Alternative Explanation of Free-Induction Decay

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***Abstract*—The terahertz pulse train resulting from absorbed rotational line spectra is modeled as the Fourier transform of a periodically filtered terahertz pulse in the frequency domain. The model is compared with previous absorption measurements in N_2O.**

I. Introduction And Background

Femtosecond pulse shaping is a well known technique of filtering in the frequency domain to generate various pulse trains in the time domain. The Fourier transform of a frequency comb in a mode-locked laser results in a pulse train with the pulse repetition rate defined by the reciprocal of the frequency comb spacing. This research explores the complement of a frequency comb and how this impacts the interpretation of absorption spectroscopy in the terahertz region.

Pulsed terahertz systems generate a broad terahertz spectrum which can be used for spectroscopy. Experiments have demonstrated impressive rotational spectroscopy of molecules such as N_2O, MeCl and many others[2-4]. The frequency domain interpretation of these experiments is straight forward – the spectrum obtained from the gas sample is compared with a reference spectrum obtained without the gas. In the time domain, the original pulse is observed after transmission through the sample followed by a train of smaller pulses--see Fig. 1(top). This pulse train has been described as emission from the vapor, and called free-induction decay[2-4]. The following analysis demonstrates that the pulse train can be interpreted as a result of the absorption of the vapor's rotational frequency comb.

II. Results

When a frequency component is removed from a pulse spectrum, the corresponding temporal domain can be modeled as a quasi-continuous wave whose phase is shifted by π such that it coherently interferes with the pulse[5]. An experiment demonstrated this effect at optical frequencies using a pulse shaper and a 0.2 mm wire to remove 2 nm of spectral width from the optical pulse[5]. In the terahertz region, rotational absorption lines in gases can be used to remove spectral components. Since the rotational spectra are nearly equally spaced, the resulting quasi-continuous waves will interfere and result in periodic pulses. The pulse separation is equal to the reciprocal of the rotational line spacing, which is very similar to the coherent addition of modes in a mode-locked laser. Fig. 1(center) demonstrates a terahertz pulse with the N_2O rotational lines removed from its frequency spectrum. The N_2O lines are removed by using the propagator in the frequency space: $\tilde{E}(\omega, L) = \tilde{E}(\omega, 0)\exp\left(ik(\omega)L\right)$; where $k(\omega)$ contains the real and imaginary components of the wavevector due to the absorption in N_2O. A simple Lorentzian profile was used for the linewidths. To circumvent suggestions that the emission of the N_2O vapor is hidden within the complex susceptibility, the pulse train was also generated using a simple notch amplitude filter at the location of each of the rotational lines of N_2O --see Fig. 1(bottom). This models wire placed in a spectrally dispersed location to remove the spectral components (i.e. amplitude pulse-shaping) similar to previous pulse shaping experiments [5].

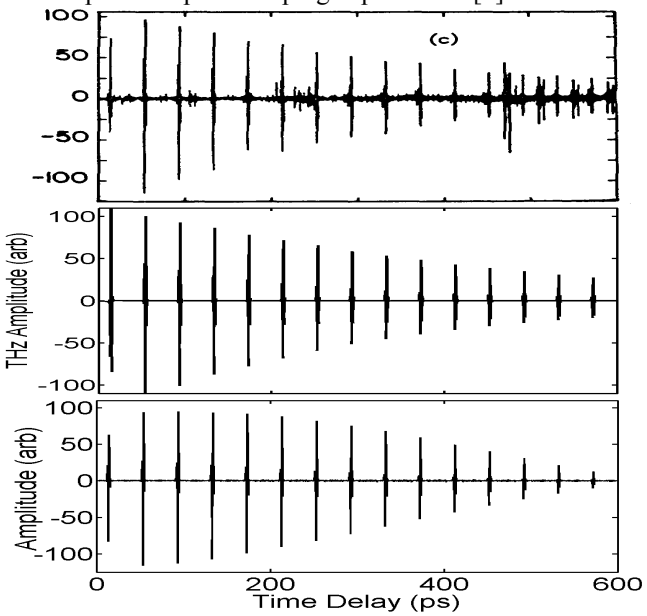

Fig. 1. (top) Experimental terahertz pulse train after propagating through N_20 [2]. (center) Model of terahertz pulse with N_2O frequency spectrum removed. (bottom) Model with spectra removed using wires in a pulse-shaper.

In addition, the phase of the modeled seventh pulse was explored and compared to the previous experimental results. Using the simple classical electrodynamic absorption model, the phase of the seventh pulse, see Fig. 2 (center), closely matches the experimental results in Fig. 2 (top).

