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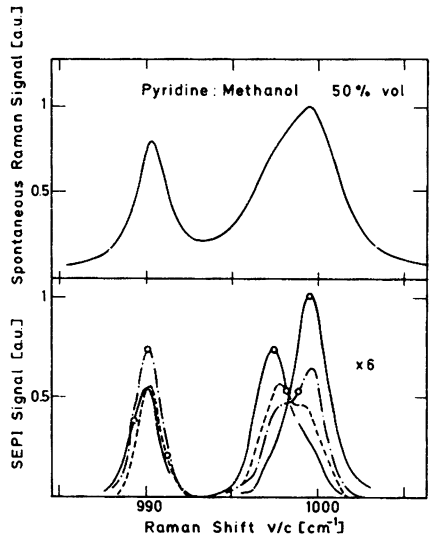

Fig. ThCC2-1. (Top) Spontaneous Raman spectrum of a mixture pyridine:methanol (50% vol), spectral resolution 0.5 cm^{-1}. (Bottom) Transient coherent Raman spectra taken at seven excitation frequencies marked by the circles.

ThCC2. Line Narrowing by Transient Coherent Raman Spectroscopy with Tunable Picosecond Excitation, W. Zinth, M. Nuss, and W. Kaiser, *Physik Department der Technischen Universität München, Arcisstrasse 21, D-8000 München 21, Federal Republic of Germany.*

Recently, a novel Raman technique was introduced based on short excitation and prolonged coherent interrogation (SEPI) of molecular transitions.[1] The experimental method allows improvement of spectral resolution beyond the limitations of spontaneous Raman spectroscopy. Up to now, discrete excitation frequencies were used, limiting wide applicability.[1,2] We present the first known data of the SEPI technique using continuously tunable excitation. The smooth tunability allows the determination of frequency positions and amplitudes of individual transitions. In congested spectral regions, new information, not available from spontaneous Raman data, is obtained.

The SEPI technique works as follows. During the transient Raman excitation process molecules are driven at the excitation frequency $\nu_E = \nu_1 - \nu_2$ by two light pulses of frequency ν_1 and ν_2. A Raman transition of frequency ν_V close to ν_E is coherently excited. At the end of the exciting process the coherent excitation oscillates at ν_V and decays with the dephasing time T_2. A third, delayed probe pulse (frequency ν_3) interacts with the coherently vibrating molecules and generates an anti-Stokes spectrum. The crucial point is the following: a narrow anti-Stokes spectrum is produced at late delay times provided a long Gaussian-shaped probing pulse is used. For pulse durations $t_p > 1.4T_2$ the SEPI spectrum is narrower than the spontaneous Raman spectrum.

The experiments are performed using two synchronously pumped dye lasers (ν_1,ν_2) excited by a mode-locked Ar^+-ion laser. The pulses from the first laser have a fixed frequency ν_1. They are used in the probing and excitation process: $\nu_3 = \nu_1$. The exciting pulses at the tunable frequency ν_2 are shorter by a factor of 2. The coherent signal is detected in a geometry for anti-Stokes phase matching.

The tunable SEPI technique is applied to the system pyridine:methanol, where strong hydrogen bonds are present. In the spontaneous Raman spectrum a band appears at 997 cm^{-1} close to the skeletal vibration of pyridine at 991 cm^{-1} [see Fig. ThCC2-1(top)]. The band at 997 cm^{-1} is broad and featureless.[3] In Fig. ThCC2-1(bottom) we show for comparison several SEPI spectra taken at a pyridine concentration of 50% by volume. The four excitation frequencies are marked by the circles. On the left-hand side we find the pyridine transition at 990.2 cm^{-1}, independent of the excitation frequency. Around 998 cm^{-1} the SEPI spectra reveal two lines that are due to hydrogen bonded aggregates. Tuning the excitation frequency we determine their frequency to $\tilde{\nu}$ = 997.3 and 999.6 cm^{-1}. A third line, found at $\tilde{\nu}$ = 1001 cm^{-1}, is not shown in Fig. ThCC2-1.

Measuring SEPI spectra for several concentrations with excitation frequencies between 985 and 1005 cm^{-1} we obtain the following picture: (a) The broad band around 998 cm^{-1} is built up by three transitions at 997.3, 999.6, and 1001 cm^{-1}. (b) The frequencies do not depend on concentration. (c) The amplitudes of the components vary with concentration allowing us to estimate the structure of the different hydrogen bonded complexes. (12 min.)

[1] W. Zinth, M. C. Nuss, and W. Kaiser, Chem. Phys. Lett. **88,** 257 (1982).
[2] W. Zinth and W. Kaiser, Proc. Third New Zealand Symposium on Laser Physics, Hamilton, in *Lecture Notes in Physics*, J. D. Harvey and D. F. Walls, eds. (Springer, New York, 1983), Vol. 182, p. 152.
[3] B. P. Asthana, H. Takahashi, and W. Kiefer, Chem. Phys. Lett. **94,** 41 (1983).

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