Raman and AFM Profiling of Quantum Dot Multilayers

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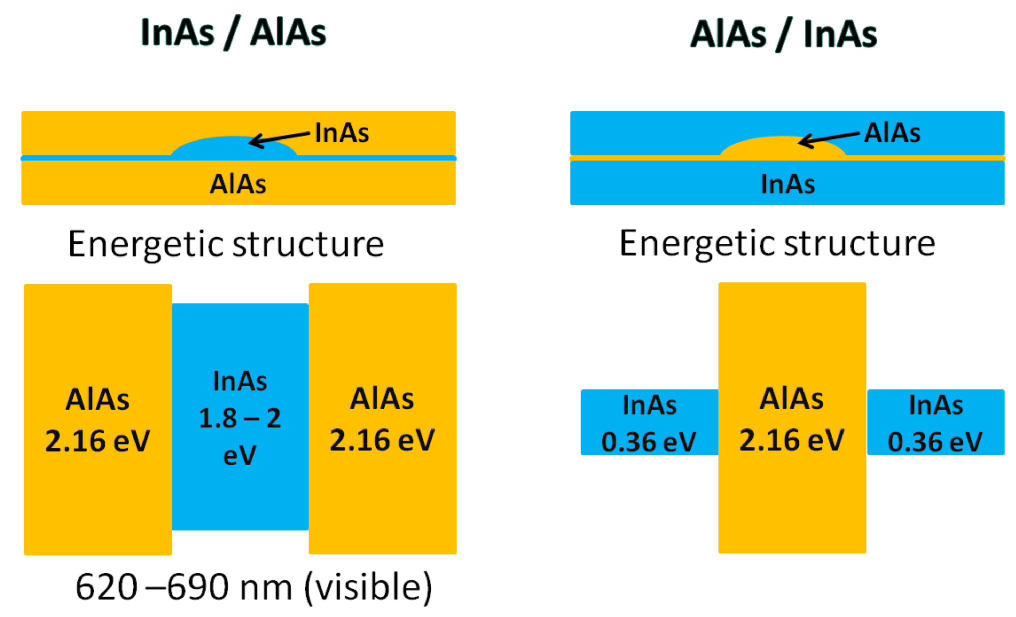
Introduction

●InAs quantum dot (QD) systems are used in photodetectors, high temperature lasers, and single photon sources [1, 2]. They are considered as promising candidates to improve efficiency of III-V solar cells [3].
●The AlAs matrix allows tuning the InAs QD energy gap in the range of 1.8 – 2 eV (620 – 690 nm) [4].
●The InAs/AlAs system is poorly studied in comparison to the InAs/GaAs one. Having in mind device fabrication and optimization, knowing the properties of the QDs, including morphology and phonon spectra governing electron scattering is of great importance.

At the same time, the AlAs matrix is prone to oxidation [5] and therefore cannot be handled the same way as GaAs. In this work we discuss approaches for spatially resolved study of InAs QD multilayer structures incorporated in AlAs or $Al_{0.75}Ga_{0.25}As$ matrix using atomic force microscopy (AFM) and Raman spectroscopy as well as inverted structures (AlAs QDs in InAs matrix) under ambient conditions.



Experimental Details

InAs/Al(Ga)As (AlAs/InAs) QDs were grown with molecular beam epitaxy (MBE) on n+ (001)-oriented GaAs. The total amount of InAs (AlAs) was 2.2 (2.4) ML for the QD growth. The substrate temperature was 500ºC (460ºC). GaAs interlayers were grown after every 500 nm of the QD layers.

AFM measurements were performed in tapping mode (Agilent 5420) from the side. Sample preparation:
●Cleaving (Fig. 1a) results in atomically flat surfaces along the <110> direction.
●Ion beam cutting (Fig. 1b) (Leica EM TIC 3X) with Ar^+ ions at 6 keV energy, and room temperature, -80, and -130° C. Samples were covered by a protective mask.

Raman measurements (Horiba JY, LabRam HR800). 514.5 and 514.7 nm excitation wavelengths, power of 0.1 – 0.5 mW, spectral resolution of 2.5 cm^{-1}.
Sample preparation (Fig. 1c): Wedged surfaces were prepared by polishing the samples at an angle of 5 to 7 degrees with respect to the layer plane. Subsequent polishing in water with a diamond foil of different grain size (3 µm, 1 µm, 0.5 µm, 0.1 µm).

