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ORGANIC SPINTRONICS

Pumping spins through polymers

Spin pumping and spin-to-charge conversion in hybrid metal–organic devices reveal the physical mechanisms at work in semiconducting polymers.

Bert Koopmans

Organic semiconductor devices have become extremely successful in opto–electronic applications such as OLED displays, and they have huge potential for applications in other areas, for instance solar cells and disposable electronics. Beyond these well-known routes, a new direction is emerging, in which the long spin-lifetimes in organic materials — due to their low-mass atoms — is being exploited. This field of organic spintronics aims to implement magnetic- or spin-functionality in (hybrid) organic devices. Although the first prototype devices have been reported, sustained progress is hindered by intrinsic problems associated with the disordered nature, and therefore low conductivity, of organic (polymer) materials.

Writing in *Nature Physics*, Shun Watanabe and co-workers report an alternative approach[1], transferring subtle yet robust mechanisms known from conventional metal-based spintronics to the realm of organics. Their approach, in which a pure spin current is 'pumped' from a magnetic electrode into the organic medium and converted into a measurable charge current in a metallic counter electrode, bypasses formerly identified obstacles. It therefore provides direct access to the physics of spin transport and spin relaxation in organic semiconductors.

Transport of charge and spin in organic semiconductors is carried by spin-1/2 polarons, which are localized electrons dressed up by a molecular deformation. Conduction occurs due to the hopping of polarons between trapping sites. The spin of these polarons can be explicitly exploited in organic spintronics, which is considered an attractive route because of the weak spin–orbit coupling, making spin a robust and long-lived quantity.

Indeed, exciting developments have been reported over the past decade. For example, giant magnetoresistance has been observed in organic spin-valves — devices consisting of an organic semiconductor thin-film sandwiched between two ferromagnetic electrodes[2,3]. However, the low conductivity of organic materials leads to a huge resistance mismatch. Even if spins are successfully injected into the organic medium — and there is no reason to doubt that this can be successfully done — the theory predicts a vanishingly small magnetoresistance because of the absence of 'spin accumulation'. Altogether, researchers in the field are in a mixed state of excitement and confusion. Some claim that devices may work because conventional theories are not applicable to hopping transport in organic devices, whereas others are sceptical about the experimental results and present alternative explanations for some of the observed effects.

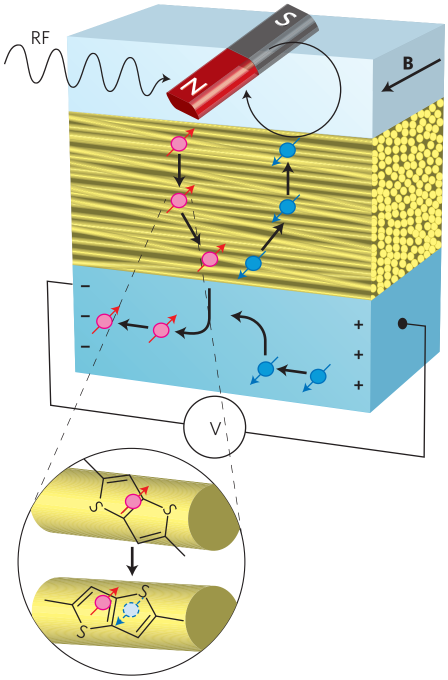

Figure 1 | Measurement of pure spin currents through a polymer thin-film. The top electrode is driven into ferromagnetic resonance by exposure to radiofrequency (RF) radiation in an applied magnetic field. A pure spin current is pumped into the polymer film (represented by downward-moving electrons with spin pointing backwards, and upward-moving electrons with spin pointing forwards). Within the polymer the spin current is carried by polarons. In the bottom electrode — made of a heavy metal to increase spin–orbit coupling — the spin current is converted into a charge current. This so-called inverse spin Hall effect causes the spin-polarized electrons to change direction depending on their spin orientation, leading to a net signal. The inset illustrates spin loss — due to a combination of spin–orbit coupling and different molecular orientations at two trapping sites — during the hopping from one site to another. This is identified as the leading contribution to spin relaxation.

