Ultrafast Interactions in Condensed Matter as Source of Terahertz-Frequency Electromagnetic Radiation

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Terahertz (THz, 10^{12} Hz) frequency radiation has long proven to be elusive relative to microwaves at lower frequencies and infrared radiation at higher frequencies. We have investigated several solid-state materials as candidate terahertz-frequency emitters under excitation by ultra-short (12 fs) pulses of near-infrared radiation, and present the results here with a comparison between different emission mechanisms.

Introduction

Terahertz (THz, 10^{12} Hz) frequency photons have energies in the meV range and so are useful for probing phenomena in solids including phonons, impurity states, and cyclotron resonance [1]. Their ability to penetrate dry non-polar materials, such as paper and plastic, coupled with their sensitivity to resonances characteristic of complex materials, opens up applications ranging from biomedicine to security. In this context THz imaging plays an important role [2]. All applications will benefit from better emitters of THz radiation. To this end we have undertaken an investigation of solid-state materials that may serve as sources of THz radiation under excitation by ultra-short pulses of near-infrared radiation.

Broadly speaking, terahertz emission under excitation from ultra-short pulses can be classified into three separate classes:

Photoconductive (PC) Emitters:

In a photoconductive emitter, the ultra-short pulses create electron-hole pairs in the semiconductor target, and an externally applied electric field accelerates the newly created charge carriers. A short terahertz pulse is emitted, due to the short carrier lifetime in the material (e.g. GaAs).

Transient-Current (TC) Emitters:

In contrast to photoconductive emitters, transient-current emitters do not require an externally applied electric field to operate. Instead, electron-hole pairs created near the surface contribute to terahertz-frequency emission through either the Photo-Dember effect, where differing hole and electron diffusion rates lead to a transient dipole at the semiconductor surface, or surface-field emission, which results from band-bending at the surface.

Optical Rectification (OR) Emitters:

Terahertz emission can also occur without an external bias in the case of optical rectification, where an intense optical beam induces a polarisation in a non-linear material, in analogy to rectification by non-linear devices (e.g. diodes) in an electric circuit. Terahertz radiation is emitted due to difference-frequency generation from the different frequency components present in the near-infrared beam.

In contrast to the behaviour expected from TC emitters, emission from OR emitters exhibits a strong and predictable azimuthal-angle dependence. On the other hand, applying a magnetic field perpendicular to the surface normal is expected to have an effect on TC

emission, but not OR emission, as the magnetization contribution to the nonlinear optical susceptibility is expected to be small. We can therefore distinguish between OR and TC emission [3].

Measurements were taken using the technique of time-domain spectroscopy. With time-domain spectroscopy, ultra-short pulses of near-infrared radiation are split into two paths – a pump beam, which is responsible for the emission of short terahertz pulses by interacting with the target sample; and a probe beam, which is subjected to a variable delay. When the emitted terahertz beam and the probe beam are brought to coincidence, the electric field of the terahertz beam modulates a measurable property in the probe beam. Since the variable-delay probe beam "gates" the electric field at different times, a time-domain profile of the signal can be obtained.

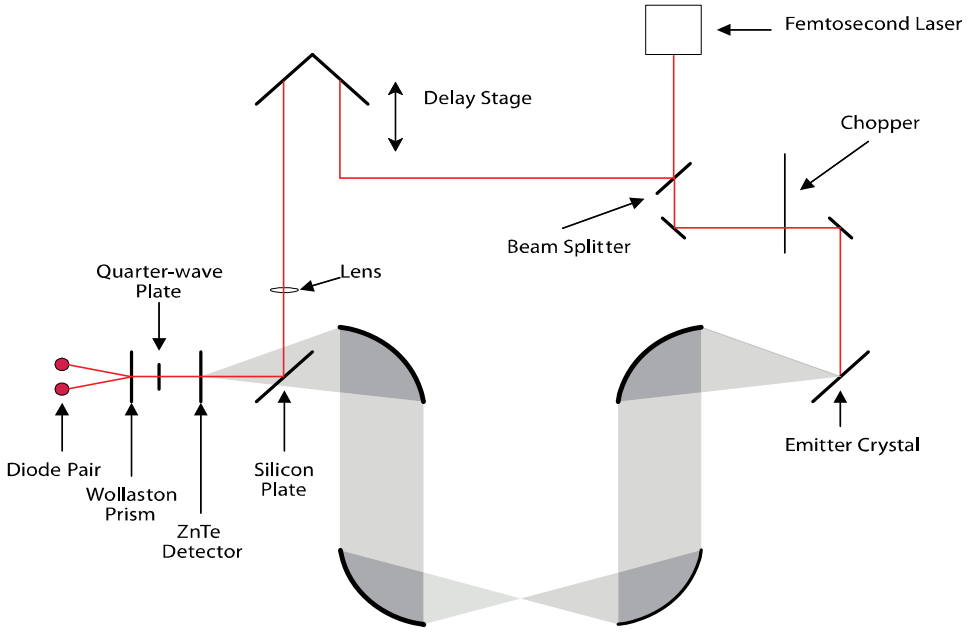

Figure 1. Diagram of the time-domain spectroscopy apparatus.

