O 33: Photonics and Nanooptics I: Infrared Spectroscopy

Time: Tuesday 10:30–13:30 Location: H4

O 33.1 Tue 10:30 H4

Directional Pt-Re nano-antennas for surface-enhanced Raman spectroscopy of suspended ultraclean carbon nanotubes — •Christian Bäuml, Nicola Paradiso, Tobias Korn, Christian Schüller, and Christoph Strunk — University of Regensburg

We developed a new kind of directional optical nano-antennas based on arrays of nano-strips of platinum-rhenium bilayers. The arrays are designed in order to display a plasmonic resonance when excited with light at around 633 nm with polarization directed orthogonally to the strips. We demonstrate that these structures retain their plasmonic resonance even if subjected to very high temperatures (850°C). This feature allowed us to dramatically amplify the Raman signal on carbon nanotubes (CNTs) grown on top of the antennas via chemical vapor deposition. Such overgrowth technique is nowadays the standard method to produce clean devices for quantum transport experiments. Our antennas are thus a promising building block for hybrid devices that enable amplification of optical signals detected on the very CNT portion investigated in transport experiments by the same device.

O 33.2 Tue 10:45 H4

Plasmonic Enhancement of Infrared Vibrational Signals: Nanoslits versus Nanorods — •Christian Huck[1], Jochen Vogt[1], Michael Sendner[1,2], Daniel Hengstler[1], Frank Neubrech[1,3], and Annemarie Pucci[1,2] — [1]Kirchhoff Institute for Physics, Heidelberg — [2]InnovationLab GmbH, Heidelberg — [3]4th Physics Institute and Research Center SCoPE, Stuttgart

We report on investigations of nanoslit arrays as a substrate for surface-enhanced infrared absorption (SEIRA). Nanoslits, fabricated by electron beam lithography and subsequent argon ion etching, feature strong resonances in the infrared spectral range, according to Babinet's principle. The plasmonic resonances are compared to those of solid nanoantennas, with exactly the same dimensions. Compared to the nanoantenna, a nanoslit can feature several advantages, as will be discussed in the talk. The SEIRA performance is compared by investigating a monolayer of octadecanethiol. Furthermore, the slit geometry is optimized by systematically examining all important parameters. We show, that the slit width has a strong influence on the near-field intensity and therefore also on the SEIRA performance. Finally, the transversal and longitudinal coupling of nanoslits is studied and the response is compared to the excitation of a single nanoslit. A strong coupling behavior is observed, and the periodicity is optimized, leading to the maximum SEIRA signal. The coupling condition is compared to that of solid nanostructures, revealing several differences. Our results give access to general design rules for any sensing application based on the use of inverse nanostructures.

O 33.3 Tue 11:00 H4

Monocrystalline Gold Structures Applied for SEIRA Spectroscopy — •Jochen Vogt[1,2], Christian Huck[1], Bettina Frank[3], Xinghui Yin[3], Frank Neubrech[1,3], Harald Giessen[3], and Annemarie Pucci[1,2] — [1]Kirchhoff Institute for Physics, Im Neuenheimer Feld 227, Heidelberg, Germany — [2]InnovationLab GmbH, Speyerer Str. 4, Heidelberg, Germany — [3]4th Physics Institute and Research Center SCoPE, University of Stuttgart, Pfaffenwaldring 57, Stuttgart, Germany

Electrochemically grown monocrystalline gold triangles that can be produced easily at low costs are beneficial substrates for surface-enhanced infrared absorption (SEIRA) spectroscopy as we have proven for vibrational modes of a self-assembled monolayer of octadecanethiol. Moreover, their SEIRA performance exceeds that of polycrystalline, electron beam lithographic (EBL) triangles. Produced planar but randomly orientated on a substrate, the plasmonic resonances of the monocrystalline triangles can be spectrally tuned via their edge length. It might be interesting for applications that the plasmonic response and the SEIRA signals are almost independent on the light polarization under normal incidence, which is related to the equilateral triangular shape. However, because polarization dependent resonances of linear antennas are expected to deliver stronger SEIRA enhancement, such structures were cut out from the monocrystalline platelets by means of focused ion beam. Also for the linear antennas the monocrystalline ones show a clearly stronger vibrational signal enhancement in comparison to the EBL fabricated counterparts.

