Broadband Nonlinear Spectroscopy of Hydrogen-Like Levels in Ge:As

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Abstract—We present broadband nonlinear THz spectroscopy and saturation of hydrogen-like electronic states of Arsenic ions, trapped in a solid Geranium substrate. The broadband pulses are generated by a photoconductive array.

I. INTRODUCTION

ECENTLY, developments in high field THz sources Rhave opened new avenues for research into nonlinear material responses, such as coherent and incoherent material processes[1], transport dynamics in graphene[2] and plasmas in superconductors[3]. Free electron lasers (FELs) were the first systems capable of producing the necessary high fields, generating monochromatic radiation from a large tunable range from ~1-100 THz with fields in the region of 100 MV/cm[4]. The advent of chirped pulse amplification, has allowed for relatively compact laser systems with large pulse energies (up to ~15 mJ) and small pulse widths (<50 fs)[5] to become widely available and are ideal for nonlinear THz spectroscopy systems on a tabletop scale. Nonlinear crystals are popular choices for generation of THz radiation for these systems [6, 7] but suffer from problems, such as low damage thresholds, growth uniformity, or phase considerations. Photoconductive antennae are an alternative source suitable for THz generation and have certain advantages over nonlinear crystals, due to their greater efficiency, broad bandwidth of ~5 THz and the ability to electrically control the output THz field, removing the need for optical chopping. Until recently, the peak fields that could be generated from these sources was relatively low, however we have recently shown [8] it is possible to generate fields >100 kVcm⁻¹ from GaAs array emitters (PCAs). In this work, we apply this source to impurity centers in semiconductors [9, 10] which show very long-lived states. These materials are typically studied using free electron lasers (FELs) and pump-probe techniques [11]. While the FELs are well suited to the narrow spectral features, these measurements are limited to pumping/probing at a single frequency, potentially missing correlations scattering channels between states. By using a broadband source, such as a PCA, the effect of the pump on the whole spectrum can be measured.

II. RESULTS

The sample investigated was a 3 mm thick Ge substrate with a 9x10¹⁴ cm⁻³ As doping concentration. The sample was placed in a cryostat in a time-domain spectroscopy system, utilising a 145mW beamline from a Spitfire Ace PA amplified laser [5], with the PCA in a focus-through configuration described in our previous work [8]. As the feature of interest only appears at low temperatures, measurements were taken at a temperature of 10 K whilst a reference measurement was taken at 100 K. At

both temperatures the strength of the incident THz field was then varied by changing the electrical bias across the PCA. A strong absorption was seen at 2.28 THz corresponding to the 1s-2p₀ transition. Fig. 1 shows the absorption at 2.28 THz can be fitted to the expected saturation behavior of an inhomogenously broadened transition with a saturation intensity corresponding to a peak field between 15-20 kVcm⁻¹.

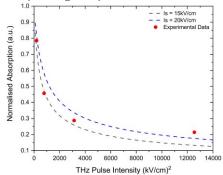


Fig. 1. Absorption at 2.28THz (1s-2p₀) of the Ge:As sample as a function of estimated intensity. Broken lines show the response of an inhomogeneously broadened transition with a saturation intensity relating to a field between 15 and 20 kVcm⁻¹.

III. SUMMARY

In summary, a system has been developed using our large area interdigitated photoconductive arrays, allowing for saturation of electron transitions in a doped semiconductor sample to be observed. We are grateful to Sergey Pavlov of DLR, Berlin for suppling the sample used for this work.

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