Watanabe and colleagues chose to access spin transport in organic semiconductors using a new route that is not hampered by the resistance mismatch problem. In their ferromagnet/polymer/non-magnetic metal trilayer architecture, the authors exploit spin pumping[4], an effect reciprocal to the well-known current-driven spin torque oscillator. A precession is externally driven by a radiofrequency field, leading to a pure spin current — a current of counterpropagating opposite spins carrying no net charge — being pumped into the organic film. After the polaron-mediated spin current has traversed the organic film, within the non-magnetic electrode it is converted into a charge current by the inverse spin Hall effect (ISHE). An essential ingredient for the occurrence of the ISHE is spin–orbit coupling, and it can be explained in a hand-waving way as a preference for spin-up (down) electrons to make left (right) turns (or vice versa). In the non-magnetic electrode this leads to a net charge current as schematically shown in Fig. 1.

The team show that at ferromagnetic resonance an ISHE signal is picked up in their device, which consists of a PBTTT polymer film with $Ni_{0.8}Fe_{0.2}$ and Pt electrodes. For the first time in organic spintronics research, a clear fingerprint of the precessional nature of polaron spins in an applied magnetic field is demonstrated. The absence so far of this so-called Hanle effect had been worrying the community[5].

Carefully excluding spurious contributions of non-ISHE origin, Watanabe and colleagues claim to have developed a reliable technique for measuring spin currents in a (disordered) organic medium. The approach was used to address a fundamental and intensely debated issue in organic spintronics: the origin of spin relaxation as polarons move through an organic film. Is it due to the precession of polaron spins in the presence of small random hyperfine fields while they reside on a local site[6,7], or is it due to the randomization of the spin information by the likewise small spin–orbit coupling[8,9]? In the latter case, is the spin relaxation caused by the coupling with molecular vibrations while residing on a trapping site, or are spins flipped while charge carriers hop between two sites? A temperature-dependent study of the attenuation of ISHE signals as a function of film thickness provided a spin diffusion length of approximately 200 nm, and, most strikingly, showed that the attenuation is almost independent of temperature between 200 and 300 K. Based on these findings Watanabe and colleagues conclusively identify a spin-relaxation scenario — in the experiments where polarons move between two trapping sites spin relaxation is dominated by spin–orbit coupling.

Surely this is not the last word. To drive the ferromagnetic resonance, the experiments were performed in the presence of a magnetic field (~100 mT) that was significantly larger than the random hyperfine field experienced by the polarons (~1 mT). As such, the results do not exclude that spin relaxation mediated by the hyperfine field may become dominant for smaller fields. It is also noteworthy that the PBTTT polymer used here is special. Owing to its relatively well-ordered nature in thin-film form, it has a large in-plane field effect mobility of around 1 $cm^2\ V^{-1}\ s^{-1}$, while having just a moderate mobility ($10^{-5}\ cm^2\ V^{-1}\ s^{-1}$) for the perpendicular configuration employed here. This means that in the diffusive regime a large number of in-plane hops will occur between two successive perpendicular hops, raising interesting questions about the effects on spin transport.

Finally, the validity of conventional spin-diffusion theories, as used by Watanabe and colleagues to describe the case of non-doped organic semiconductors in the space-charge-limited hopping regime, is far from trivial. Clearly, understanding spin transport is still in its infancy, and — as the team also stress — a more serious microscopic framework has yet to be developed.

Despite these critical notes, the pioneering work by Watanabe *et al.* will certainly fuel the further development of organic spintronics. It will not solve all of the outstanding problems of organic spin-valves, but will surely increase the understanding of them. Eventually, the findings may inspire researchers to think about organic spintronic applications based on pure spin currents, avoiding the resistance mismatch altogether. ❐

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References

1. Watanabe, S. *et al. Nature Phys.* http://dx.doi.org/10.1038/nphys2901 (2014).
2. Xiong, Z. H. *et al. Nature* **427,** 821–824 (2004).
3. Dediu, V. A. *et al. Nature Mater.* **8,** 707–716 (2009).
4. Saitoh, E. *et al. Appl. Phys. Lett.* **88,** 182509–182509 (2006).
5. Grünewald, M. *et al. Phys. Rev. B* **88,** 085319 (2013).
6. Bobbert, P. A. *et al. Phys. Rev. Lett.* **102,** 156604 (2009).
7. Nguyen, T. D. *et al. Nature Mater.* **9,** 345–352 (2010).
8. Yu, Z. G. *Phys. Rev. Lett.* **106,** 106602 (2011).
9. Nuccio, L. *et al. Phys. Rev. Lett.* **110,** 216602 (2013).