Systematic study of broadband terahertz gas sensor

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Broadband terahertz wave detection through field-induced second harmonic generation was systematically investigated using selected gases. The dependences of the detected second harmonic intensity on probe pulse energy, bias field strength, gas pressure, and third order nonlinear susceptibility are systematically investigated with xenon, nitrogen, SF₆, and alkanes. Experiment results reveal that the detected second harmonic intensity quadratically depends on the third order nonlinear susceptibility of the gas. Two orders of magnitude enhancement in the dynamic range of broadband terahertz wave detection are observed with alkane gas $C_4H_{10}$ sensor. © 2008 American Institute of Physics. [DOI: 10.1063/1.3056119]

The intense terahertz wave generated from laser-induced plasma in the gaseous medium provides a promising broadband terahertz source for sensing and imaging techniques. However, simultaneously coherent and broadband detection remains challenging. Work with EO crystals and photoconductive antenna detectors has been reported to reach frequencies above 20 THz, but there are some gaps in their spectra due to phonon resonance. Different from solid state sensors, gaseous media have no phonon resonances, only relatively weak vibrational/rotational resonances in molecular gases, exhibit much lower dispersion, and are continuously renewable. Although broadband terahertz wave detection has been demonstrated using ambient air, the dynamic range in these earlier experiments was limited by laser noise and detector background noise.

Terahertz detection using a gaseous medium is governed by a third order nonlinear process: generating second harmonic photons when mixing one terahertz photon and two fundamental photons together. Initially, quasicoherent terahertz detection through gaseous media was reported via terahertz field-induced second harmonic generation in laser-induced air plasma with ultrashort laser pulses. Recently, heterodyne terahertz wave detection has been demonstrated to improve the detection in the coherent region by introducing a modulated local bias-induced second harmonic oscillator. However, the efficiency of second harmonic generation in this method is much less compared to electrooptical detection. In this letter, we perform a systematic study of the field-induced second harmonic generation to optimize terahertz wave detection using selected gases. Our study shows that in such heterodyne detection, the detected second harmonic intensity depends quadratically on the third order nonlinear susceptibility of the gas, providing an approach to enhance detection efficiency. Among all selected gases, $n$-butane is an efficient gas sensor covering over 10 THz with two orders of magnitude improvement on dynamic range.

The experiment was performed with a Spectra-Physics Hurricane amplified laser system, which delivers 800 nm, 80 fs, 650 μJ pulses at a 1 kHz repetition rate. The laser pulses were split into pump and probe beams by a 60%–40% broadband beam splitter. A 100 μm thick type-I beta barium borate crystal is used to generate the second harmonic pulses. Both fundamental pulses and second harmonic pulses are focused with a 125 mm focal length lens. After focusing, the radiated terahertz wave is collected by a 90° off-axis parabolic mirror and focused again by another parabolic mirror. The probe beam is sent through a time delay stage and then focused by a 125 mm lens through a hole in second parabolic mirror. Thus, the terahertz wave propagates collinearly with the probe beam and is focused into a gas cell at the same spot. The second harmonic signal is passed...
The data from SF6 and nitrogen are multiplied by a factor of 10 for clarity.

bias field of 7.5 kV.

through a pair of 400 nm bandpass filters and detected by a photomultiplier tube (PMT).

Figure 1(a) is the illustration of the gas cell used in our experiment. Our gas cell body is made of stainless steel. The entrance and exit windows are made, respectively, of 2 mm and 3 mm thick fused quartz with 25 mm diameter. The longitudinal length of the cell is 50 mm. The bottom of the cell is connected to a gas distribution system, which can pump the cell to vacuum (below 20 mtorr) or introduce a certain amount of each gas in to cell. The pressure inside can be precisely monitored with a Kurt. J. Lesker pressure gauge. A pair of electrodes with a 1.8 mm gap are placed at the focal spot through an electrical feedthrough connected to an ac bias modulator which is synchronized with the laser, providing tunable voltage up to 3.2 kV at a frequency of 500 Hz. By referring the lock-in amplifier to the 500 Hz bias modulation frequency, the measured second harmonic intensity ($I_{2\omega}$) can be expressed as:

$$I_{2\omega} \propto [\chi^{(3)}I_{laser}]^2 E_{bias} E_{THz}.$$  \hspace{1cm} (1)

For our heterodyne detection, the main noise is from laser fluctuation and PMT detected background noise. From Eq. (1), the laser fluctuation-induced noise can be expressed as

$$\delta I_{2\omega} \propto 2N_{laser}[\chi^{(3)}I_{laser}E_{bias}]^2.$$  \hspace{1cm} (2)

where $N_{laser} = \delta I_l / I_l$ is the laser fluctuation. Thus, the dynamic range of this detection method could be expressed as $DR = I_{2\omega} / \sqrt{\delta I_{2\omega}^2 + NEP^2}$, where $\sqrt{\delta I_{2\omega}^2 + NEP^2}$ is the total noise in detected second harmonic intensity and NEP is the PMT detected background noise. This expression predicts that under certain conditions, increasing $I_{2\omega}$ is the only efficient approach to enhance the dynamic range.

