Ultrabroadband infrared pump-probe spectroscopy using chirped-pulse upconversion

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Abstract: We have demonstrated infrared pump-probe spectroscopy using chirped-pulse upconversion with a nonlinear mixing in a gas. Ultrafast dynamics of free carrier in Ge was clearly observed in the range 200–5000 cm^{-1}.

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Ultrabroadband mid-infrared (MIR, 500–4000 cm^{-1}) coherent light is highly attractive for studies in molecular science and semiconductor physics since number of molecular vibrations and intraband transition of free carriers have resonance in this wavelength region. Such a light source has been provided with nonlinear frequency conversion through two-color filamentation in gases [1]. Recently, the light source have been used for a femtosecond pump-probe spectroscopy [2].

Single-shot detection of MIR spectra with a dispersive spectrometer is essential for fast acquisition at the pump-probe spectroscopy. However, one has to suffer from low pixel count, reduced sensitivity, and high cost of multichannel infrared detectors. Chirped-pulse upconversion (CPU) is a technique to solve the problem. A MIR pulse is converted to visible with a nonlinear frequency mixing between the MIR pulse and a chirped pulse, and the visible spectra are detected with a high quality optical multichannel analyzer [3]. Very recently, Ultrabroadband detection of infrared spectra with CPU has been demonstrated by using a gas medium for the frequency conversion [4].

In this contribution, we report an application of the ultrabroadband CPU to infrared pump-probe spectroscopy for the investigation of ultrafast carrier dynamics in Ge (germanium). We have succeeded in measuring free carrier dynamics of Ge in an entire MIR range (200–5000 cm^{-1}).

The ultrabroadband infrared pump-probe spectroscopy with CPU was realized with the system shown in Fig. 1(a). The light source was based on a Ti:sapphire multi-pass amplifier system (800 nm, 30 fs, 0.85 mJ at 1 kHz, Femtopower compactPro, FEMTOLASERS). The output pulse was split into three, the first pulse was for infrared generation, the second pulse was the pump pulse for the pump-probe spectroscopy, and the third pulse was used as a chirped pulse.

The ultrabroadband infrared pulse for the probe pulse at the pump-probe spectroscopy was generated by using four-wave mixing of fundamental and second harmonic of Ti:sapphire amplifier output through filamentation in nitrogen.

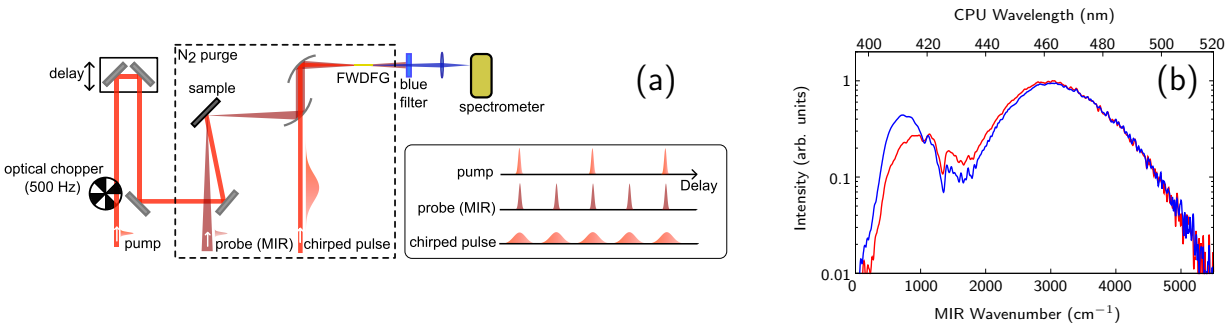

Fig. 1. (a) Schematic illustration of the experimental setup. (b) Typical upconverted spectra of the MIR pulse reflected by the sample (Ge) with (blue curve) and without (red curve) the pump pulse, respectively.

