Development of a Stacked-Pulse Ringdown Cavity for Spectroscopy on Self-Assembled Quantum Dots

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High-sensitivity absorption measurements on self-assembled InAs quantum dots are of importance for the investigation of all-optical nonlinearities. We introduce cavity-ringdown spectroscopy into the field of quantum dot physics. With the use of classical cavity-ringdown and the stacked-pulse technique, the time-integrated and the time-resolved absorption spectra can be determined. An enhancement by a factor of $2{\times}10^3$ *is expected with respect to the traditional differential transmission techniques with a cavity finesse of 3140.*

Introduction

The technological evolution towards nanoscaled devices encourages the research of low-dimensional structures: e.g. self-assembled quantum dots (SAQDs). This includes the improvement of the fabrication techniques and the control but also the enhancement of the knowledge of the optical properties of these structures. Especially the discrete energy spectra of SAQDs, due to the 0-D confinement, make the use of SAQDs in quantum dot lasers [1,2] interesting which encourages further research.

The optical characterization of SAQDs usually involves the photoluminescence (PL) spectra and not the investigation of the absorption spectra. In order to obtain a complete insight in the carrier dynamics and (multi-)exciton formation, techniques like pump-probe transmission and differential absorption, which all rely on absorption measurements, are also required. In this presentation we introduce a novel pump-probe optical absorption technique for the investigation of quantum dots (QDs). The absorption coefficient, α, is normally in the order of 10^3-10^4 cm^{-1}. Due to the small height, $d \approx 5$ nm, and coverage, $\varepsilon \approx 1\%$, it is rather hard to measure the absorption. The common approach is to determine the pump-induced absorption by measuring the differential transmission of the probe signal [3-5], $\left|\frac{I - I_0}{I_0}\right|$, which is in the order of 10^{-7}-10^{-5} depending on the dot-size and dot-density.

Differential transmission signals in the order of 10^{-7}, which are required, are hard to measure with this method, which encourages the search for alternative techniques. Samples with stacked QD layers increase the differential transmission signal proportional to the number of layers, as has been used by Birkedal *et al*. [5]. However, the disadvantages of this method are the sample demands involving the homogeneity, thickness and dot density of each layer. These disadvantages can be overcome if a multi-pass pump-probe measurement technique is used. The probe-pulse passes the sample multiple times after a single pump-pulse, similar to a N-stacked layer structure [5]. A drawback of the multi-pass method, however, is the loss of synchronization between pump and probe pulses. In order to overcome this drawback, we introduce stacked-pulse cavity-ringdown (SP-CRD) spectroscopy which preserves the time information of the optical processes and therefore allows us to determine the absorbance, both time-integrated and time-resolved. To our knowledge the SP-CRD technique has never been used before in the field of quantum dot physics.

From Classical towards Stacked-Pulse Cavity-Ringdown

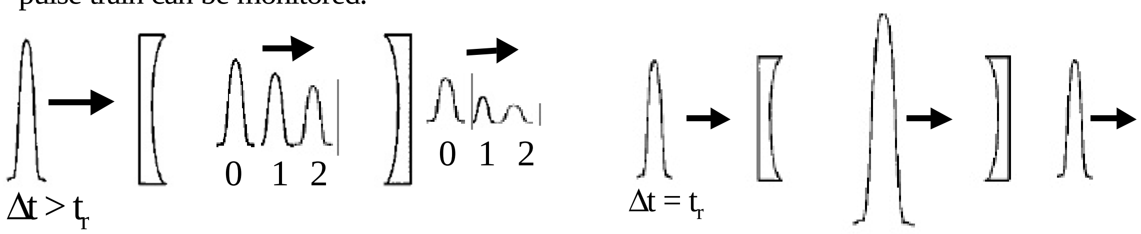
Stacked-pulse cavity-ringdown spectroscopy is based on the non-stacked ringdown technique which will be referred as the classical ringdown technique.

