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Coherent anti-Stokes Raman scattering spectroscope/microscope based on broadband laser source.

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Introduction

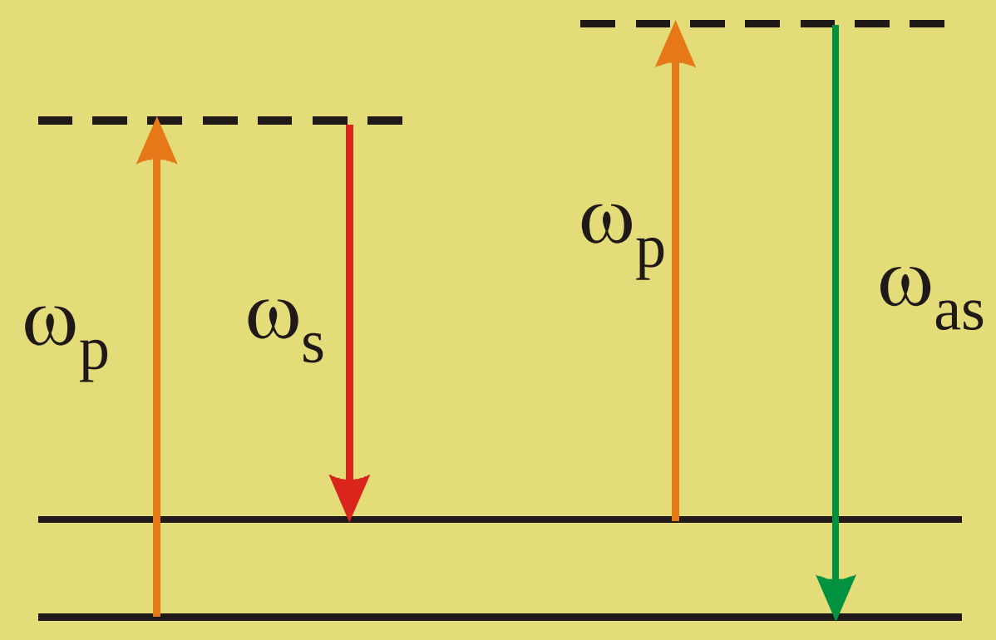

Fig. 1 Energy diagram

High contrast chemical imaging based on Coherent Antistokes Raman Scattering (CARS) phenomenon is one of advanced imaging techniques [1]. The CARS signal is obtained at frequency $\omega_{as} = 2\omega_p - \omega_S$, where ω_p and ω_S are pump and Stokes field frequencies, respectively (Fig.1). Different light sources were used in this technique from tightly synchronized two Ti:sapphire lasers to parametric optical oscillators providing both pulses [2].

Here we describe application of a compact optical parametric generator (OPG) pumped by the second harmonic of Nd: YVO_4 laser for the CARS spectrometer.

[1] Ji-Xin Cheng, A. Volkmer, and X. S. Xie, J. Opt. Soc. Am. B Vol. 19, No. 6, 1363-1375 (2002).
[2] http://bernstein.harvard.edu/research/carssources.htm

Laser

The laser consists of picosecond laser and optical parametric generator (OPG) (see Fig.2). The tuning range of the OPG signal wave extends from 860 nm to 1030 nm and correspondingly from 1100 to 1400 nm for the idler wave. This enables to probe 700 – 4500 cm^{-1} range of vibrational frequencies.

