understanding and application of such spin phenomena is a promising future direction for organic semiconductors. ❐

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More than spectroscopy

To the Editor — While we appreciate the attempt by Boehme and Lupton to outline several failures of research efforts in organic spintronics[1], on some issues we have different opinions and judgements.

Boehme and Lupton criticize conclusions on spin-related phenomena in organic semiconductors based on magnetoresistance measurements, which are linked to the reversal of the electrodes' magnetization. Indeed, there are still many unsolved puzzles in the results provided by this approach. For example, in the prototypical vertical device composed of a $La_{0.7}Sr_{0.3}MnO_3$/tris(8-hydroxyquinolinato)aluminium(Alq3)/Co multilayer, spintronic effects are detectable in a voltage range from 1–10 mV to 1 V (ref. 2), whereas for Alq3 the injection barriers to the lowest unoccupied molecular orbital or the highest occupied molecular orbital conducting levels is about 1–2 eV (ref. 3). This demonstrates the need to consider intragap states (or possibly a band) caused by either defects or impurities, but a fully fledged debate on this topic has not started yet. Another example is the very origin of the magnetoresistance, which is neither clearly formalized nor understood yet. The absence of the Hanle effect further complicates matters. In inorganic spintronics the Hanle effect had the role of ultimate proof of spin injection in the spin-transporting medium[4]. Unexpectedly, the absence of the Hanle effect was firmly established in devices involving several organic materials[5,6]. Thus, a lively debate on this topic has spread within the community and a number of scientists are at work to reveal the new physics at the basis of this and other unusual observations in organic spintronics. Along this track Zigang Yu[7] has recently advanced the hypothesis of charge–spin separation in organic conducting media, which implies the absence of the Hanle effect in some conduction regimes and gives a theoretical justification to the presence of magnetoresistance.

Transport investigations have also revealed other unforeseen properties such as easy tuning and control of magnetoresistance by the application of a voltage[8], suggesting new device concepts such as the magnetically modulated memristor. Besides fundamental interest, focus on magnetoresistance is justified by the fact that most of the foreseeable applications are based on it or on variations thereof. From this point of view, Boehme and Lupton listed several shortcomings that organic semiconductors have in comparison with inorganic spin-transporting media; we would like to strike one out. The spin–lattice relaxation time T_1 is about 1 ns in Si (ref. 9), whereas it is of the order of 1 μs for organic semiconductors[10]. This contradicts the statement that, in organic semiconductors, T_1 too falls short of the performance of inorganic spin-transporting media.

Summarizing, we definitely support the call by Boehme and Lupton for more spectroscopic studies. Optical, X-ray and tunnelling spectroscopies have already been intensively employed to reveal strong and captivating spin-filtering effects at organic/ferromagnetic interfaces[11,12], not in contrast with but greatly enhancing the understanding of spin-transport results[13]. To make further and decisive steps towards understanding of spin-related effects in organic semiconductors the community needs perhaps to strengthen all its tools, including spectroscopic and transport studies, and try to increase the efforts in achieving the highest possible overlap between them. Studying spin behaviour inside organic materials by spectroscopy does not solve the question of whether organic spintronics will be limited to the manipulation of spin species intrinsic to the material, or will be able to exploit spin injection and transport, so as to work far beyond the equilibrium regime. ❐

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Standardization should come first

To the Editor — We agree with Boehme and Lupton[1] that the organic spintronics community should focus more on scrutinizing the existing models and to carry out detailed spectroscopic experiments. However, there are other elements to take into consideration. In addition to distinguishing whether the bipolaron model or polaron-pair model applies when interpreting the magnetotransport properties of organic materials, another important task is to identify the contributions of the factors that cause spin relaxation, that is, the hyperfine field and spin–orbit coupling. The hyperfine field is considered the dominant factor, due to the lack of heavy atoms and the presence of hydrogen in organic

materials[2]. However, spin–orbit coupling too is found to be a significant source of spin relaxation[3].

