EXTREMELY-NARROW LINEWIDTH OF THE CONFINED EXCITONS IN CuCl QUANTUM DOTS

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Homogeneous linewidth of the excitons confined in CuCl quantum dots embedded in a NaCl crystal was studied by heterodyne-detected accumulated photon echo technique. At 2K, photon echo signal shows an exponential decay with a decay time of 160ps except for a slow rise. The homogeneous linewidth calculated from the dephasing time was found to be much smaller than that reported previously by the other methods. At 1.5K, we observed the linewidth of 1.6μeV. As far as we know, this is the narrowest exciton linewidth in quantum dots.

1 Introduction

In nanometer-sized semiconductor crystallites, three-dimensional quantum confinement modifies electronic structure of the material remarkably. Namely, continuous energy bands become to atomic-like discrete energy levels. Therefore quantum dots are expected to show ultra-narrow optical spectra. However, usual semiconductor quantum dot sample shows large inhomogeneous broadening because of the large size-distribution of the quantum dots. Recently, single quantum dot spectroscopy made it possible to observe luminescence from a quantum dot, and it was found that their luminescence linewidth was very narrow (< 0.1 meV) compared with previously reported homogeneous linewidth[1]. Since their linewidth are so narrow, it is difficult to evaluate the homogeneous width correctly using usual spectrometer. On the other hand, time domain measurements such as photon echo are useful in this case. In this paper, we report on the measurement of the phase relaxation time of the lowest excited state (Z_3-exciton) of the CuCl quantum dots embedded in a NaCl crystal by using heterodyne-detected accumulated photon echo method[2,3].

2 Experimental Procedure

Accumulated photon echo uses accumulation effect by the bottleneck states to enhance echo signal[2], so it is suitable for the system which have bottleneck states whose lifetime is longer than the period of the repetitive excitation laser pulses. In CuCl quantum dots, persistent spectral hole burning phenomena occurs, and this indicates the existence of some bottleneck states[4]. Moreover in this method excitation laser pulses are phase modulated by a piezoelectric actuator and the echo signal is measured by using interference with laser pulses[3]. As a result, it becomes very sensitive method, which allows us to measure the echo signal under very weak excitation energy density (~100pJ/cm^2). Since phase relaxation time is known to strongly depend on the carrier density, it is important to reduce the excitation energy density for our purpose.

The sample used here was a CuCl quantum dots embedded in a NaCl crystal, which was made by the heat treatment on a Cu^+ doped NaCl crystal. The light source was a ps Ti:sapphire laser system whose repetition rate was 82 MHz. The second harmonics was used as the excitation pulse.

3 Results and Discussions

Figure 1 shows photon echo signals at various temperatures. Excitation energy density was so small that the echo signal does not depend on the excitation density. Echo signals can be fitted by a single exponential decay except for the initial part, whose rise time depends on the temperature and can be assigned to the anti-hole structures in the hole burning spectrum. The dephasing time T_2 was determined by the decay time constant t by the relation, $T_2=2t$. The dephasing time decreases rapidly with the temperature rise.

In fig. 2, temperature dependence of the homogeneous linewidth was shown. The homogeneous width agrees with that measured by persistent spectral hole burning (PSHB) at the high temperature region (~50K) shown by open circles[5]. However, at low temperature region (below 50K) there is a large discrepancy between our data and PSHB data. The most remarkable difference is that the former continue decreasing with decreasing temperature even below 5K, while the

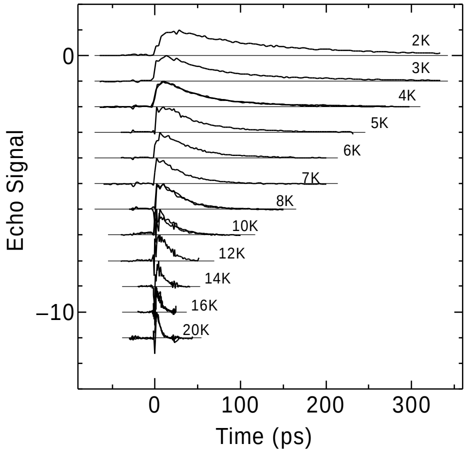

Fig. 1 Echo signals of CuCl quantum dots at various temperatures. The radius of the quantum dots is 3.1nm. The decay time becomes shorter with the temperature rise. Slow rise observed in the initial part come from the antihole structure in the persistent spectral hole burning.