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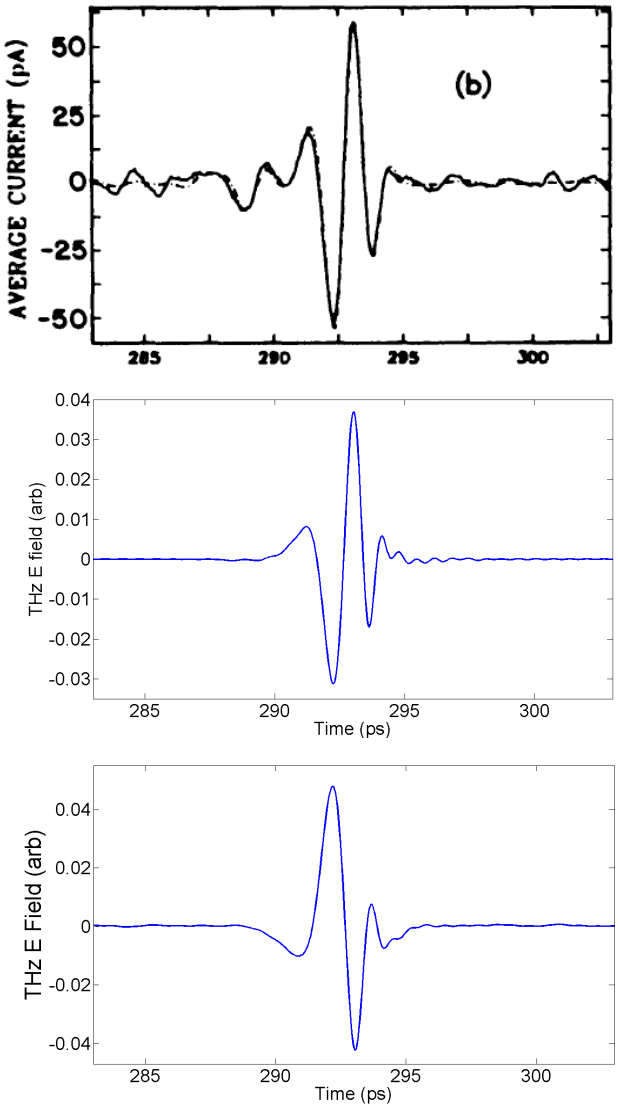

Fig. 2. (top) Expanded view of seventh pulse in the pulse train from experimental data [2]. (center) Classical absorption model. (bottom) Pulse generated from reradiated polarization.

In the theory of free induction decay, the molecules coherently reradiate the absorbed radiation. The induced polarization in the vapor will be proportional to the applied electric field, $\vec{P} = \varepsilon_0 \chi \vec{E}$. By multiplying the spectrum of the incident pulse by the spectrum of the vapor, the induced polarization can be obtained. If this polarization is then reradiated, a pulse train similar to the observed experimental pulse train is obtained; however, the phase of this pulse is in phase with the applied electric field, which is 180^0 out of phase with the observed phase, see Fig. 2 (bottom).

It is instructive to consider the effect in the frequency domain. By plotting the initial pulse spectrum and the spectrum after the absorbing vapor, see Fig. 3,

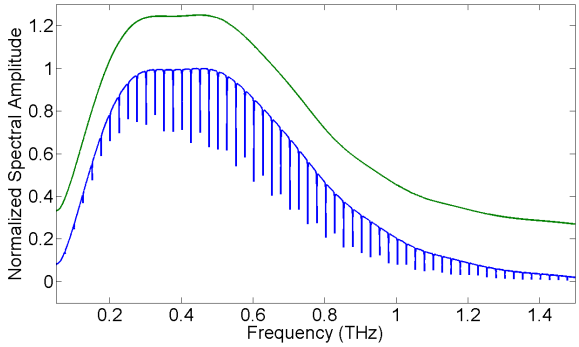

Fig. 3 (top) Initial pulse amplitude spectrum (offset by 0.25 for clarity). (bottom) Pulse amplitude spectrum after propagation through absorbing gas.

it is clear that the absorption lines have created an inverted frequency comb similar to frequency combs observed in mode-locked lasers; however, since the comb is inverted, the phase of the pulses generated by this inverted comb are 180^0 out of phase with respect to the incident pulse, which is precisely what is observed experimentally, see Fig. 2. This classical explanation of the pulses resulting from the absorbed spectral comb is, at least to this author, more intuitive than the explanation offered by free-induction decay. In summary, the pulse train from a terahertz pulse was modeled using classical absorption. The absorbed frequency comb provides an intuitive description of the resulting pulse train similar to the frequency comb observed in mode-locked lasers.

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