Summary of thesis:
Excitonic fine structure and nonequilibrium phase transition of the electron-hole system in diamond

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The contrast between equilibrium and nonequilibrium phenomena is one of the central issues in contemporary physics, and has been studied in various systems including high-T_c superconductors and polariton superfluids. Among such systems, the many-body system of electrons and holes excited in indirect-gap semiconductors with light is an ideal platform to investigate a gas-liquid phase transition in an open system. For one thing, this is because the system is intrinsically dissipative because particles annihilate due to recombination processes. In addition, the gas-liquid phase transition between an excitonic gas and electron-hole liquid (EHL), the Fermi liquid composed of electrons and holes, is realized below critical temperature T_c. Similarly to other systems such as water, transitions from an excitonic gas to EHL are initiated by nucleation into small clusters. Finally, the system becomes spatially inhomogeneous, where the gas and the liquid coexist.

The properties of gas-liquid phase transitions would drastically change if systems are driven out of equilibrium. It was predicted [1] that the phase separations disappear when the nucleation is hindered by the dissipation of particles. As a result, clear distinction between the gas and the liquid was expected to be lost. Recently, polyexcitons, or N-body bound states of electron-hole pairs, were observed in diamond [2], which in fact would be the manifestation of nonequilibrium nature of the system.

In this thesis, I present a novel phase diagram of an electron-hole system out of equilibrium in diamond, whose carrier lifetime is substantially short compared to those of conventional semiconductors such as silicon and germanium. Before reaching our key scientific theme, however, there were a couple of technical challenges unique to diamond to be overcome.

We first study the structure of the exciton, the simplest entity appearing as the result of the Coulomb interaction [3] between an electron and a hole. We perform high-resolution spectroscopy on an ultrapure diamond in order to clarify the origin of the fine structure in the excitonic energy levels. We

find that the fine structure is caused by the competition between three effects: the spin-orbit interaction on the hole, the exchange interaction between the electron and the hole, and the mass anisotropy at the conduction-band minima.

By using the knowledge on the fine structure, we then propose and test a new excitation method necessary to create a dense and cold electron-hole system which is crucial for investigating nonequilibrium phenomena [4]. We find that the effective temperature of excitons can be lowered to 15 K even at the high density of 10^{19} cm^{-3}.

Finally, we investigate the nonequilibrium gas-liquid phase transition between excitons and the EHL. We perform photoluminescence spectroscopy at various temperatures and densities by using our excitation method.
We observe polyexcitons below 30 K in a wide range of densities even though equilibrium thermodynamics predicted the stabilization of the EHL. By analyzing the size distribution function of polyexcitons, we find an unprecedented signature of a transition without a phase separation while a phase separation is expected below T_c. The comparison with a numerical simulation reveals that the unconventional transition is caused by the effect of finite carrier lifetime.

Our findings will contribute to further development of the many-body physics in excited states mainly in two-fold way. First, our result on the fine structure will boost the study on the structure of polyexcitons and their relation to the valley degrees of freedom. Second, the observation of a transition without a phase separation below T_c will benefit the long-lasting quest for the comprehensive framework on nonequilibrium thermodynamics.

References

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