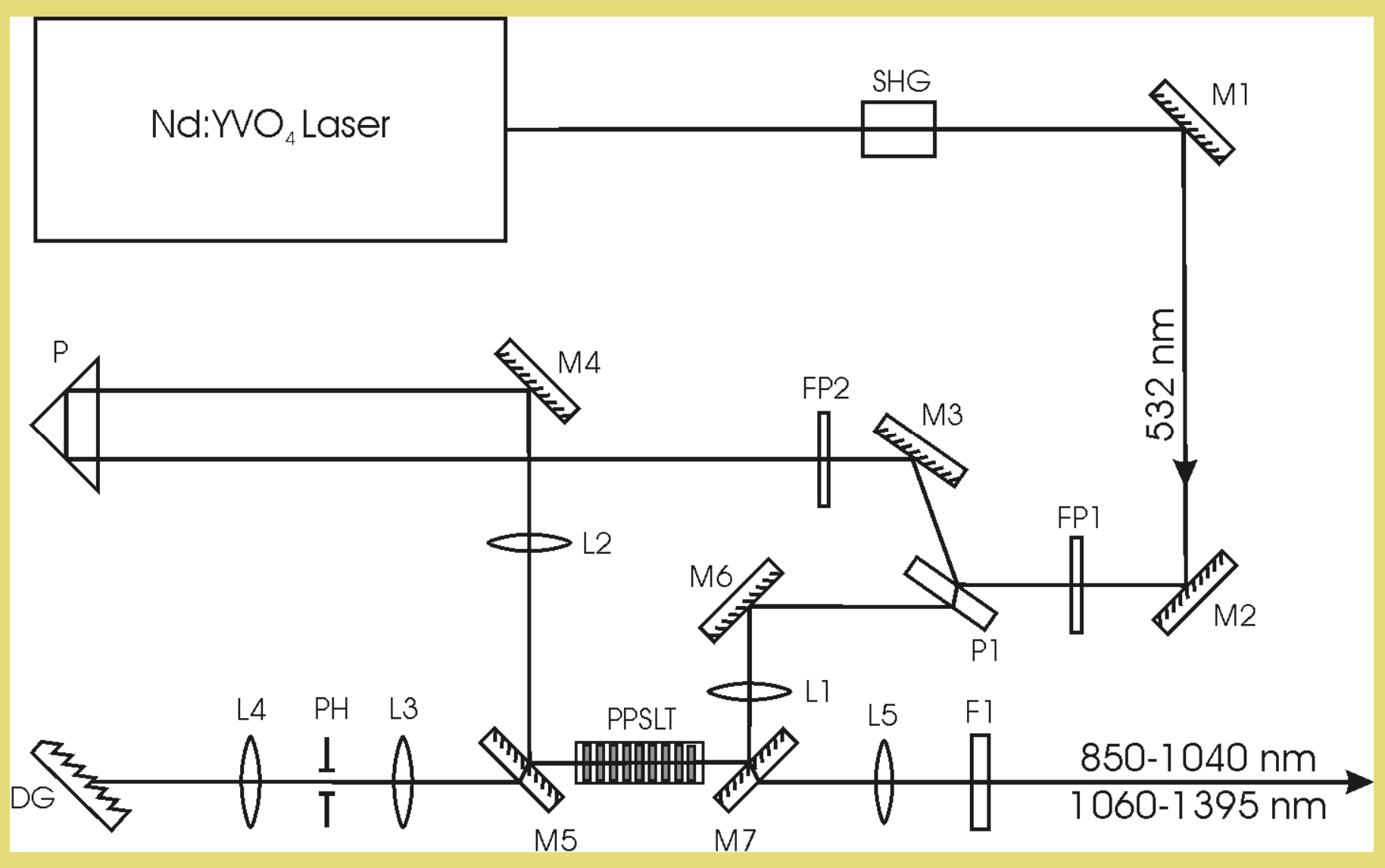

Fig. 2 Principal scheme of the parametric generator

Picosecond laser. A fiber-coupled laser diode delivers pump to the picosecond laser operating in a CW passive mode-locking mode. The cavity dumping regime is realized by means of a high speed electro-optical system. At the repetition rate of 1 MHz the output pulses of about 1 μJ energy and 1 W average power with shot to shot stability better than 1% and the pulse duration of 7 ps were obtained. The second harmonic converter provides 52% conversion efficiency.

The OPG setup utilizes a temperature tuned periodically-poled stoichiometric lithium tantalate (PPSLT) crystal. A single crystal double pass scheme in a form of a master oscillator – power amplifier has been employed. The setup was adjusted for the maximum spectral purity and beam quality. The wavelength tuning curves for both signal and idler waves are presented in Fig.3.

