Solid-state physics

An historic experiment redesigned

Large quasiparticles known as Rydberg excitons have been detected in a natural crystal of copper oxide found at the Tsumeb mine in Namibia. The result may find use in applications such as single-photon logic devices. See Letter p.000

An exciton is a quasiparticle in a solid-state system comprising an electron and a hole — the absence of an electron. It has an energy spectrum akin to that of the hydrogen atom, so it may be considered as an artificial hydrogen atom in a solid-state environment, with the hole playing the part of the proton. The concept of excitons has first been formulated in the early 1930s by Yakov Frenkel, who predicted their existence in molecular crystals. A few years later, Gregory Wannier[2] and Nevil Mott[3] described these electron–hole bound states for inorganic semiconductors. In 1952 Evgeniy Gross and Nury Karryjew discovered[**4**] such Wannier–Mott excitons experimentally in a copper oxide semiconductor. On page 000 of this issue, Kazimierczuk *et al.*[5] now report how they have redesigned this historic experiment to find excitons in a natural crystal of copper oxide that extend more than tens of billions of lattice sites of the crystal.

Gross's and Karryjew's historic discovery marked the beginning of 'excitonics' — an area of solid-state physics which holds promise for applications in optoelectronics and information and communication technologies[6]. Gross and Karryjev selected for his studies crystals of copper oxide (Cu_2O). Using a spectrograph, they could identify eight dark lines in the material's transmission spectrum. Such absorption dips were associated with the energies of optical transitions from a ground state of the crystal, which corresponded to the absence of excitons, to exciton states with principal quantum numbers $n = 2, 3, \ldots, 9$ (Fig. 1). The transition energies scaled with n in a similar fashion to those of the hydrogen atom. This result proved that

hydrogen-like quasiparticles, the excitons, can be generated in these semiconductor crystals by photoabsorption.

In their study, Kazimierczuk *et al.* performed high-resolution transmission spectroscopy of a record-quality natural crystal of copper oxide found at the Tsumeb mine in Namibia using laser light of tunable frequency and ultralow spectral linewidth (corresponding to roughly 1.2 megahertz). Taking advantage of such narrow linewidth and the extremely high purity of the crystal, which has a thickness of 34 micrometres, the authors have been able to measure transmission spectra of the material with a spectral resolution of 5 nanoelectronvolts — a value that is extremely high for optical spectroscopy experiments. Analysis of the spectra revealed spectral absorption lines associated with the formation of excitons that have principal quantum numbers as large as $n = 25$. The size of an exciton increases as the second power of n, with $n = 25$ corresponding to a huge quasiparticle filling a sphere that has a diameter of more than 2 μm — that is, ten times the wavelength of light which is needed to create this exciton (see Figure 1 of the paper[5]). Moreover, the authors found that excitons formed at different locations in the crystal had, within the experimental accuracy of the measurements, identical spectral lines. This observation confirmed the extraordinary quality of the sample.

Such enormous quasiparticles, called Rydberg excitons owing to their large n, exhibit unusual quantum phenomena. One of them is giant diamagnetism, which results in a blueward shift of the exciton spectral lines and is related to the ability of the excitons to counteract an applied magnetic field. The giant diamagnetism in copper oxide was first observed by Gross and colleagues[7] and has been confirmed in the present experiments. Another phenomenon is Rydberg blockade, which has now been observed by Kazimierczuk and co-workers[5] and manifests as a saturation of the increase of the amplitudes of the absorption lines that correspond to large n.

Rydberg blockade means that only a limited number of large Rydberg excitons is permitted within a given volume in the crystal. The effect could be used to make nonlocal all-optical switches and single-photon logic devices.

