

Rydberg excitons in cuprous oxide

Tomasz Kazimierczuk^{1,2}, Dietmar Fröhlich², Johannes Thewes², Julian Heckötter², Marc Aßmann², Stefan Scheel³, Heinrich Stolz³, M. A. Semina⁴, M. M. Glazov⁴, and Manfred Bayer^{2,4}

¹ Institute of Experimental Physics, Faculty of Physics, University of Warsaw, Poland

² Experimentelle Physik 2, Technische Universität Dortmund, Germany,

³ Institut für Physik, Universität Rostock, Germany,

⁴ Ioffe Institute, Russian Academy of Sciences, St.-Petersburg, Russia

Finding exact solutions of Schrödinger equation is virtually impossible in solid state physics due to extremely large number of involved particles. As a remedy, a selection of different approximate models were introduced to describe various effects in crystals. A particular example of such a model is a Wannier-Mott exciton. Such an exciton – a bound state of an electron and a hole – is a solid-state analogy of the hydrogen atom. As a consequence, the energy spectrum of an exciton is described by a Rydberg formula: $E_n = E_0 - Ry/n^2$ with effective Rydberg constant on the scale of 1-100 meV. However, in the typical case only few (e.g., up to $n = 3$ in the prototypical GaAs) excitonic lines can be resolved in the absorption spectrum.

In our work we demonstrate hydrogen-like excitonic states in bulk Cu₂O with principal number n as high as 25 [1]. Such states require particularly high quality crystals, since the spatial extension of the $n = 25$ state exceeds 1 μm. The energy positions as well as the linewidths of the excitons are found to follow power-law dependence in accordance with the model predictions. The oscillator strengths of the transitions are consistent with the predictions for $n < 17$, while for higher excited states additional quenching of the oscillator strength is observed. We discuss the origin of this quenching in terms of a dipole blockade between the excitons in the sample.

Our results show remarkable agreement between the experimental results and the theory. Nevertheless, the high-resolution absorption measurements allowed us to find also the effects beyond the predictions of the hydrogen-like model. Apart from the main P-series of the excitons, we identify in the absorption spectrum a series of weaker lines corresponding to higher angular momentum states. The lack of degeneracy with respect to angular momentum quantum number L is a direct consequence of the coupling with other bands, which does not have analogy in the actual hydrogen atom. Similarly, a splitting within a subspace of F-states is a result of valence band warping and does not exist without the anisotropic environment of the crystal lattice.

Altogether, our results show unprecedented agreement between the hydrogen-like model and the excitonic absorption spectrum, as well as the limitations of such a description. The observation of the high excitonic states and the dipole blockade effect opens the way to combine tools of atomic physics with the advantages of the solid state system.

[1] T. Kazimierczuk *et al.*, *Nature* **514**, 343–347 (2014)