Study of the Exciton-Polariton Relaxation in CuCl by Picosecond Time-Resolved Spectroscopy(Abstracts of Doctral Dissertations)

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Tsuyoshi IKEHARA

Department of Physics

Abstract

Chap. I. Introduction

Exciton polariton state has been confirmed to be a good eigenstate in semiconductor crystals in which an exciton has strong interaction with a photon. The temporal variation of the exciton-polariton energy distribution has been measured in several materials by the picosecond time-resolved spectroscopy. However, the relaxation probability of the exciton polariton at different energies has not been measured directly and could not be compared with theoretical one calculated from various exciton-phonon interactions.

Chap. II. Experimental Procedures

Since the semiconductor CuCl has the simplest exciton system, it is appropriate to study its detailed dynamics. However, no high-repetitive picosecond laser was available at the exciton energy (near uv) of CuCl, so far. In this study, a high-repetitive (76 MHz) picosecond pulse is generated by doubling the frequency of a cw mode-locked dye laser light. This light source allows us to measure the exciton luminescence at picosecond domain, without any high-density excitation effect. The detection system consists of the combination of a subtractive double monochromator and a synchroscan streak camera, in which high resolutions on time and energy are compatibly realized.

Chap. III. Propagation and Relaxation of the Exciton Polariton which Appear in the Temporal Response of the Exciton Luminescence

The exciton luminescences are investigated under the excitation of an absorption band lying 70 meV higher than the lowest exciton. The temporal response of the exciton resonant luminescence shows many pulsed structures which appear at different time depending on the measurement configuration and the observation energy. Analyzing the time of the pulse emergence, the exciton polaritons are found to propagate coherently with their group velocity, for the distance longer than 10 μm. The temporal width and the intensity of this pulsed structure are analyzed in detail. In particular, the intensity of the pulsed structure reflects directly the relaxation of the exciton polariton during the propagation in the crystal. The energy-dependent relaxation probability of the exciton polariton is determined to be $10^9 \sim 10^{10}$ s^{-1} within the longitudinal-transverse splitting energy region.

Chap. IV. Picosecond Time-Resolved Measurement of the Resonant Light Scattering

To estimate the scattering probability in the energy region wider than Chap. III, the temporal response of the resonant 2LO scattering and the resonant Brillouin scattering are measured and analyzed. Theoretically, it is shown that the decay time of the one-phonon scattering light is described by the scattering probability and the group velocity of the exciton polaritons at the energy of both the incident and scattered light. Using this relation, the scattering probability of the exciton polariton is evaluated in the energy region between the transverse exciton energy − 3 meV and the longitudinal exciton energy + 8 meV. A large contribution of the radiative decay is proved below the transverse exciton energy.

Chap. V. Discussion on the Relaxation Probability of the Exciton Polariton

The experimental results are compared with the theoretical probability of the exciton acoustic phonon scattering. The energy dependence qualitatively shows good agreement with the scattering probability through the piezoelectric interaction, rather than through the deformation potential. The decay time of the total number of the exciton polaritons is also analyzed and found to be determined by the trapping of the exciton polaritons to crystal imperfections, not by the radiative decay.

Chap. VI. Correlation between Absorption Spectrum and Exciton Polariton Relaxation

In the polariton picture, the relaxation of the exciton polariton will be the absorption of light. One can reproduce an absorption spectrum from the measured relaxation probability. However, within the longitudinal-transverse splitting energy this spectrum has large discrepancy from the absorption spectrum obtained from the conventional transmission measurement. In this study, the effect of a fine surface roughness is verified to be one of the reason for this discrepancy. The angle-resolved transmission measurement shows that the transmitted light beam in this energy region is scattered by the surface roughness because of a very large refractive index of the lower branch exciton polariton.

Chap. VII. Space-Resolved Spectroscopy using Group-Velocity Propagation of the Exciton Polariton

Applying the nature of long propagation of the exciton polariton with the group velocity, a spatial structure of micrometer order in a crystal is observed by projecting it on the picosecond time axis. It is shown that a layer structure exists in several crystals, where the exciton polariton is mostly reflected. The spatial distribution of several kinds of imperfections is also observed by the time-resolved measurement of the bound-exciton luminescences.

Chap. VIII. Conclusions

The results are summarized. The dynamics of the exciton polariton is analyzed in the polariton-picture framework where the coherent propagation and the radiation at the crystal surface are most important for the exciton polariton. The energy-dependent relaxation probability of the exciton polariton is measured in three methods and compared with the existing theory of exciton-phonon interaction.