Air lasing: A new tool for atmospheric detection
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Experimental setup. M1, M2: reflective mirror; L1-L5: lens; SP: sapphire plate; BBO: beta barium borate; DM1-DM3: dichroic mirror; GT: Glan-Taylor prism; F1: narrowband filter with the central wavelength of 428 nm and the bandwidth of 1 nm; F2: combination of variable filters for recording the Raman signal at different wavelengths. Schematic diagram of the polarization states and time sequences of pump, seed and air lasing is shown in inset. Credit: Ultrafast Science (2022). DOI: 10.34133/2022/9761458

Ultrafast laser technologies provide new strategies for remote sensing of atmospheric pollutants and hazardous biochemical agents due to their unique advantages of high peak power, short pulse duration and broad spectral coverage.

Particularly, air lasing shows promise in atmospheric remote sensing due to its ability to generate cavity-free light amplification in the open air. It is suitable as a probe for atmospheric diagnosis.

Recently, a research team from the Shanghai Institute of Optics and Fine Mechanics (SIOM) of the Chinese Academy of Sciences (CAS) proposed an air-lasing-assisted coherent Raman spectroscopy, which realizes quantitative measurement and simultaneous detection of two greenhouse gases, as well as identification of CO₂ isotopes. The detection sensitivity reaches 0.03% and the minimum signal fluctuation is about 2%.

The work was published in Ultrafast Science on April 8.

The extremely nonlinear interaction of the femtosecond laser with air molecules excites the optical gain of molecular nitrogen ions and achieves a seed amplification of more than 1,000 times, resulting in 428 nm air lasing with a linewidth of 13 cm⁻¹.

Meanwhile, the spectral width of the pump laser has reached 3800 cm⁻¹ after nonlinear propagation, which is more than one order of magnitude broader than the spectrum of the incident laser.

It thus enables the excitation of the molecular coherent vibrations of most pollutants and greenhouse gases. When air lasing encounters coherently vibrating molecules, it will effectively produce coherent Raman scattering. By recording the frequency difference of Raman signal and air lasing, namely the Raman fingerprint, the molecular identity information can be determined.

Air-lasing-assisted coherent Raman spectroscopy combines the advantages of femtosecond laser and air lasing. Femtosecond laser has a broad spectral coverage and a short pulse duration, which can excite coherent vibrations of many molecules at the same time. Air lasing has a narrow spectral width, enabling the distinguishing of the Raman fingerprints of different molecules. Therefore, this technique can meet the needs of multi-component measurement and chemical specificity.

Furthermore, the researchers demonstrated that the technique can be applied for multi-component simultaneous measurement and distinguishing ¹²CO₂ and ¹³CO₂. The simultaneous measurement of various pollutants and greenhouse gases as well as the detection of CO₂ isotopes are of great significance for tracing the sources of air pollution and studying the carbon cycling.

However, for realistic application of trace gas
remote detection, it is necessary to improve the detection sensitivity to the ppm or even ppb level, as well as extend the detection distance from the laboratory scale to the kilometer scale.