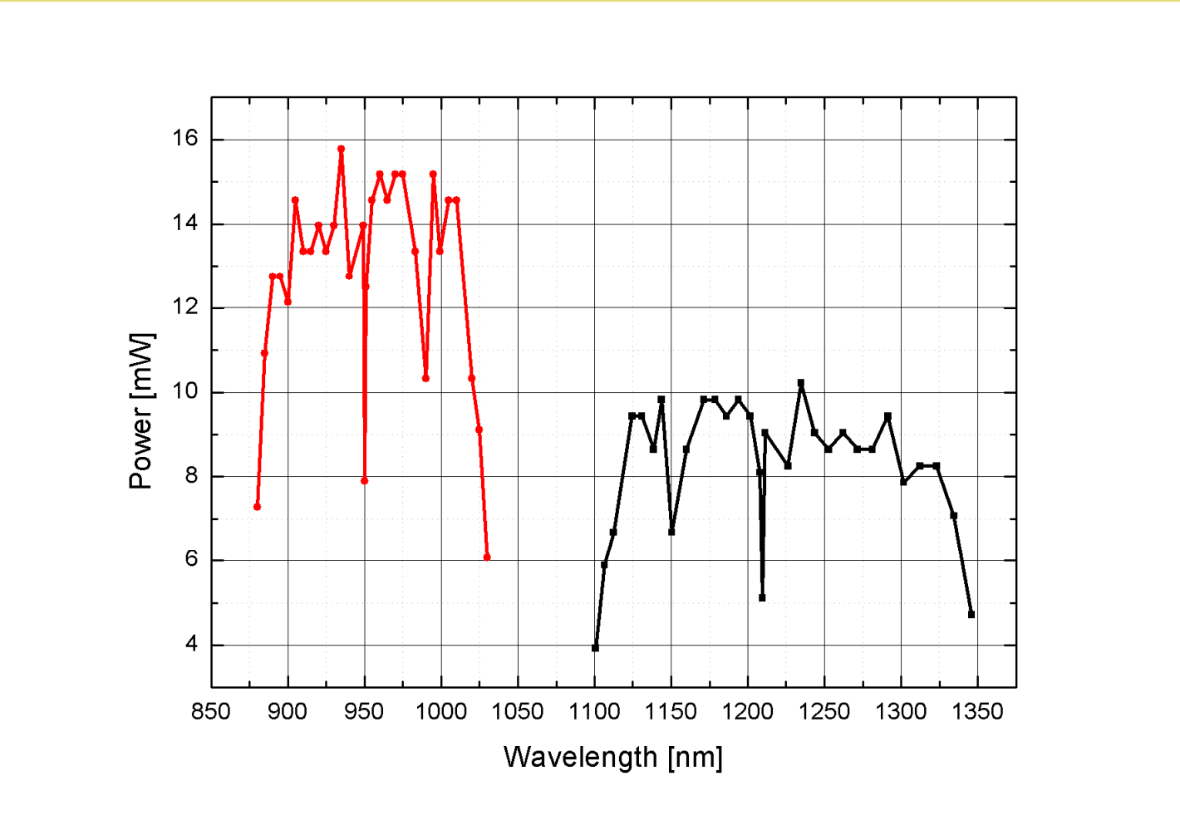

Fig. 3 Output power versus wavelength

The overall conversion efficiency is low (around 6%), but this is compensated by enhanced spectral and spatial properties of the beam. The spectrum width remains almost constant within the OPO turning range (Fig. 4). The spectral profile at one of individual wavelengths is presented in Fig.5.

