MO 23: Molekülphysik Poster 2

Zeit: Donnerstag 16:30–19:00 Raum: VMP 8 Foyer

MO 23.1 Do 16:30 VMP 8 Foyer

Alkali-helium snowballs formed on helium nanodroplets — Severin Müller, •Raphael Katzy, Marcel Mudrich, and Frank Stienkemeier — Physikalisches Institut, Universität Freiburg, 79104 Freiburg, Germany

Ions implanted in suprafluid helium significantly influence the surrounding quantum liquid. The strong electric field gradient exerted by the charge induces a severe increase in the helium density leading to the localization of helium atoms in layers around the ion. Since the density in these layers commonly exceeds the one of solid helium, such structures are named snowballs. Monte Carlo calculations provide numbers of helium atoms N_1 for shell closures around alkali ions [1].

We systematically study the formation of alkali-helium snowballs $Ak^+_{1,2}He_N$ (Ak=Na, K, Rb, Cs). Helium nanodroplets are multiply loaded with alkali atoms before being photo-ionized by a Ti:Sa fs laser. The distribution of snowballs $Ak^+_{1,2}He_N$ is analyzed by means of a quadrupole mass spectrometer. The experimental data show snowball formation around all investigated alkali species. Agreement of N_1 with theoretical predictions is found in the case of Rb^+ and Cs^+. Evidence of a subshell closure for Cs^+ is explained in line with single-atom evaporative energies taken from theory.

[1] M. Rossi, M. Verona, D.E. Galli, L. Reatto: *Phys. Rev. B*, 69:212510, 2004.

MO 23.2 Do 16:30 VMP 8 Foyer

Vibrational wave packet dynamics of Rb_2 formed on helium nanodroplets — •Philipp Heister, Thomas Hippler, Marcel Mudrich, and Frank Stienkemeier — Physikalisches Institut, Universität Freiburg

Superfluid helium nanodroplets are a valuable tool to investigate alkali dimers at temperatures in the millikelvin range. Using fs-pump-probe spectroscopy we study the dynamics of wave packets created by the coherent superposition of vibrational states of triplet Rb_2 molecules. Upon excitation, desorption of Rb_2 off the helium droplet leads to an undamped wave packet motion, which is mass selectively detected in the photo ionization signal. We are able to observe wave packet dynamics up to 2 ns. This allows us to study the de- and rephasing mechanism of wavepackets of Rb_2 initially coupled to the helium droplet environment. Fourier analysis provides high resolution vibrational spectra of triplet ground and first excited states, which are compared to *ab initio* calculations.

MO 23.3 Do 16:30 VMP 8 Foyer

High resolution electronic spectroscopy of porphyrine derivatives in superfluid helium nanodroplets — •Ricarda Riechers and Alkwin Slenczka — University of Regensburg, Germany

A combination of laser induced fluorescence excitation and dispersed emission spectra delivers insight into intramolecular dynamics of molecules inside superfluid helium nanodroplets. Characteristic features in the electronic spectra such as fine structure in vibronic transitions as well as the frequency shift of the 0^0_0 transition in excitation and emission are indicative for dynamic processes either within the embedded molecule or within its closer environment. This experimental approach was applied to several porphyrine derivatives as well as to molecular aggregates consisting of one porphyrine molecule and either an argon atom or a water molecule. In the case of chloroaluminiumphthalocyanine (AlClPc) the van der Waals complexes with argon showed a rich spectrum of sharp transitions. By help of the dispersed emission spectra the signals of isomeric complexes could be identified. Particularly, the emission spectra of some AlClPc-Ar_1-complexes showed a rich fine structure of the emission origin which repeats in the vibrational transitions. This can be ascribed to a relaxation of the configuration of the solvated complex upon electronic excitation. Regarding AlClPc-$(H_2O)_n$ in helium droplets, one can study the complexation of two polar molecules under low temperature conditions. By using Stark-field induced optical anisotropy measurements structural information about the possible complex isomers becomes accessible.

