New laser-based instrument designed to boost hydrogen research

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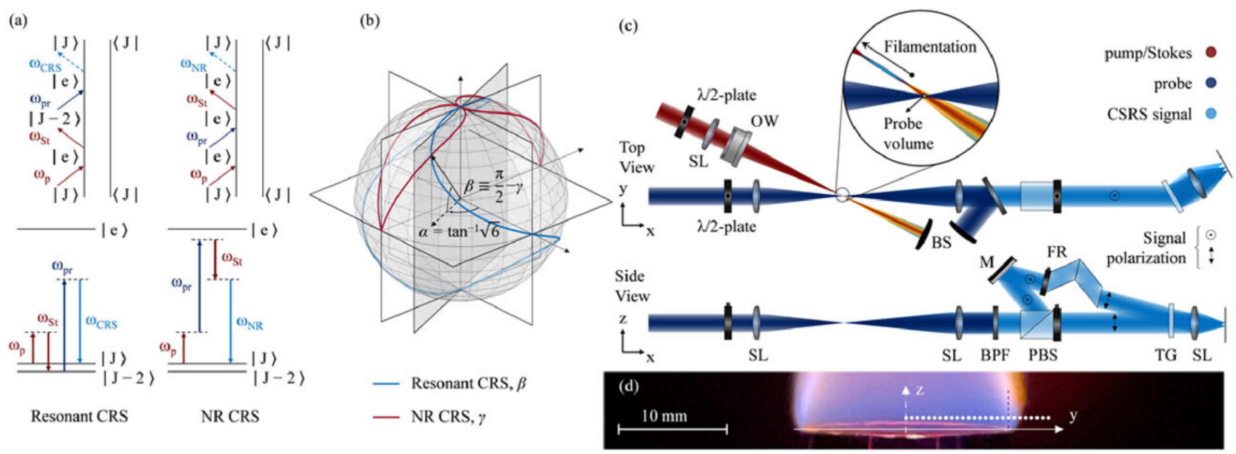

(a) Energy and Feynman diagrams of a resonant (left) and a non-resonant (right) CRS pathways. (b) Polarization angles of the resonant (blue line) and non-resonant (red line) CRS signals, β and γ, represented as the elevation angle on the unit sphere as a function of the relative polarization angle (azimuthal angle) of the pump/Stokes and probe fields, α. (c) Schematic of the polarization-sensitive coherent imaging spectrometer. OW, optical window; SL, spherical lenses; M, mirror; BPF, band-pass filter; PBS, polarization beam splitter; FR, Fresnel rhomb; BS, beam stop. Inset: probe volume. The probe crosses the ultrabroadband pump/Stokes beam ~2mm after the end of the filament. The increment of input energy results in the elongation of the filament towards the focusing optics (arrow direction) (d) Measurements points across the H2/air flame front, the dashed red line identifies the location of the burner rim at y = 9.5 mm. Credit: *Optics Express* (2022). DOI: 10.1364/OE.465817

Researchers have developed an analytical instrument that uses an ultrafast laser for precise temperature and concentration measurements of hydrogen. Their new approach could help advance the study of greener hydrogen-based fuels for use in spacecraft and airplanes.

"This instrument will provide powerful capabilities to probe dynamical processes such as diffusion, mixing, energy transfer and chemical reactions," said research team leader Alexis Bohlin from Luleå University of Technology in Sweden. "Understanding these processes is fundamental to developing more environmentally friendly propulsion engines."

In *Optics Express*, Bohlin and colleagues from Delft University of Technology and Vrije Universiteit Amsterdam, both in the Netherlands, describe their new coherent Raman spectroscopy instrument for studying hydrogen. It was made possible due to a setup that converts broadband light from a laser with short (femtosecond) pulses into extremely short supercontinuum pulses, which contain a wide range of wavelengths.

The researchers demonstrated that this supercontinuum generation could be performed behind the same type of thick optical window found on high-pressure chambers used to study a hydrogen-based engine. This is important because other methods for generating ultrabroadband excitation don't work when these types of optical windows are present.

"Hydrogen-rich fuel, when made from renewable resources, could have a huge impact on reducing emissions and make a significant contribution to alleviating anthropogenic climate change," said Bohlin. "Our new method could be used to study these fuels under conditions that closely resemble those in rocket and aerospace engines."

Getting light in

There is much interest in developing aerospace engines that run on renewable hydrogen-rich fuels. In addition to their sustainability appeal, these fuels have among the highest achievable specific impulse—a measure of how efficiently the chemical reaction in an engine creates thrust. However, it has been very challenging to make hydrogen-based chemical propulsion systems reliable. This is because the increased reactivity of hydrogen-rich fuels substantially changes the fuel mixture combustion properties, which increases the flame temperature and decreases ignition delay times. Also, combustion in rocket engines is generally very challenging to control because of the extremely high pressures and high temperatures encountered when traveling to space.

"The advancement of technology for sustainable launch and aerospace propulsion systems relies on a coherent interplay between experiments and modeling," said Bohlin. "However, several challenges still exist in terms of producing reliable quantitative data for validating the models."

One of the hurdles is that the experiments are usually run in an enclosed space with limited transmission of optical signals in-and-out through optical windows. This window can cause the supercontinuum pulses needed for coherent Raman spectroscopy to become stretched out as they go through the glass. To overcome this problem, the researchers developed a way to transmit femtosecond pulsed laser through a thick optical window and then used a process called laser induced filamentation to transform it into supercontinuum pulses that remain coherent on the other side.

Studying a hydrogen flame

To demonstrate the new instrument, the researchers set up a femtosecond laser beam with the ideal properties for supercontinuum generation. They then used it to perform coherent Raman spectroscopy by exciting hydrogen molecules and measuring their rotational transitions. They were able to demonstrate robust measurements of hydrogen gas over a wide range of temperatures and concentrations and also analyzed a hydrogen/air diffusion flame similar to what would be seen when a hydrogen-rich fuel is burned.

The researchers are now using their instrument to perform a detailed analysis in a turbulent hydrogen flame in hopes of making new discoveries about the combustion process. With a goal of adopting the method for research and testing of rocket engines, the scientists are exploring the limitations of the technique and would like to test it with hydrogen flames in an enclosed slightly pressurized housing.

More information: Francesco Mazza et al, Coherent Raman spectroscopy on hydrogen with in-situ generation, in-situ use, and in-situ referencing of the ultrabroadband excitation, *Optics Express* (2022).
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