Quantum Droplets of Electrons and Holes in GaAs Quantum Wells

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Abstract: We present evidence for electron-hole quantum droplets in GaAs quantum wells using transient-absorption spectroscopy. Quantum droplets have a correlation function characteristic of a liquid, but have quantized binding energy, unlike macroscopic droplets.

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Many-body dynamics can be greatly simplified by the identification of appropriate quasiparticles. In semiconductors near the ground state, interactions between free electrons and the crystal lattice can be neglected by considering quasiparticle electrons and holes with modified mass and, for holes, charge. Electrons and holes can bind to form excitons [1], and two opposite-spin excitons can bind to form biexcitons. In indirect semiconductors, such as silicon and germanium, macroscopic droplets of electrons and holes in thermal equilibrium with an electron-hole plasma have been observed [2]. Evidence for polyexcitons has also been reported [3]. We present experimental and theoretical evidence for a new quasiparticle, which we call a quantum droplet [4]. A quantum droplet is a bound state of a small number of electrons and holes with no pairwise correlations, as in a polyexciton, but instead a two-particle correlation function characteristic of a liquid. Quantum droplets can form in GaAs quantum wells (QWs) under sufficiently high excitation density, and exist on a picosecond time scale, long before the system has reached thermal equilibrium.

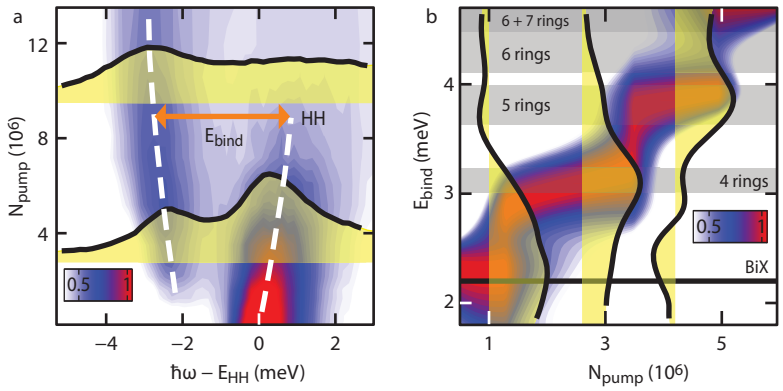

Fig. 1: (a) Probe $1 - T(\omega)$ for a delay of 2 ps as a function of pump photon number. White dashed lines track the peaks of the two main resonances. The HH exciton blue shifts with increasing number of pump photons. The lower-energy resonance unexpectedly red shifts. (b) Projection of probe $1 - T(\omega)$ to a "slanted-Schrödinger's-cat" state, which enhances the contribution of $\geq$ 3-photon correlations. The binding energy increases in a series of quantized steps.

In the transient-absorption experiment, a high-intensity pump pulse resonantly excites the heavy-hole (HH) QW exciton in GaAs multiple quantum well sample. The number of photons in the pump pulse, N_{pump}, is systematically varied while the average number is locked with a feedback circuit. A low-intensity probe pulse, with opposite circular polarization, arrives after a time delay of zero to several picoseconds. We measure the spectrum of the transmitted probe light and calibrate the probe transmission $T(\omega)$ in absolute units.

Figure 1a shows the probe $1 - T(\omega)$ for a probe delay of 2 ps over a range of N_{pump}. The HH resonance is a peak at $\hbar\omega = E_{\text{HH}}$. As N_{pump} is increased, the HH resonance blue shifts because of bandgap renormalization, screening, and Pauli blocking of the low-energy states [5–7]. At the same time, the lower-energy resonance, which we naively identify as a biexciton, red shifts, indicating an increase in the binding energy E_{bind}. However, this binding energy should respond similarly to the HH binding energy. The unexpected red shift suggests that the lower-energy resonance cannot be due to biexcitons alone.

More information about the many-body aspects of the sample response is obtained by projecting out the part of the measured spectra caused by correlations of three or more photons (i.e., higher-order correlations than biexcitons) [7]. Figure 1b shows the resulting differential spectra, for a probe delay of 8 ps. As in the unprojected data, the resonance red shifts with increasing N_{pump}, but the projection reveals a stepped behavior between quantized energy levels.

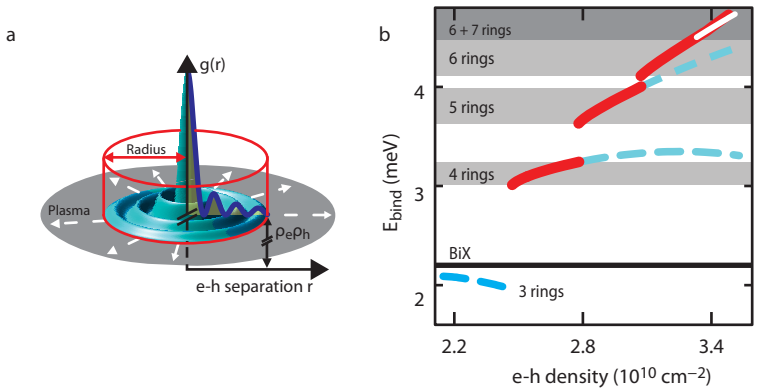

Fig. 2: (a) Electron-hole correlation $g(r) = \Delta g(r) + \rho_e\rho_h$ for a quantum droplet, as a function of e-h separation r. (b) Calculated E_{bind} for quantum droplets, as a function of e-h density.

Quantum droplets explain both the red shift of the resonance, and the quantized steps evident in Fig. 1b. The electron-hole correlation function of a quantum droplet is shown in Fig. 2a. The correlation function has a sharp peak in the middle because of the attractive Coulomb interaction between electrons and holes. The rings in the correlation function are separated approximately by the mean electron-hole separation in the droplet, and vanish beyond the droplet-plasma boundary at radius R. Figure 2b shows the calculated binding energy of droplets as a function of electron-hole density, which closely matches the measured binding energy from Fig. 1b.

To test the identification of this resonance as quantum droplets, we performed two control experiments. First, we repeated the transient absorption measurement with co-circular polarizations for pump and probe. Thus, selection rules prohibit excitation of biexcitons or polyexcitons, but we still see the quantum droplet resonance. Second, we raised the sample temperature to 70 K, where phonon scattering should prevent the formation of biexcitons or quantum droplets. In this case, the quantum droplet signature is absent from both the recorded data and the projection. Thus, we conclude that all observations are consistent with identifying the low-energy resonance of Fig. 1a as a quantum droplet.

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