Extra sulphur improves electronic structure of quantum dots

September 29 2017

Dr. Maria Antonietta Loi of the University of Groningen. Credit: Sylvia Germes

Quantum dots are nanometre-sized semiconductor particles with potential applications in solar cells and electronics. Scientists from the University of Groningen and their colleagues from ETH Zürich have now discovered how to increase the efficiency of charge conductivity in lead-sulphur quantum dots. Their results will be published in the journal *Science Advances* on 29 September.

Quantum dots are clusters of some 1,000 atoms which act as one large "super-atom." The dots, which are synthesized as colloids, i.e. suspended in a liquid like a sort of paint, can be organized into thin films with simple solution-based processing techniques. These thin films can turn light into electricity. However, scientists have discovered that the electronic properties are a bottleneck. "Especially the conduction of holes, the positive counterpart to negatively charged electrons," explains Daniel Balazs, Ph.D. student in the Photophysics and Optoelectronics group of Prof. Maria A. Loi at the University of Groningen Zernike Institute for Advanced Materials.

Stoichiometry

Loi's group works with lead-sulphide quantum dots. When light produces an electron-hole pair in these dots, the electron and hole do not move with the same efficiency through the assembly of dots. When the transport of either is limited, the holes and electrons can easily recombine, which reduces the efficiency of light-to-energy conversion. Balazs therefore set out to improve the poor hole conductance in the quantum dots and to find a toolkit to make this class of materials tunable and multifunctional.

"The root of the problem is the lead-sulphur stoichiometry," he explains. In quantum dots, nearly half the atoms are on the surface of the super-atom. In the lead-sulphur system, lead atoms preferentially fill the outer part, which means a ratio of lead to sulphur of 1:3 rather than 1:1. This excess of lead makes this quantum dot a better conductor of electrons than holes.

Thin films

In bulk material, transport is generally improved by "doping"the material: adding small amounts of impurities. However, attempts to add sulphur to the quantum dots have failed so far. But now, Balazs and Loi have found a way to do this and thus increase hole mobility without affecting electron mobility.

Many groups have tried to combine the addition of sulphur with other production steps. However, this caused many problems, such as disrupting the assembly of the dots in the thin film. Instead, Balazs first produced ordered thin films and then added activated sulphur. Sulphur atoms were thus successfully added to the surface of the quantum dots without affecting the other properties of the film. "A careful analysis of the chemical and physical processes during the assembly of quantum dot thin films and the addition of extra sulphur were what was needed to get this result. That's why our group, with the cooperation of our chemistry colleagues from Zürich, was successful in the end."

Devices

Loi's team is now able to add different amounts of sulphur, which enables them to tune the electric properties of the super-atom assemblies. "We now know that we can improve the efficiency of quantum dot solar cells above the current record of 11 percent. The next step is to show that this method can also make other types of functional devices such as thermoelectric devices." It underlines the unique properties of quantum dots—they act as one atom with specific electric properties. "And now we can assemble them and can engineer their electrical properties as we wish. That is something which is impossible with bulk materials and it opens new perspectives for electronic and optoelectronic devices."

More information: Daniel M. Balazs, Klaas I. Bijlsma, Hong-Hua Fang, Dmitry N. Dirin, Max Döbeli, Maksym V. Kovalenko, Maria A. Loi: Stoichiometric control of the density of states in PbS colloidal quantum dot solids Electronic structure engineering in quantum dot solids. *Science Advances*, 29 September 2017, DOI: 10.1126/sciadv.aao1558

Provided by University of Groningen