dependence in NiO thickness rules out an explanation for the effect based on conduction electron tunnelling [24], or purely on the magnetic susceptibility of NiO [9, 21]. Indeed, the NiO susceptibility at GHz frequencies is low, it should not vary with thickness and a hypothetical amplification at the YIG interface cannot be expected considering the absence of magnetic coupling. Another very interesting experimental observation in Fig. 2 is the decrease of spin conductance for the thinnest 2 nm thick layer. We underline here that this is a real effect as any problem of layer discontinuity would instead significantly enhance the signal through the regions with direct YIG|Pt contact. Reduced magnetic quality or/and spatial confinement induced overlap of antiferromagnetic magnons might be responsible for this loss of angular momentum transfer. Clearly, the behaviour of these entities in these thin layers need to be theoretically studied in

the future. The transparency of the YIG|NiO interface can be inferred from the extrapolation of the exponential behaviour in Fig. 2 to zero thickness, which is found to be twenty times smaller than the ISHE voltage in the YIG|Pt reference sample. Thus, the total $G_{\uparrow\downarrow} \simeq 5.10^{12}\ \Omega^{-1}\cdot\mathrm{m}^{-2}$, which can be interpreted as resulting from the spin resistances of the two interfaces in series. We argue that the critical interface is the YIG|NiO one where the transfer of angular momentum cannot rely anymore on exchanging conduction electrons. Instead, the magnons in YIG have to transform into antiferromagnetic magnons in NiO, which are to be found on average at a much higher frequency/energy and with a dispersion linear in wave vector [9, 21]. This mismatch is likely to be the bottleneck of the transmission problem but this is not demonstrated here and should be tested in the future. Moreover, the growth of micro-crystalline NiO on top of the excellent quality YIG crystal are very likely not optimal. Hence, the YIG|NiO interface transparency to the diffusion of angular momentum can be estimated to be around 0.05 (attributing the original $G_{\uparrow\downarrow}$ value to the NiO|Pt interface). There is an obvious need for a deeper theoretical understanding of the problem, but this is beyond the scope of the present study.

Beside the availability of good angular momentum conductors, the design of useful devices also requires the identification of good insulators. These should be non-magnetically ordered in order to suppress the magnon channel. However, this is probably not enough as, although often forgotten, phonons are also able to carry angular momentum [25] (like in the Einstein-de Haas effect), especially in materials with strong spin-orbit coupling elements. Therefore, light oxides, free from the existence of magnetic moments and spin-orbit interactions, look like good candidates. Here we select SiO_2. Remarkably, as shown in Fig. 3, SiO_2 layers as thin as 2 nm are able to completely suppress the spin current propagation in our YIG|SiO_2|Pt trilayers. This is an impressive effect showing that angular momentum cannot even 'tunnel' through such a thin layer. Thus, SiO_2 can be considered an excellent angular momentum insulator, which we argue comes from the absence of magnetic excitations in this compound, together with the light O and Si atoms reducing spin-orbit interactions. Beyond these concepts, it would be very interesting to study in more details the behaviour of phonons through the interface and their role in conducting angular momentum.

In conclusion, we show here that antiferromagnets can be classified as spin conductors, probably because their antiferromagnetic magnons can propagate angular momentum. On the other hand, light non magnetic insulators are good spin insulators because their phonons

are inefficient to carry orbital momentum. The diffusion length measured in antiferromagnetic NiO is however inconsistent with a picture of mobile magnons diffusing from the spin source to the spin sensor. Instead, it seems that angular momentum crosses through confined non-propagating antiferromagnetic magnons. The YIG|NiO interface transparency is found to be of the order of 5 %, possibly reflecting the magnon energy mismatch as well as the non optimal interface quality as NiO is not epitaxially grown on single crystalline YIG. Therefore, beyond the conceptual advance brought by the present results, there is room for optimization on the materials' side and a critical need for theoretical understanding of angular momentum transfer through interfaces in the absence of conduction electrons.

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Author contribution. MV, GdL, OK, CH and JBY planned and designed the experiment. JBY produced and characterized the samples. CH, VN and GdL performed the experiments. MV, GdL and OK wrote the manuscript. All authors discussed the results and commented on the manuscript.