O 33.4 Tue 11:15 H4

Towards Resonant Plasmonic Antenna-Enhanced Terahertz Spectroscopy — •Ksenia Weber[1], Frank Neubrech[1], Mario Hentschel[1], Michael Scherer[2], and Harald Giessen[1] — [1]4th Physics Institute and Research Center SCoPE, University of Stuttgart, Stuttgart — [2]InnovationLab GmbH, Heidelberg, Germany

Terahertz spectroscopy is a technique with the potential for a vast range of sensing applications based on material-specific absorption features of molecular vibrations. However, the low absorption cross section of these excitations strongly limits its sensitivity. The possibility to increase the sensitivity of spectroscopic methods via the enhanced electromagnetic near fields provided by plasmonic nanoantennas has been shown before for surface-enhanced infrared spectroscopy (SEIRA). Our goal is to transfer the concept of SEIRA to terahertz frequencies. Here, we take a first step towards resonant antenna-enhanced terahertz spectroscopy by using plasmonic nanoantennas for the enhancement of molecular vibrations with frequencies towards the terahertz regime. We therefore fabricated arrays of rectangular gold antennas by electron beam lithography and coated them with a 30 nm thick layer of the fullerenes C60 and C70, respectively. The resonances of the structures were tailored to match the molecular vibrations located around 16 THz. The samples were investigated with Fourier transform infrared spectroscopy using a bolometer as detector. Compared to reference measurements with a pure molecular layer, an enhancement factor of four orders of magnitude is found. Our technique is a first step towards plasmon-enhanced terahertz sensing with unprecedented sensitivities.

O 33.5 Tue 11:30 H4

Investigating diffractive coupling of fs-laser induced standing gold antennas for surface enhanced IR spectroscopy — •Ruslan Röhrich[1], Tobias W. W. Mass[1], Martin Reininghaus[2], Reinhart Poprawe[2], and Thomas Taubner[1] — [1]Institute of Physics (IA) RWTH Aachen — [2]Fraunhofer Institute for Laser Technology ILT

IR resonant metallic nanoantennas induce enhanced and strongly localized electromagnetic fields. These nearfield enhancements are exploited in surface enhanced infrared absorption (SEIRA) spectroscopy to detect smallest quantities of molecules.

In this work micrometer-sized upright standing gold antennas are fabricated by irradiating a thin gold film with single femtosecond laser-pulses [1]. Due to image charge formation in the surrounding gold film, the resonance wavelength of the antennas is proportional to 4 times the antenna length [2]. The hot spot of the structure is distant from the substrate which is beneficial for SEIRA applications compared to other geometries.

By arranging these antennas in arrays, coupling between multiple antennas occurs. Collective resonances can be used to achieve stronger nearfield enhancements [3]. FTIR measurements in grazing incidence geometry and simulations demonstrate that diffractive coupling plays a significant role in the spectral response of standing antenna arrays.

[1] Wortmann et al., J. Laser Appl. **24**, 042017 (2012)
[2] Reininghaus et al., Opt. Express **21**, 32176-32183 (2013)
[3] Adato et al., PNAS **106**, 19227-19232 (2009)

O 33.6 Tue 11:45 H4

Surface-enhanced infrared spectroscopy for poly-peptide sensing on single nanostructures — •Rostyslav Semenyshyn[1], Frank Neubrech[1], Mario Hentschel[1], Xinghui Yin[1], Jochen Vogt[2], Felix Weiher[1], Christoph Stanglmair[3], Claudia Pacholski[3], and Harald Giessen[1] — [1]4th Physics Institute and Research Center SCoPE, University of Stuttgart — [2]Kirchhoff Institute for Physics, University of Heidelberg — [3]Max Planck Institute for Intelligent Systems, Stuttgart

Metal nanowires and nanoslits with plasmon resonances tuned to the infrared spectral region are ideally suited for surface enhanced infrared absorption (SEIRA) spectroscopy. Since its discovery, SEIRA has been applied to chemical analysis and biochemical sensing.

In our present work, we will demonstrate how SEIRA is used for the detection of peptides and proteins in minute amounts, and sensing of temperature-induced structural changes. For this purpose, nanostructures were functionalized by mercaptoundecanoic acid in order to selec-

tively bind poly-L-lysine to the gold surface. Additionally, we covered other nanoantennas with specially designed collagen peptides. The plasmonic resonances of such nanostructures were tuned to the amide I and II bands of the respective poly-peptides. A tailored flow cell was used to allow the control of molecular adsorption on the nanostructures and in-situ SEIRA in aqueous solutions. Following this approach, the spectroscopic detection of about 13000 poly-peptides on one single slit could be achieved. In further experiments, structural changes of the poly-peptides due to temperature changes were observed.