There are more than 30 million organic compounds differing in structure and composition, resulting in a multitude of features found in organic materials. Even when the same material is used in different experiments, its purity can vary, and any impurities can substantially affect the data. We note that two independent groups used deuterated materials to explore the hyperfine field effects on the electron spin relaxation, and reached opposite conclusions[2,4]. Furthermore, divergent conclusions from similar experiments[5] could also be the consequence of dissimilar processes for sample preparations and of the lack of consistency in the testing instruments of different groups, notably in the organic magnetoresistance subfield.

Therefore, priority should be given to standardizing data. Researchers should carefully record experimental procedure, material and so on, and possibly share them in a database repository, so that results from different laboratories can be compared. Such a repository would also be useful in testing applicability and accuracy of theoretical models. ❐

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Focusing on the molecular scale

To the Editor — The call of Boehme and Lupton[1] for the increased use of spectroscopy to study spin phenomena in organic spintronics is valuable to the community. However, there are instances where spectroscopy techniques such as X-ray absorption, X-ray magnetic circular dichroism, X-ray photoemission, two-photon photoemission and muon spin-rotation spectroscopy are not suitable investigative tools because they provide only an averaged-out response of interface and bulk phenomena. One example is the study of ferromagnet/organic semiconductor interfaces, for which the above methods are not sufficiently sensitive for locally probing the magnetic field-dependent response of the interface. I would argue that the organic spintronics community should tackle the challenges in the field starting from the molecular scale working towards building a theoretical framework of the fundamental physical mechanisms.

For example, the emergent subfield of interface molecular spintronics has a strong theoretical hold supported by advanced spectroscopy tools to probe molecular spin responses. Although the observation of spin-conserved tunnelling through organic semiconductors is accepted in theory, questions remain about the sign and magnitude of tunnelling magnetoresistance in organic tunnel junctions. Spin-polarized scanning tunnelling spectroscopy offers direct access to the spin states of the organic molecules on magnetic surfaces[2]. Supported by computational *ab initio* studies, such exploration provides direct insight on the role of local surface chemistry in the magnetic response of an interface. Evidence of inversion and amplification of interface spin-polarization may explain the reported discrepancy in tunnelling magnetoresistance values of macroscopic organic tunnelling devices. Recently, reports of induced molecular-magnetism and the spin-filter response at the interface in macroscopic organic devices[3] have been confirmed using such techniques[4]. They offer a way forward in our understanding and may be technologically relevant for the development of molecular spintronic devices in sensor, memory and computing applications[3]. Also, similar approaches for probing the organic magnetoresistance effect at lower dimensions may help to appropriately scrutinize its origin.

In addition to the message of Boehme and Lupton, other important criticisms[5] of the field are the occasional improper device fabrication practices for soft and porous organic semiconductors, misinterpretation of experimental reports and the poor reproducibility of the experimental data from different research groups, which raise concerns about the technological applicability of organic spintronic devices. Channelling our research efforts towards the molecular scale may present a promising future for the field. ❐

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An agnostic approach

To the Editor — The current situation of research in organic spintronics as outlined by Boehme and Lupton[1] may be due in part to the early successes[2] and subsequent heavy reliance on the theory and techniques inherited from the more mature field of inorganic semiconductors. However, there are differences between organic and inorganic semiconductors that reflect in the microscopic reality of spin-dependent processes. Models that account for these differences are therefore necessary to advance our understanding of spin-based phenomena in organic semiconductors.

Discriminating between different models of spin transport and dynamics in organic semiconductors has proven challenging. Recent progress, however, is promising. Harmon and Flatté[3] have predicted signatures for the three dominant spin-relaxation mechanisms for organic semiconductors: intrasite relaxation, hyperfine interactions and spin–orbit coupling. Similarly, Janssen *et al.*[4] have developed a unified model of organic magnetoresistance, which includes polaron-pair recombination, bipolaron spin blocking and triplet exciton–polaron quenching mechanisms. These amalgamated approaches force the characteristics of