MO 23.4 Do 16:30 VMP 8 Foyer

A damping model for molecular dynamics on helium nanodroplets — •Martin Schlesinger[1], Walter Strunz[1], Marcel Mudrich[2], and Frank Stienkemeier[2] — [1]Institut für Theoretische Physik, TU Dresden, 01062 Dresden — [2]Physikalisches Institut, Universität Freiburg, 79104 Freiburg

Superfluid helium nanodroplets provide an ideal "refrigerator" for high precision spectroscopy of embedded species. Recent experiments study vibrational dynamics of dimers attached to helium nanodroplets, employing femtosecond pump-probe spectroscopy [1]. Unexpected features in the spectra exhibit the influence of the helium environment on the dimer dynamics. We propose that the most important effect is a general damping of vibrational wave packets. Numerical calculations allow us to reproduce and explain crucial experimental findings, unseen in gas-phase experiments. Remarkably, best agreement with experiment is found if one allows for undamped motion of slowly moving wave packets, which might be attributed to the superfluid properties of the host.

[1] P. Claas, G. Droppelmann, C. P. Schulz, M. Mudrich, and F. Stienkemeier, J. Phys. B: At. Mol. Opt. Phys. **39**, 1151 (2006).

MO 23.5 Do 16:30 VMP 8 Foyer

Spektroskopie an dotierten Heliumtropfen — •Sebastian Göde, Andreas Przystawik, Josef Tiggesbäumker und Karl-Heinz Meiwes-Broer — Universität Rostock, Institut für Physik, Universitätsplatz 3, 18051 Rostock

Bei einer Düsenstrahlexpansion von Helium ins Vakuum entstehen ultrakalte Heliumtröpfchen, die beim Passieren einer Pickup-Zelle Atome aus einem Dampf niedriger Dichte aufnehmen können. Die Wechselwirkung der aufgenommenen Atome mit dem Helium entscheidet darüber, ob sich die Atome nach dem Pickup auf der Tropfenoberfläche oder im Tropfen befinden. Für das Verhalten der Elemente der Erdalkalimetalle ist eine genaue Vorhersage im Rahmen des Modells von Ancilotto [1] nicht möglich. Experimente haben gezeigt, dass die Elemente Sr und Ba einen stabilen Zustand nahe der Oberfläche der Heliumtropfen haben [2].

In diesem Beitrag werden Resultate von Mg und Ca vorgestellt. Verschiedene experimentelle Techniken wie resonante Zwei-Photonen-Ionisation, laserinduzierte Fluoreszenz sowie Elektronenstoßionisation wurden gezielt eingesetzt, um die Löslichkeit der Atome bei unterschiedlichen Quellenbedingungen zu bestimmen. Für Mg sind sowohl Signaturen oberflächennaher als auch gelöster Atome nachweisbar. Weiterhin zeigen die Ergebnisse, daß mehrere Magnesiumatome in einem Tropfen einen besonderen Bindungszustand aufweisen [3].

[1] F. Ancilotto et al., J. of Low Temp Phys. **101**, 1123 (1995)
[2] F. Stienkemeier et al., Eur. Phys. J. D **9**, 313 (1999)
[3] A. Przystawik, S. Göde et al., Phys. Rev. A **78**, 021202 (2008)

MO 23.6 Do 16:30 VMP 8 Foyer

Calculation of resonant interatomic Coulombic decay widths of inner-valence-excited states delocalized due to inversion symmetry — •Sören Kopelke[1], Kirill Gokhberg[1], Lorenz Cederbaum[1], and Vitali Averbukh[2] — [1]Theoretische Chemie, Physikalisch-Chemisches Institut, Universität Heidelberg, Im Neuenheimer Feld 229, D-69120 Heidelberg, Germany — [2]Max Planck Institute for the Physics of Complex Systems, Nöthnitzer Str. 38, D-01187 Dresden, Germany

Inner-valence-excited states of clusters can decay by electron emission via several of mechanisms, the leading ones being intra-atomic autoionization and resonant inter-atomic Coulombic decay. Recently, we have derived the Wigner-Weisskopf theory for the calculation of the decay widths of the inner-valence excitations [J. Chem. Phys. **124**, 144315 (2006)]. While the new method has been successful in producing the decay rates of heteronuclear diatomic clusters, it can not be applied to systems possessing inversion symmetry, e.g. to homonulcear diatoms, due to delocalization of the molecular orbitals involved in the decay processes. In the present work, we show that the Wigner-Weisskopf theory of the decay of inner-valence-excited states can be generalized to systems with inversion symmetry using a technique of adapted final states [J. Chem. Phys. **125**, 094107 (2006)]. The same technique can be employed when going beyond the Wigner-Weisskopf theory. We consider the experimentally relevant case of competing resonant interatomic Coulombic decay and autoionization in neon dimer.