O 33.7 Tue 12:00 H4

Ultrafast Vibrational Ladder Climbing on a Surface with 2D ATR IR Spectroscopy — •Jan Philip Kraack and Peter Hamm — Department of Chemistry, University of Zürich, Winterthurerstrasse 190, CH-8057

Vibrational dynamics of molecules at solid-liquid and solid-gas interfaces are important for various technical and scientific applications. In order to directly obtain multi-dimensional infrared (IR) spectra of adorbats at surfaces we have recently developed 2D Attenuated Total Reflection (ATR) IR spectroscopy.(1,2) Surface-enhancement effects in 2D ATR IR spectroscopy, stemming from plasmonic nano-structured noble-metal surfaces, have recently been characterized.(3) Here, we demonstrate optimized surface-enhancement for organic molecules on ultrathin (nm) metal surfaces which allows for vibrational ladder climbing (LC) in the electronic ground state potential of IR labels with even low extinction coefficients ($<$ 200 M-1 cm-1). LC in such samples enables the characterization of ultrafast vibrational relaxation within the electronic ground state potential of a broad range of chemical bonds and is likely to be exploitable in coherent control(4) of surface-reactions.

(1) J.P. Kraack, D. Lotti and P. Hamm, J. Phys. Chem. Lett., 2014, 5, 2325-2329.

(2) J.P. Kraack, D. Lotti and P. Hamm, J. Chem. Phys., 2015, 142, 212413.

(3) J.P. Kraack and P. Hamm, submitted, 2015.

(4) T. Witte et al., J. Chem. Phys., 2003, 118, 2021-2024.

O 33.8 Tue 12:15 H4

Surface Phonon Polariton Localization in SiC Nano Pillars Probed by Second Harmonic Spectroscopy — •Alexander Paarmann[1], Ilya Razdolski[1], Joshua D. Caldwell[2], Alexander J. Giles[2], Sandy Gewinner[1], Wieland Schöllkopf[1], Stefan A. Maier[3], and Martin Wolf[1] — [1]Fritz-Haber-Institut, Berlin, Germany — [2]Navy Research Laboratory, Washington D.C., USA — [3]Imperial College, London, UK

Surface Phonon Polaritons (SPhP) have recently emerged as novel building block for mid-infrared nanophotonics, promising to possibly overcome the intrinsic loss problem of plasmonics associated with the short life time of plasmonic excitations [1]. SPhPs arise in polar dielectrics from optical phonon resonances causing negative permittivity between transversal and longitudinal optical phonon frequencies. Specifically, the long life times of optical phonons allow for remarkable degrees of SPhP localization in sub-wavelength nanostructures.

Here, we show that mid-infrared second harmonic generation (SHG) spectroscopy [2] provides the most direct experimental approach to study SPhP field localization. Our experiments employ intense, tunable, and narrowband mid-infrared pulses from a free-electron laser to acquire SHG excitation spectra from SiC nanopillars. We observe sharp enhancement of the SHG yield for various resonant modes of our nanostructures. Most importantly, the measurements show much increased sensitivity of the SHG yield to field localization, as compared to linear optical approaches. [1] Caldwell et al., Nanophotonics 4, 1 (2015), [2] Paarmann et al., APL 107, 081101 (2015)

O 33.9 Tue 12:30 H4

Second Harmonic Phonon Spectroscopy of α-Quartz — •Christopher Winta, Sandy Gewinner, Wieland Schöllkopf, Martin Wolf, and Alexander Paarmann — Fritz-Haber-Institut der Max-Planck-Gesellschaft

We experimentally study the efficiency of second harmonic generation (SHG) from optical phonon resonances in crystalline α-quartz, employing intense, tunable picosecond mid-infrared laser pulses generated from the FHI free-electron laser (FEL). Due to its numerous optical phonon branches, α-quartz exhibits multiple Reststrahl bands with high reflectivity, located between the respective transversal and longitudinal optical phonon resonances. For each of these spectral regions, the real part of the dielectric function is negative, resulting in rapidly attenuated evanescent waves which make these experiments sensitive to a thin near-surface layer.