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FIGURES

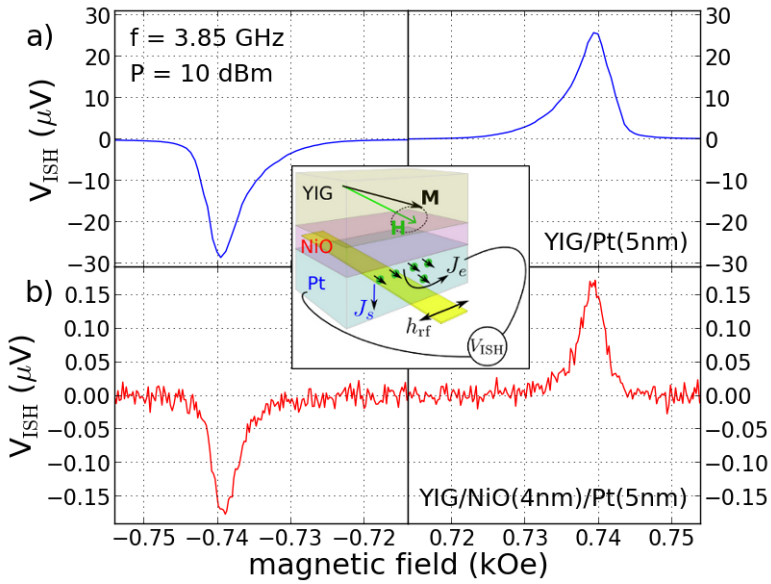

FIG. 1. (a): ISHE signal measured in a YIG|Pt bilayer generated by the spin current emitted by the YIG at resonance at a frequency of 3.85 GHz and a power of 10 dBm. (b) : Similar signal when a 4 nm NiO layer is inserted between YIG and Pt. Note the difference in scale for the measured ISHE voltage. Inset: geometry of the measurement showing the induced spin current in the tri-layer geometry.

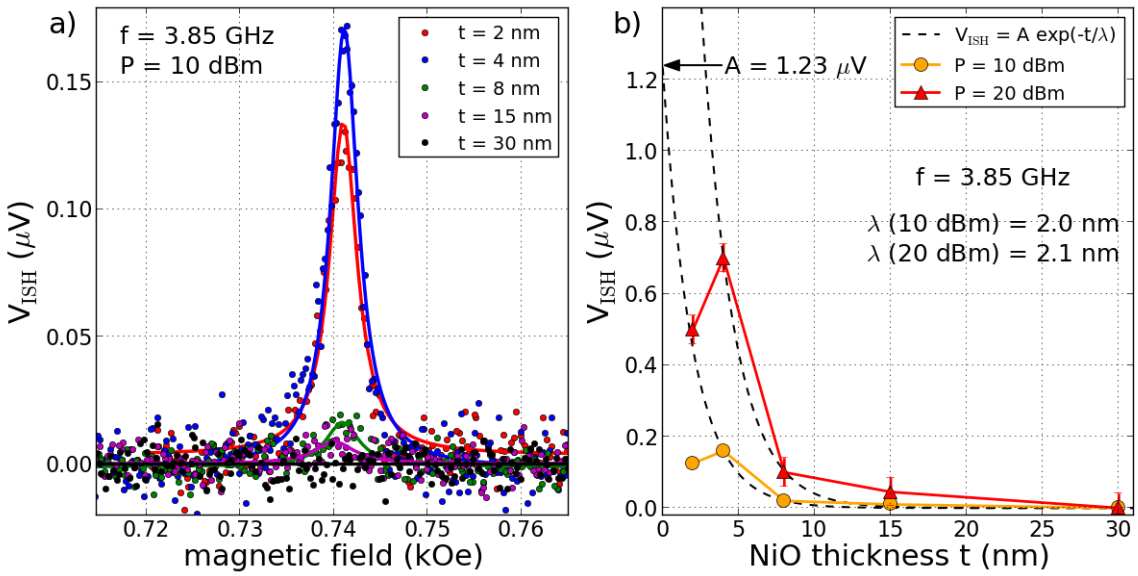

FIG. 2. NiO thickness dependence of the angular momentum transfer in YIG|NiO(t)|Pt devices at 3.85 GHz and two different powers: 10 and 20 dBm. (a): Raw measurements at 10 dBm and positive field. (b): Summary of the NiO thickness dependence of the measured spin Hall Voltage at 10 dBm (orange) and 20 dBm (red). The diffusion length from the exponential fits is close to 2 nm. The significant difference between the signal extrapolated to zero NiO thickness and that of the pure YIG|Pt sample, gives an estimate interface YIG|NiO transparency of 0.05.

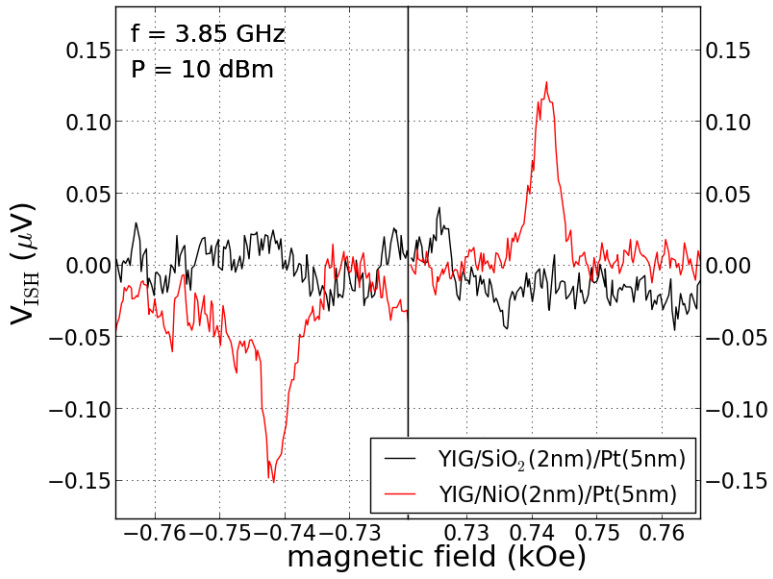

FIG. 3. Comparison of the effect of the insertion of a 2 nm layer of NiO and SiO_2 between source and sensing layer demonstrating that SiO_2 is an efficient angular momentum insulator.