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Microphotoreflectance spectroscopy – a modulation technique with high spatial resolution

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There is presented an experimental setup for the measurements of photomodulated reflectivity spectra of low-dimensional semiconductor structures with a micrometer spatial (plane) resolution. The setup has been developed as an extended and improved version of a standard bright configuration, *i.e.*, where the probe beam is provided directly by a broad band light source (*e.g.*, halogen lamp) and then it is dispersed after being reflected off the sample. It gives typically the plane resolution, expressed by the spot size of the beams on the sample surface, on the level of single millimetres. Introducing optics, based on a long working distance and a high numerical aperture microscope objective, has allowed decreasing the spot size by three orders of magnitude into the micrometer range for both the probe and the pump beams. The optimization of microphotoreflectance signal to noise ratio has made it possible to detect the normalized reflectivity coefficient changes ($\Delta R/R$) from an ultrathin single quantum well formed of the wetting layer in the structure with self-assembled InAs/GaAs quantum dots and from single pillar microresonators of the lateral sizes in the range of single micrometers.

Keywords: photoreflectance, high resolution, microphotoreflectance.

1. Introduction

Quickly emerging new semiconductor technologies enabling an artificial creation of objects in a micro- or even nanoscale are the main driving forces in the development of high spatial resolution optical measurement techniques, which give the spectroscopic information on single individuals and at the same time a possibility of testing their uniformity over a larger area. Typically, it concerns photoluminescence (PL) as it is easily applicable and has the spectral response in the form of single peaks, which makes it directly interpretable and hence available for a very wide range of research fields (it can be even exploited by researchers being non-experts in spectroscopy). However, PL measurements suffer from a number of disadvantages and limitations (these are: *i*) probing the lowest energy states only, *ii*) non-reflecting either the real density of

states or oscillator strength of the optical transitions) and there are many specific cases where it needs to be supported or replaced by more sophisticated methods. One of the solutions is a modulation spectroscopy, *i.e.*, an absorption-like technique probing always the whole band structure (including the higher order state transitions) with an extraordinary sensitivity due to its derivative nature of the spectra (making it possible to observe low intensity transitions, like those which are nominally forbidden). However, performing modulation spectroscopy measurements with a high spatial resolution causes specific technical problems which need to be overcome: *i*) the probe beam is usually a non-coherent light which is difficult to focus to a small enough diameter without loosing much of its intensity; *ii*) in the most direct experimental configuration (photoreflectance – PR) there are two beams which are required to be controlled and provided independently and then focused to the same size in the same place on the sample surface; *iii*) increasing the spatial resolution (so decreasing the spot size) reduces the total light intensity reaching a detector. The latter impairs the signal to noise ratio of the measured quantity ($\Delta R/R$), which, however, can be improved by some modifications in the optical path (which will lead to a better light collection on the sample; nevertheless, the power density on the sample surface cannot be increased too much, due to the risk of local heating) or by improvements in the detection part of the setup.

Photoreflectance has been demonstrated to be very sensitive to optical transitions in small quantum-dot-like objects, whether it is used either for a large number of dots [1–4] (the signal from each dot contributes to the final spectral response, which, however, suffers from the inhomogeneous broadening due to the non-identical properties of the dots within the whole ensemble, which conceals the single dot features) or for single dots, which requires further experimental enhancement and improvements (*e.g.*, AC voltage modulation via electric contacts and exploiting the Stark shift effect [5]). On the other hand, a high resolution modulation spectroscopy has been successfully used to investigate the properties of the device structures (such as transistors or lasers), including QW or bulk-like layers, in a dark experiment configuration (already dispersed probe beam comes from the monochromator onto the sample) where the pump beam is focused to a range of micrometers [6, 7] or for the mapping of a whole wafer properties (for a sample with an ensemble of self-assembled QDs even [8]) but with a millimetre resolution.

Hereby, there is presented the most direct approach to photoreflectance measurements (or other modulation spectroscopy) with micrometer spatial resolution. The setup design is based on a standard bright configuration macro photoreflectance setup, developed into a high resolution version (with shifting both the probe and the pump beam to a microscale). We show not only the details of the experimental configuration but also the emerging problems which need to be solved and possible limitations. By making a series of tests with different resolutions and detection systems, the prospects for the development and use of the technique in the future are discussed.

