Two-Dimensional Time-Domain Spectroscopy for Determination of Energy and Momentum Relaxation Rates of Hydrogen-Like Donor States in Germanium

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Abstract (300 words max):

Impurity centres in semiconductors have gained attention in the THz frequency region for applications including optically pumped lasers [1], and coherent control of atomic orbitals [2], which has potential applications for quantum computing [3]. For these applications, state lifetime knowledge is critical for determining parameters such as optical gain and timescales which atomic states can be manipulated to perform quantum computations. Single frequency pump-probe techniques, common at free-electron laser facilities, are often used to measure these lifetimes [4], but the temporal resolution is limited by pulse duration (5-100 ps) and multiple measurements are required to interrogate multi-pathway decays in materials with a complex energy spectrum (Fig. a, b). Two-dimensional time-domain spectroscopy (2D-TDS) provides a potential alternative technique, as it has been shown to simultaneously acquire carrier and polarisation lifetimes of multiple decay pathways on fs timescales for quantum well systems [5].

We present measurements of coherence times of excited states of hydrogen-like arsenic impurities in germanium (Ge:As) using a table-top 2D-TDS system. We observe coherent population transfer and decay of transitions from the 2p0 and 2p± states by fitting the detected coherent nonlinear response with the known intracenter transition frequencies. Coherences between excited electronic states that are not visible via conventional single frequency pump-probe measurements are also observed in the off-diagonal resonances in the 2D frequency-domain map (Fig. c).

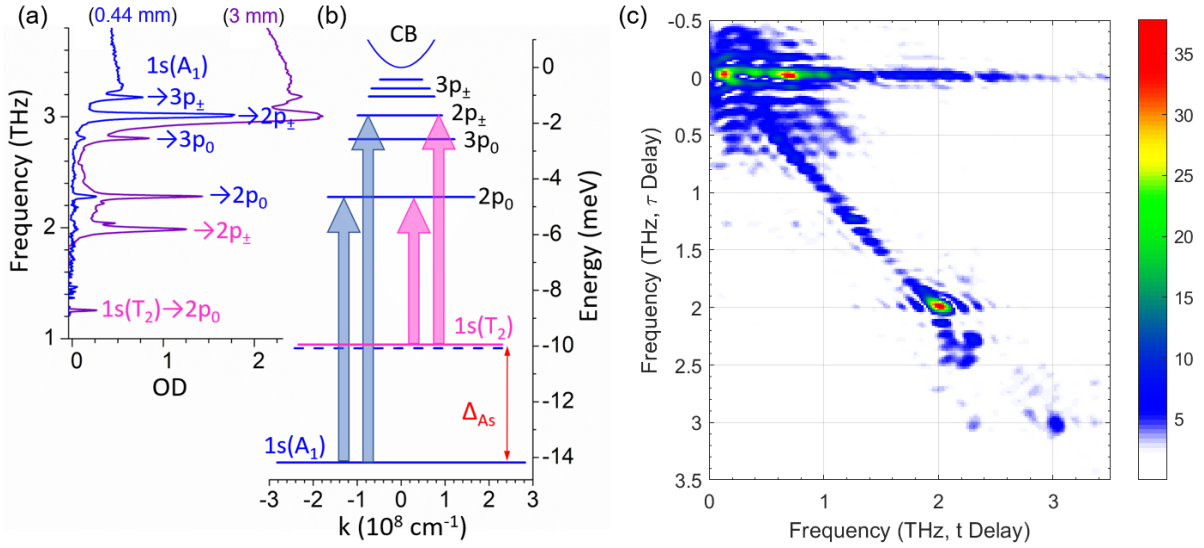

Figure: (a) FTIR absorption spectra of Ge:As samples of different thicknesses. (b) Ge:As energy level diagram, showing the observed intracenter transitions. (c) The 2D FFT of the measured 2D THz time-domain nonlinear response of Ge:As, excited by two intense broadband THz pulses.

References

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