Generation of femtosecond vacuum-ultraviolet optical pulses by four-wave Raman mixing in molecular hydrogen.

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Introduction

A femtosecond laser emitting in the deep-ultraviolet (DUV) and the vacuum-ultraviolet (VUV) regions has attracted many researchers' attention due to a variety of applications. In fact, numerous ultrafast reactions and dynamics can be studied using a laser with a pulse width shorter than their processes. Especially, such an ultrashort DUV-VUV optical pulse can successfully be used in mass spectrometry (MS) as an ionization source, since many organic molecules have strong absorption bands below 200 nm. Ionization of these molecules using single- or even multi-photons process would prove itself as a useful means for practical trace analysis.

In this study, we report herewith an optical system based on four-wave Raman mixing (FWRM) to generate multiple-color emission extending from the near-infrared (NIR) to the VUV region.

Experiment

As shown in Fig. 1, the fundamental beam of a Ti:sapphire laser (800 nm, 35 fs, 1 kHz, 3.9 mW, Elite, Coherent) was introduced into an optical parametric amplifier (OPerA, Coherent) to generate a femtosecond laser emitting at 1200 nm. A remaining part of the Ti:sapphire laser emitting from the OPA was split into two parts. One beam with an output power of 1.6W was used for fourth harmonic generation (FHG) by means of three BBO crystals (NC1-3) and two optical delay lines consisting of 8 dichroic mirrors (DM1-4 HR267 nm; DM5-8 HR800 nm). The dispersion and the polarization are compensated by using a calcite plate (DP) and two half-wave plates (HWP).

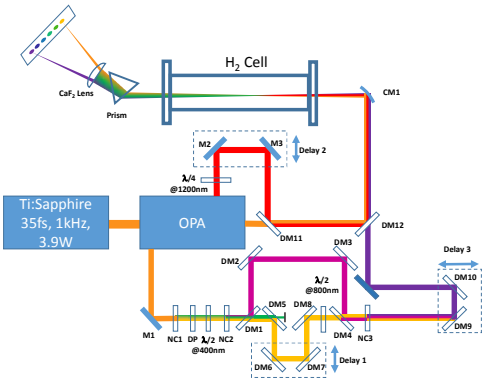

Figure 1: *Experiment configuration*

Two pump beams (800 nm and 1200 nm) are overlapped to each other in time and space using M2 and M3. An UV-enhanced concave mirror (CM1, f = 500 mm) was employed to focus the beams into a capillary waveguide in the Raman cell filled with a hydrogen gas. The time delay was carefully optimized by adjusting delay 2 to obtain the highest intensity ratio between the first anti-Stokes (601 nm) and the fundamental beams (800 nm). The probe beam was, then, introduced into the waveguide in the gas cell. Delay 3 was used to control the temporal overlap of the pump and probe pulses. Using a combination of a UV-grade fused-silica prism and a CaF_2 lens (f = 180), a spectrograph was obtained by projecting the beam onto a white paper with no coating.

Results and discussion

Figure 2 shows the spectrograph observed in this study: this picture was taken by attenuating the intensity of the strong emission and the individual brightness of the beam does not reflect the intensity of the Raman emission. The VUV Raman emission (the first anti-Stokes emission) was clearly observed at 185 nm, although it is strongly attenuated by passing it through an ambient air: oxygen in the air absorbs the light significantly at below 200 nm. For the evaluation of the light intensities for anti-Stokes beams, it is required to use a VUV monochromator equipped with a photomultiplier with sufficient spectral response in the VUV region. In principle, however, the intensity of the anti-Stokes beam (185 nm) should be identical to that of the Stokes beam (218 nm), suggesting a sufficient efficiency for the generation of the anti-Stokes beam. As demonstrated, a technique of FWRM would provide an ultrashort VUV pulse, which could be used as a light source in mass spectrometry for monitoring of numerous pollutants in the environments.

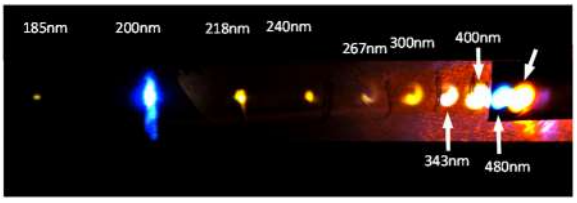

Figure 2: *FWRM lines ranging from 185nm to 601nm*