2. Experimental setup

The setup is schematically shown in Fig. 1. It is a modified and extended version of the setup for low resolution (*i.e.*, in a millimetre scale) measurements. The white light probe beam comes from a tungsten halogen lamp (HL), which is focused on a pinhole (PH), in order to create a small size spot, imaged on the sample surface. A quasi-parallel beam is formed by a long focal length achromatic lens in front of it, introduced into the main optical axis by a 50:50 Ag-based beam splitter and then it is focused in the sample plane (cryostat mount – CM) by a high numerical aperture and achromatic microscope objective (Ob) with a long working distance. The final size of the spot on the sample surface depends on the diameter of the pinhole and the ratio of the focal lengths of the lens (making the quasi-parallel beam) and the objective. However, the lowest spot diameter is restricted by two factors: the diffraction limit and the minimum power density reaching the sample surface, below which the signal is too weak to be detected. Due to this compromise there has been achieved the spot diameter of single micrometers for the white light. The laser (pump beam) spot size, which is much easier to bring to the range of diffraction limit sizes (approximately 2 μm in our case), does not determine the real signal collection area. The effects of carrier diffusion make the real modulation area significantly larger than the pump beam illumination area, therefore, for the realistic and controlled spatial resolution, both the probe and the pump beam need to be focused to the micrometer range. As a pump beam there is applied a 660 nm line of a semiconductor laser (La), which is

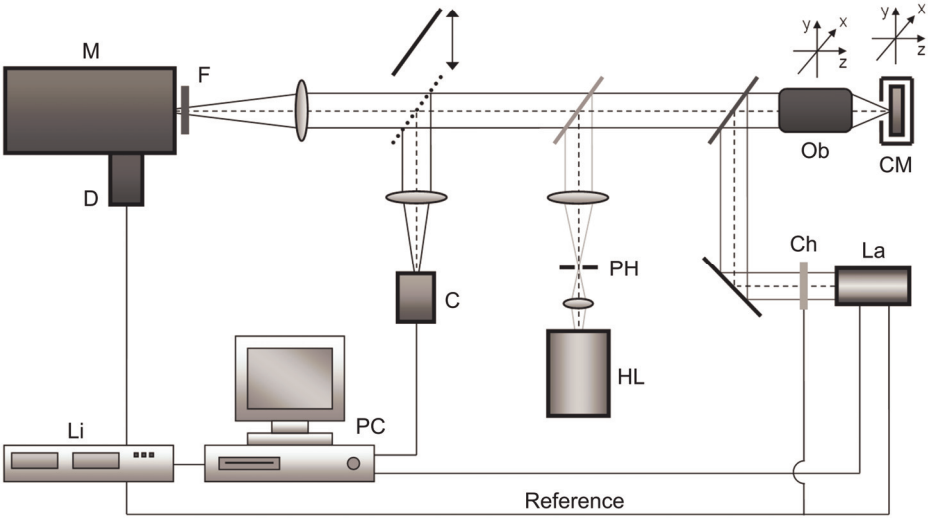

Fig. 1. Microphotoreflectance setup scheme: CM – sample (cryostat mount), Ob – microscope objective, La – semiconductor laser, Ch – optical modulator ("chopper"), HL – tungsten halogen lamp, PH – pinhole, C – monitoring camera, F – red glass filter, M – monochromator, D – detector, Li – lock-in amplifier, PC – personal computer.

