Light-Induced Electron Pairing in Laser-Excited Semiconductor-Metal Heterostructures

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Laser excited quasi-2D heterostructures of transition metal dichalcogenides (TMDCs) have been shown to allow for quite a few higher order excitonic bound states such as trions (charged excitons), biexcitons (excitonic molecules), charged biexcitons, and more [1-5]. Such a large variety of coupled electron-hole quasiparticle excitations opens the door to a variety of new laser-driven phenomena in these systems, including metal-insulator transitions and Wigner crystallization, Bose-Einstein condensation (BEC), and even unconventional superconductivity [6-10]. Recently [5,10], an atom-like excitonic complex was reported experimentally in laser excited bilayer TMDCs in accord with theory predictions – the quaternion, the tightly bound complex of a free charge carrier in the top layer coupled to a like-charge trion in the bottom layer – provided that the entire heterostructure is placed close to a metallic surface to screen the excessive repulsive interaction in the system. Because such quaternions carry two net charges and are also bosonic, BEC of these quasiparticles would be a superfluid and therefore also a Schafroth superconductor [11]. Here, we develop a theoretical framework to explain the latest experimental observations of the Zeeman effect for quaternion complexes in perpendicular magnetostatic field [10]. Our theory is based on group theoretical analysis and spin-Hamiltonian formalism. We show that, contrary to the linear Zeeman shift known for excitons and trions in TMDC monolayers [12], the quaternion ground state is the spin-triplet to exhibit a quadratic magnetic field shift similar to that known for hydrogen-like atoms (whose ground state is singlet). In addition to prospective laser-driven BEC and superconductivity of bosonic quaternion excitations, another fascinating possibility for quaternions is that, as these bound four-particle doubly charged complexes repel each other, they could form a bosonic Wigner crystal. Such a light-induced quasiparticle crystal would be an 'atom-like' supersolid inside of the crystalline material. The process of Wigner crystallization is controlled by the ratio of Coulomb repulsion energy to average single-particle kinetic energy of an ensemble of charge carriers [13,14]. Due to the double charge and quadruple mass as compared to electrons, this ratio is at least 10 times greater for quaternions, suggesting higher crystallization temperature than that of the order of 10 K reported for quasi-2D electrons in TMDC nanostructures [15,16].

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