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LETTER

Very long terahertz free induction decay in gaseous hydrogen bromide

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Abstract
Free induction decay radiation was observed in low pressure hydrogen bromide that continued for more than 180 ns after excitation by a terahertz pulse from a free electron laser. The signal was measured in real time using an ultrafast Schottky diode detector. Such a signal corresponds in frequency domain spectral resolution to about 10 MHz, which allows for resolution of the isotopic and quadrupole structure of the absorption line.

(Some figures may appear in colour only in the online journal)

Terahertz time-domain spectroscopy (THz-TDS) has developed into a versatile characterization tool in the far-infrared spectral region [1]. In such experiments, changes in the temporal structure of the short pulses of broadband terahertz radiation are used for investigation of a test medium. These changes can be described in terms of free induction decay (FID) of the medium [2]. The FID signal generated in absorbing media by a short optical pulse contains the complete information on absorption spectra within the frequency range of the input pulse.

In conventional THz-TDS, temporal profiles of FID are measured by scanning the time interval between the THz pulse and probe pulse using a mechanical translation stage. The spectral resolution in THz-TDS is defined as a reciprocal of the maximum time delay of the probe pulse. A typical maximum delay is several hundred ps and the resolution is several GHz [3]. A commercially available instrument [4] has a resolution of 7.5 GHz. An alternative approach is the asynchronous optical sampling (AOS) technique [5] which requires two mode-locked lasers with slightly different repetition rates. The time delay in the AOS technique can be longer than the one achieved by the conventional mechanical stage; a spectral resolution of 80 MHz has been reported [6].

Direct detection of the FID signal using ultrafast terahertz detectors [7] does not require delayed pulses; furthermore, the signal can be recorded in a longer time interval. Using this technique allowed the observation of an FID signal in low pressure hydrogen bromide (HBr) gas which continued for 180 ns, corresponding to a spectral resolution of about 10 MHz which is better than can be delivered by any other time-domain spectroscopy method. Note that long FID signals, formed by Stark switching of absorption lines, were observed in experiments with a continuous wave CO_2-laser [8], but this technique has limited application for time-domain spectroscopy.

As a source of terahertz radiation a Novosibirsk Free Electron Laser (NovoFEL) [9] was used. The laser emits a continuous sequence of pulses at a repetition frequency of 5.6 MHz with an energy of 5–20 μJ and pulse duration of 100–150 ps. The width of the laser spectrum was Fourier limited. The complete experimental setup is described in [7]. It consists of a gas cell 20 cm in length and a detection system. The latter has a rise time of better than 30 ps and includes custom-made ultrafast Schottky diode detectors [10] and a 30 GHz LeCroy direct oscilloscope. The laser beam passed through the gas cell filled with HBr gas to the detector. When the center of the laser line was tuned to 66.7 cm^{-1}, an FID

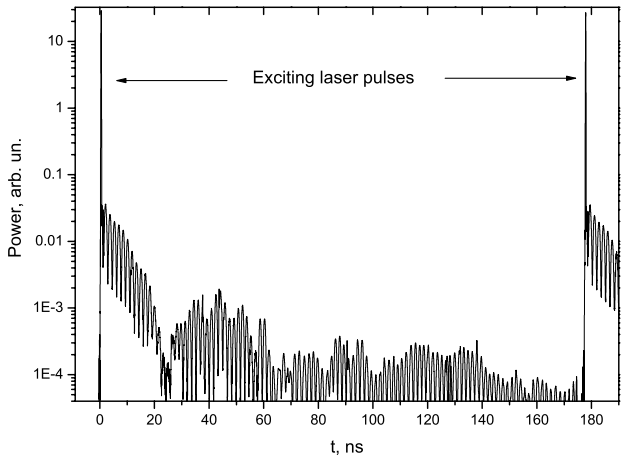

Figure 1. FID signal observed at 0.045 Torr HBr.