MO 23.7 Do 16:30 VMP 8 Foyer

Fusion and scattering mechanism in C_{60}-C_{60} collisions — •Jan

Handt and Rüdiger Schmidt — Institut für Theoretische Physik, Technische Universität Dresden, D-01062 Dresden

We present a theoretical study of C_{60}-C_{60} collisions without and with electronic excitation (by a laser) in the impact energy up to 150eV (center of mass frame). As is well known, fusion and inelastic scattering occur in this energy range. Fusion events are more favoured in the higher energy regime whereas scattering in the lower one. Our molecular dynamics calculations show that the individual event depends strongly on the initial orientation of the collision partners. On the other hand analyzing the vibrational dynamics similarities can be found in the collision dynamics for all kinds of events. A coupled spring model is presented which allows us to determine analytically the fusion threshold and its dependence on the vibrational constants. The model results agree well with the molecular dynamics simulations.

MO 23.8 Do 16:30 VMP 8 Foyer

Spectroscopy of thin molecular films under ultrahigh vacuum conditions using tapered optical fibres — •Ariane Stiebeiner, Olga Rehband, David C. Papencordt, Ruth Garcia-Fernandez, and Arno Rauschenbeutel — Abteilung QUANTUM, Institut für Physik, Johannes Gutenberg-Universität Mainz, 55099 Mainz

The guided modes of sub-wavelength-diameter air-clad optical fibres exhibit a pronounced evanescent field. The absorption of light by molecules deposited at the fibre surface is therefore readily detected by measuring the fibre transmission. We have shown that the resulting absorption for a given surface coverage can be orders of magnitude higher than that for conventional surface spectroscopy. The measurements were performed on sub-monolayers of 3,4,9,10-perylene-tetracarboxylic dianhydride (PTCDA) molecules at ambient conditions, revealing the agglomeration dynamics on a second to minutes timescale [1].

We set up a new apparatus integrating the ultrathin fibre under ultrahigh vacuum (UHV) conditions in order to gain better control over the system. Firstly, this arrangement enables us to produce an homogeneous flux of molecules for deposition onto the ultrathin fibre waist. Furthermore, it allows us to desorb pollutants from the fibre and thus to work with a better defined surface. The measured absorption spectra of the deposited molecules and their time evolution are compared with the results obtained at ambient conditions.

We gratefully acknowledge financial support by the Volkswagen Foundation, the ESF, and the EC.

[1] F. Warken et al., Opt. Express 15, 11952-11958 (2007)

MO 23.9 Do 16:30 VMP 8 Foyer

Starkeffekt-induzierte Prädissoziation von H_3 — •Frank Baumgartner, Peer Fechner und Hanspeter Helm — Universität Freiburg

Aufgrund seiner einfachen Struktur eignet sich das neutrale H_3 als Modellsystem zur Untersuchung dynamischer Vorgänge in Molekülen. Besonderes Interesse kommt dabei dem Dreiteilchenzerfall in die Fragmente H(1s)+H(1s)+H(1s) zu, der einen direkten Einblick in die Wirkung nicht-adiabatischer Prozesse erlaubt. Im vorgestellten Experiment werden Moleküle im metastabilen 2p Niveau prädissoziiert, indem ihnen über den Starkeffekt ein regulierbarer Anteil des kurzlebigen 2s Zustandes beigemischt wird. Mit Hilfe einer Dreifach-Koinzidenz-Technik können die Impulsvektoren aller Fragmente sowie die räumliche Ausrichtung des Moleküls erfasst werden. Unsere Ergebnisse belegen eindrucksvoll, dass sich mit steigender Feldstärke das Zerfallsverhalten des 2p Zustands kontinuierlich mit dem des ungestörten 2s Niveaus mischt. Im Übergangsbereich treten Interferenzeffekte in den Dalitzplots auf, die von der unterschiedlichen Entwicklung der beiden Wellenfunktionsanteile auf der Grundzustandsfläche herrühren.

Darüber hinaus zeigen unsere Ergebnisse, dass bevorzugt die in Feldrichtung orientierten 2p Zustände dissoziiert werden, während senkrecht zum Feld ausgerichtete Moleküle erhalten bleiben. Der vorgestellte Aufbau eignet sich damit auch zur Erzeugung eines orientierten Molekülstrahls.