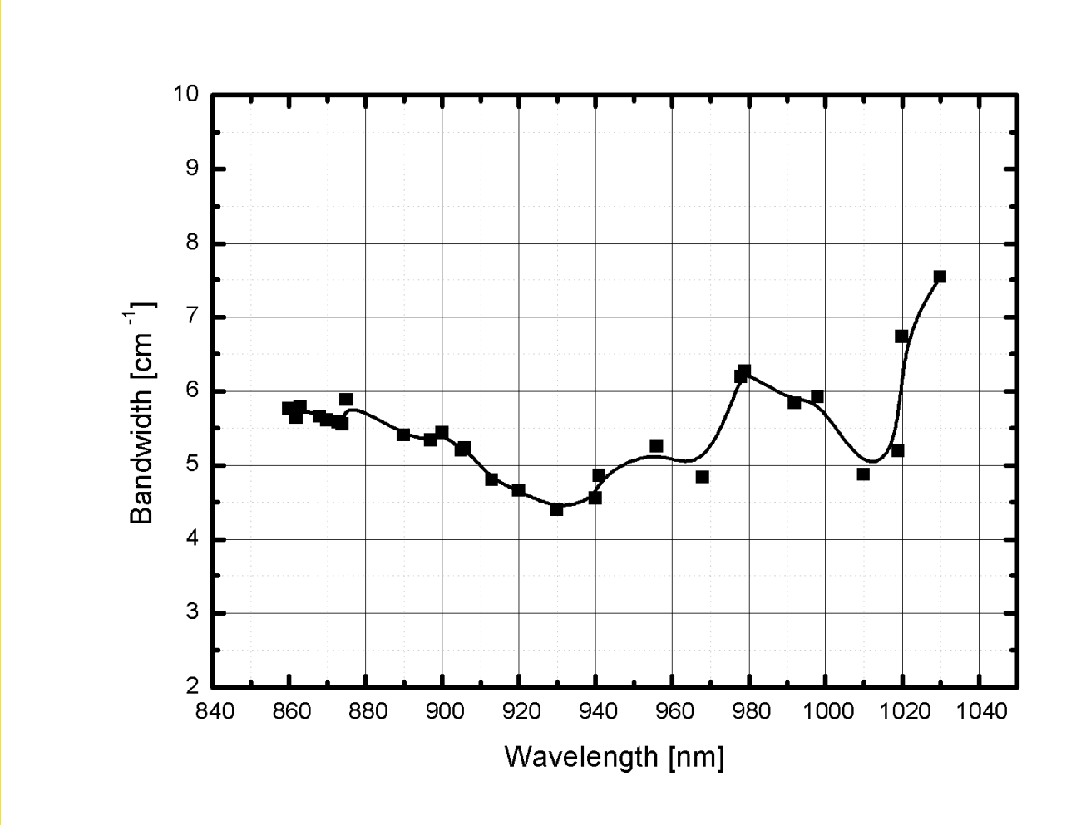

Fig. 4 Spectrum width versus wavelength

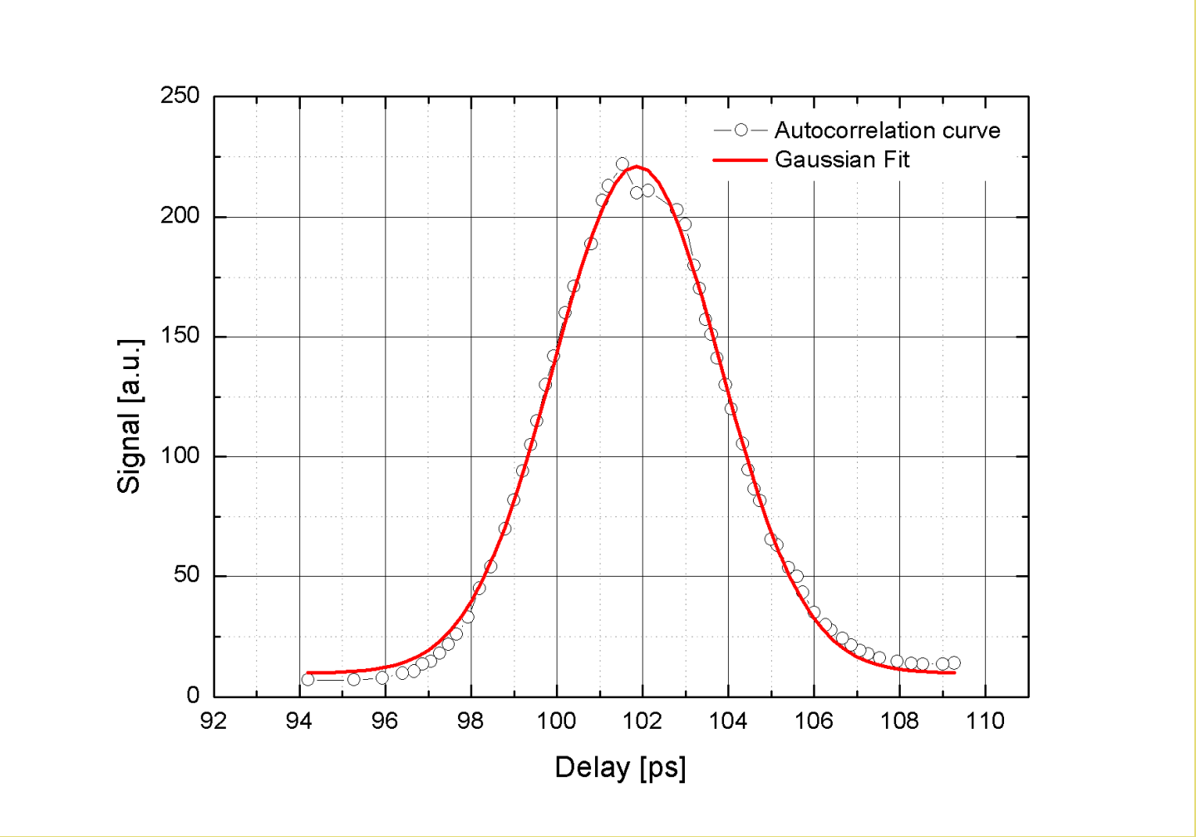

Fig. 5 Spectral profile of output pulse

From the autocorrelation trace (presented in Fig.6) we deduced the pulse length of 3.3 ps for the signal wave, assuming a Gaussian pulse shape. The pulse duration is independent of the operation wavelength. Thus, the time-bandwidth product less than 0.8 has been obtained within all the operation wavelength range with typical values of 0.6.