A diagram of our system is shown in Figure 1. In our system, a (110) ZnTe crystal is used as a detector – the terahertz-frequency electric field induces a change in the polarisation of the probe beam, which is detected using a quarter-wave plate, a Wollaston prism and a photodiode pair. Near-infrared pulses are provided by a FemtoSource mode-locked Titanium-Sapphire laser, which produces pulses of 12 fs duration at a centre-wavelength of 790 nm and a repetition rate of 75 MHz.

We have tested a number of different samples in the system – Be-doped GaAs and GaAsSb have been tested as photoconductive emitters, as well as for emission without bias. We have also tested InAs, InP, ZnTe, as well as multiple GaAs samples cut along different crystallographic planes.

In addition to time-domain measurements, the emitted terahertz intensity for a number of samples has been estimated with a golay cell.

Results

We find that, while low-temperature grown GaAsSb exhibits TC effects, high-temperature grown GaAsSb serves only as a photoconductive (PC) emitter [4]. The generation of THz radiation by PC in Be-doped GaAs varies systematically with optical, electrical and materials factors [4, 5].

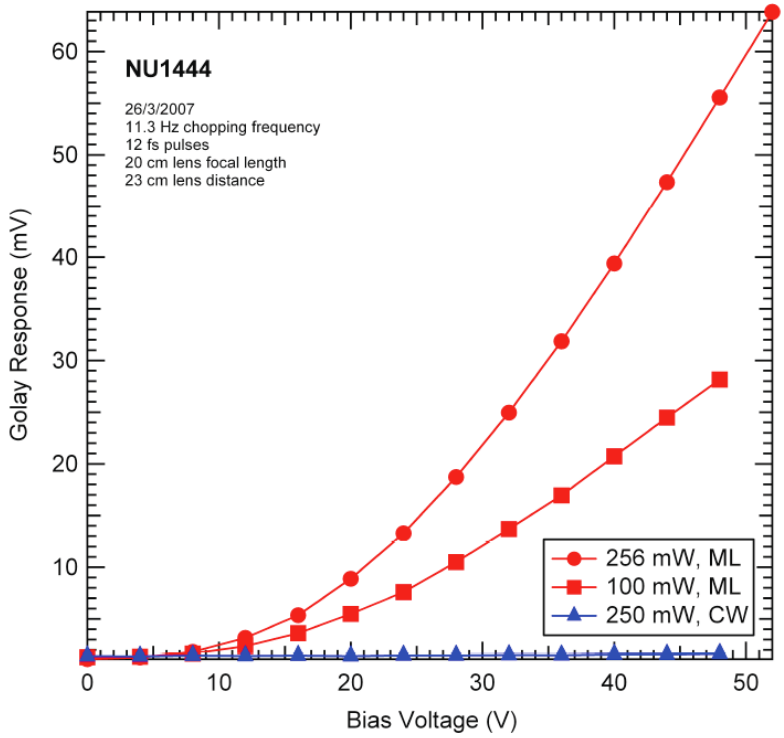

Figure 2. Terahertz output power as a function of bias voltage, GaAs:Be.

Figure 2 provides an example of variation in emitted terahertz output power with both bias voltage and optical excitation density in GaAs:Be, as measured with a golay cell. Output power varies approximately quadratically with bias voltage, and approximately linearly with the optical excitation density. To confirm that measurements are of coherent terahertz generation and not a result of heating, measurements were repeated with the laser operating in continuous-wave mode, under which no output was observed.

