Four wave mixing studies of polaron dephasing in InAs/GaAs self-assembled quantum dots

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Abstract— **We will present a detailed investigation of intraband polaron dephasing processes in n-type InAs quantum dots. Excellent agreement is found between our energy and temperature dependent, far-infrared degenerate four wave mixing (FWM) measurements and calculations of the FWM dynamics. We find that the presence of the weak acoustic phonon sidebands results in strongly damped oscillations shortly after resonant excitation of the ground to first excited conduction band state. Long (~90ps) dephasing times are measured and a clear change from phonon-mediated to Auger-mediated dephasing is observed as the dot carrier population increases.**

Index Terms— **Four-wave mixing, intraband transitions, far-infrared spectroscopy, quantum dots, polarons.**

I. Introduction

RECENT experimental [1] and theoretical [2, 3] studies have demonstrated that the interaction between electrons and phonons in quantum dots (QDs) can only be described by considering them to be in the strong coupling regime, forming polarons. To date, ultrafast measurements in this area have concentrated on studying the polaron decay process [4], which has been shown to occur on a long timescale (tens of ps), relative to intraband carrier relaxation in quantum wells (ps timescale). These studies have also identified the polaron decay mechanism and show a monotonic increase of the decay time with increasing energy, providing clear evidence of a strong coupling regime in which the 'phonon-bottleneck' does not exist. The long relaxation time highlights the potential of QD-based mid-infrared intraband detectors and emitters, which require long excited state lifetimes for efficient operation.

We will present a detailed investigation of polaron dephasing processes in n-type InAs quantum dots (QDs) using energy and temperature dependent, far-infrared degenerate four wave mixing (FWM). Our measurements allow us to determine the polaron dephasing time and hence the homogeneous linewidth. At these far-infrared wavelengths it is not possible to obtain the homogeneous linewidth by direct, single QD measurements due to the small QD absorption and relatively insensitive detectors. We find that the presence of the weak acoustic phonon sidebands results in strongly damped oscillations shortly after resonant excitation of the ground (*s*) to first excited (*p*) conduction band state. This is in strong contrast to previous *interband* studies [5] which show a pronounced fast decay component in the FWM signal, due to the larger lattice distortion following interband excitation. At low temperatures we measure polaron dephasing times, T_2 ~90 ps, (Γ_{hom}~20 μeV) close to those expected from our independent polaron decay studies ($2T_1$ ~100 ps) [4].

II. Results and discussion

Coherent polaron polarizations in QDs were studied using a standard two-pulse photon echo arrangement in a non-collinear geometry. The phase-matched signal was detected in the $2\cdot\mathbf{k}_2-\mathbf{k}_1$ direction. The far-infrared time-integrated FWM measurements were carried out using the Dutch free electron laser (FELIX) which provides tunable far-infrared laser pulses of ~1ps duration.

For excitation in resonance with the QD ground to first excited conduction band transition, we find low temperature dephasing times T_2 ~90 ps (Fig. 1a), approximately two orders of magnitude longer than T_2 for intersubband transitions in quantum wells. From the measured T_2 time at 10K, we extract a homogenous linewidth Γ_{hom}~20 μeV, approximately 0.4% of the predominantly inhomogeneously broadened linewidth of (Γ_{inhom}~5 meV) measured using linear absorption.

We also found that the polaron dephasing time decreases from ~90 ps at excitation energy $\hbar\omega \approx 53$ meV to ~60 ps at $\hbar\omega \approx 48$ meV. This is consistent with the energy dependence of the polaron decay time [3, 4], which decreases as the polaron energy approaches that of the LO-phonon.

We have calculated the FWM dynamics for excitation in resonance with the *s* to *p* transition, which take into account polaron decay processes as well as the interaction with

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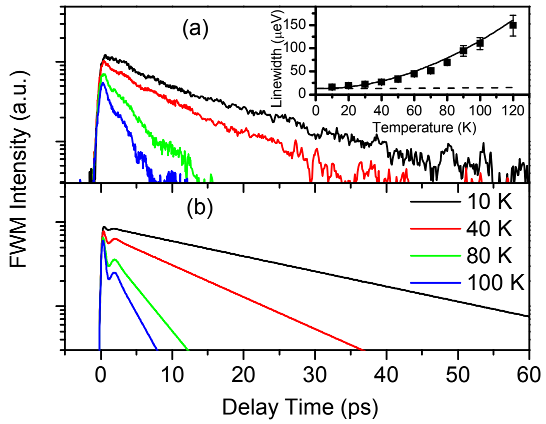

Fig. 1: Temperature dependent four wave mixing signals: experiment (a) and simulations (b). The inset shows temperature dependence of the polaron linewidth: experiment (closed squares) and calculation (solid line). Dashed line exhibits the temperature dependence of the population relaxation.

acoustic phonons which result in acoustic phonon sidebands as replicas to the zero-phonon line (independent boson model) [6]. We also include real and virtual [7] transition towards the higher energy p_+ state, where the p_-, p_+ energy separation is ~5 meV. Our calculations reveal the presence of the initial decoherence oscillations between 0 and 5 ps due to the presence of acoustic phonon sidebands (see Fig.1b). These oscillations, also observed experimentally as a shoulder at ~2.5 ps, become more prominent with increasing temperature since the population of the acoustic phonons increases (Fig.1a). The latter also results in the coherent polaron polarization decay to be dominated by pure dephasing at temperatures >80 K due to real and virtual absorption/emission of acoustic phonons with energy close to p_-, p_+ splitting (solid line in Fig.1 inset). This behaviour is very different from the weak temperature dependence of the polaron decay time (T_1) over the same energy range (dashed line in Fig.1 inset), which we have previously shown arises from polaron decay to two high energy acoustic phonons [4].

The use of intersublevel excitation also allows us to study excited state dephasing for samples doped with two or more electrons per dot, a regime which is difficult to access using interband excitation due to Pauli blocking of the QD ground state. We find a dramatic reduction in T_2 for samples doped to contain more than one electron per dot (Fig. 2). This result gives a clear indication that there is a transition from a phonon mediated dephasing process to one which is due to carrier-carrier interactions as the QD population increases, in contrast to quantum wells where intersubband dephasing is always dominated by carrier-carrier interactions.

In summary, we have shown that the polarization decay in InAs quantum dots following intraband excitation shows strong differences compared with the decay following interband excitation [5], due to the smaller lattice distortion in the former case. Furthermore, compared with interband studies, intraband investigations allow a clear picture of dephasing mechanisms in n-doped dots to be obtained as a result of the well-resolved 3 level conduction band system (with no dark states), providing a deep insight with relevance to the development of quantum information processing schemes.

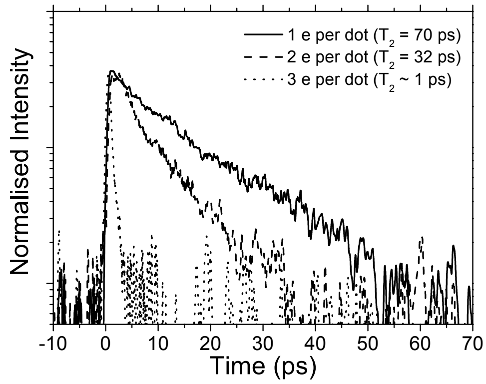

Fig.2: Normalized four wave mixing signals for samples doped to contain 1 electron (solid line), 2 electrons (dashed line) and 3 electrons (dotted line) per dot.

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