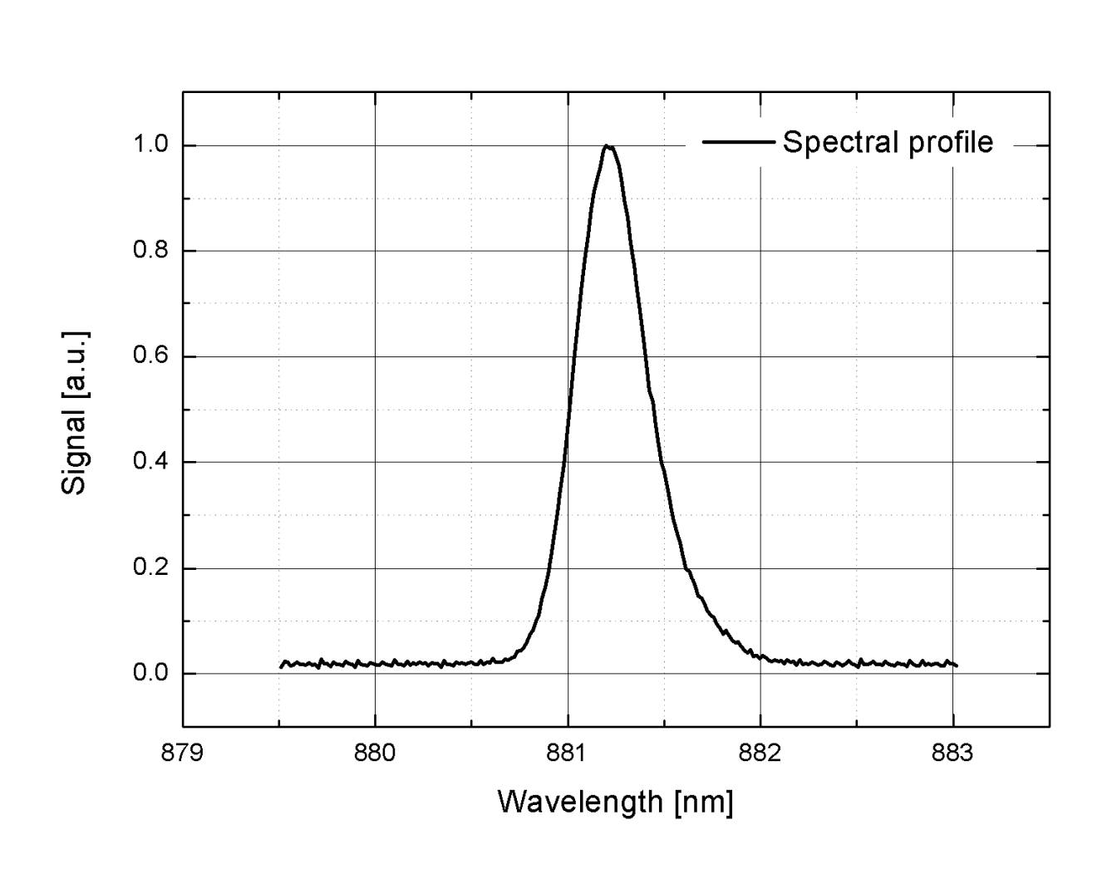

Fig. 6 Autocorrelation trace of output pulse

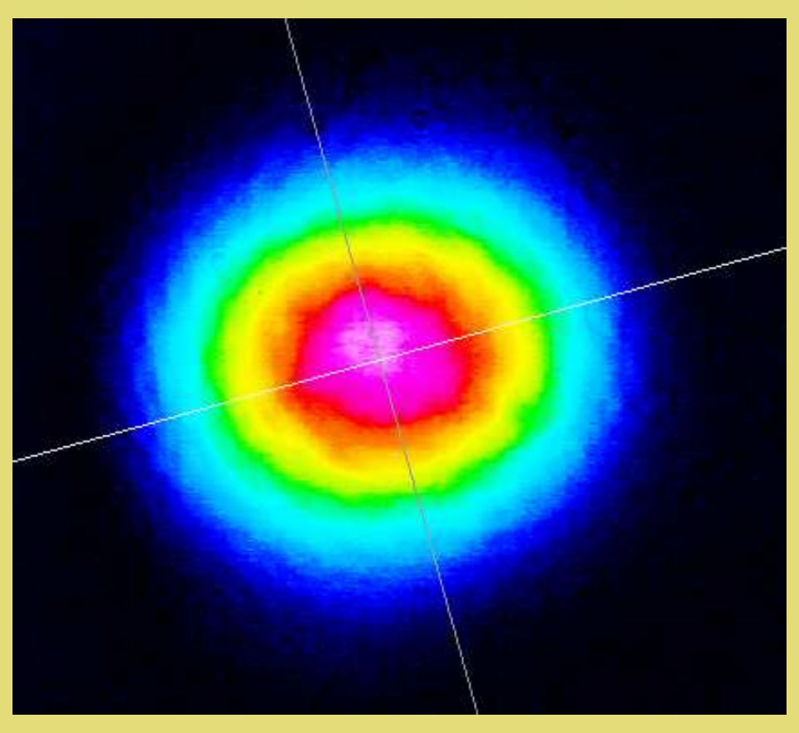

Fig. 7 Signal wave (λ=900 nm) beam profile.

CARS microscope

The CARS-microspectrometer installation was implemented in a conventional manner. Optical scheme of the CARS- microscope setup is depicted on Fig. 8.

