PROC. 15TH INT. CONF. PHYSICS OF SEMICONDUCTORS, KYOTO, 1980
J. PHYS. SOC. JAPAN **49** (1980) SUPPL. A p. 401–404

TWO NEW METHODS OF POLARITON SPECTROSCOPY TESTED WITH CdS : MAGNETO BRILLOUIN SCATTERING THIN PRISM REFRACTION

I. Broser and M. Rosenzweig
Institut für Festkörperphysik, Technische Universität Berlin
R. Broser, E. Beckmann and E. Birkicht
Fritz-Haber-Institut, Max-Planck-Gesellschaft, Berlin-Dahlem

Brillouin scattering in high magnetic fields has been measured for the first time. The singlet-triplet-splitting of the A-exciton of CdS, unknown up to now, is determined to be $\Delta = 0.2$ meV.
Refraction of light by thin prismatic CdS crystals has been used to obtain dispersion curves for the A- and B-polariton. With this direct and precise method the knowledge of the optical behaviour of polaritons has been improved essentially.

To determine the behaviour of polaritons several spectroscopic methods have been applied such as reflectivity, absorption (two-photon), emission and resonant scattering (two-photon, Raman, Brillouin). Nevertheless, there are still open problems some of which could be solved by the two methods reported here.

Magneto Brillouin Scattering

Resonant Brillouin scattering is one of the most direct methods to determine the dispersion curves of excitonic polaritons. We have used Brillouin scattering for the first time in a magnetic field to analyse the Zeeman-splitting of free excitons.

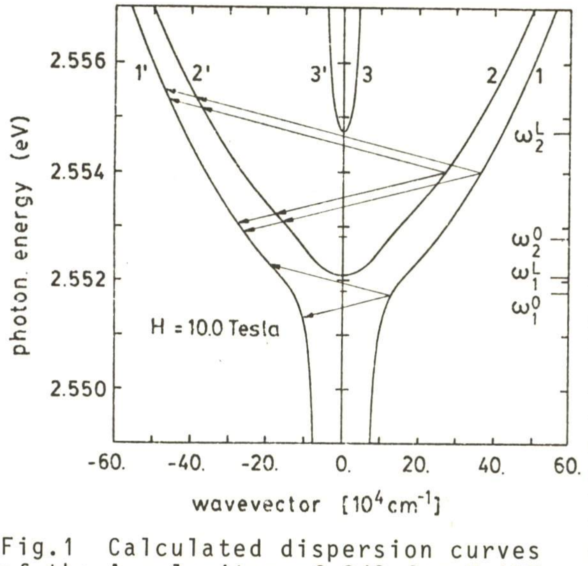

Fig.1 Calculated dispersion curves of the A-polariton of CdS for H=10T

In CdS the optically allowed Γ_5-states and the forbidden Γ_6-states, energetically separated by the singlet-triplet-splitting (Δ), are split and mixed by an external magnetic field $H \perp c$, proportional to the g-value of the conduction band [1] :

$$E_{\pm} = E_0 - \frac{1}{2} \left(\Delta \mp \sqrt{\Delta^2 + g_e^2 u_B^2 H^2}\right)$$

The ratio of oscillator strengths of the two excitons f_-/f_+ increases from zero to unity with increasing field strength. In the model of excitonic polaritons for H=0 only one oscillator exists, but for $H \neq 0$ two excitonic oscillators interact with photons. As demonstrated in Fig.(1) the number of scattering lines changes from one to four for $\omega_1^L < \omega < \omega_2^L$.

The CdS crystals (thin platelets with smooth surfaces parallel to the c-axis) were placed in a superconducting magnet with split coils, cooled in vacuum to approximately 8K and excited by a cw-dye-laser. The scattered light was analysed by a 0.75m double monochromator and detected by a photomultiplier and a lock-in amplifier. The spectral resolution of the set-up was 0.04 meV.

In Fig.(2) typical scattering spectra for different magnetic fields are shown. The Stokes lines of the LA- and the TA-phonons [2,3] are observable. The LA-line splits into three sub-lines with increasing field strength, while the corresponding splitting of the TA-line cannot be resolved as a result of its much smaller sound velocity. For all laser energies one Stokes and one anti-Stokes line appear (arrows) which shift linearly with H. The g-value $g=1.75\pm0.05$ is that of the conduction band. We interpret these lines as the result of a spin-flip Raman process of free or loosely bound electrons.