In our experiments, we employ a non-collinear, reflective geometry which allows for a background-free detection of the SHG signal, even for strongly absorbing samples. Making use of the large tuning range of the FEL, we can investigate essentially all optical phonon resonances of α-quartz. We observe sharp resonances in the SHG at the frequencies of transverse optical phonons due to a resonant enhancement of the nonlinear susceptibility. At each resonance, the SHG signal also strongly depends on the azimuthal orientation of the crystal as a consequence of its trigonal crystal symmetry. We show that this feature can be exploited to selectively investigate the different contributions of the nonlinear susceptibility tensor to the SHG signal.

O 33.10 Tue 12:45 H4

Phonon control in a 2D phononic crystals — •Yuning Guo and Thomas Dekorsy — Department of Physics and Center of Applied Photonics, University of Konstanz, 78457 Konstanz, Germany

Due to the ability to confine and mold acoustic waves in periodic nanostructure, the acoustic properties of phononic crystal (PnC) are tailored by designing various kinds of defects, which is suitable for a wide range of applications from transducer technology to filtering and guidance of acoustic waves. The ways of control phonons in 2D PnC are studied in this report. Channel drop filters can access one channel of a wavelength division multiplexed signal while leaving other channels undisturbed, which are essential components of acoustic communication systems. The target dropping frequency is selected and the transmission efficiency is improved by designing different waveguides and a series of cavities in PnC. The self-collimation of acoustic wave can be regarded as a kind of channelless phononic waveguide, which provides another way to control the propagation of phonons. The self-collimation beam can propagate with almost no diffraction along the PnC, which can be used to acoustic imaging, negative refraction etc.. When the self-collimation beam propagates in a PnC with certain defects, those defects behave like mirrors which lead to acoustic wave bending, splitting and phonons trapping. By analyzing the equi-frequency contour, the properties of waves propagating are deduced and the frequency of self-collimation is determined.

O 33.11 Tue 13:00 H4

Large-Area Fabrication of TiN Nanoantenna Arrays for Refractory Plasmonics in the Mid-Infrared. — •Shahin Bagheri[1], Christine M. Zgrabik[2], Timo Gissibl[1], Andreas Tittl[1], Florian Sterl[1], Ramon Walter[1], Stefano De Zuani[3], Audrey Berrier[3], Thomas Stauden[4], Gunther Richter[5], Evelyn L. Hu[2], and Harald Giessen[1] — [1]4th Physics Institute and Research Center SCoPE, University of Stuttgart, Pfaffenwaldring 57, Stuttgart, Germany. — [2]School of Engineering and Applied Sciences, Harvard University, Cambridge, MA 02138, USA. — [3]1st Physics Institute and Research Center SCoPE, University of Stuttgart, Pfaffenwaldring 57, Stuttgart, Germany. — [4]Fachgebiet Nanotechnologie, Technische Universität Ilmenau, Ilmenau, Germany. — [5]Max-Planck-Institut für Intelligente Systeme, D-70589 Stuttgart, Germany.

We utilize fast and wafer-scale fabrication methods such as direct laser writing and laser interference lithography to fabricate large-area nanoantenna arrays of TiN. Our lithographic tools allow for fast and homogeneous preparation of nanoantenna geometries on a polymer layer, which is then selectively transferred to TiN by subsequent argon ion beam and chemical wet etching process. Tailoring of the TiN antenna geometry enables precise tuning of the plasmon resonances from the near to the mid-infrared spectral range. The antennas are protected by an additional Al2O3 layer which allows for high-temperature annealing in argon flow without loss of the plasmonic properties.

O 33.12 Tue 13:15 H4

Resonances from thermal waves using photothermal deflection microscopy — •André Heber, Markus Selmke, and Frank Cichos — Universität Leipzig, Exp. Phys. I, Molecular Nano-Photonics Group, 04103 Leipzig, Germany

A gold nanoparticle is heated by a harmonically modulated pump laser which is fixed in space. This configuration results in a thermal wave centred around the nanoparticle. The thermal wave is exponentially damped with a decay-length corresponding to a single thermal wavelength. We co-align a second laser beam which is non-absorbed and steerable to measure deflections induced by refractive index changes due to the optical heating. This setup enables the detection of the thermal wave's resonances in the components in phase and out-of-

phase with the pump. The deflection signal depends on the distance. The externally controllable offset provides a well-defined length scale which can be used to quantify the thermal diffusivities from these resonances in combination with a ray optics model.