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Observation of Exciton Polariton Dispersion in CuCl†

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The upper branch of an exciton-polariton curve is measured directly for the first time. The two-photon spectrum of the first exciton line of CuCl shows a strong dependence on the total K vector of the two beams. The resulting dispersion curve can be quantitatively explained using one-photon data.

Polaritons result from the strong coupling between the polarization in a crystal and the electromagnetic radiation field.[1] They describe the normal modes of the coupled system. Depending on the mechanism of polarization, phonon and exciton polaritons are possible. Polaritons are already inherent in the solution of Maxwell's equations for a medium characterized by a dielectric constant, and thus are an intrinsic feature of classical crystal optics. In the quantum mechanical description of the optical dispersion and absorption process they provide a more detailed view in terms of elementary excitations.[2] As pointed out by Hopfield,[3] polaritons are a particularly useful concept when discussing experiments in which the polarization is driven by a weak coupling force different from the strong couplings already included in the polariton states. These kinds of interactions may arise in special situations of crystal optics,[4] in Raman scattering,[5] and in other fields of nonlinear optics including two-photon absorption.

The polariton effect in Raman scattering has become an especially useful tool for studying the dispersion relation of phonon polaritons.[6] In view of the close relationship between Raman and two-photon spectroscopy, the latter should be an equally suited method for measuring exciton-polariton dispersion. This measurement may be carried out by changing the angle between the two photon beams and thus tuning the wave vector of the excited polariton. It is the purpose of this Letter to present the first two-photon detection and dispersion measurement of the exciton polariton. We have chosen CuCl as the material since it is optically isotropic and possesses sharp 1s-exciton lines which are clearly separated from the rest of the electronic spectrum.[7] Because of the lack of inversion symmetry in its point group (T_d), the polariton may be excited in the two-photon absorption process.

Two-photon spectra are measured with an automatic recording spectrometer[8] which provides an energy resolution of $\Delta E/E = 10^{-5}$. Figure 1 shows the experimental results in the neighborhood of the first exciton band of CuCl. In an unoriented crystal, two bands appear in this spectral region. The oscillator strengths and the energy separa-

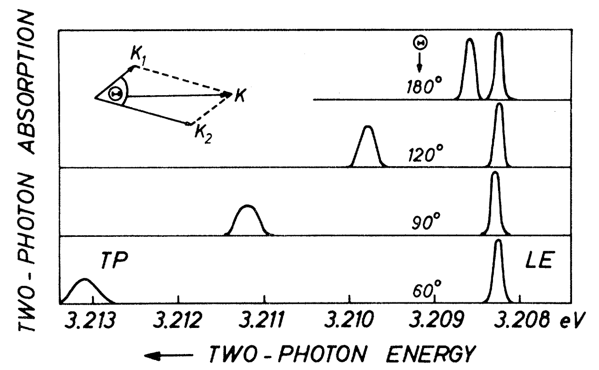

FIG. 1. Two-photon absorption bands of CuCl near the first exciton for different angles Θ between the two beams, at 1.5°K. The areas underneath all the bands are normalized to the same value despite their different polarization dependences.

tion between these bands depend on the total K vector of the two beams. The oscillator strengths also depend on the beam polarization. These bands are plotted in Fig. 1 as a function of two-photon energy and the angle Θ between the photon beams in the crystal. Different parts of the figure thus refer to different values of the total K vector. The low-energy peak is independent of Θ; the higher band is broadened and drastically shifted as Θ is varied. Additional information on the degeneracy of the two-photon excited states is obtained by measurements in a magnetic field (Faraday configuration, 50 kG). The lower peak does not split in a magnetic field, but the second band is split into two components. On the basis of these results we identify the lower energy band as the longitudinal (nondegenerate) exciton (LE) state which is allowed in two-photon absorption, in contrast to one-photon absorption. The higher energy band is attributed to the transverse (twofold degenerate) polariton (TP). This state belongs to the upper branch of the polariton dispersion curve originating from the first exciton line of CuCl. The broadening of the polariton bands for large Θ is probably due to the spread of the total K value which is caused by the aperture of the tunable light source. Because of the increasing steepness of the dispersion curve for large K values, the spread of the total K value results in an increase of the broadening of the measured polariton band for these K values.