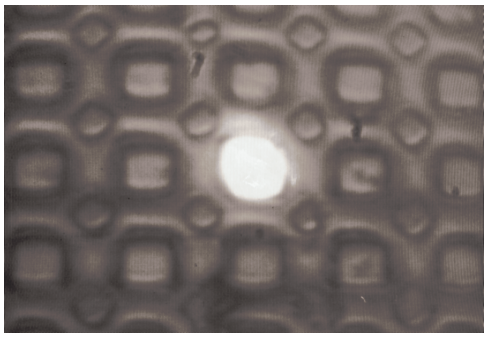

Fig. 2. Monitoring camera view of the sample surface with mesas (pillar microresonators of 10 μm lateral size) with the white light spot visible in the middle. A sample with different mesa sizes has been used for the white light spot diameter evaluation.

put into the optical axis by a cold mirror (reflecting the laser beam, but at the same time being transparent for the infrared light of the probe beam reflected off the sample) and focused in the sample plane by the same microscope objective (Ob) as the white light beam. The laser beam spot size on the sample surface is naturally of the order of 2 μm and it can be enlarged, to increase the overlap area with the white light spot, by introducing additional optics into the laser principal axis. The microscope objective (Ob) collects the light reflected off the sample and forms a quasi-parallel beam again, which is then focused on the monochromator entrance slit by an achromatic lens. The cutoff filter (F) is applied to prevent the disturbance of the detected signal by the scattered laser light. Additionally, a monitoring camera is installed (the light can be introduced by a swing mirror), which gives the possibility of direct observation of the sample surface (Fig. 2) and a very precise selection of the investigated area (*e.g.*, single patterned mesas or specific device parts). A basic selection of the place investigated on the sample surface is obtained by a cryostat (CM) movement with a micrometer resolution. The exact control of the position of spots is achieved by slight movements of the microscope objective, which is mounted on a table with a manual micrometer regulation and a piezo-control giving overall accuracy position selection of 20 nm. The light is dispersed by a single grating 0.5 m focal length monochromator and detected by a photodetector (D) taking advantage of a lock-in amplifier (Li) technique. The pump laser beam modulation is accomplished by an optical chopper (Ch) placed in the laser path or by a computer controlled internal modulation of the semiconductor laser (La). The detection system is based on two thermoelectrically cooled pin diodes with additional preamplifiers: InGaAs and Si. They differ in sensitivities and spectral range coverage, so a compromise is always necessary. The Si diode works in a spectral range of 200–1100 nm and longer wavelengths require switching to the InGaAs diode (800–1600 nm) which is, however, approximately by two orders of magnitude less sensitive.

3. Test measurements

There have been performed a few test measurements to check if the system works properly and to evaluate the collected signal quality. The first group of mensurations have been done on the previously studied sample [9] with a single layer of InAs self-assembled quantum dots, based on a GaAs substrate and designed for a long wavelength emission. The overall structure characterization, based on an ordinary (*i.e.*, macro) photoreflectance and photoluminescence measurements, is presented in Fig. 3. The PR spectrum reveals strong GaAs bulk feature and two optical transitions in the quantum well (WL1 and WL2) formed from a thin layer of a quantum dot material (1.5 ML ~ 5 Å thick InAs wetting layer). The lowest transitions originate from quantum dots ground and excited states (QD1–QD5) and are confirmed in a photoluminescence emission spectrum. Due to the much higher sensitivity of the Si diode, which covers spectral range above 1.2 eV, and relatively strong photoluminescence intensity from quantum dots, which is a spurious background for the photoreflectance spectrum, the tests in a high spatial resolution setup have been performed in the range of InAs wetting layer quantum well and bulk GaAs optical transitions, *i.e.*, in the spectral working range of the two detector diodes where no PL background is present. The results are shown in Fig. 4. The uppermost spectrum has been obtained as a reference in a standard macro configuration with a spot diameter size of 5 mm on the InGaAs detector and it is actually a part of the PR spectrum presented in Fig. 3. Consecutive spectra have been obtained in the high resolution system with different experimental details. For the spot size of 100 μm the signal has been measured on both detectors (Si and InGaAs diodes) under the same experimental conditions. One can clearly observe the two transitions in a wetting layer

