Non-Perturbative Terahertz Sideband Generation from Bulk GaAs

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Abstract Using intense picosecond pulses of coherent terahertz (THz) radiation, we have investigated time-resolved non-resonant THz sideband generation from bulk GaAs. The THz power dependence clearly reveals a non-perturbative strong-field regime. In addition to the expected ω_{-2} sideband, we detected the ω_{-1} (or odd) sideband which has previously been observed only in a quantum well system where the inversion symmetry was intentionally broken.

1 Introduction

Semiconductors subject to an intense electric field exhibit phenomena that cannot be understood by treating the field as a small perturbation. Intense fields can strongly modify the lattice periodic potential, exerting significant influences on optical processes. Exploring these electro-optical effects in high frequency ac fields, especially in the terahertz (THz) range, is not only important technologically, but also allows us to explore new multi-photon, strong-field phenomena that cannot be probed by other traditional methods.

Here, we study THz sideband generation [1]. In this multi-photon process, a weak optical beam passes through a THz-driven semiconductor, acquiring sideband frequencies separated by integer multiples of the THz frequency:

$$\omega_n = \omega_{NIR} + n\omega_{THz}; \quad n = \pm 1, \pm 2, \pm 3.... \qquad (1)$$

These lines completely dominate the near-bandedge emission properties of THz-driven semiconductors [1]. Recent theoretical studies predict new phenomena for THz sidebands such as dynamical symmetry breaking [2], non-monotonic power dependence [3], chaotic behavior [4], sideband disappearance under cyclotron resonance [5], and THz-induced subband hybridization [6]. However, none of these has been observed.

Previous observation of THz sidebands relied on *resonant enhancement* using magnetoexcitons [1] or quantum well subbands [7], and the main results were successfully explained by *perturbation* theory [8]. In [7] the inversion symmetry of the system was intentionally broken, allowing the detection of first-order ($n = \pm 1$) sidebands, whereas in [1] only even ($n = \pm 2, \pm 4$) sidebands were observed. In either case, no evidence of *bulk* $\chi^{(2)}$ contribution was observed. Finally, these previous studies were essentially CW, and thus did not provide any information on the time evolution of the sdiebands. In what follows, we present results of *time-resolved* spectroscopy of *non-perturbative*, *off-resonance* THz sideband generation in *bulk* GaAs.

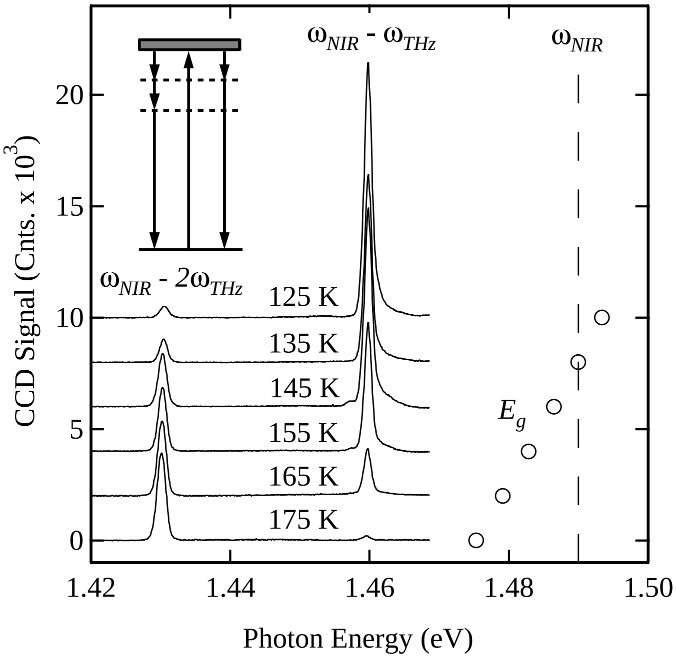

Fig. 1 Solid lines: offset for clarity sideband spectra at different T; dashed line: position of the fundamental line; open circles: band edge position calculated for each T

2 Experimental Results and Discussion

We utilized the wide tuning range (5 to 100 THz or 3 to 70 μm), short pulse duration ($\geq$ 0.6 ps), and high peak powers ($\leq$ 2 MW) of the Stanford Free Electron Laser (FEL). We combined the THz pulses from the FEL with the near-infrared (NIR) pulses from a mode-locked Ti:Sapphire laser using a Pelicle plate and then focused the collinear beams onto the sample placed in a variable temperature (20 - 300 K) cryostat. The transmitted and emitted NIR radiation was then sent into a grating spectrometer/CCD combination. The temporal overlap between the two beams was adjusted via a delay stage. Most measurements were made with the sample oriented at 45° with respect to the incident beams. While similar results were obtained for a variety of THz and NIR wavelengths, here we present the data only for $\lambda_{NIR} = 832$ nm (1.49 eV) and $\lambda_{THz} = 42$ μm (0.03 eV).