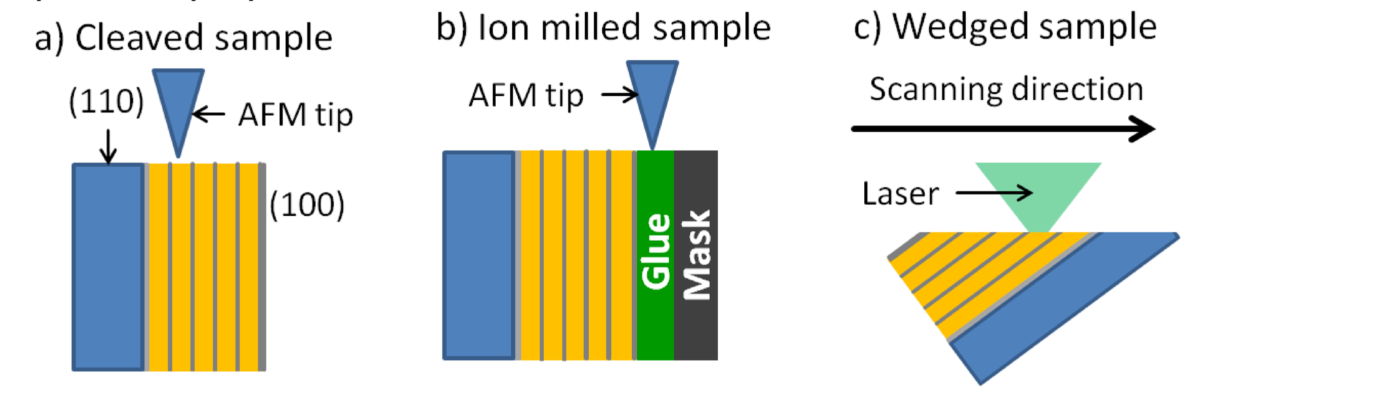
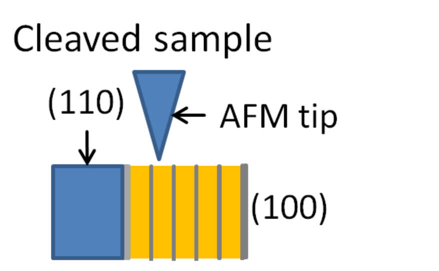


Figure 1. Structure of samples for AFM and Raman measurements.

AFM Study

Cleavage (InAs QDs in Al(Ga)As matrix)



● AlAs matrix volume increases due to oxidation (height up to 15 – 30 nm with respect to the substrate).
●$Al_{0.75}Ga_{0.25}As$ matrix is much less prone to oxidation and the increase in matrix height is only 1 nm.
●Minima correspond to the QD position.