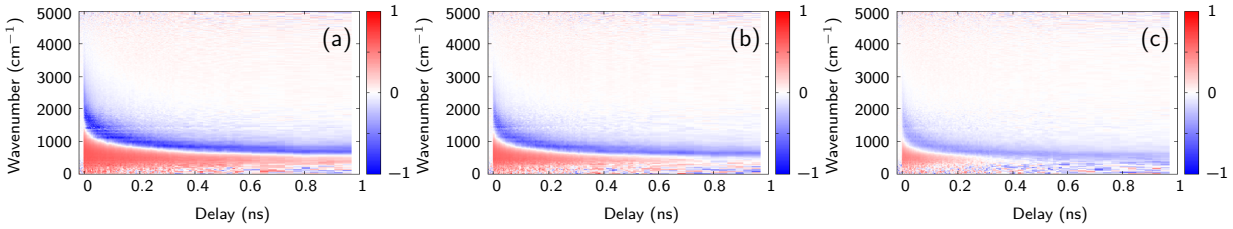

Fig. 2. Ultrafast reflectivity-change signal $\Delta R/R$ of Ge under variable excitation density: (a) 202 μJ/cm^2 (b) 135 μJ/cm^2 (c) 67 μJ/cm^2.

The pulse duration and spectral range of the infrared pulse were 7 fs and 200–5000 cm^{-1}, respectively. The infrared pulse was reflected by several substrates coated with indium tin oxide (t=300 nm) to reduce the residual visible beam along the infrared pulse. The infrared pulse was focused onto a sample, a Ge substrate, by using a concave mirror (f=0.75 m). The diameter of the infrared beam on the sample was 0.4 mm.

The pump pulse (800 nm, 30 fs) was collimated down to 4 mm diameter and overlapped to the infrared pulse on the sample. For shot-to-shot data acquisition of reflectivity-change signals, every second pump pulse was blocked by using a mechanical chopper, which is synchronized with a half-frequency of the repetition rate of the laser pulse train.

The third beam was chirped by passing through several dispersive media, BK7 and ZnSe substrates. The pulse duration of the chirped pulse became ∼0.4 ps, which corresponds to the frequency resolution of 41 cm^{-1} at the upconversion spectroscopy. The chirped pulse was combined with the infrared pulse reflected by the sample through a mirror with a hole. The combined beam was focused into nitrogen with a parabolic mirror (f = 50 mm) and upconverted into a visible beam (ω_2, 400–500 nm) through four-wave difference frequency generation (FWDFG, $\omega_1 + \omega_1 - \omega_0 \rightarrow \omega_2$) of the chirped pulse ($\omega_1$) and the infrared pulse (ω_0).

The FWDFG signal generated at a fixed delay between the infrared and chirped pulses was sent to a ∼ 0.3 m focal length spectrometer with a 300 grooves/mm grating in combination with a 1600×400 pixel EMCCD camera (SP-2358 with ProEM+1600, Princeton Instruments). The upconverted spectrum was obtained by accumulated 100 pairs of with/without-pump infrared spectra at each delay time between the pump and probe pulses to achieve a reasonable signal-to-noise ratio. Figure 1(b) shows a pair of with/without-pump infrared spectra after removing cross-phase modulation due to CPU [5].

Figure 2 shows the ultrafast reflectivity-change signal, $\Delta R/R$, of Ge for the excitation density of 202 μJ/cm^2(Fig. 2(a)), 135 μJ/cm^2(Fig. 2(b)) and 67 μJ/cm^2(Fig. 2(c)). One trace can be obtained within 20 minutes. The positive and negative reflectivity-changes correspond to the bleaching of the phonon absorption (around 300 cm^{-1}) and induced free carrier absorption (1000–5000 cm^{-1}), respectively. These signals were detected in this region at the same time by using the ultrabroadband infrared pump-probe spectroscopy. Decay time of the signal due to the phonon absorption becomes shorter with lower excitation density, while that of the free carrier absorption does not depend on the excitation density. At the conference, we plan to show the detail of the dynamics.

References

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