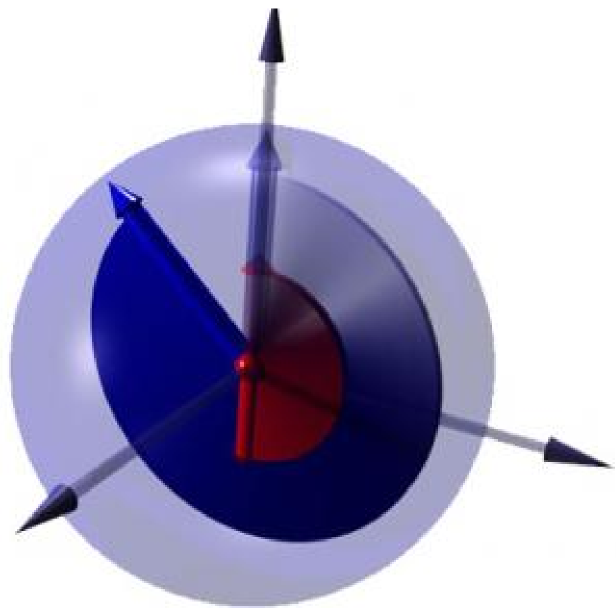
Researchers road-test powerful method for studying singlet fission

October 18 2016



Spin, an intrinsic property of electrons, is related to the dynamics of electrons excited as a result of singlet fission – a process which could be used to extract energy in future solar cell technologies. Credit: Leah Weiss

In a new study, researchers measure the spin properties of electronic states produced in singlet fission – a process which could have a central role in the future development of solar cells.

Physicists have successfully employed a powerful technique for studying electrons generated through singlet fission, a process which it is believed will be key to more efficient solar energy production in years to come.

Their approach, reported in the journal *Nature Physics*, employed lasers, microwave radiation and magnetic fields to analyse the spin of excitons, which are energetically excited particles formed in molecular systems.

These are generated as a result of singlet fission, a process that researchers around the world are trying to understand fully in order to use it to better harness energy from the sun. Using materials exhibiting singlet fission in solar cells could make energy production much more efficient in the future, but the process needs to be fully understood in order to optimize the relevant materials and design appropriate technologies to exploit it.

In most existing solar cells, light particles (or photons) are absorbed by a semiconducting material, such as silicon. Each photon stimulates an electron in the material's atomic structure, giving a single electron enough energy to move. This can then potentially be extracted as electrical current.

In some materials, however, the absorption of a single photon initially creates one higher-energy, excited particle, called a spin singlet exciton. This singlet can also share its energy with another molecule, forming two lower-energy excitons, rather than just one. These lower-energy particles are called spin "triplet" excitons. Each triplet can move through the molecular structure of the material and be used to produce charge.

The splitting process - from one absorbed photon to two energetic triplet excitons - is singlet fission. For scientists studying how to generate more solar power, it represents a potential bargain - a two-for-one offer on the amount of electrical current generated, relative to the amount of light put in. If materials capable of singlet fission can be integrated into solar cells, it will become possible to generate energy more efficiently from sunlight.

But achieving this is far from straightforward. One challenge is that the pairs of triplet excitons only last for a tiny fraction of a second, and must be separated and used before they decay. Their lifespan is connected to their relative "spin", which is a unique property of elementary particles and is an intrinsic angular momentum. Studying and measuring spin through time, from the initial formation of the pairs to their decay, is essential if they are to be harnessed.

In the new study, researchers from the University of Cambridge and the Freie Universität Berlin (FUB) utilised a method that allows the spin properties of materials to be measured through time. The approach, called electron spin resonance (ESR) spectroscopy, has been used and improved since its discovery over 50 years ago to better understand how spin impacts on many different natural phenomena.

It involves placing the material being studied within a large electromagnet, and then using laser light to excite molecules within the sample, and microwave radiation to measure how the spin changes over time. This is especially useful when studying triplet states formed by singlet fission as these are difficult to study using most other techniques.

Because the excitons' spin interacts with microwave radiation and magnetic fields, these interactions can be used as an additional way to understand what happens to the triplet pairs after they are formed. In short, the approach allowed the researchers to effectively watch and manipulate the spin state of triplet pairs through time, following formation by singlet fission.

The study was led by Professor Jan Behrends at the Freie Universität Berlin (FUB), Dr Akshay Rao, a College Research Associate at St John's College, University of Cambridge, and Professor Neil Greenham in the

Department of Physics, University of Cambridge.

Leah Weiss, a Gates-Cambridge Scholar and PhD student in Physics based at Trinity College, Cambridge, was the paper's first author. "This research has opened up many new questions," she said. "What makes these excited states either separate and become independent, or stay together as a pair, are questions that we need to answer before we can make use of them."

The researchers were able to look at the spin states of the triplet excitons in considerable detail. They observed pairs had formed which variously had both weakly and strongly-linked spin states, reflecting the co-existence of pairs that were spatially close and further apart. Intriguingly, the group found that some pairs which they would have expected to decay very quickly, due to their close proximity, actually survived for several microseconds.

"Finding those pairs in particular was completely unexpected," Weiss added. We think that they could be protected by their overall spin state, making it harder for them to decay. Continued research will focus on making devices and examining how these states can be harnessed for use in solar cells."

Professor Behrends added: "This interdisciplinary collaboration nicely demonstrates that bringing together expertise from different fields can provide novel and striking insights. Future studies will need to address how to efficiently split the strongly-coupled states that we observed here, to improve the yield from singlet fission cells."

Beyond trying to improve photovoltaic technologies, the research also has implications for wider efforts to create fast and efficient electronics using spin, so-called "spintronic" devices, which similarly rely on being able to measure and control the spin properties of electrons.

More information: Leah R. Weiss et al. Strongly exchange-coupled triplet pairs in an organic semiconductor, *Nature Physics* (2016). DOI: 10.1038/nphys3908

Provided by University of Cambridge