In Eq. (1), $I_{2\omega}$ is proportional to $[\chi^{(3)}]^2 I_{laser}$, and $E_{bias}$. As a result, enhancing the dynamic range should be possible by using gases with high third order nonlinear susceptibility, ionization potential, and breakdown voltage. Among all our selected gases, xenon (high $\chi^{(3)}$) (Ref. 13) and SF6 (high breakdown voltage) are two of the better candidates for gaseous sensors. Figure 1(b) shows a terahertz spectrum and waveform obtained with xenon as the sensing medium. With 80 fs laser pulses, the spectrum covers up to 10 THz with high dynamic range, providing a detector available for nonlinear spectroscopy covering the entire terahertz gap. There is no significant spectrum changes observed with various gases.

Figure 2(a) shows the dependence of $I_{2\omega}$ on the probe pulse energy. The detected second harmonic intensity $I_{2\omega}$ was calculated by integrating the terahertz wave spectra from 0.3 to 10 THz. As can be seen in Fig. 2(a), the measured second harmonic signal evolves quadratically for low probe pulse energy, as is expected from Eq. (1). This behavior is modified at higher probe pulse energy because of intensity clamping which occurs during and after plasma formation.\textsuperscript{15} In Fig. 2(a), there is no minimum requirement for probe pulse energy, which is different from the quasicoherent detection reported previously.\textsuperscript{3}

Due to the limitation of probe pulse energy resulting from intensity clamping, increasing bias field strength is another approach to enhance $I_{2\omega}$. Figure 2(b) shows the dependence of $I_{2\omega}$ on the bias field using xenon, SF6, and air as the sensing media, together with linear fits (dashed lines). To eliminate the complicated effects present at high pressure, tests were performed at a pressure of 100 torr. In additional tests performed at 1 atm (756 torr), under the same condi-
tions, the detected terahertz signal from xenon is an order higher than that from air.

The effect of the third order nonlinear susceptibility on this detection scheme is investigated by performing measurements with a series of gases. The absorption of alkane gases is ignorable for such short terahertz path length. Figure 3 shows $I_{2h}$ as a function of normalized third order nonlinear susceptibility $\chi^{(3)}$ (Ref. 13) together with quadratic fits (dashed line). The measurements were performed at pressure of 100 torr, a probe pulse energy of 15 $\mu$J, and a bias field of 5 kV/cm to eliminate phase mismatch, intensity clamping, and electrical breakdown for all the gases. The ionization effect on $\chi^{(3)}$ can be ignored, as the probe pulse energy is lower than ionization threshold. We find that $n$-butane generates nearly two orders higher second harmonic signal than nitrogen.

From the microscopic origin of the third order nonlinear susceptibility, $\chi^{(3)}$ is proportional to molecular density. Thus, $I_{2h}$ should be proportional to the square of pressure. This identifies the pressure of the detection medium as an important parameter for gas sensing. Figure 4 shows $I_{2h}$ as a function of gas pressure. From Fig. 4(a), in the low pressure regime (below 300 torr), our measurement of the second harmonic varies quadratically with pressure. As the pressure increases, the behavior starts to deviate from the quadratic pressure dependence. The different turning points for different gases indicate that this phenomenon is related to the laser pulses propagating in a dispersive medium. To verify this, we repeated the pressure dependence experiment at a probe pulse energy of 100 $\mu$J [Fig. 4(b)]. The deviation points for each gas move toward lower pressure for increasing probe pulse energy. When the pulse energy is far above the ionization threshold (>150 $\mu$J), a linear relation is established instead of a quadratic relation. It is evident that the plasma density has a significant effect on the propagating laser pulse, confirming that the intensity clamping effect limits the efficiency of gas sensing. Especially, xenon shows an unusual decreasing behavior at high pressure which is from phase mismatch due to high index of refraction, and the peak moves toward low pressure with increasing power.

In conclusion, we report systematic study of terahertz wave detection using gases as sensor through the field-induced second harmonic generation. Nearly two orders of magnitude enhancement of the dynamic range are observed using alkane gases as sensor. This high efficiency sensing capability allows simultaneously coherent and broadband spectroscopic measurements across the full terahertz range.

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FIG. 4. (Color online) Dependence of $I_{2h}$ on the gas media pressure at probe pulse energies of (a) 50 $\mu$J and (b) 100 $\mu$J. Due to limitation of breakdown voltage, the lowest achievable pressure for xenon gas is around 350 torr at 11.5 kV/cm bias field.