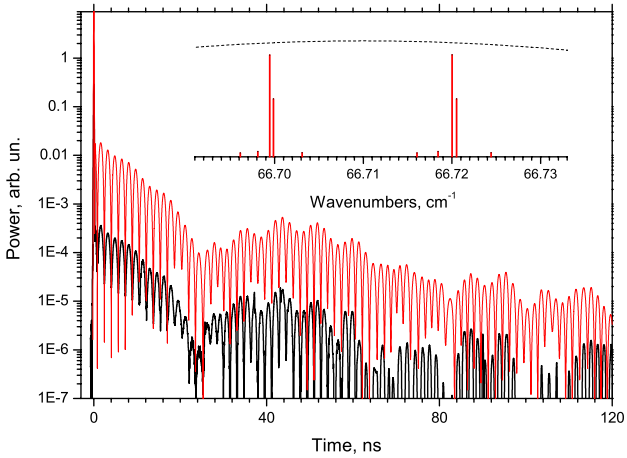

Figure 2. Thick black line: part of the experimental FID signal observed in HBr at 0.045 Torr; thin red: calculated signal. The calculated curve is shifted up for clarity. In the inset: isotopic and quadrupole structure of the rotation line of HBr near 66.7 cm^{-1}. The dashed line shows the laser spectrum in this region.

signal that looked like an oscillating tail was observed after the laser pulse. The duration of the FID signal depended on the HBr gas pressure; at low pressure the signal became very long. It was possible to detect the FID signal in the whole 180 ns time range corresponding to the period between laser pulses (figure 1).

Figure 2 shows part of the experimental signal and calculations based on available spectroscopy information. In this experiment the laser line (width 0.15 cm^{-1}) overlaps the absorption line ($J = 4 \leftarrow 3$) in the rotation spectra of HBr. The absorption line splits into several components due to ^{79}Br and ^{81}Br isotopes and the quadrupole moment of Br nuclei [11]. The isotopic splitting is 0.021 cm^{-1}, while the quadrupole splitting is one order of magnitude lower. The insert in figure 2 shows the structure of this absorption line. The intensities of the spectral components and their positions are taken from [12]. The temporal behavior of the FID signal was calculated using this spectral pattern (the calculation procedure is described in [7]). The calculated signal is shown in figure 2 by a red line, demonstrating a good correspondence to the experiment in most of the details.

The time-domain FID signal contains information on absorption spectra. As mentioned in [13], certain information can be gained directly from the time-domain waveform, without the need for a Fourier transformation. In our case such information can be obtained from FID oscillations. The period of the main oscillations is 0.16 ns which corresponds exactly to the isotopic splitting. Slower oscillations originate from the quadrupole splitting and from the 'top hat' shape of absorption lines [7].

In the current experiment the detected signal was proportional to the intensity of the FID and did not have information on the phase. Additional assumptions are necessary in order to obtain the spectra from this signal by Fourier transformation. Figure 3 shows the Fourier transformation of the experimental FID which was made using the phase information from the calculated signal. In this approximation the transformation does not deliver the absolute value of the frequency, therefore only relative positions of the absorption lines are presented.

