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Excitons in cuprous oxide

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Document Version

Publisher's PDF, also known as Version of record

Publication date:

2008

[Link to publication in University of Groningen/UMCG research database](#)

Citation for published version (APA):

Fishman, D. (2008). Excitons in cuprous oxide. s.n.

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Introduction

In 1931 Yakov I. Frenkel proposed that it would be possible to excite charges in materials without influencing their electrical conductivity [1]. This suggestion can be marked as the birth of the exciton [1, 2], which turned out to be crucial in understanding many of the optical properties of condensed matter. The exciton can be imagined as the electron-hole analog of a hydrogen atom, where the light hole takes over the role of massive positron. More than 20 years later, there were two independent experimental observations of a hydrogen-like absorption spectrum in Cu_2O in visible energy range by Gross *et al.* [3], and Hayasi *et al.* [4]. This was the first experimental proof of the actual existence of excitons, even though at the time Hayasi *et al.* did not relate the observed absorption lines to the exciton suggested by Frenkel. Now, we are many years later, and a vast experimental literature of precise experiments on excitonic properties of many materials exists. Arguably, the best documented case is Cu_2O . All classes of transitions predicted by the theory for exciton spectra are observed in different parts of the, mostly visible, energy region have been observed in this compound. It might therefore not be surprising that cuprous oxide was the favorite semiconductor among many physicists in the pre-silicon era.

There are a number of well established phenomena which evidence that composite bosons made of even number of fermions, such as for example He-4 atoms, Cooper pairs, or alkali-metal atoms, behave under some conditions like ideal bosons showing a spectacular phase of large ensemble of these later particles: a macroscopic quantum state known as Bose-Einstein condensate. The exciton, being composed of two fermions, is an integral-spin particle, *i.e.* a composite boson. As a many particle system, these composite bosons show a variety of intriguing high density phenomena, including formation of excitonic molecules [5], electron-hole droplet and plasma formation [6], and even Bose-Einstein condensation [7]. Strong indications exist that Bose-Einstein condensation of excitons, predicted a long time ago [8], indeed occurs but, weirdly and wonderfully enough, in a peculiar system of the bilayer two-dimensional gases [9] in which excitons are formed under the equilibrium condition. The appearance of Bose-Einstein condensation of optically-created, finite-lifetime excitons is less evident [10] as only complex systems such as, for example, excitonic polaritons in semiconductor microcavities [11] provide signatures of a condensate phase. Since excitons in Cu_2O are strongly bound their bosonic character persists up to high densities and, due to light masses of particles, their condensate form is expected at appealingly high temperatures [7]. These are the lowest energy state excitons (singlet or so-called paraexcitons of the yellow series) that are expected to condense. Remarkable, these excitons are optically inactive and therefore characterized of

very long lifetimes provided the crystal is of a perfect quality. Their significant population can be achieved via laser pumping of higher-energy, optically-active states and the subsequent efficient relaxation processes towards the lowest singlet state. Advantageous on one side, the optical darkness of singlet excitons does not, however, allow to easily probing their properties and this has been one of the main obstacles in searching of their possible condensate form.

This work revisits the excitonic properties of Cu_2O as observed with (time resolved) optical techniques, making use of current days experimental capabilities providing very precise measurements, with high energy and/or time resolution, using a variety of high power coherent pulsed light sources, intense magnetic fields, and cryogenic temperatures.. Besides short reviews of what has done in the past, a number of new experiments are discussed aiming at the determination of the exciton lifetime and the time evolution of statistical properties (temperature, density, chemical potential) of an exciton gas created by a short light pulse. In doing so, the reader is also introduced to the principles of ultrafast time-resolved spectroscopy and time-resolved luminescence spectroscopy. Particular attention will be paid to the question whether it is possible to create the proper conditions for an excitonic Bose-Einstein condensate. The presence of an external magnetic field drastically changes the properties of the excitons, leading for instance to fairly complicated excitonic magneto-absorption spectra, which are discussed in terms of a model which is also applicable to hydrogen in the presence of extremely high magnetic fields as found in the atmosphere of neutron stars [12]. One other consequence of the presence of a magnetic field is that excitons which are not active in optical experiments may be activated by field induced mixing with optically active excitons. Since the optically inactive excitons generally have a very long recombination time, ranging up to microseconds, this is of particular interest in view of a possible Bose-Einstein condensation of excitons. Finally, the last chapter discusses some surprising observations in the optical properties of Cu_2O in visible-pump, far infra-red (about 1 THz) probe experiments are discussed, which deviate substantially from the usual Drude like behaviour observed in semiconductors.

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