MO 23.10 Do 16:30 VMP 8 Foyer

Characterization and optimization of a femtosecond high-order harmonic VUV photon source for investigating ultrafast molecular dynamics — •Torsten Leitner[1], Philippe Wernet[1], Kai Godehusen[1], Olaf Schwarzkopf[1], Tino Noll[1], Jerome Gaudin[3], Andrei Sorokin[2], Henrik Schoeppe[2], Mathias Richter[2], and Wolfgang Eberhardt[1] — [1]Helmholtz-Zentrum Berlin - Elektronenspeicherring BESSY II — [2]Physikalisch-Technische Bundesanstalt, Berlin — [3]DESY/ European XFEL Project Team, Hamburg

High-order harmonic generation (HHG) with femtosecond lasers in rare gas media has recently emerged as a promising tool to produce bright atto- and femtosecond vacuum-ultraviolet and soft x-ray pulses. These pulses can be used to study ultrafast molecular dynamics in a pump-probe electron spectroscopy configuration. To allow for appropriate experimental conditions the key parameters of the photon source like flux, spot size and shape have to be optimized. This was done in recent experiments at the BESSY femtosecond HHG source. A Gas Monitor Detector was used to investigate the absolute photon flux of the source and validate the photon numbers derived from a calibrated GaAsP diode. The beam divergence and transverse shape were investigated with a CCD camera showing, that optimizing the source to the highest photon flux does not necessarily result in optimal transverse beam profiles as needed for further pump-probe experiments on molecular dynamics. Therefore the experimental setup was extended by a quasi-online monitoring system enabling fast optimization of the light source.

MO 23.11 Do 16:30 VMP 8 Foyer

Development of broadly tunable cw narrowband mid-IR laser source for molecular spectroscopy — •Sergey Vasilyev[1], Stephan Schiller[1], Alexander Nevsky[1], Arnaud Grisard[2], David Faye[2], Eric Lallier[2], Zhaowei Zhang[3], Morten Ibsen[3], Andrew Clarkson[3], Peter Kaspersen[4], Axel Bohman[4], and Peter Geiser[4] — [1]Institut für Experimentalphysik, Universität Düsseldorf, 40225 Düsseldorf, Germany — [2]Thales Research and Technology, 91767 Palaiseau cedex, France — [3]Optoelectronics Research Centre, University of Southampton, SO17 1BJ, UK — [4]Norsk Elektro Optikk A/S, N-1473 Lørenskog, Norway

We are developing a continuous-wave, widely tunable, single frequency mid-IR laser source based on a down conversion of 1.5-2.0 μm laser radiation to mid-IR spectral region using difference frequency generation (DFG) and optical parametric generation (OPO) in a Orientation-Patterned Gallium Arsenide (OP-GaAs) crystals.

A mid-IR source with sub-mW output based on DFG between a narrowband broadly tunable high power EDFA and a thulium doped fiber laser MOPA has been developed [1]. DFG output wavelength was tunable via computer control in 7.6-8.2 μm range with the picometer precision. The DFG source was integrated in a compact multi-gas spectrometer for environmental gas sensing applications.

We are currently upgrading the DFG source to higher output power and broader tuning range, and exploring the cw OP-GaAs OPO source using a high power (10 W) Tm- fiber laser pumping.

[1] S.Vasilyev et al., Optics Letters 33, 413 (2008)

MO 23.12 Do 16:30 VMP 8 Foyer

A new XUV Autocorrelator for Pump-Probe Experiments and Pulse Diagnostics at FLASH — Michel Toppin[1], Oliver Herrwerth[2], Matthias Lezius[2], Matthias Kling[2], Moritz Kurka[1], Artem Rudenko[1], Claus Dieter Schröter[1], Yuhai Jiang[1], Kai-Uwe Kühnel[1], Lutz Foucar[3], Rolf Treusch[4], Michael Gensch[4], •Robert Moshammer[1], and Joachim Ullrich[1] — [1]Max-Planck-Institut für Kernphysik, Saupfercheckweg 1, 69117 Heidelberg — [2]Max-Planck-Institut für Quantenoptik, Hans-Kopfermann-Str. 1, D-85748 Garching — [3]Institut für Kernphysik, Universität Frankfurt, D 60486 Frankfurt — [4]DESY, Notkestrasse 85, 22607 Hamburg

A split-mirror setup for the focussing of XUV radiation was designed and brought into operation at the free-electron laser at Hamburg (FLASH). It consists of a spherical multi-layer mirror (50 cm focal length) that is cut into two identical half-mirrors ("half-moon"geometry). The whole mirror is operated in on-axis back-reflection mode. While one half-mirror is mounted at a fixed position, the other one is movable by means of a high precision piezo-stage along the beam axis. This way a time delay between both reflected light beams is adjustable within a range of $\pm$1500 fs at a resolution of better than 1 fs. The foci of both mirrors are merged inside a dilute, well localized (less than 1 mm diameter) and cold atomic or molecular beam in the centre of a Reaction Microscope for momentum resolved detection of created electrons and ions. First results will be presented.