There is no doubt that Kazimierczuk and colleagues' discovery of giant Rydberg excitons opens new avenues for the field of excitonics. Standard descriptions of light–matter interaction in a regime where the size of the exciton that results from the interaction exceeds the wavelength of light used to create it need to be revised. The old ansatz of additional boundary conditions on the exciton wavefunction, such as Pekar boundary conditions[8], that must be included into the classical calculation of optical excitations in crystal slabs acquires new importance in view of the nonlocal electrical properties of the slabs introduced by giant Rydberg excitons: creation of such an exciton by light at a point A in a crystal may strongly modify the electrical properties of the crystal at a point B separated from A by more than a micrometre. Furthermore, the fine structure of the absorption lines associated with excitons of large n may be rich and unusual owing to multiple emission and absorption of virtual photons by the excitons[9] and interactions between the spins and orbital momenta of the excitons' electrons.

Finally, a question arises: if $n = 25$ is now achieved, can researchers hope to observe excitons with principal quantum numbers as large as $n = 50$, for which the exciton would have a diameter of about a millimetre, and so would, in principle, become visible to the naked eye? Answering this question would require performing Kazimierczuk and colleagues' experiment at millikelvin temperatures (the present experiments have been done at 1.2 K) as well as attaining a spectral resolution of about 1 neV. This seems to be challenging but not impossible.

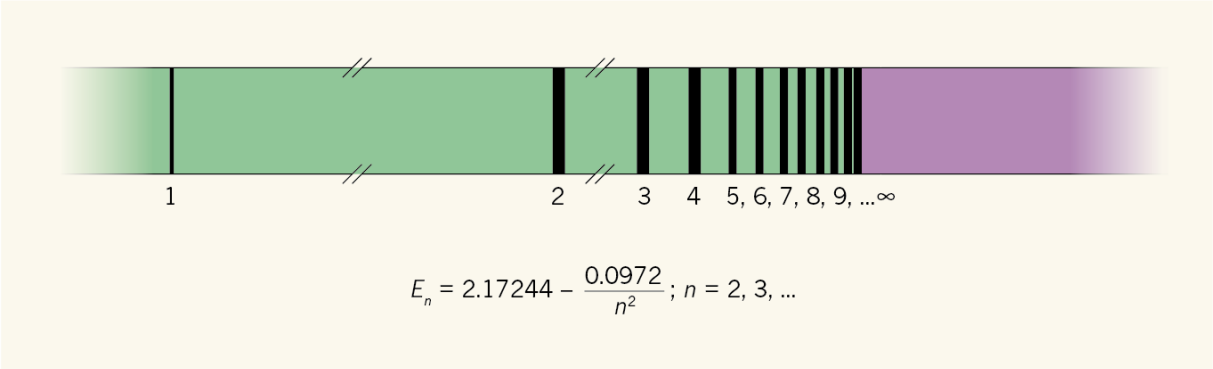

Figure 1 | The spectral signatures of excitons. This diagram shows the spectral absorption dips (black) associated with the hydrogen-like transitions of Wannier–Mott excitons in a copper oxide crystal. In their historic 1952 experiment, Gross and Karryjew identified[4] the eight dips that correspond to transition energies E_n with n = 2, 3, … , 9. The transition with the energy E_1 is forbidden by optical selection rules and could not be observed. Kazimierczuk *et al.*[5] have observed excitons in this crystal with n as large as 25. The energies are given in electronvolts.

Questions/comments about the figure/caption that need to be addressed:

1) Who should the credit for this graphic be? Alexey Kavokin? And on which of the Gross papers is it based? Please clarify.

Yes, based on Ref. 4. See also email and remark on Prof. Ruben Seisyan..

2) The paper seems to give different values for the numbers in the formula: 2.17208 eV and 92 meV. Please clarify as the reader will wonder.

Gross and Karryjew used slightly different energies to fit their spectra

3) Why are there two different colours in the spectrum, green and violet? Please clarify in the caption.

The violet colour shows energies of the interband optical absorption. The spectrum darkens at these energies.

4) The reader wonders about E_1. Please introduce this quantity in the caption too.

Done

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