Summary of thesis

Localization dynamics of paraexcitons and their lattice relaxation at oxygen vacancies in cuprous oxide

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An ensemble of excitons or bound electron-hole pairs in a semiconductor is predicted to undergo a transition into a Bose-Einstein condensate (BEC) phase at low temperatures. The required condition is that the excitonic lifetimes should be longer than the relaxation time in order to achieve thermal equilibrium. Due to their long lifetime, 1s paraexcitons in cuprous oxide (Cu_2O) have been believed to be a good candidate to realize excitonic BEC. However, the lifetime of the 1s paraexciton is limited by some unknown non-radiative recombination processes, and the measured lifetimes of paraexcitons widely scatter depending on the measurement method or on the sample quality. In this thesis, I describe spectroscopic studies investigating dynamics and mechanism of the lifetime shortening of paraexcitons in Cu_2O.

In recent investigations on excitonic BEC, there is a trend of performing experiments at sub-Kelvin temperatures. This is to suppress the two-body collisions (also known as Auger recombination) of paraexcitons by lowering the critical density for BEC transition. At such ultralow temperatures, the dephasing time and lifetime of paraexcitons are limited by extrinsic sources like impurities and crystal defects even in high-quality natural Cu_2O crystals. Therefore, it is an important and urgent task to clarify dynamics and mechanism of paraexciton trapping at defect centers.

In the former part of the thesis, I describe our study on the trapping dynamics of free excitons into crystal defects in Cu_2O. By using high-grade samples containing small amounts of oxygen vacancies, we find the direct correlation between the lifetimes of free and trapped paraexcitons. We establish a four-level model to explain the temperature dependent lifetimes, and find out that trapping of paraexcitons into oxygen vacancies is the main channel of lifetime shortening in high-quality samples. Based on the temperature dependence of transients and the photoluminescence intensities, we have obtained the activation energy of E_A = 33 meV for trapping of free excitons into oxygen vacancies.

The measured value of the activation energy is much smaller than the energy

separation of ΔE = 0.30 eV between the photoluminescence peaks due to free and trapped excitons. Therefore, we further explore the photoluminescence (PL) and photoluminescence excitation (PLE) spectroscopy on trapped excitons to clarify the trapping potential and the relaxation process based on an adiabatic potential model.

In the latter part of the thesis, I describe our investigation to extract the exciton-phonon coupling strengths and the effective frequencies of the trapping potentials by analyzing the temperature dependence of the widths of the PL and PLE spectra of trapped excitons. We obtain the Stokes shift of 0.27 eV, which agrees with the energy difference $\Delta E - E_A$. This indicates that the large residual energy is released to the vibrational modes by the trapped excitons at localized centers. The frequencies of the vibrational modes of the trapping potentials are found to be close to that of optical phonons of Γ_4^- symmetry, which are infrared active in Cu_2O. Also, our finding of medium coupling between excitons and phonons (with Huang-Rhys factors ~ 8) indicates efficient lattice relaxation at defect centers.

Since trapping to oxygen vacancies is found to be the main decay channel of free paraexcitons, this calls for necessity of actively controlling the trapping at low temperatures toward stable formation of paraexcitons and realization of BEC. The small activation energy revealed by our analysis means that thermal ionization occurs from the trapped state to the free exciton state at a rate higher than that previously expected. The shallowness of the trapped state opens up a new possibility of optically controlling the activation of free excitons out of the defect levels. For example, by using a pump beam in the terahertz region (c.f., 33 meV corresponds to 8 THz), activation of trapped excitons into the free exciton state could be possible. One could also be able to prevent further trapping of paraexcitons by saturating the defect levels by pumping at 640 nm (1.94 eV) below the exciton resonance, where the maximum of the absorption has been found in our experiments. Thus, outcome of this thesis provides detailed strategies to control paraexciton lifetimes and to realize excitonic BEC in Cu_2O.