In Fig. 1 we show typical sideband spectra acquired at different temperatures T. The vertical dashed line represents the fundamental NIR frequency and the open circles show the bandedge of GaAs E_g for each T determined by FTIR interband absorptin spectroscopy.

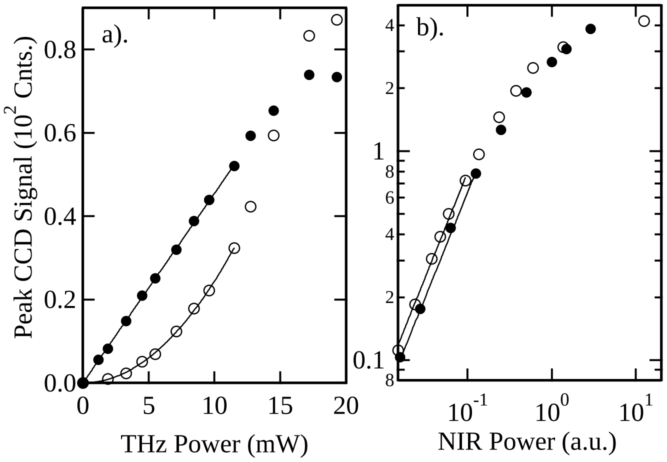

Fig. 2 Filled (open) circles: ω_{-1} (ω_{-2}) sideband intensity; solid curves: fit

Two strikingly strong lines are observed. They are separated by the energy of the THz quantum $\hbar\omega_{THz}$, and the higher energy line is precisely one THz quantum below the fundamental, identifying themselves to be the ω_{-2} and ω_{-1} sidebands, respectively (see Eqn. 1). The energy level diagram in Fig. 1 illustrates the experimental situation; the dashed lines are the intermediate virtual levels and the shaded area represents the continuum of real states above the bandedge. With our available THz intensities and detector sensitivity, we were not able to detect third or higher order sidebands.

The intensity of the sidebands is strongly temperature-dependent, as seen in Fig. 1. As T increases from 125 K to 175 K, the ω_{-2} sideband increases while the ω_{-1} sideband decreases and finally disappears. At higher T (not shown) ω_{-2} sideband weakens and vanishes at around 230 K. This behavior can be qualitatively explained by the interplay between the absorption of the fundamental and that of the sidebands. While a larger density-of-states at the fundamental frequency is desirable for more intense sidebands, no density-of-states is desirable at the sideband frequencies. The inset in Fig. 1 represents the results of our calculations (Section 3), which qualitatively agree with experimental observation.

There are three new features that are worth pointing out. First, unlike the previous studies [1,7], the sidebands appear at energies *below* the bandgap where there are no real states and therefore are not resonantly enhanced. Second, we observe an odd sideband (i.e., the ω_{-1} sideband) without intentional breaking of the inversion symmetry. We found that the intensity of this odd sideband sensitively depends on the crystal orientation with respect to the laser beams, indicating that the origin of this sideband is the lack of inversion symmetry of the GaAs crystal. In particular, it disappeared for normal incidence. Third, since we used picosecond THz pulses, we were able to monitor the evolution of the sidebands directly in the time domain, allowing us to measure the temporal profile of the THz pulse using a Si CCD detector.

Finally, we studied power dependence (Fig. 2(a) and 2(b)). Here, the sideband intensity is plotted vs. THz and NIR powers, respectively. At low THz powers the ω_{-1} (ω_{-2}) sideband depends on the THz power linearly (quadratically), indicating a perturbative $\chi^{(2)}$ ($\chi^{(3)}$) process involving one (two) THz photons and one NIR photon. At higher THz powers, these dependences show noticeable deviation from the perturbative behavior, indicating the entrance into the strong THz field regime. The saturation at high NIR powers is probably caused by free carrier absorption of THz radiation since the NIR photon energy is close to E_g.

3 Theoretical Model

The experimental observation of the odd sideband implies the lack of inversion symmetry. While the observed angle dependence on the incident beams can be explained by a simple bulk $\chi^{(2)}$ consideration, it is also possible that the surface is playing a role via band-bending due to Fermi-level pinning and the presence of residual carriers. In order to model this effect we assume that a static electric field is present in addition to the THz and NIR fields. Since the static field is involved, the first sideband can be explained as a $\chi^{(3)}$ process involving one NIR, one THz plus one zero-frequency photon of the static field. Using non-equilibrium Green function techniques developed in [9] and assuming a simple two band model, we calculated sideband intensities. The main results are shown in the insets of Fig. 1 and Fig. 2 and show good qualitative agreement with experiment. In particular, the theory accounts for both the T dependence of the sidebands and the THz power dependence, including the departure from the perturbative regime.

4 Summary

We have performed THz optical sideband generation from bulk GaAs. We observed sidebands below the band gap where there are no states. We demonstrated a new scheme to measure the temporal profile of the THz pulses with a NIR detector. The THz power dependence clearly revealed a non-perturbative strong field regime. Finally, we detected an odd sideband, which has previously been observed only in an asymmetric quantum well system where the inversion symmetry was intentionally broken.

References

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