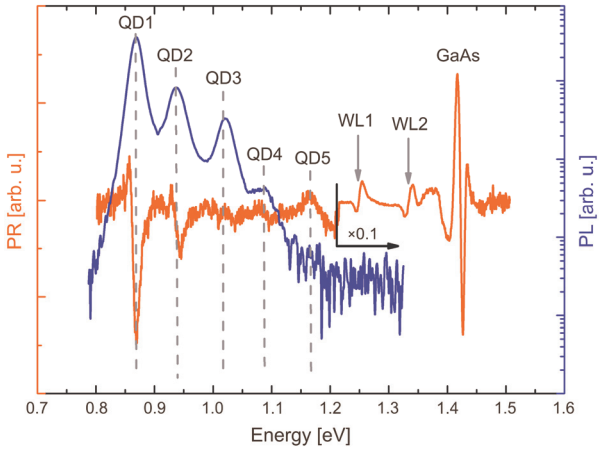

Fig. 3. Room temperature macro photoreflectance and photoluminescence spectra of the GaAs based structure with self-assembled InAs quantum dots on a thin wetting layer [9].

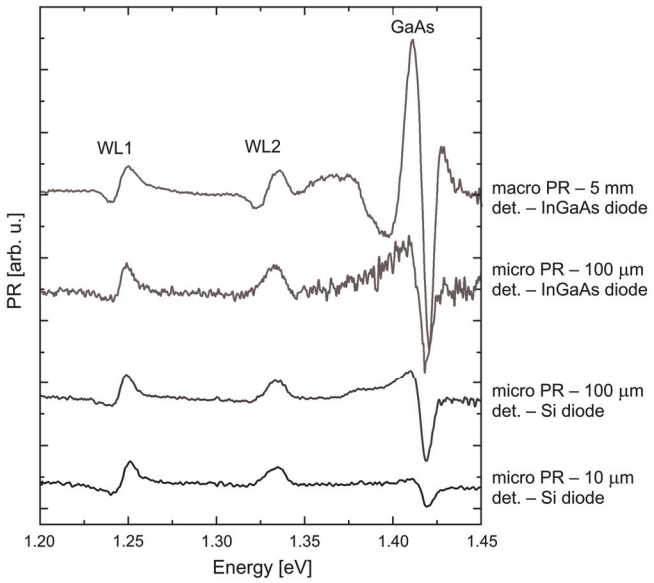

Fig. 4. Room temperature photoreflectance (PR) spectra obtained in a macro PR setup (the upper one) and in microPR measurements with different experimental configurations (the remainder) on the GaAs based structure with self-assembled InAs quantum dots on a thin wetting layer.

quantum well and the strong resonance corresponding to the bulk transition in GaAs, as in the macro PR experiment. The spectra have been normalized to the WL ground state optical transition intensity, due to the substantial differences between macro- and microphotoreflectance (μPR) measurement conditions (such as power densities of both beams on the sample surface or their spots overlapping), which would make the direct spectra comparison difficult. There are no significant differences between both spectra for the 100 μm spot, besides the apparent improvement in the signal to noise ratio in favour of the Si diode. Since it is more effective, the consecutive trial measurements with smaller white light spot sizes have been performed on the Si detector only. A selected spectrum for a 10 μm spot diameter is presented at the very bottom of Fig. 4. With such a small spot diameter it has been still possible to detect a strong photoreflectance signal with distinctly visible optical transitions from the quantum well and the bulk material. Moreover, taking advantage of the measurement time constant increase (compensating in part the ΔR and R signal decrease for smaller spot sizes), it was possible to achieve a good signal to noise ratio, comparable with the one for the spectrum obtained with a 100 μm spot size. This shows that it is possible to have a PR signal for the spot size diameters even below 10 μm. Nevertheless, our experiments have shown that it results then in a very strong signal to noise ratio decrease and the overall spectrum quality deterioration.

It is worth noting here that although the QD transitions occur in the spectral range not covered by the more sensitive Si detector, in general, our approach should also be effective for QD states investigation. Nevertheless, beside the importance of the spectral

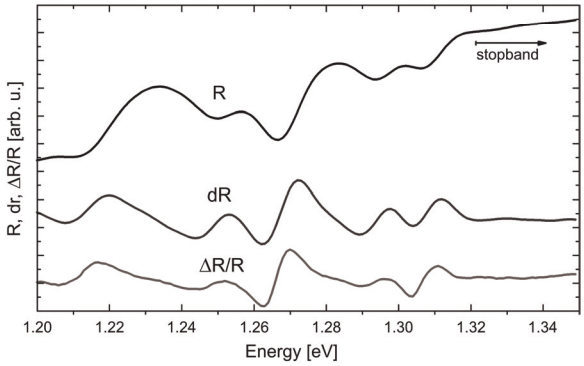

Fig. 5. Comparison of reflectivity *R*, its numerical derivative d*R* and microphotoreflectance spectra for a 10 μm lateral size square shaped GaAs/AlAs Bragg reflector pillar microresonator (microcavity) in the range below the stopband.