This may be clearly seen in Fig. 2 where we plot the energies of the measured bands in the familiar dispersion diagram of polaritons versus wave vector K. The magnitude of K is easily calculated as a function of Θ from the diagram shown in Fig. 1 in terms of the wave vectors $K_1 = n_1\omega_1 c^{-1}$ and $K_2 = n_2\omega_2 c^{-1}$ of the two beams. We have used a refractive index[9] $n_1 = 1.89$ for the laser energy $\hbar\omega_1 = 1.1648$ eV and a refractive index $n_2 = 1.97$ for a representative energy $\hbar\omega_2 = 2.045$ eV of the second photon.

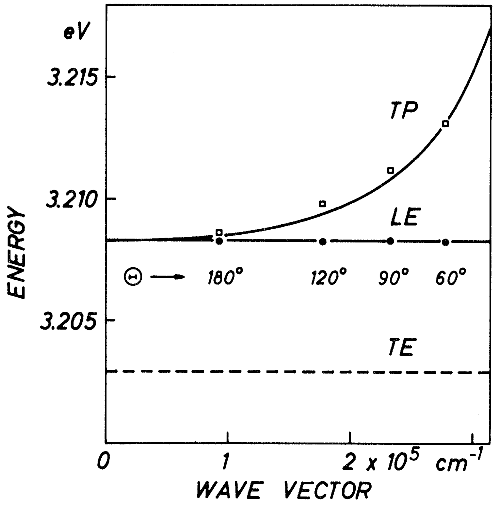

FIG. 2. Polariton diagram for the first exciton of CuCl: closed circles, longitudinal exciton (LE); open squares, upper branch of transverse exciton polariton (TP); solid line, dispersion curves calculated from reflectivity data; dashed line, transverse exciton (TE).

It is informative to compare the experimental two-photon dispersion with the dispersion relation calculated from one-photon optical constants. If the optical properties are expressed by the dielectric function $\epsilon(\omega, K)$ the polariton dispersion $\omega(K)$ is determined by the equation

$$c^2K^2 = \omega^2\epsilon(\omega, K).$$

In our case we may neglect the K dependence (spatial dispersion) of ϵ because the two-photon measured longitudinal branch shows no dispersion within the experimental accuracy ($\pm 2 \times 10^{-5}$ eV). This can be understood in view of the rather large exciton mass of CuCl ($m \sim 13$ electron masses[10]). We have used values for $\epsilon(\omega)$ calculated from the 4.2°K reflectivity data of Staude[11] via a Kramers-Kronig transformation. The solutions of the dispersion equation and the longitudinal exciton branch are shown in Fig. 2 as full lines.[12] The dashed line corresponds to the position of the transverse exciton resonance. Coincidence of the measured and calculated dispersion is very satisfactory if one takes into account the different origins of the data which are compared with each other. The two-photon spectra are measured from the bulk of the crystal, whereas the reflectivity data, on which the Kramers-Kronig procedure is based, may depend on the quality of the sample surface.

Finally we would like to point out some consequences of the preceding results for the interpretation of two-photon experiments. In calculations of the polarization dependence of excitonic two-photon absorption,[13] it has been assumed that the exciton states reached in the process transform according to the irreducible representations of the point group of the crystal. This has to be altered in the case of two-photon transitions which are also one-photon active. From the foregoing it is evident that these states should be classified according to the group of the polariton wave vector K, and the polariton should be regarded as the final state of the transition. Because the degeneracy between longitudinal and transverse states is lifted, a more complicated angular dependence of the two-photon absorption should result. These features will be demonstrated in a forthcoming paper.

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Quantum Interference of Electron Waves in a Normal Metal*

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Using a very pure single crystal of magnesium we have observed large-amplitude oscillations in the transverse magnetoresistance which result from the direct interference of normal-state electron waves. The amplitudes of these interference oscillations appear to be independent of the kT broadening of the Fermi distribution. The primary result of our experiment shows that these waves are phase coherent at 1.2°K over macroscopic distances of at least 0.01 mm; this corresponds to a quasiparticle lifetime of at least 2×10^{-11} sec.

In our study of the transverse magnetoresistance of magnesium we have observed large-amplitude oscillations resulting from the direct interference of electron waves. Unlike the de Haas-van Alphen and de Haas-Shubnikov oscillations, which are consequences of the phase coherence and quantization of electron states on closed paths, these oscillations are due to the quantum interference of electrons on different branches of an open trajectory. The effect we have observed is the physical equivalent of the Mercereau experiment,[1] in which the interference