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Non-linear infrared properties of InAs/GaAs self-assembled quantum dots

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Abstract

We have investigated various optical non-linearities in resonance with intersublevel transitions in InAs/GaAs self-assembled quantum dots. The infrared pulses were delivered by CLIO at Orsay (France) and enabled (i) the evidence of second-harmonic generation with giant susceptibilities, (ii) the first measurement of a 70 ps long polaron lifetime through noise improved pump-probed experiments and (iii) a photon echo experiment with a 15 ps long extracted dephasing time.

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1. Introduction

Semiconductor quantum dots are nanostructures that present a three-dimensional confinement for the carriers (electrons, holes). InAs/GaAs self-assembled quantum dots are a model system that can be grown spontaneously by molecular beam epitaxy with high densities and good size uniformity. Because of the discrete delta-like density of states, they are often referred to as “artificial

atoms” inserted into a solid matrix. The high power, short time scale and tuning capabilities of infrared pulses delivered by CLIO enable the original and enlightening investigation of non-linear infrared properties of these nanostructures.

2. Spectroscopy of InAs quantum dots

2.1. Linear spectroscopy

The electronic structure (level energy position, oscillator strength, polarization) can be investi-

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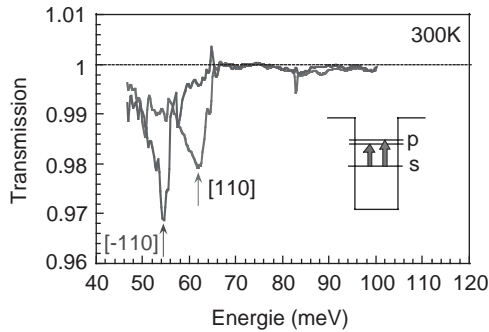


Fig. 1. Infrared absorption of n-doped quantum dots polarized along $[1\ 1\ 0]$ and $[\bar{1}\ \bar{1}\ 0]$ attributed to the s-p transition.

gated through Fourier Transform Infrared Spectroscopy in n-doped or p-doped quantum dots in the mid-infrared spectral range corresponding to intersublevel transitions [1]. In Fig. 1, the absorption of the s-p polaron transition is evidenced around 55 meV.

2.2. Non-linear spectroscopy

Frequency doubling was achieved at $22\ \mu\text{m}$ in resonance with this s-p transition and a $10^{-6}\ (\text{m/V})^2$ second-order non-linear susceptibility was measured [2]. This giant value results mainly from the realization of a double resonance between the pump wave and s, p and high-energy confined d-level.

3. Polaron dynamics

For the first time the polaron dynamics was investigated, using pump-probe spectroscopy of the s-p transition [3]. Special care was devoted to the improvement of the signal-to-noise ratio. The stability of CLIO allowed the measurement of less than 2% transmission modulation (Fig. 2). A polaron lifetime as long as 70 ps at low temperature was measured. The polaron lifetime decreases as CLIO wavelength increases as a consequence of the polaron nature of the elementary excitation in the quantum dots. The lifetime was investigated up to room temperature where its value is still 37 ps. The rather robust temperature dependence of the

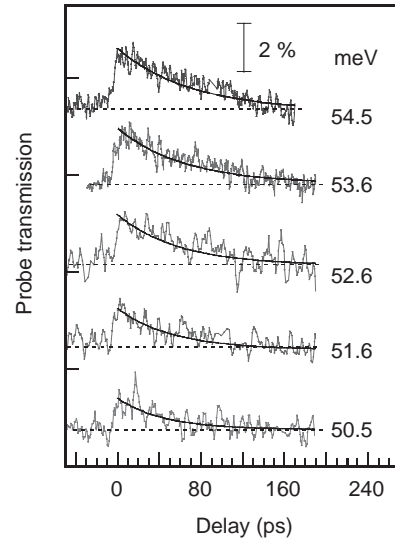


Fig. 2. Polaron time-resolved relaxation at various energies.

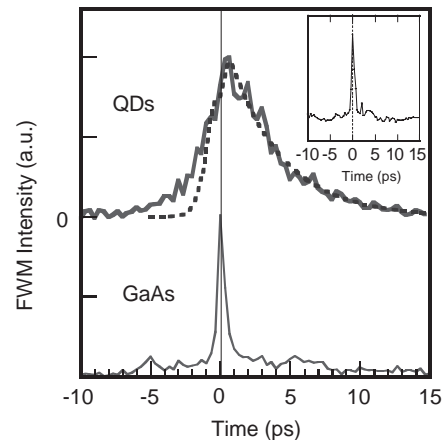


Fig. 3. Four-wave mixing signal at $7.4\ \mu\text{m}$ wavelength in resonance with a quantum dot transition (QDs) and in reference bulk GaAs. The dashed line corresponds to the theoretical FWM signal associated with a dephasing time of 15 ps. Inset: signal at $7.0\ \mu\text{m}$ away from the transition in the quantum dot sample.

lifetime showed that the polaron existence is driven by the instability of its phonon component.

4. Photon echo

We have performed time-integrated four-wave mixing experiment at $7.4\ \mu\text{m}$ wavelength in

resonance with a intersublevel transition between the confined discrete valence states of p-doped InAs/GaAs quantum dots [4]. The quantum dots were inserted into a mid-infrared wave-guide to improve the interaction between the incident pulses and the quantum dots. A dephasing time T_2 of 15 ps is deduced from the delay dependence of the echo signal at low temperature, as seen in Fig. 3.

The non-linear response of the system was calculated to consider a double resonance present in the system and to account for the signal measured at negative delay. The finite 0.5 ps duration of the CLIO pulses was also included in the equations. Because of the inhomogeneous broadening of the transition, a $\exp(-4t/T_2)$

dependence is calculated at positive delays (dashed line). An ultrashort response was obtained out of resonance at $7.0\ \mu\text{m}$ (inset). The dephasing time, corresponding to a homogeneous broadening of $87\ \mu\text{eV}$, is nearly constant with temperature suggesting that dephasing does not occur through phonon scattering.

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