Shaped Femtosecond Pulses for Remote Chemical Detection

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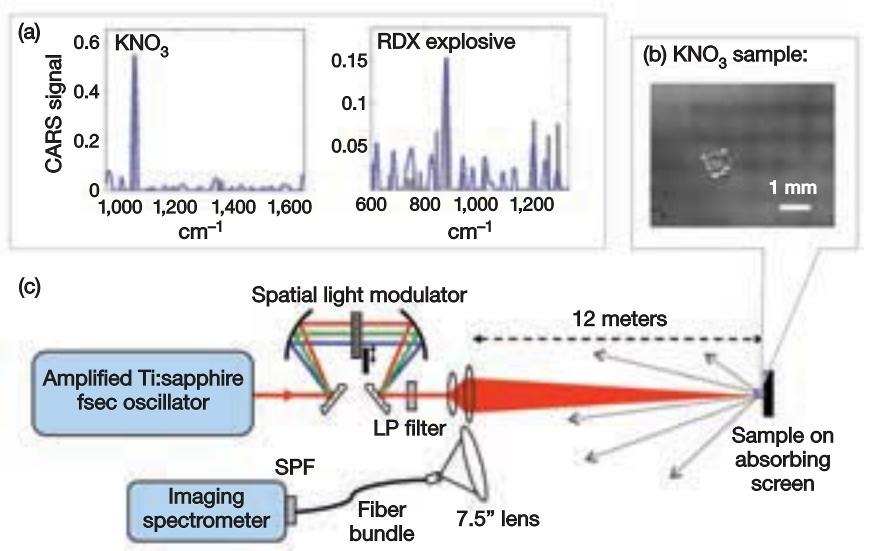
The vibrational spectrum of molecules can be used to detect hazardous materials at standoff distances. One of the best optical pathways for revealing a molecule's vibrational spectrum is through coherent anti-Stokes Raman scattering (CARS) nonlinear spectroscopy. In CARS, a molecule's vibrational modes are excited into motion by the simultaneous interaction of pump and Stokes photons. This excitation is subsequently probed by a narrow-frequency probe beam, and the vibrational level energies are resolved by measuring the amount of blue-shift of the scattered anti-Stokes photons.

CARS has become the method of choice in nonlinear optical spectroscopy and microscopy. However, due to its multi-wavelength nature, it is typically carried out as a multi-beam, multi-source technique. The need to maintain spatial and temporal overlap of the beams set hurdles on its use as an applicable tool outside the laboratory.

We recently reported our demonstration of a single-beam coherently controlled femtosecond pulse CARS technique for remote detection and identification of minute amounts of solids and liquids at a standoff (>10 m) distance.[1] We have succeeded in rapidly resolving the vibrational spectrum of trace amounts of contaminants, such as explosives and nitrate samples, from the weak backscattered photons under ambient light conditions. In our single-beam technique, we carried out the multiplex measurement of characteristic molecular vibrations using a single broadband phase-shaped femtosecond laser pulse, which supplies the pump, Stokes and probe photons simultaneously.[2,3]

Naturally, the wide bandwidth of the femtosecond pulse limits the spectroscopic resolution. However, through careful spectral-phase shaping, we selected a narrowband probe within the wide pulse spectrum and maintained a spectroscopic resolution more than 30 times better than the pulse bandwidth. Li et al. reported similar findings using spectral polarization-shaping.[4]



(a) Resolved femtosecond CARS vibrational spectra for two trace samples obtained at a standoff distance of 12 m (<1000 μg crystallized KNO_3 particle and RDX explosive particles with a total mass of <4 mg). Each spectrum was resolved from a single measurement with an integration time of <3 s. The known vibrational lines are plotted in gray bars. (b) The KNO_3 contaminant sample, placed 12 m from the measuring system. (c) The experimental setup. The laser source is an amplified femtosecond Ti:sapphire laser (0.5 mJ, 30 fs, 1 kHz repetition rate). Pulses are shaped using an electronically controlled liquid-crystal spatial light modulator. The beam is focused on a distant sample through a telescope, and the backscattered radiation is collected to a spectrometer with a 7.5" diameter lens. (LP/SPF—long/short pass filter.)

A main difficulty in using ultrashort, high-peak-power pulses for nonlinear spectroscopy arises from the strong nonresonant four-wave-mixing signal, which in most cases obscures the vibrationally resonant CARS signal. In our phase-contrast technique, instead of struggling to eliminate this strong nonresonant background, we harnessed its coherent nature to amplify the weak, backward-scattered vibrationally resonant radiation by interfering them together coherently. The combination of the considerable amplification in resonant signal and the polarization insensitivity within a multiplex single-beam scheme make it attractive for standoff probing applications.

Furthermore, femtosecond CARS spectroscopy exhibits higher efficiency at low average powers compared to the longer (ns) pulses used in conventional CARS techniques—a merit for the nondestructive probing of sensitive samples such as explosives. ▲

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