Freeze-out of difference-phonon modes in ZnTe and its application in detection of THz pulses

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Generation by optical rectification and detection by electro-optic sampling of freely propagating subpicosecond electromagnetic pulses (terahertz pulses) in ZnTe is limited by two broad difference-phonon absorption bands in the spectral region below the transverse optical resonance at 5.32 THz. In this letter, we show that, at temperatures below 80 K, these difference modes are frozen out and consequently ZnTe becomes completely transparent within our detection bandwidth. We utilize this effect to obtain a three-fold increase in detection efficiency at frequencies above 3 THz in a thin ZnTe detector operated at 80 K, compared to the performance at room temperature. © *2000 American Institute of Physics.* [S0003-6951(00)02944-2]

Since its introduction a few years ago, free-space electro-optic sampling[1] has become one of the most important methods for the detection of ultrashort terahertz (THz) pulses, competing with traditional photoconductive sampling in both signal-to-noise ratio and bandwidth. Whereas a photoconductive antenna normally displays a smooth frequency response, the detection efficiency of an electro-optic detector crystal is in general a complicated function of frequency.

Owing to its favorable properties, ZnTe has become the first choice of detector crystal. Therefore, we concentrate on this crystal in this letter. The detection capability of electro-optic crystals is determined mainly by the temporal walkoff between the probing near-infrared (IR) pulse and the frequency components of the THz pulse. Following the treatment by Gallot and Grischkowsky[2] this walkoff can be represented as a frequency-dependent response function

$$W(\nu)=\{\exp[i\Delta k_+(\omega_0,\nu)l]-1\}/i\Delta k_+(\omega_0,\nu), \qquad (1)$$

where ω_0 is the center frequency of the probe field, ν is a frequency component of the THz field, and l is the crystal thickness. The (complex) phase difference $\Delta k_+ \approx 2\pi\nu(n-n_g)/c+i\alpha(\nu)/2$, where n and α are the refractive index and absorption coefficient at frequency ν, respectively, and n_g is the group refractive index at the frequency ω_0 of the probe field. Δk_+ describes the difference in propagation speed between the optical pulse and the frequency components of the THz field through the crystal. In the case of a transparent crystal Δk_+ is real and Eq. (1) displays local minima at $\Delta k_+ l=0,\pi,2\pi,\ldots$. The first minimum (in the following the cutoff frequency) will be at $\nu_{\mathrm{cut}}=c/\{2[n(\nu_{\mathrm{cut}})-n_g(\omega_0)]l\}$. Apart from phase-matching requirements the bandwidth of the detected THz pulse is limited by the frequency dependence of the second-order nonlinearity $\chi^{(2)}(\nu)$ and by the duration of the probe pulse. In ZnTe below the fundamental transverse optical (TO)-phonon resonance at 5.32 THz,[3] and for probe pulses shorter than 100 fs, these two effects can be neglected. For further important references on the detection properties of ZnTe, see Ref. 4.

In this letter we demonstrate that if a thin ZnTe crystal is cooled to below 80 K the detection capability is significantly enhanced. We demonstrate the difference between cooling of a thick crystal ($l=2$ mm) and a thin crystal ($l=0.25$ mm), and find that there are limited benefits of cooling a thick crystal. A thin crystal, on the other hand, displays up to a three-fold detection enhancement above 3 THz. This enables reproducible spectroscopy up to approximately 4 THz in a spectrometer driven by a low-repetition-rate, amplified laser system. The main advantage of a spectroscopy system based on optical rectification and electro-optic sampling is the commercial availability of emitter and detector materials, as opposed to the highly specialized semiconducting dipole antennas used in photoconductive spectrometers. Since a high-power femtosecond laser is used to drive the electro-optic setup, it is also possible to perform optical pump-THz probe experiments.[5]

The experimental setup is illustrated in Fig. 1. The setup is driven by pulses from a regeneratively amplified Ti:sapphire oscillator operating with a center wavelength of 797

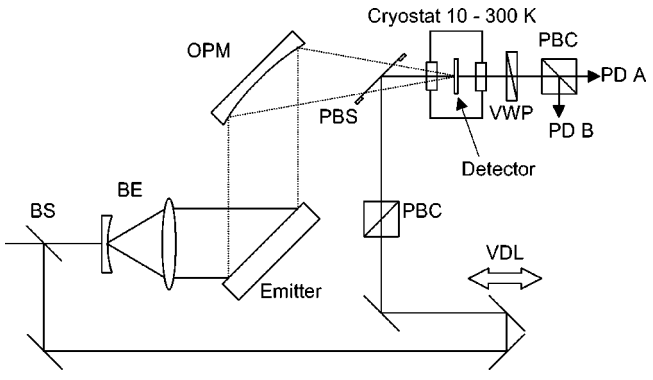

FIG. 1. Experimental setup. BS–beam splitter, BE–beam expander, VDL–variable delay line, OPM–off-axis paraboloidal mirror, PBS–pellicle beam splitter, VWP–variable wave plate, PBC–polarizing beam cube, and PD–photodiode.