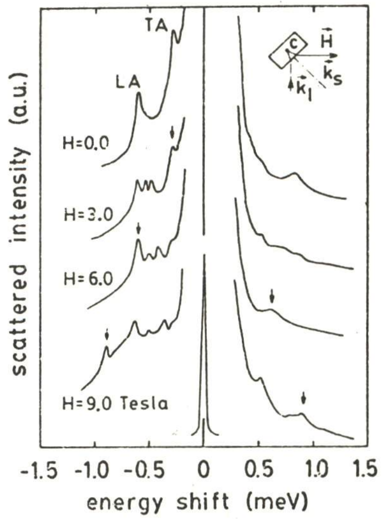

Fig.2 Measured Brillouin scattering spectra

In Fig.(3) experimental Stokes Brillouin shifts are plotted versus laser-energies for H=0 and H=10 Tesla. The calculated Brillouin shifts are the lines fitted using the parameters for best agreement to the experimental results at all fields. Note: The calculated shifts of the (1-2') and the (2-1') transitions are nearly identical, which explains the observation of only three lines instead of the expected four.

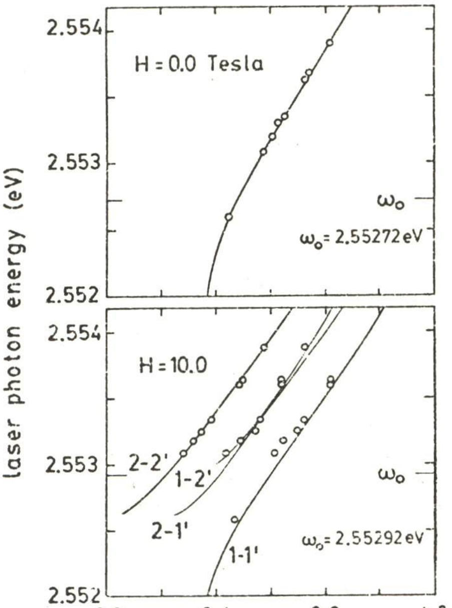

Fig.3 Measured and calculated Brillouin shifts

The sound velocity $v=4.25\cdot10^5$cm/s has been taken from literature [2]. The parameters $\varepsilon_0=8.1$, $m_\perp=0.9m_0$, $\omega_0=2.5527$eV and $4\pi\alpha_0=0.01335$ have been derived from our zero field Brillouin scattering measurements. They agree satisfactorily with those in literature [2,3]. The dispersion parameters of phonons, photons and excitons have been assumed to be field independent. The diamagnetic shift of $(2.\pm0.2)$ μeV/Tesla^2 is in agreement with the results of others. An increase of total oscillator strength with H as predicted from theory has not been observed. The g-value $g=1.75\pm0.05$ results from the spin-flip Raman process for the same experimental conditions. The still remaining splitting parameter is Δ. This constant, unknown up to now, can be determined best from the fitting of the (2-2') transition for small fields, when the field energy $(g_e\mu_B H_\perp)$ is comparable to Δ. The analysis yields $\Delta=(0.2\pm0.03)$ meV. This value coincides with theoretical estimations in literature [4].

In conclusion, magneto Brillouin scattering allows the direct measurement of magneto optical parameters of free excitons. In the special case of the A-exciton of CdS the singlet-triplet splitting could be determined for the first time.

Thin Prism Refraction

Polariton dispersion curves are normally obtained by a complicated line shape analysis of experimental data and the knowledge of several parameters, gotten from other measurements.

The most direct method for the determination of the refractive index is the observation of refraction by a prism. However in the polariton region the transmission is very small, so that up to now this kind of polariton spectroscopy was not performed. Using very thin prismatic crystals with small prism angles, we have overcome this difficulty.

We report on measurements with two as-grown CdS prismatic platelets having mean thicknesses of 16 and 80 μm resp. and prism angles of 0.53 and 2.02^o. The light of a Xenon high pressure lamp passing a high resolution monochromator was focussed on a narrow spot of an optical multichannel analyser. The deflection of this light beam by a CdS prism could be measured with high precision. The real part of the refractive index, Re(n), could then easily be calculated from the geometry. The imaginary part of the refractive index, Im(n), is obtained simultaneously from the transmission which we could measure down to values of 10^{-4}.

Fig.(4) shows results of such a thin prism refraction experiment for the plane of incidence perpendicular to the c-axis. The two regions of resonance for the A- and B-exciton are clearly visible. As expected a pronounced birefringence occurs. The two branches for E$\perp$c (A- and B-polariton overlapping) could be investigated in the range $1.6<n<6.4$ ($0.6<k/k_o<2.3$), the two branches for E$\parallel$c (B-polariton) however are measurable due to higher absorption only in a smaller region ($2<n<3.5$).

