maximized and Doppler-free spectra of all three energy levels can be obtained.

The authors are indebted to Dr. S. Stenholm for stimulating our interest in this problem.

A7 TWO-PHOTON ABSORPTION MEASUREMENTS WITH ULTRASHORT BROAD BAND LIGHT CONTINUA

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A new technique of two-photon spectroscopy is described. The spectral attenuation of a broad frequency pulse is studied in the presence of a second intense monochromatic pulse. The method has several important advantages: (i) The two-photon absorption spectrum can be measured by a single light pulse. (ii) It is readily possible to distinguish between two-photon absorption and two-step transitions by adjusting the temporal overlap of the two picosecond pulses. (iii) High light intensities can be applied to the samples without material damage.

In our experiments a mode-locked Nd–glass laser was used. Single picosecond light pulses were selected from the early part of the pulse train and amplified to an energy of ~3 mJ ($\Delta t_L \simeq 6$ ps fwhm, $\Delta\tilde{\nu}_L \simeq 3$ cm^{-1} fwhm). The picosecond broad-band light continua were generated by parametric four-photon interaction of the single laser pulse in liquid water [1].

In the new technique of picosecond two-photon spectroscopy, the intense monochromatic laser pulse and the broad-band picosecond light continuum pass simultaneously through the two-photon absorbing sample. With optical multi-channel analysers the two-photon absorption spectrum was measured over a wide frequency range in a single shot.

As an example the nonlinear absorption of CdS single crystals was measured. The two-photon absorption cross-sections in the energy range between 2.4 and 3.5 eV were determined. New information is obtained of the two-photon absorption of CdS on account of the improved accuracy and higher sensitivity of our technique.

As an application of two-photon absorption we have investigated a system where the intensity of ultrashort light pulses is directly determined. The one-to-one correspondence between the energy transmission through a two-photon absorbing medium and the input peak intensity of the laser pulse was utilized to determine calibration curves for intensity determination. Experimentally straightforward energy transmission measurements are made. In a first experiment we used rutile crystals as nonlinear absorbers. The input peak intensity was determined by simultaneously measuring the energy, duration and beam diameter of the light pulses. Two-photon absorption measurements with picosecond ligth continua showed a smooth absorption behavior in the spectral range between 5300 and 6260 Å. The obtained calibration curves may be used for intensity detection of the second harmonic of mode-locked Nd–glass laser pulses and of mode-locked dye laser pulses.

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A8 LINESHAPE ANOMALY IN INFRARED-MICROWAVE DOUBLE RESONANCE

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The infrared-microwave double resonance effect has been observed in H_2CO with a Zeeman-tuned 3.51 μm He–Xe laser [1]. The effect of double resonance is observed as a variation of the infrared absorption when the microwave frequency is swept over the rotational transition of H_2CO. The observed pressure dependence of the signal shows that the inversion of the signal occurs at a certain pressure where the anomalous lineshapes are observed. Fig. 1 shows the observed lineshapes with the double resonance cell in the laser cavity. Fig. 1b de-

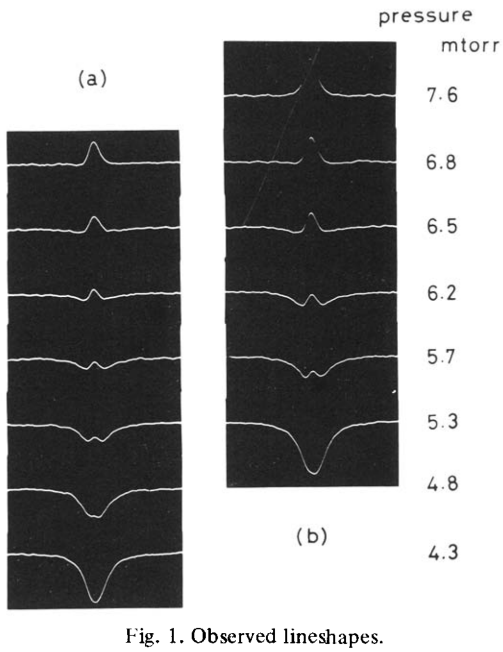

Fig. 1. Observed lineshapes.