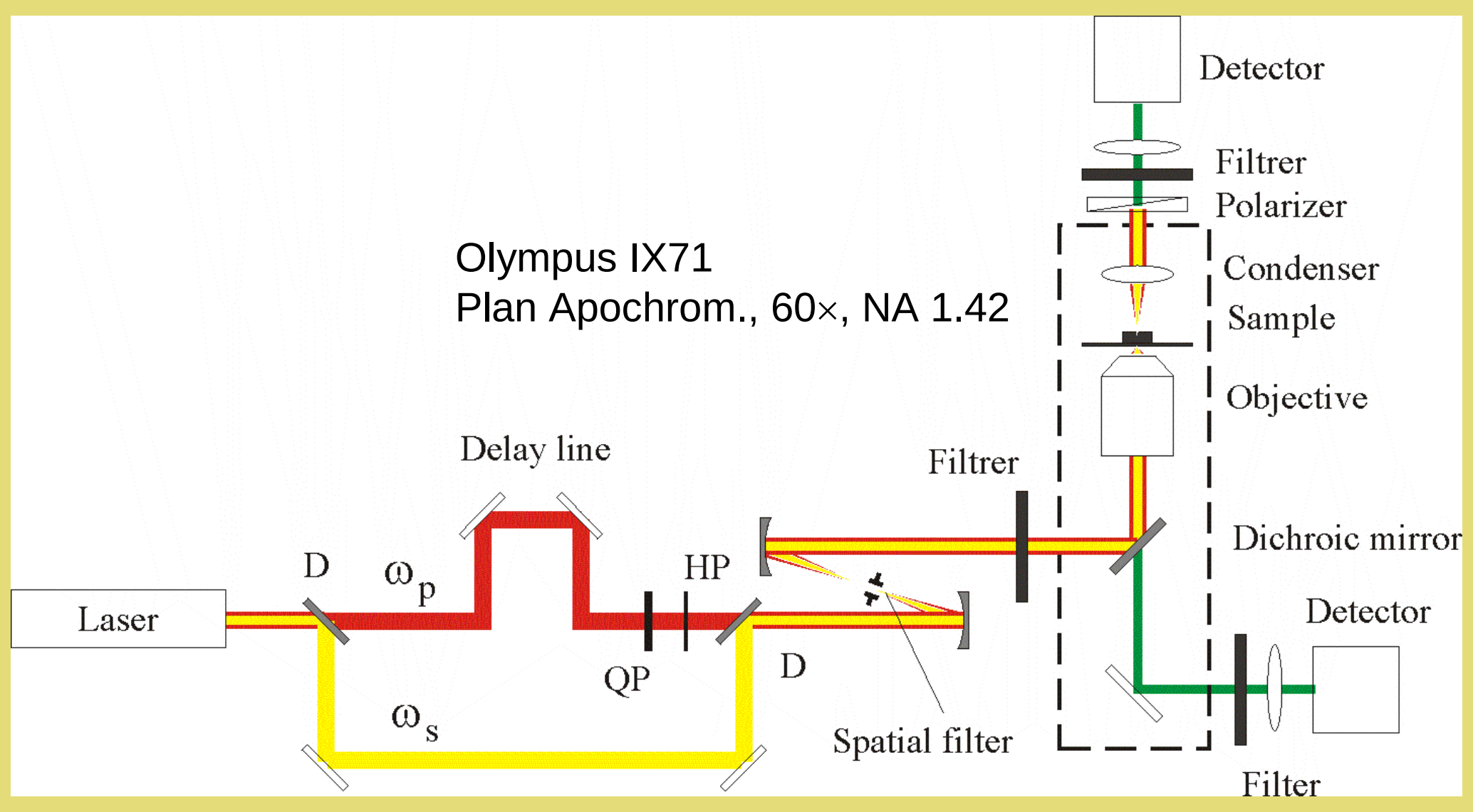

Fig. 8 Experimental setup of CARS microscope

Both epi-detection (E-CARS), when CARS radiation propagating in backward direction is detected and forward-detection (F-CARS) were implemented. In case of forward detection a P-CARS scheme, where pump and probe beams and CARS detection had some particular polarization directions was also realized in order to obtain nonresonance-CARS free spectrum. Long-pass and short-pass filters are used as a blocking tool for better spectral separation of the CARS signal. To improve the beam quality a spatial filter between laser and microscope was utilized. Collected CARS signal was detected to the avalanche photodiode (SPCM-AQRH-14), which was connected to a photon counter (Stanford Research SR400). To obtain images the sample was scanned with a piezo-driven stage (Physik Instrumente P-517.3LC). Scanning, data processing and laser wavelength tuning was controlled by computer.