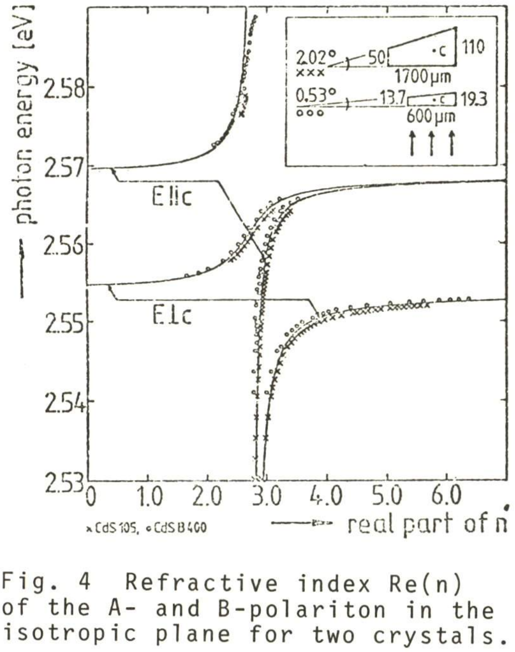

Fig. 4 Refractive index Re(n) of the A- and B-polariton in the isotropic plane for two crystals.

In the excitonic part of the dispersion curves the transmission is very small. For the A-polariton, active only for E$\perp$c, there exists the possibility to diminish the oscillator strength and to enlarge the transmission considerably by oblique incidence of light. Fig.(5) shows for this mixed mode case the real and imaginary part of n obtained simultaneously. It can be seen that for an angle of 18^o even at strongest resonance a measurable part of light is still transmitted (see Fig.5, right part). In this region a new kind of birefringence can be observed (Fig. 5, left part):

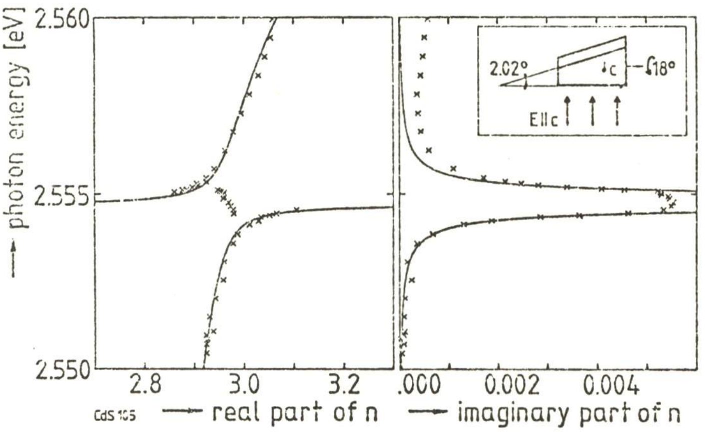

Fig. 5 Refractive indices Re(n) and Im(n) for the A-polariton in the mixed mode case

An additional wave appears having a refractive index close to that for E∥c. We believe that this phenomenon can be explained by interference effects of the two waves which are produced as a result of spatial dispersion. This opens a new way to solve the problem of additional boundary conditions in the polariton region.

The values of the refractive indices in Fig.(4) and Fig.(5) can be accurately described by the known polariton formalism [5,6] (full line).

Comparing theory and experiment the following parameters were determined:

$\omega_0^A = 2.5528$ eV, $4\pi\alpha_0^A = 0.013$, $\varepsilon_0^A = 7.6$, $m_\perp^A = 0.9\ m_0$, $\Gamma^A = 3\cdot 10^{-5}$ eV,

$\omega_0^B = 2.5680$ eV, $4\pi\alpha_0^B = 0.010$, $\varepsilon_{0\parallel}^B = 7.6$.

Most of these are consistent with values from other experiments [7]. The value of ε_0 however has to be corrected from about 8.1 to 7.6 as a consequence of our observation, because a considerable overlap of the A- and the B-polariton has now been taken into account.

In conclusion we have shown that the thin prism refraction is a powerful and yet simple method not only to obtain improved polariton parameters but also to investigate the behaviour of polaritons in general. We believe that this method will be useful for many other solids to study optical properties and their dependence on temperature, electric and magnetic fields, and on intensity, especially at high densities of excitation.

References:

1) K. Cho: Phys. Rev. B 14, 4463 (1976).
2) G. Winterling, E. Koteles: Solid State Commun. 23, 95 (1977).
3) G. Winterling, E. Koteles, M. Cardona: Phys. Rev. Lett. 39, 1286 (1977).
4) J. Voigt, F. Spiegelberg, M. Senoner: Phys. stat. sol.(b) 91, 189 (1977).
5) S. I. Pekar: Soviet Phys.-J.exper. theor. Phys. 6, 785 (1958).
6) J.J. Hopfield, D.G. Thomas: Phys. Rev. 132, 563 (1963).
7) I. Broser, M. Rosenzweig, R. Broser, M. Richard, E. Birkicht: phys. stat. sol. (b) 90, 77 (1978).