Classical Cavity-Ringdown (CRD) spectroscopy is a high-sensitively technique for absorption measurements and was first demonstrated by O'Keefe and Deacon [6] in 1988. The ringdown cavity is based on the Fabry-Pérot etalon principle and is constructed by a pair of highly reflective mirrors (R > 99.9%) a distance L apart. The injected light, cw [7] or pulsed [6,8], reflects back and forth between the two mirrors, and the intensity decays exponentially with the cavity ringdown time. The ringdown time of an empty cavity, t_0, depends on the finesse of the cavity, $\mathcal{F}$, and is given by:

$$t_0 = \frac{2L}{nc}\frac{\mathcal{F}}{\mathrm{R}} = \frac{2L}{nc}\frac{1}{1-\mathrm{R}^2}, \qquad (1)$$

with n the refractive index of the medium inside the cavity, c the velocity of light and $R=|R|^2$ the reflectivity of the mirrors. With the presence of an absorbing medium the ringdown time decreases allowing to determine the absorbance. Spectroscopy where one measures the cavity ringdown time is insensitive to amplitude fluctuations of the light source, which is a key factor for improved sensitivity.

If a pulsed light source is used for the excitation of the optical cavity, the injected pulses will circulate in the cavity with the cavity round trip time, $t_r = \frac{2L}{nc}$. If the time between two successive pulses, Δt, is shorter or longer than the round trip time, a train of pulses is formed in the cavity, Fig. 1a. A small part of the pulse intensity escapes such that behind the mirrors the pulse train can be monitored.



a) b)

Figure 1. Excitation of a non-resonant cavity by a pulsed light source a) and the excitation of a resonant cavity forming stacked-pulses b). The numbers represent the number of roundtrips of each pulse.

If we now want to use the CRD technique in combination with pump-probe induced absorption measurements, such that the probe-pulse is stored in the cavity, the synchronization is lost except when, $t_r = \Delta t$, the pulses will be stacked on top of each other, i.e. pulse stacking [9], forming Stacked-Pulse Cavity-Ringdown.

In **Stacked-Pulse Cavity-Ringdown** spectroscopy, the length of the cavity around the absorbing medium has exactly the same length as the cavity of the mode-locked laser. In the frequency domain, the comb of laser modes of the mode-locked laser is tuned into resonance

with the Fabry-Pérot resonances of the measurement cavity. In this case, all the injected pulses are stacked on top of each other to form a single pulse as depicted in Fig.1b.
The number of pulses inside the optical cavity depends on its finesse and stability. If we assume a stable two mirror confocal cavity [10] which is excited by a train of pulses along the optical axis of the empty cavity, the electrical field inside the cavity will be a summation of the individual fields of the pulses, $E_{in,i}(w,t) = E_{0i}e^{j(w_i+f_i)t}$, weighted by the mirror reflectivity and transmittance. With the injection of n pulses, such that the spatial separation of the individual pulses in the external pulse train is equal to the cavity round trip length, of equal amplitude E_0, frequency ω and phase φ the field can be written as

$$E_{cav}(w,t) = \lim_{n\to\infty} \frac{1-\mathrm{R}^{2(n+1)}}{1-\mathrm{R}^2} \mathcal{T} E_{in}(w,t) = \frac{\mathcal{T} E_{in}(w,t)}{1-\mathrm{R}^2}. \quad (2)$$

In the case $T=|\mathcal{T}|^2 = 0.001$, $R=0.999$ and non-absorbing mirrors, the intensity stored inside the optical cavity is a factor 10^3 higher than the intensity of a single incident pulse. The electrical field behind the second mirror becomes, $E_{out}(w,t) = \mathcal{T} E_{cav}(w,t) = E_{in}(w,t)$. Notice that the cavity has a transfer function of unity.
With the inclusion of an absorbing medium with an absorption α, thickness d and coverage ε, a transmittance e^{-ade} is introduced such that eq.(2) becomes:

$$E_{cav}(w,t) = \frac{\mathcal{T} E_{in}(w,t)}{1-\mathrm{R}^2 e^{-2ade}}. \quad (3)$$