Results

CARS spectroscope/microscope capabilities were tested by measuring well known polystyrene material. Fig.9 shows CARS spectrum of a single polystyrene bead of 1.1 μm in diameter measured at epi- and forward and P-CARS detection schemes. The spectrum was recorded by continuous tuning the OPG with step corresponding to the 5 cm^{-1} in Stokes frequency. The typical spectrum scan speed is about 100 cm^{-1}/100 s. The spectra obtained by all three detection schemes contain well distinguished resonance of aromatic C–H stretch at 3055 cm^{-1} and aliphatic $–CH_2$ (2840 cm^{-1}) and $–CH_3$ (2890 cm^{-1}) stretches. However, in case of forward detection a strong nonresonsnt background appears.

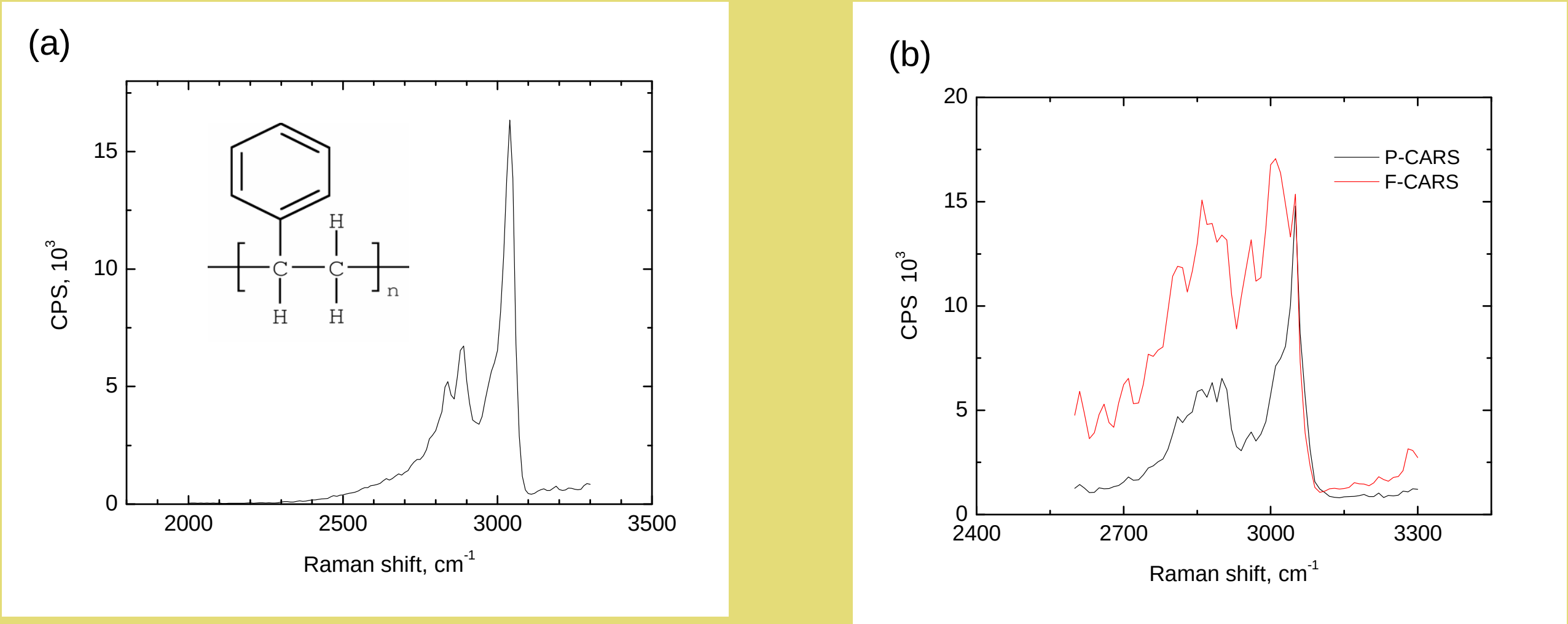

Fig. 9 E-CARS (a), F-CARS, P-CARS (b) spectra of a polystyrene bead (1.1 μm in diameter) on a coverslip. The average pump and Stokes powers were 0.26 mW and 0.6mW, respectively.

The ability to record CARS images was demonstrated by raster scanning the sample. The image was taken at a Raman shift of polystyrene aromatic C–H stretch (3055 cm^{-1}) that corresponds to the excitation wavelengths of 915.25 and 1270 nm for Pump and Stokes beams respectively. Figure 10 (a) shows an E-CARS image of group 1.1-μm-diameter polystyrene beads under glass coverslip. Additional scanning of a single bead (Fig. 10 (b)) demonstrates spatial resolution of the CARS microscope. For the tight focusing conditions the fwhm of intensity profiles across the bead are smaller than the diameter of the bead (1.1 μm) by about √2 times that reflects the quadratic dependence of signal on pump field intensity. The scanning test confirms that the laser beam quality is close to perfect providing CARS microscopy with a high 3D sectioning capability.