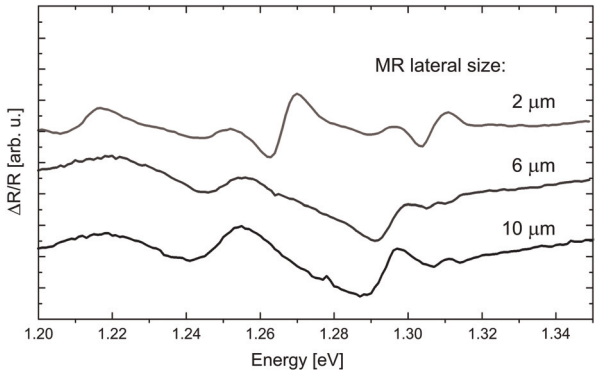

Fig. 6. Microphotoreflectance spectra for single pillar microresonators of various lateral sizes.

position with respect to the setup detectivity characteristics, issues such as strong photoluminescence background versus weak photoreflectance response related to QDs, result in additional difficulties which need to be overcome. One of the solutions is to apply a double modulation system [10] (both the probe and the pump beam are mechanically chopped, whereas the signal is detected at the sum-frequency). This allows subtracting entirely any kind of background as it has a different frequency than the one of interest, even if its intensity (amplitude) exceeds one of the photoreflectance signals by a few orders of magnitude. Capabilities of this solution as regards application to quantum dot structures are under test at our laboratory and are going to be a subject of an independent publication. Another option, which could be helpful in the case of microphotoreflectance of quantum dots, is the use of a tunable laser as a probe beam (providing good beam focusing possibility), which allows the monochromator to be removed from the setup and makes photoluminescence background featureless and easy extractable as a signal offset (to its certain values).

Other tests have been performed on a structure with patterned mesas which are GaAs/AlAs Bragg reflector square-shaped single lambda GaAs microcavities (microresonators – MR) of different lateral sizes ranging from 2 to 10 μm (similar to those whose photoluminescence properties have been reported previously [11]). Figure 5 shows a comparison of reflectivity spectrum R, its numerically taken derivative dR and microphotoreflectance (*i.e.*, experimentally obtained derivative) for a single 10 μm large pillar MR. It concerns the range below the stopband where a set of oscillation-like features (of interference origin) appears in reflectivity. It is apparent that the dR and $\Delta R/R$ spectra are basically the same. This proves the effectiveness of microphotoreflectance as a modulation kind of an absorption-like experiment on a single microcavity. Such a signal could be obtained from smaller size MRs going down to 2 μm (see Fig. 6 showing μPR spectra for three different cavities).

4. Conclusions

There has been constructed a high spatial resolution setup for photoreflectance measurements, based on a bright configuration macro photoreflectance system. For the sample surface probing area of single micrometers there have been obtained a good signal to noise ratio modulated reflectivity signal of optical transitions from a thin InAs wetting layer quantum well and a bulk GaAs in a planar QD structure and from single pillar microresonators. The results prove the capabilities of microphotoreflectance for small-scale area optical investigation (*e.g.*, applied to small patterned objects or for structure planar uniformity mapping). The main limitation has appeared to be the probe beam spot size and the total signal intensity competition. However, the increase in the detection efficiency together with the limitation to the light loss in the optical path, can bring hope for further improvements and open up the way into the sub-micrometer resolution photoreflectance measurements (*e.g.*, in a solid immersion lens approach [12]). The subsequent development of high resolution photoreflectance spectroscopy can give the possibility of studying the single quantum dot or single nanocrystal optical properties, or investigating the solid state quantum electrodynamics effects in single semiconductor microcavities.

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