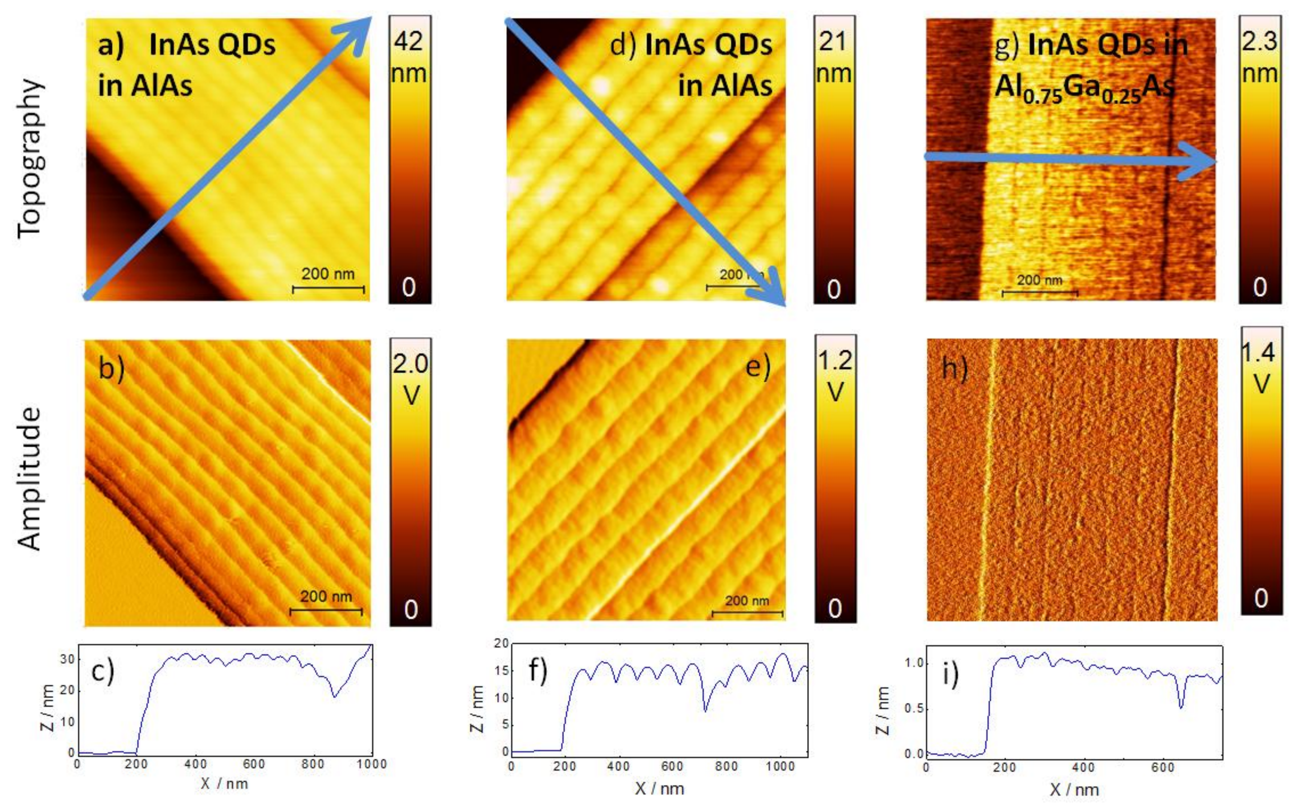
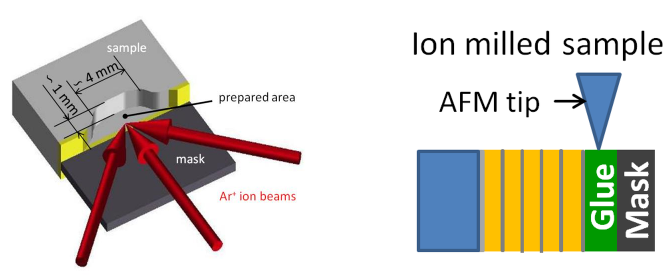


Figure 2. AFM images of cleaved InAs/Al(Ga)As samples.

Ion Beam Cutting (AlAs QDs in InAs matrix)

Cleavage fails due to defects. Ion beam cutting used. Cooling in order to minimize expected sample damage by Ar^+ ions.



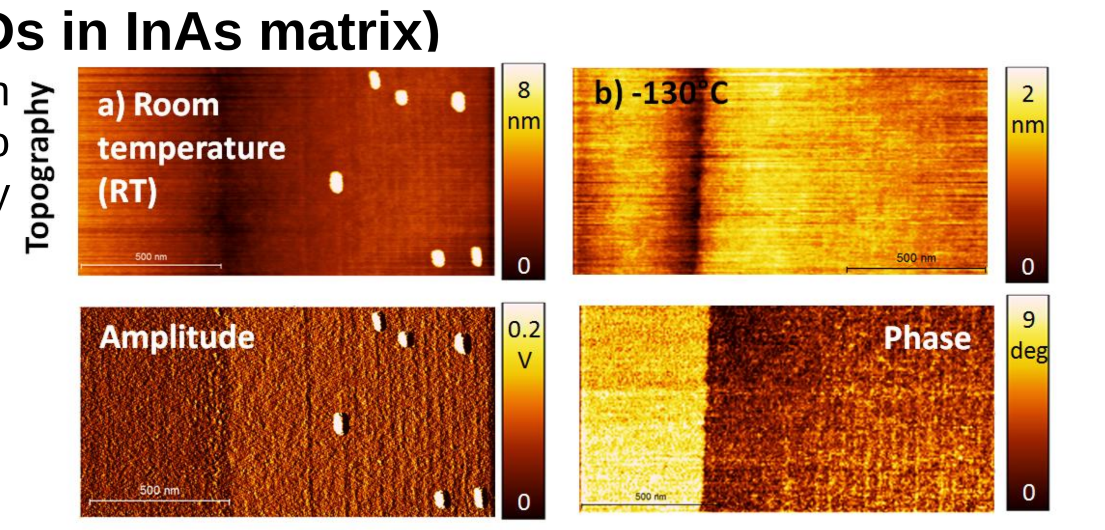


Figure 3. AFM images of AlAs/InAs samples ion milled at room temperature and under cooling (ion energy 6 keV).