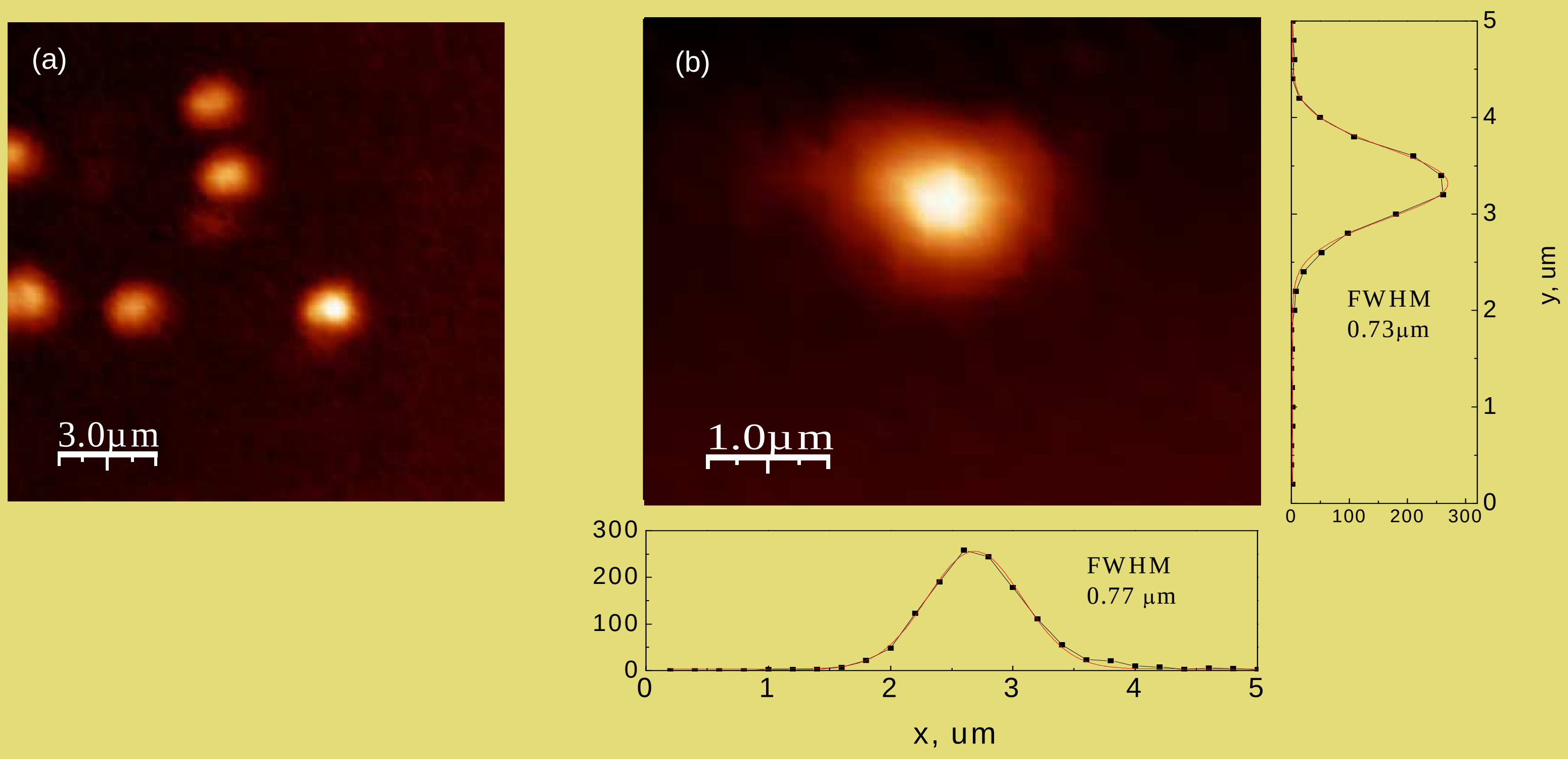

Fig. 10 E-CARS image of polystyrene beads of 1.1 μm in diameter (a). Image of a single polystyrene bead taken at tight focusing conditions (b). FWHM of lateral profiles of CARS intensity are 0.77 and 0.73 μm along the X and Y axis respectively. The average Pump and the Stokes powers were 0.3 and 0.5 mW, respectively. Images taken at the pulse repetition rate of 1MHz, and the pixel dwell time was 5.55 ms.

Acknowledgments

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