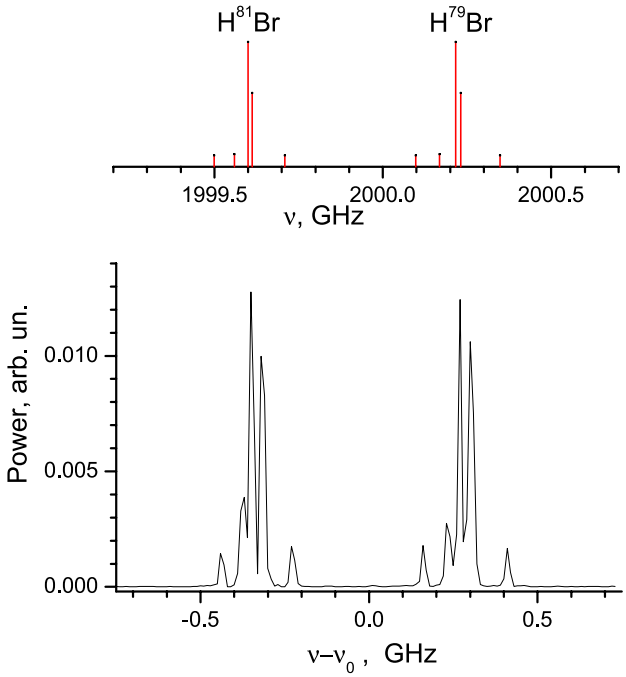

Figure 3. Lower panel: spectra obtained by Fourier transformation of the experimental FID (only relative positions of the absorption lines are shown). Upper panel: literature data of the absorption spectra of HBr in this region.

Line positions reported in the literature [12] are plotted in the top panel of figure 3. All lines are observed in the reconstructed spectrum. The frequency differences of the strong lines of $H^{81}Br$ and $H^{79}Br$ are 12 and 15 MHz respectively. In the reconstructed spectrum these pairs appear resolved although their positions are slightly shifted. The real resolution can be estimated from the width of the individual lines in the reconstructed spectrum which is about 20 MHz. Maximal resolution for our experiments can be estimated as twice the pulse repetition of the FEL which is 5.6 MHz.

A probable origin of the distortion of the spectrum and the decrease of the resolution are the approximations made in the procedure for the reconstruction of the spectrum. A proper procedure for the spectrum acquisition using Fourier transformation would require the measurement of the electric field of the FID, which in principle is possible using heterodyne detection with a terahertz gas laser as a local oscillator.

Acknowledgments

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References

[1] Nuss M C and Orenstein J 1998 Terahertz time-domain spectroscopy (THz-TDS) *Millimeter and Sub-Millimeter-Wave Spectroscopy of Solids* ed G Grüner (Heidelberg: Springer)
[2] Jacobsen R H, Mittleman D M and Nuss M C 1996 *Opt. Lett.* **21** 2011–3
[3] Hoffmann M C 2006 Novel techniques in THz-time-domain-Spectroscopy—a comprehensive study of technical improvements to THz-TDS *PhD Thesis* University of Freiburg, Germany
[4] 2008 *TPS Spectra 3000 Datasheet* (Cambridge: TeraView) www.teraview.com
[5] Yasui T, Saneyoshi E and Araki T 2005 Asynchronous optical sampling terahertz time-domain spectroscopy for ultrahigh spectral resolution and rapid data acquisition *Appl. Phys. Lett.* **87** 061101
[6] Yasui T, Kabetani Y, Saneyoshi E, Yokoyama S and Araki T 2006 *Appl. Phys. Lett.* **88** 241104
[7] Chesnokov E N, Kubarev V V, Koshlyakov P V and Kulipanov G N 2012 *Appl. Phys. Lett.* **101** 131109
[8] Gol'dort V G, Ledovskikh D V, Khvorostov E B and Rubtsova N N 2008 *Laser Phys. Lett.* **5** 197
[9] Gavrilov N G *et al* 2007 *Nucl. Instrum. Methods* A **575** 54
[10] Kubarev V V, Ovchar V K and Palagin K S 2009 Ultra-fast terahertz Schottky diode detector *Conf. Digest of the 34th Int. Conf. on Infrared, Millimeter and Terahertz Wave (Busan, Sept.)*
[11] Di Lonardo G and Fusina L 1991 *J. Mol. Spectrosc.* **148** 86
[12] Pickett H M, Poynter R L, Cohen E A, Delitsky M L, Pearson J C and Muller H S P 1998 Submillimeter, millimeter, and microwave spectral line catalog *J. Quant. Spectrosc. Radiat. Transfer* **60** 883–90
[13] Mittleman D M, Jacobsen R H, Neelamani R, Baraniuk R G and Nuss M C 1998 *Appl. Phys.* B **67** 379–90