●Periodic structure observed corresponds to AlAs QD layers separated by InAs.
●At -130ºC AlAs QDs are not clearly detected (surface roughness is 0.1 nm, the noise limit of the AFM).
●Raman spectroscopy analysis indicates damage introduced by ions.
●LO modes forbidden, selection rules are lifted for ion milled samples. Cooling reduces this effect (reduces damage of the sample).

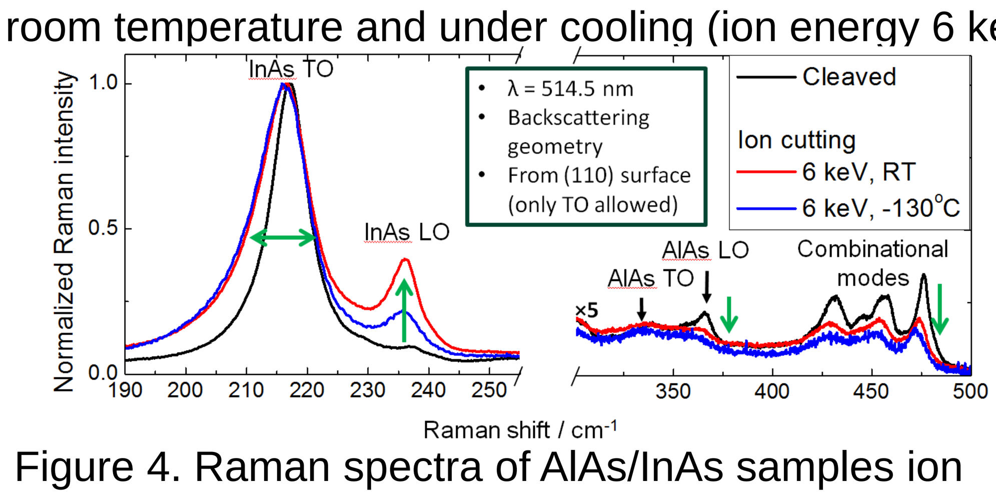
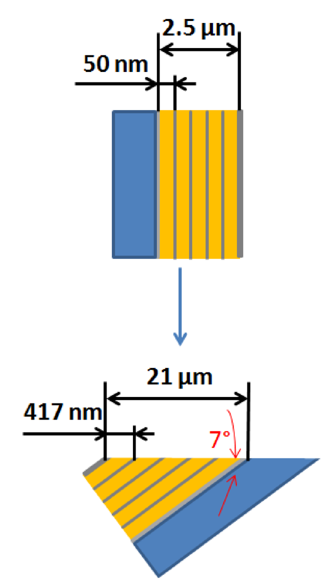


Figure 4. Raman spectra of AlAs/InAs samples ion milled at room temperature and under cooling.
→ - evidence of defects introduced by ion cutting.

Raman Profiling

50 nm nominal period of QD layers

Wedging at an angle of 5–7 degrees with respect to the sample plane increases the effective period of the QD multilayers above the resolution of the micro-Raman system (500 nm). Allows for simultaneous measurement of LO and TO modes.



●The Raman profile matches the optical image.
●AlAs phonon profile is out of phase with that of GaAs interlayer.
●The QD signal is correlated with the matrix phonon.
●Individual QD layers are not resolved.