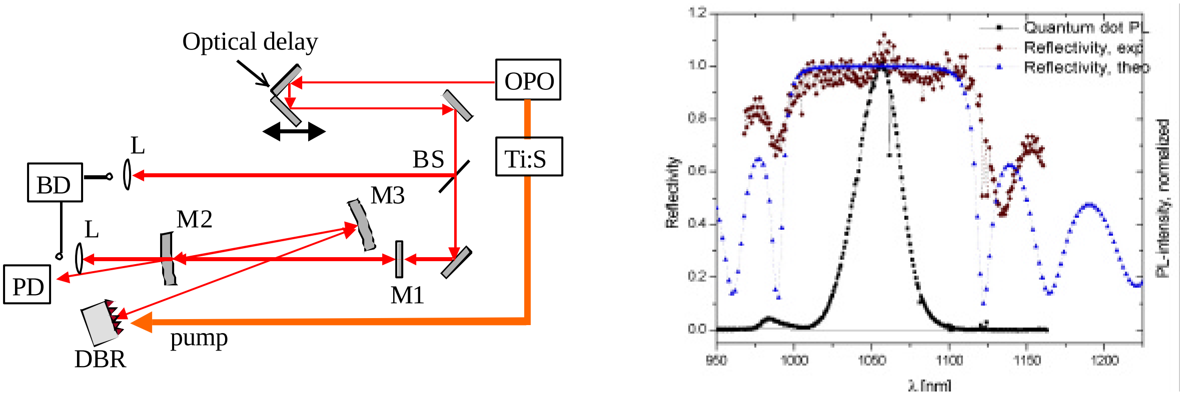
Notice, the probe pulse will pass through the absorbing medium twice which introduces a quadratic contribution. To derive the influence of the absorption we measure the differential transmission of the light leaking out of the cavity, $\left|\frac{\Delta I}{I_0}\right| = \left|\frac{I_{abs} - I_0}{I_0}\right|$, with I_{abs} and I_0 the measured intensities behind the second mirror with and without absortion. The differential transmission can be written as function of the finesse of the cavity,

$$\left|\frac{\Delta I}{I_0}\right| = \left|\left[\frac{p\mathrm{R}}{\mathcal{F}} \cdot \frac{1}{1-\mathrm{R}^2 e^{-2ade}}\right]^2 - 1\right| \approx \left|\left[\frac{p\mathrm{R}}{\mathcal{F}} \cdot \frac{1}{1-\mathrm{R}^2(1-2ade)}\right]^2 - 1\right|. \quad (4)$$

For R = 0.999, $\mathcal{F}$ = 3140, and $2ade$ = 10^{-5}, single pass absorption [6], the differential transmission signal for the stacked-pulse ringdown cavity becomes $2{\cdot}10^{-2}$. An enhancement by a factor of $2{\cdot}10^3$ with respect to a single pass is obtained.

Experimental realization

The quantum dot sample under examination is grown by molecular beam epitaxy (MBE) and consists of a 20-periods GaAs/AlAs multilayer structure which forms a high-reflective diffracted Bragg reflector and a single self-assembled InAs quantum dot layer on top. This configuration allows the integration of the sample into the optical cavity as a high reflective end mirror. Hereby, complicated anti-reflection coatings on the sample as well as transmission of the probe pulse through the substrate are avoided.
A schematic lay out of the set up and the DBR reflectivity are depicted in Fig. 2. We use a picosecond Optical Parametric Oscillator (OPO) tunable between 1050 and 1330 nm to probe the SAQD sample. The pulses from the Ti:Sapphire laser which are used to pump the OPO, will also be used as the pump source for the measurements. A balanced



a) b)

Figure 2. Illustration of the experimental stacked-pulse ringdown cavity for the spectroscopy on quantum dots a) and the characterization of the DBR reflectivity including the photoluminescence of the quantum dots b).

detector (BD) is used to cancel the laser noise, >70dB noise reduction. The photo-detector in combination with an electronic feedback loop is needed to stabilize and to lock the measurement cavity to the laser modes.

In figure 2b, the reflectivity of the DBR and the PL-spectrum of the quantum dots are depicted. The peak of the PL-spectrum is located at the center of the DBR waveband, which ensures a good optical interaction of the quantum dots with the DBR. The feature on top of the measured reflectivity is due to photoluminescence of the quantum dot layer.

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