MO 23.13 Do 16:30 VMP 8 Foyer

Lifetime vibrational interference during the CO $1s^{-1}$ π^* (v_r) resonance excitation investigated by the CO^+ ($A\ ^2\Pi \rightarrow X\ ^2\Sigma^+$) dispersed fluorescence — •Philipp Reiss[1], Witoslaw Kielich[1], Andre Knie[1], Rainer Hentges[1], Irina Haar[1], Philipp V. Demekhin[2], Ivan D. Petrov[2], Viktor L. Sukhorukov[2], and Arno Ehresmann[1] — [1]Institute of Physics and CINSaT, University of Kas-

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Dispersed fluorescence from fragments formed after the de-excitation of the $1s^{-1}\pi^*$ resonances of C*O and CO* molecules has been measured by photon-induced fluorescence spectroscopy (PIFS) in the spectral range from 378 nm to 578 nm.

The excitation was performed with synchrotron radiation from the U49/2 PGM1 beamline at BESSY II.

The investigated fluorescence range is dominated by lines of atomic carbon and oxygen fragments and by the A $^2\Pi(v') \rightarrow$ X $^2\Sigma^+(v'')$ fluorescence bands of CO^+ ion resulting from the participator Auger decay of the $1s^{-1}\pi^*$ resonances.

The ratios of the natural lifetime width of the $1s^{-1}\pi^*$ resonances, Γ_R, and the energy spacing between their vibrational levels, ω_e, are equal to the 0.32 and 0.95 for the C*O and CO* resonances, respectively, providing a prototype for an expected strong (CO*) and weak (C*O) lifetime vibrational interference.

MO 23.14 Do 16:30 VMP 8 Foyer

Lifetime vibrational interference during the CO $1s^{-1}\pi^*$ (v_r) resonance excitation investigated by the CO^+ (A $^2\Pi \rightarrow$ X $^2\Sigma^+$) dispersed fluorescence — •Philipp Reiss[1], Witoslaw Kielich[1], Andre Knie[1], Rainer Hentges[1], Irina Haar[1], Philipp V. Demekhin[2], Ivan D. Petrov[2], Viktor L. Sukhorukov[2], and Arno Ehresmann[1] — [1]Institute of Physics and CINSaT, University of Kassel, Heinrich-Plett-Str. 40, 34132 Kassel, Germany — [2]Rostov State University of Transport and Communications, 344038 Rostov-on-Don, Russia

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MO 23.15 Do 16:30 VMP 8 Foyer

Absorption and energy transfer of molecular aggregates — •Jan Roden[1], Walter Strunz[2], John Briggs[3], and Alexander Eisfeld[1] — [1]MPIPKS Dresden — [2]TU Dresden, Theoretische Quantenoptik — [3]Uni Freiburg, Theoretische Quantendynamik

The coupling of electronic excitation to vibrational degrees of freedom strongly influences characteristic properties of molecular aggregates (e.g. optical properties and energy transfer dynamics) [1]. We treat this exciton-phonon coupling using a non-Markovian stochastic Schrödinger equation [2]. Solving a Holstein-type model, we determine optical and transfer properties for exciton dynamics coupled to a realistic, complex phonon bath such that energy dissipation to the phonons is fully included. Our approach captures uniformly the transition from fully coherent to incoherent excitation transfer. As a specific example demonstrating the capability of this approach, we investigate J-band and H-band spectra and energy transfer dynamics of mesoscopic molecular aggregates. The results are compared with that of the well established CES approximation (Coherent Exciton Scattering) [3] and the exact solution for the extreme case when the complex spectral density is replaced by that of a single harmonic oscillator.

[1] A. Eisfeld, J.S. Briggs, Phys. Rev. Lett. 96, (2006) 113003; M. Bednarz, V. A. Malyshev, J. Knoester, J. Luminescence 112, (2005) 411; S. Kirstein, S. Daehne, Int. J. Photoenergy 2006, (2006) 20363.
[2] T. Yu, L. Diosi, N. Gisin, W. Strunz, Phys. Rev. A. 60, (1999) 91
[3] J.S. Briggs, A. Herzenberg, J. Phys. B 3, 1663 (1970)