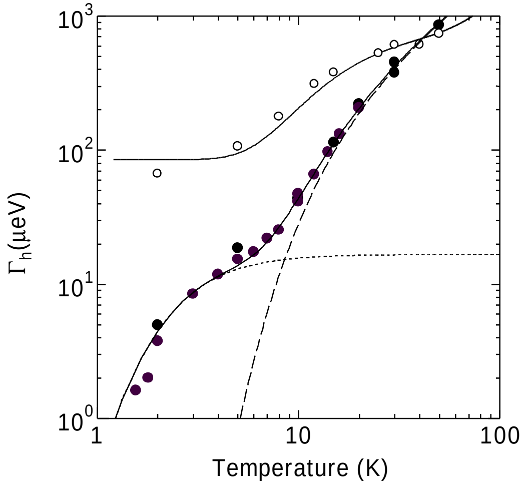

Fig.2 Temperature dependence of the exciton homogeneous linewidth from the accumulated photon echo data. At high temperature, our data correspond to the previously reported homogeneous linewidth measured by persistent spectral hole burning [5].

PSHB data shows constant value below 5K. In the PSHB case, the spectral resolution of the monochromator determines low temperature limit of the homogeneous width and spectral diffusion may affect the experimental results. So we believe that we have measured true homogeneous width by the photon echo method.

We observed extremely long decay time of the photon echo signal at low temperature. For example, decay time of the echo signal is 410ps at 1.5K, which corresponds to the homogeneous linewidth of 1.6μeV. As far as we know, this is the narrowest homogeneous width of the confined exciton in quantum dots. Since the lifetime of the excitons in CuCl quantum dots in a NaCl crystal is about 2ns[6] and the corresponding lifetime-limited linewidth is below 1μeV, more narrower linewidth may be observed in this system. Observed linewidth of the CuCl quantum dots is much less than that in bulk CuCl, which was measured by two-photon absorption of longitudinal exciton by M. Kalm[7] and found to have constant value of 140μeV below 10K. The long coherence time of excitons in quantum dots may be due to the disappearances of the interactions with any disturbance by the spatial confinement.

The temperature dependence of the hole width in PSHB measurement was explained by thermally excited confined acoustic phonon and longitudinal optical phonon[5,8]. Obviously, one cannot explain simply this anomalous temperature dependence in low temperature region of our data by these two phonon modes. We tried to reproduce the temperature dependence at high temperatures (>10K) by the interaction with the confined acoustic phonon mode of the 3.1nm radius quantum dots, and succeeded to some extent as fitted in fig. 2 by a dashed line. As the other process, one candidate may be two level system (TLS) which was used to explain temperature dependence of the homogeneous linewidth of impurity in glass matrix

[9] or dye molecules in polymer. Temperature dependence by the interaction with TLS is expressed in the form of $cosh^{-2}$ *(hn/2kT)*, here hν is the energy of TLS. We can reproduce the experimental data by the sum of these two processes assuming hν to be 0.44meV. However, the low temperature data seems to depend on the sample quality, the dephasing mechanism at low temperature is not clarified yet.

4 Conclusion

Temperature dependent homogeneous linewidth of the excitons confined in CuCl quantum dots embedded in a NaCl crystal was investigated. Measurements of the linewidth was performed by the heterodyne-detected accumulated photon echo technique. Under very weak excitation condition and at low temperatures, the photon echo signal shows an exponential decay with a long decay time. It corresponds to the very sharp line width. Especially, at 1.5K, exciton line width was 1.6μeV, it is the smallest value of the linewidth of quantum dots. Further experimental study is necessary to clarify the exciton dephasing mechanism in quantum dots, especially at lower temperatures.

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