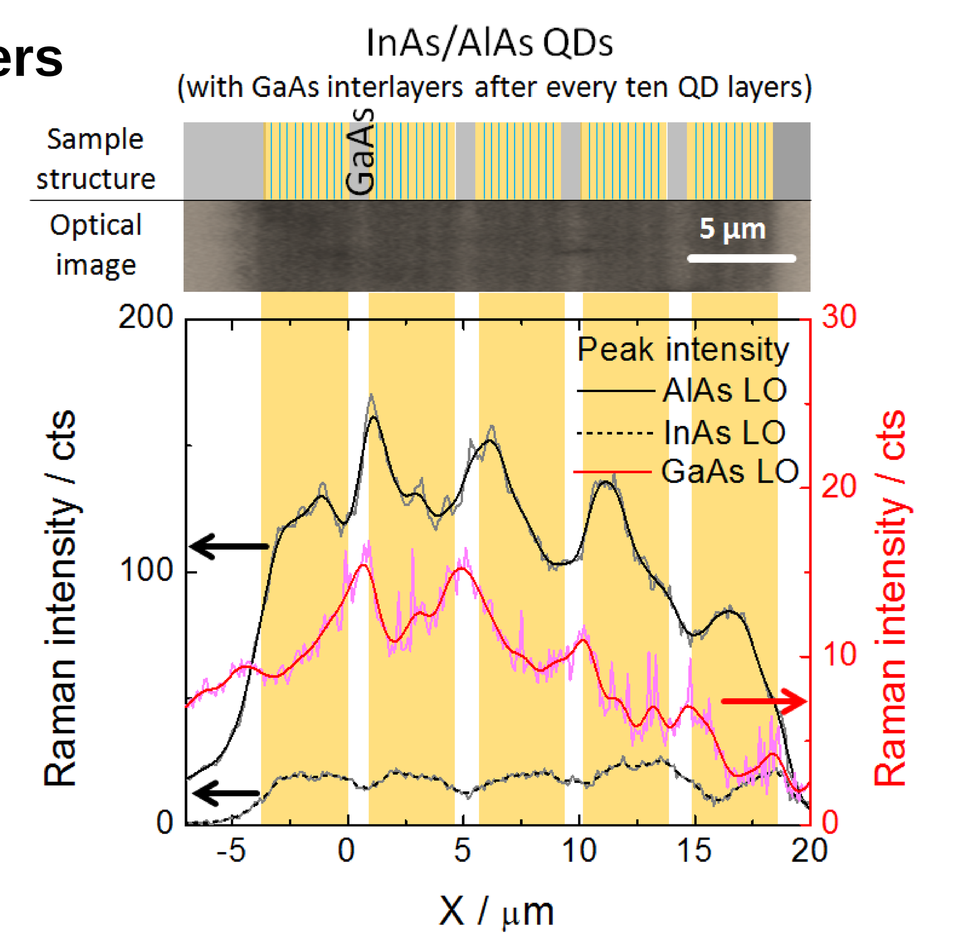
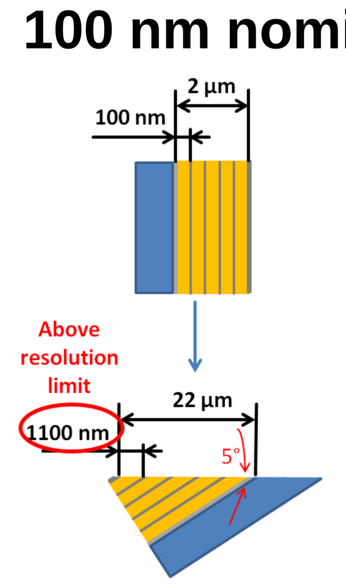


Figure 5. Raman profile of a wedged InAs/AlAs sample.
— - InAs QDs; ▇ - AlAs matrix