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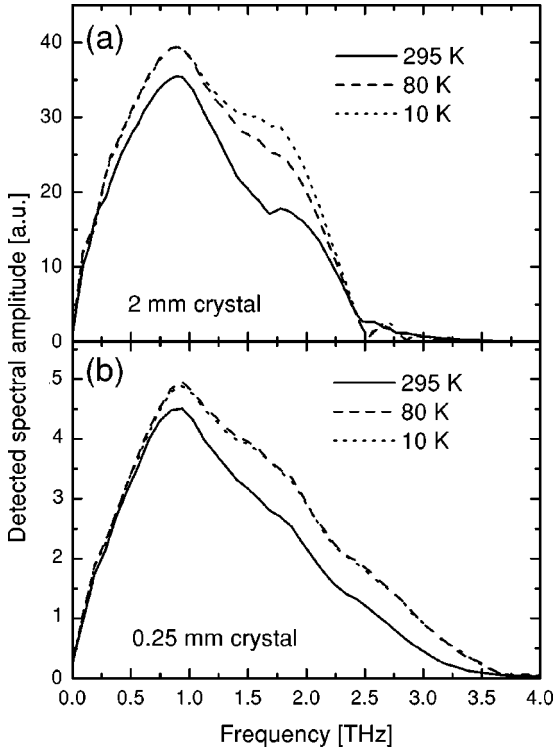

FIG. 2. Detected THz spectrum in 2 and 0.25 mm crystals, at different temperatures.

nm with a pulse duration of 80 fs and a repetition rate of 1 kHz. The THz pulses are generated in a GaAs (311) surface at a fluence of $10\,\mu J/cm^2$ by optical rectification of an expanded part of the laser beam. The resulting beam of THz pulses propagate in a highly collimated beam,[6] and is focused by an off-axis paraboloidal mirror ($f=120$ mm) into a cryostat where the (110)-cut ZnTe detector crystal is placed. A second part of the laser beam is coupled onto the THz propagation axis with a pellicle beam splitter and measures the THz pulse shape by free-space electro-optic sampling.[1] In order to couple both the THz beam and near-IR probe beam into the cryostat we use windows made of polymethylpentene (TPX).[7] This material has extremely low loss and dispersion in the THz range and has acceptable transmission around 800 nm. The temperature of the detector crystal can be varied continuously between 10 and 300 K. The phase retardation signal from the photodiodes is measured by a lock-in amplifier, which is locked to the frequency of a mechanical chopper in the THz generation beam [not shown in Fig. 1]. A typical scan is recorded with a time constant of 30 ms and averaged ten times to increase the signal-to-noise ratio.

In Fig. 2 we show the spectrum of the detected pulse under different conditions. In part (a) of the figure the pulse was detected in a 2-mm-thick crystal, at 295, 80, and 10 K. At all temperatures the spectral response is limited by the phase factor Δk_+ which apparently remains constant within our temperature range. At room temperature a strong feature at 1.6 THz is observed. The lower detection efficiency in this frequency band is caused by a combination of difference-phonon absorption bands[8] which disappear at lower temperatures. In part (b) of Fig. 2 the THz pulse is detected in a 0.25-mm-thick ZnTe crystal. The detected bandwidth is now much larger, due to the thinner crystal. More importantly we observe that the high-frequency part of the spectrum is greatly enhanced upon cooling of the crystal, and there is virtually no difference between the spectra detected at 80 and at 10 K. According to recently published measurements by Gallot and Grischkowsky[3] there is a strong absorption band centered at 3.7 THz which probably is associated with another combination of difference-phonon modes.[8] Consequently the imaginary part of Δk_+ is greatly reduced at low temperature, leading to an enhancement of the detection efficiency.

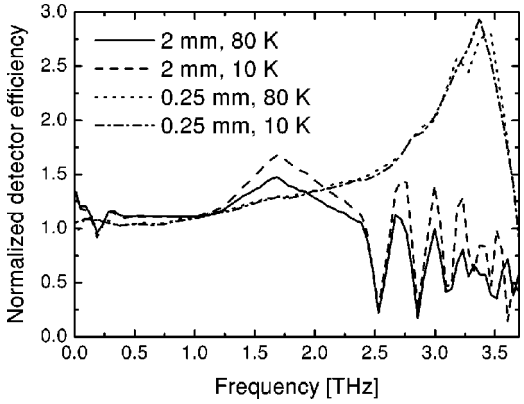

FIG. 3. Detector efficiencies of 2 and 0.25 mm crystals at 80 and 10 K, relative to efficiency at room temperature.

