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Very Low Frequency Stokes and Anti-Stokes Raman Spectra Accessible with a Single Multichannel Spectrograph and Volume Bragg Grating Optical Filters

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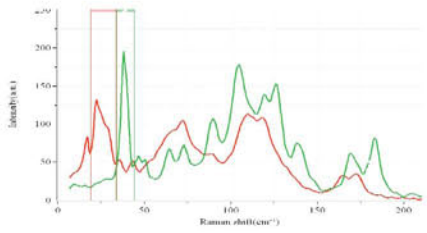
Introduction

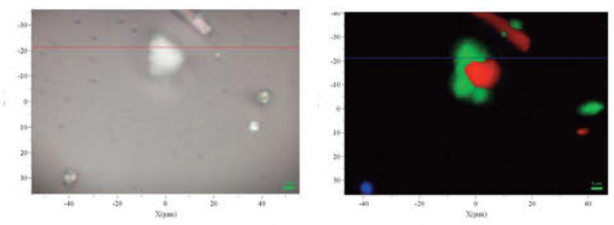
When studying Raman scattering from a material, some information can be found at very low Raman shifts (below 50 cm^{-1}).
Some chemical bonds, Longitudinal Acoustic Modes (LAM), Radial Breathing Modes (RBM) in Single Wall Carbon NanoTubes (SWCNT), Brillouin scattering… have Raman frequencies very close to the Rayleigh line.
These studies are usually done with a triple monochromator that enable proper elimination of the Rayleigh scattering while opening the measurement window very close to the shoulder of the laser (typically, 3 or 4 cm^{-1} in oblique incidence).
Progress in PhotoThermoRefractive (PTR) glass technology enable the design of low scattering, high optical density, narrow bandwidth Volume Bragg Gratings (VBG).
We show spectra obtained using a module that includes Notch VBGs in series and enables accessing very low Raman frequencies when using a single stage multichannel spectrograph (HORIBA Jobin Yvon LabRAM HR800), revealing features down to 7 cm^{-1}.

Instrument: LabRAM HR

Results

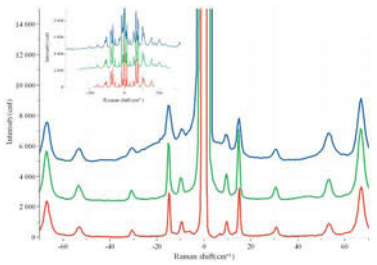
1.Chemical compounds
Carbamazepine





Mapping obtained on Raman frequencies below 50 cm^{-1}.
2 types of crystals of different chemical composition can be identified.

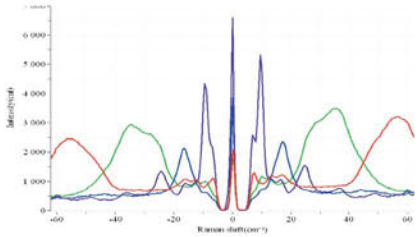
L-Cysteine excited at 488 nm, 532 nm and at 633 nm



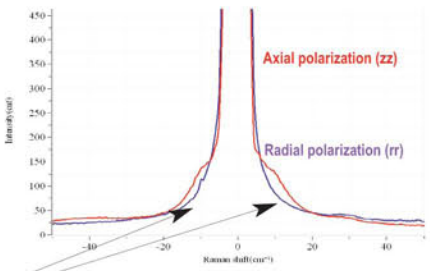
Strong points of ULF module

- Access to Raman frequencies below 10 cm^{-1}
- Stokes and Anti-Stokes frequencies readily available
- High throughput (≥ 70%)
- Unlimited lifetime
- Ease of use
- Upgrade of LabRAM HR and LabRAM ARAMIS possible

2. LAModes in Poly-Ethylene (PE)

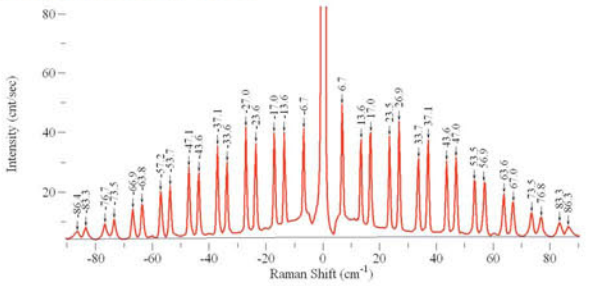


LAModes in bulk PE crystals



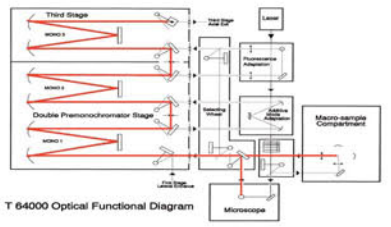
LAMode in a PE fiber

3. Ultra lattice of GeSi[a]



[a]. data courtesy of :
P. H. Tan, State Key Laboratory for SL and Microstr., Institute of Semiconductors, Beijing, 100083, P. R. China
K. Brunner, University Wuerzburg, EP 3, Am Hubland, D-97074Wuerzburg, Germany

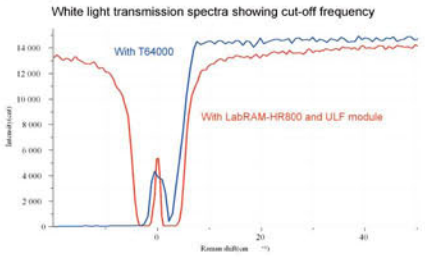
Comparison to T64000



T 64000 Optical Functional Diagram

A triple monochromator such as T64000 retains strong advantages compared to the UltraLow Frequency modules:
- It operates in the UV region
- It is continuously tunable over a very wide range (200 to 1600 nm)
- Its cutoff is even lower than the one achieved with VBGs as seen in the white light transmission spectra. This enables measurement of Brillouin scattering frequencies.

White light transmission spectra showing cut-off frequency



Conclusion

The module developed is designed for specific wavelengths. Currently, 785 nm, 633 nm, 532 nm and 488 nm have been tested.
Easy access to both Stokes and Anti-Stokes Raman signal is now enabled down to very low frequencies on a single stage multichannel spectrometer with the ULF module. The transmission window of VBG filters extends to above 2.5 micron, so the ULF accessory opens the possibility to make simultaneous measurement of low frequency Raman as well as photoluminescence signal.
Diode lasers require perfect filtering to avoid influence from the laser sidebands so Raman spectra excited at 785 nm below 20 cm^{-1} were not achieved with the laser source available at the time. Additional work is in progress to provide a solution in the short term.
Users who have applications that require access to even lower Raman frequencies, multiple excitation wavelengths, or tunability of the excitation wavelength (for resonance studies for example) are still better fitted with the use of a **Triple monochromator** such as the **HORIBA Jobin-Yvon** T64000.

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