100 nm nominal period of QD layers



- InAs QDs;
Matrices:
- AlAs;
- $Al_{0.75}Ga_{0.25}As$

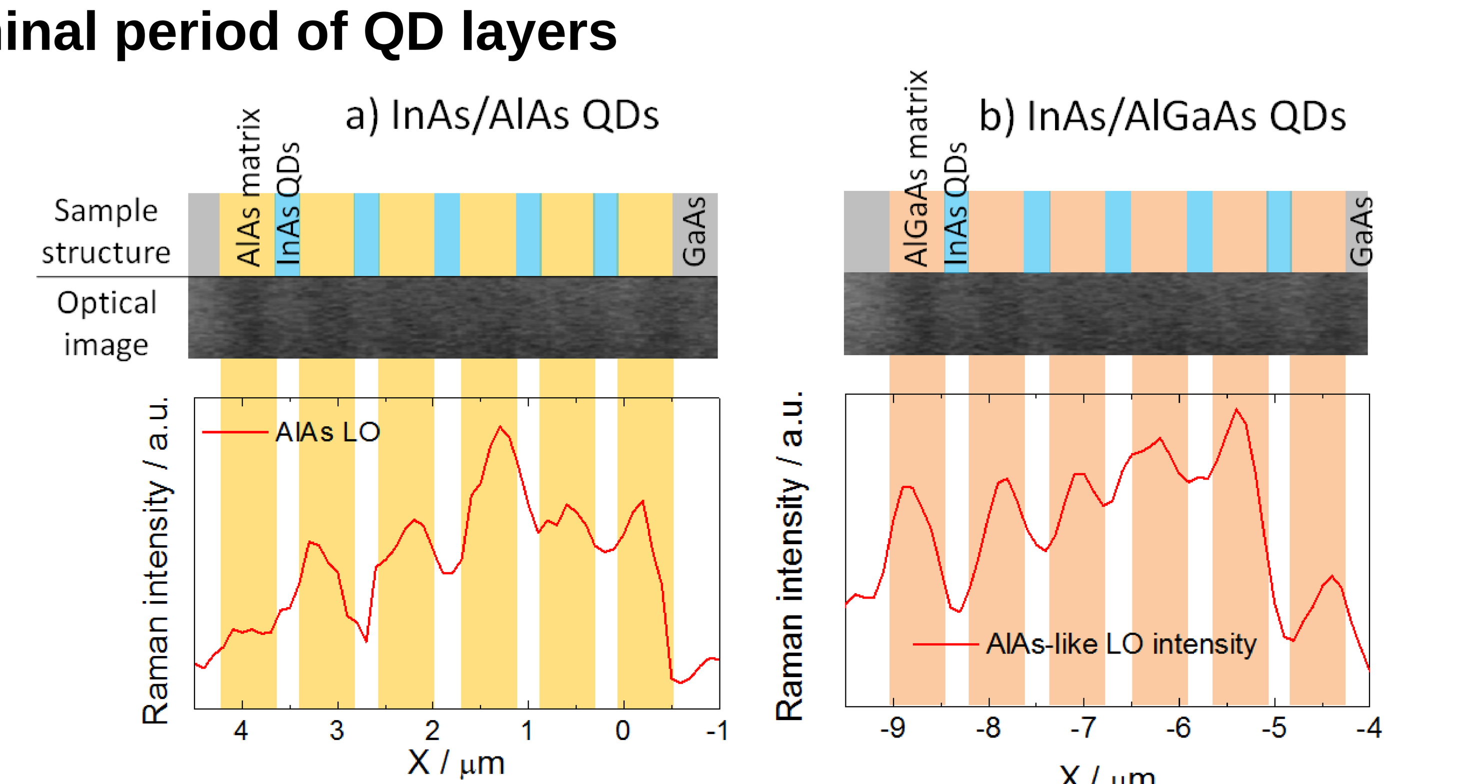


Figure 6. Raman profile of a wedged InAs/Al(Ga)As samples.

●The effective spacing between individual QD layers exceeds 1 µm.
●We can locate individual matrix and QD layers with optical microscopy and Raman.

This method would be able to provide us layer-by-layer monitoring of the changes in InAs QD phonon replica.

Summary

●AFM measurements require direct access to the QDs. We studied several preparation techniques for exposing the QDs.
●Cleaving as the most non-invasive technique is preferable for AFM study of the multilayers, but it requires a high crystalline lattice quality.
●Ion beam cutting is an alternative solution. However, ion beam cutting unavoidably introduces defects to the crystalline lattice as evidenced by Raman spectroscopy. This damage can be reduced by cooling during preparation.
●In combination with Raman spectroscopy these preparation techniques open perspectives for nano-Raman characterization of single nanostructures.
●Raman profiling is limited by the optical resolution limit. Wedging samples at a low angle with respect to the sample surface increases the effective period of the quantum dot layers allowing us to define the positions of individual quantum dot layers in the Raman profile.

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Acknowledgements This study was partially supported by the Russian Foundation for Basic Research (RFBR,Grant No. 13-02-00063), the Program of the Presidium of the RAS-2010 (project 24.27), SB RAS (Grant No. 134), DFG (project ZA146/22-1), and IRTG (Grant No. GRK 1215) and DFG-Research Unit 1713.