The phase difference Δk_+ is independent of temperature as indicated in Fig. 2, where the cutoff frequency remained constant at all temperatures. This was further confirmed in an independent measurement where a ZnTe crystal (inside the cryostat) was placed in the *optical* beam generating the THz pulse, thereby delaying the generation pulse in accordance to the group refractive index at the probe frequency. The temperature-dependent delay of the generation pulse was measured by monitoring the arrival time at the detector of the THz pulse generated by the optical pulse. The group index n_g obtained from this measurement showed the same temperature dependence as the general shift of the refractive index observed in the THz range.[8] We therefore conclude that below 300 K, Δk_+ is independent of temperature.

In Fig. 3 we have plotted the detection efficiency of the 2 mm crystal and the 0.25 mm crystal with respect to the efficiency at 295 K, calculated as the ratio of the detected spectra at the different temperatures. In the case of the 2 mm crystal, we observe an efficiency enhancement in the vicinity of the 1.6 THz absorption band. Apart from this we observe pronounced oscillations above the cutoff frequency at 2.5 THz. Using $\nu_{cut}=2.5$ THz we infer that in the 2 mm crystal $n-n_g=0.03$ at the cutoff frequency. In the 0.25 mm crystal we observe an increasing efficiency at high frequencies. At 3.5 THz the detection efficiency is approximately three times larger at 80 K compared to detection at room temperature. The cut-off frequency is at 3.7 THz, and at this frequency we therefore find that $n-n_g=0.16$. The larger index difference can be understood intuitively since the index in the THz region increases at high frequencies as a direct consequence of the presence of the TO phonon at 5.32 THz. This effect is also the reason why the cutoff frequency in ZnTe does not scale linearly with the crystal thickness.

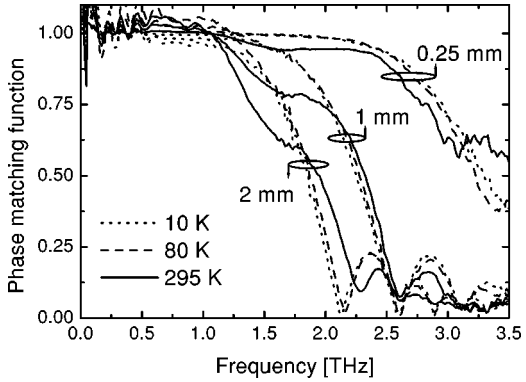

FIG. 4. Phase matching function of detector crystals of different thicknesses.

In Fig. 4 we show the temperature dependence of Eq. (1) for three different crystal lengths. The absorption coefficient $\alpha(\nu)$ and refractive index $n(\nu)$ are taken from Ref. 8. A similar figure for the room-temperature operation of ZnTe is found in Ref. 3. According to Fig. 4 the phase matching requirement should limit the bandwidth of a 2 mm crystal to approximately 2 THz, whereas we in Fig. 2 observe the cutoff at 2.5 THz. This discrepancy is probably caused by a slightly different refractive index of our particular crystal, which by coincidence enhances the detection bandwidth. Apart from the obvious bandwidth increase when using thinner crystals we observe that the influence of the 1.6 THz absorption band is largest in the thicker crystals. The curves representing the frequency response of the 0.25 mm crystal are limited by the spectral bandwidth of the measurement of n and α to below 3 THz.

In conclusion we have determined the temperature dependence of the detection bandwidth of the electro-optic crystal ZnTe. We find that for thick crystals there is limited enhancement of the performance of a cold crystal compared to operation at room temperature, except in the frequency range around 1.6 THz, where a combination of difference-phonon modes is frozen upon cooling. The upper frequency limit is in both warm and cold crystals determined by the temporal walkoff between the THz pulse and the probe pulse. In a thin crystal the situation is different, since the cutoff frequency is higher. The detection efficiency below the cutoff frequency is now limited by absorption of a much stronger difference-phonon band centered at 3.7 THz. Like the 1.6 THz mode, this mode disappears upon cooling, and the crystal becomes virtually transparent below the fundamental TO-phonon frequency. Since this resonance is extremely narrow[3] we expect that a slightly thinner ZnTe crystal than used in the present experiment, operated at 80 K, will display a featureless response from very low frequencies to at least 4 THz, much like the frequency response demonstrated in the best photoconductive THz spectrometers (see, e.g., Ref. 3). We firmly believe that this appealing feature will open up new avenues for static and transient spectroscopy based on amplified laser systems.

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