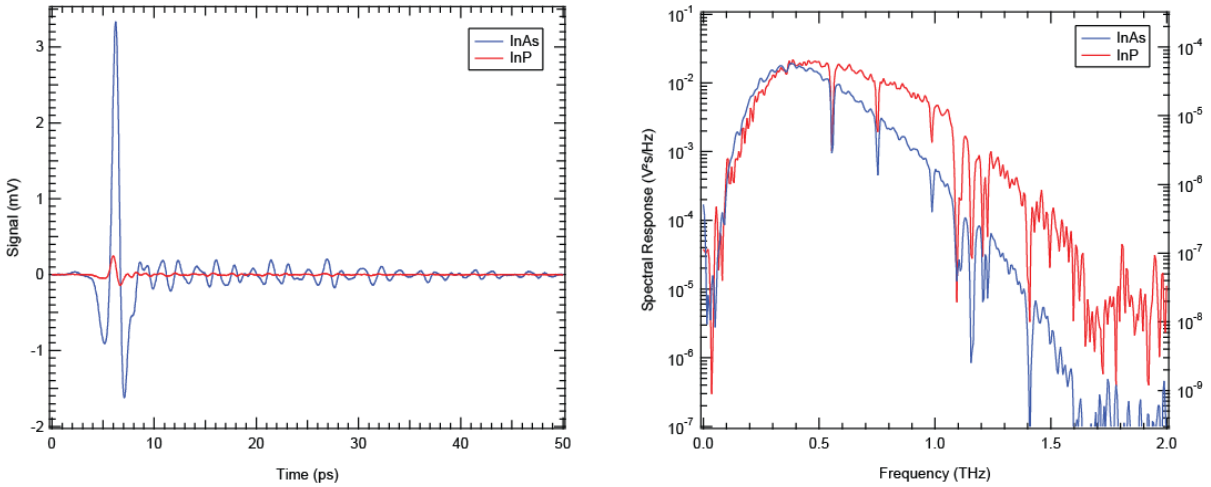

Figure 3. Time-domain spectrum of emission from InP and InAs (left) and the corresponding Fourier transform (right).

Figure 3 illustrates the difference between two transient-current emitters, InAs and InP, as well as providing an example of both the time-domain spectrum (left) and the frequency-domain Fourier transform (right). Absorption lines evident in the Fourier transform are due to absorption by atmospheric water vapour. It can be seen that even though InAs is a stronger

emitter in absolute terms, InP displays relatively stronger emission at higher spectral frequencies [3].

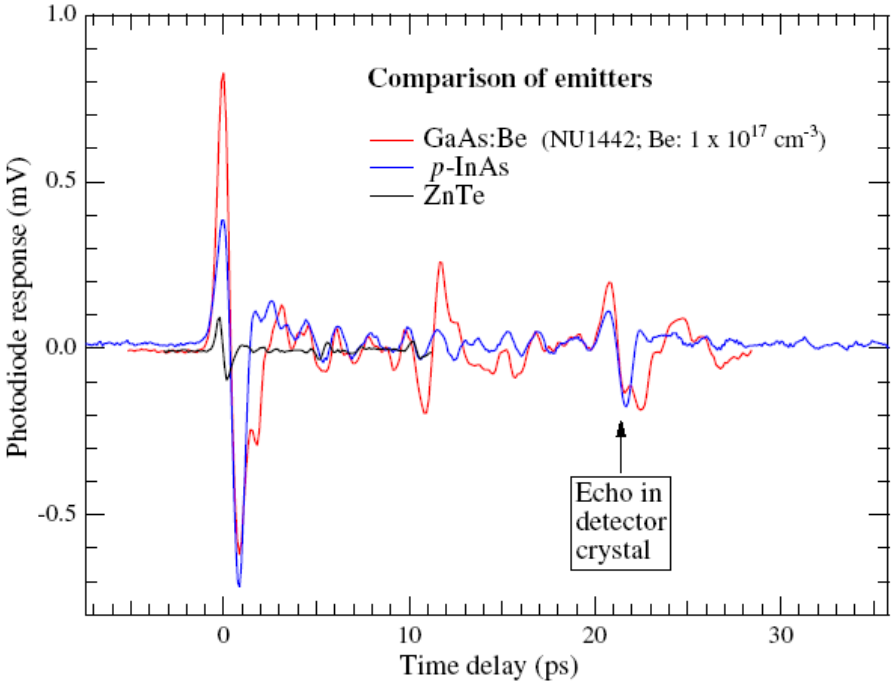

Figure 4. A comparison between the three emitter classes.

Finally, we present a comparison between the three different emitter classes in Figure 4. Emission via the electro-optic effect in ZnTe (110) was found to be relatively weak – about an order of magnitude below that of the photoconductive emitter in terms of the electric field. InAs, which produces terahertz radiation via the transient-current photo-Dember effect, was found to have a signal strength between the ZnTe and photoconductive emitters.

Conclusions

We have demonstrated terahertz emission from a number of different sources under excitation from ultra-short near-infrared pulses. We also demonstrate and distinguish between the different emission mechanisms of photoconductivity, transient current emission and optical rectification. Of these, photoconductive emission appears to be the strongest.

References

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