MO 16: Posters 3: Experimental Techniques and Theoretical Approaches

Time: Thursday 17:00–19:00 Location: P OGs

MO 16.1 Thu 17:00 P OGs

Resolving Vibrational from Electronic Coherences in Two-Dimensional Electronic Spectroscopy: The Role of the Laser Spectrum — Franco V. De A. Camargo[1], •Lena Grimmelsmann[1,2], Harry L. Anderson[3], Stephen R. Meech[1], and Ismael A. Heisler[1] — [1]School of Chemistry, Norwich Research Park, University of East Anglia, Norwich NR4 7TJ, United Kingdom — [2]Physical Chemistry II, Ruhr-University Bochum, 44780 Bochum, Germany — [3]Department of Chemistry, Chemistry Research Laboratory, University of Oxford, Oxford OX1 3TA, United Kingdom

Light-induced coherent superposition of states are observed through the presence of oscillations in two-dimensional electronic spectroscopy (2DES). The pioneering 2DES studies on coherences mostly focused on single coordinates of the real part of 2D maps, with limited scanning of the waiting time, because of the long data acquisition times. Here we use a state-of-the-art 2D spectrometer that quickly acquires data with very good signal-to-noise to recover complex-valued 2DES maps and study vibrational coherences in a zinc-porphyrin monomer. Further we show that these signatures are critically dependent on the laser spectrum, demonstrating that a blueshift filters pathways so that the final result matches exactly those expected for an electronic coherence. This result is of crucial importance for the interpretation of coherences in 2DES. Finally, we use this new understanding of the laser spectrum effects to perform a complementary 2D experiment that lifts any ambiguity in the interpretation.

MO 16.2 Thu 17:00 P OGs

Two-dimensional Electronic Spectroscopy of Controlled Isolated Systems — •Ulrich Bangert, Lukas Bruder, Marcel Binz, Daniel Uhl, Katharina Schneider, Andreas Wituschek, and Frank Stienkemeier — Institute of Physics, University of Freiburg, Germany

Two-dimensional electronic spectroscopy (2DES) is a powerful tool to study coherences and correlations on ultrafast time scales. Until now, 2DES has been limited almost exclusively to condensed phase studies. Our aim is to apply 2DES to controlled isolated systems by using doped helium nanodroplet beams. Helium nanodroplets provide the dopant with a cold environment and minimal perturbation, which are ideal conditions to study the behavior of an individual system in a well-controlled environment.

However, the target density in doped helium droplet beams is several orders of magnitudes lower than in bulk condensed phase samples. We adapt a phase modulation technique [1] to overcome this issue. Using this, we currently implement a 2DES setup combined with a doped helium droplet beam. This setup will open exciting new possibilities for future 2DES studies.

[1] P. F. Tekavec, G. A. Lott and A. H. Marcus, J. Chem. Phys. 127, 214307 (2007)

MO 16.3 Thu 17:00 P OGs

The Stability of Broadband Phase Shaping in Liquid Crystal Spatial Light Modulators — •Elisabeth Brühl, Tiago Buckup, and Marcus Motzkus — Physikalisch-Chemisches Institut, Ruprecht-Karls Universität Heidelberg, Germany

Liquid crystal spatial light modulators (LCM) have found many successful applications in physical chemistry, where they are used to modulate phase, amplitude and polarization of femtosecond (fs) laser pulses. In this regard, shot-to-shot as well as long-term modulation stability of LCMs are crucial requirements on its application in the broadband shaping of fs pulses. In this work, we show that when LCMs are applied to broadband tailoring of fs pulses (12 fs), depending on acquisition and detection conditions, noise figures up to 7 % can be observed. Our results show that spectral phase instability is originated by the LCM. In order to understand and thus minimize such instabilities, phase-sensitive measurements based on second harmonic generation of the tailored pulse, comparison of rapid-scan and step-scan results as well as careful control of the LCM temperature were implemented. The noise figure can be decreased by a factor of ten if a rapid-scan data acquisition approach is used instead of step scan. Furthermore, the study shows that the key in maximizing pulse stability and minimizing noise effects lies in careful control of the LCM operating temperature, improving the shot-to-shot stability of the output tailored pulse by a factor of two. Based on the experimental data we developed a physical model for a deeper understanding of the influence, especially the average scheme, and the level of noise.

MO 16.4 Thu 17:00 P OGs

Amplitude and phase shaping of femtosecond laser pulses in the ultraviolet with the help of an acousto optical modulator — •Sascha Bickhardt, Cristian Sarpe, Alexander Kastner, Philipp Hillmann, Arne Senftleben, and Thomas Baumert — University of Kassel, Institute of Physics and CINSaT, Heinrich-Plett-Str. 40, D-34132 Kassel, Germany

Femtosecond laser pulse shaping is the key technology in quantum control. So far, we were able to demonstrate pulse shaping with subcycle temporal accuracy making use of phase and amplitude modulation of femtosecond laser pulses in the infrared spectral region [1]. The experimental demonstration of molecular strong-field control schemes was achieved [2].

Organic molecules typically show pronounced absorption bands lying in the ultraviolet (UV) spectral region. Tailoring ultrashort UV laser pulses with respect to temporal and spectral shape opens up the possibility to investigate electron dynamics within different organic molecules, e.g. chiral ones.

We present the current status of our 4f and acousto optical modulator based setup for amplitude and phase shaping in the ultraviolet.

[1] J.Koehler *et al.*: *Optics Express* **19** (12), 11638-11653 (2011)
[2] T.Bayer *et al.*: *Physical Review Letters* **110**, 123003 (2013)

MO 16.5 Thu 17:00 P OGs

Optimization of data acquisition in time-domain spectroscopy experiments — •Daniel Uhl, Lukas Bruder, Marcel Binz, Ulrich Bangert, and Frank Stienkemeier — Institute of Physics, University of Freiburg, Germany

We are exploring different approaches to optimize the data acquisition in time-domain spectroscopy, in particular, in phase-modulated pump-probe and 2D spectroscopy experiments. On the one hand, compressed sensing can be used to significantly reduce the amount of data points, while yielding the same information content. This method allows in principle to go below the fundamental boundary of the sampling rate given by the Nyquist-Shannon-Theorem. On the other hand, we work on a software-based lock-in amplifier to improve data acquisition which represents a cost-effective alternative to commercial devices. This provides much higher flexibility: simultaneous detection with custom demodulators and filter algorithms is possible and acquisition times can be individually optimized for specific applications.

MO 16.6 Thu 17:00 P OGs

Development of a table-top soft X-ray source for probing ultrafast molecular dynamics — •S. Raabe[1], G. Goldsztejn[1], E.T. Karamatskos[2], S. Tripple[2], K. Kovács[3], E. Balogh[3], B. Major[3], V. Tosa[3], J. Küpper[2], K. Varjú[3], M.J.J. Vrakking[1], and A. Rouzée[1] — [1]Max-Born-Institute, Max-Born-Str. 2A, 12489 Berlin, Germany — [2]Center for Free-Electron Laser Science, DESY, Notkestr. 85, 22607 Hamburg, Germany — [3]Dpartment of Optics and Quantum Electronics, University of Szeged, Szeged, Hungary

High harmonic generation (HHG) is a powerful technique to generate ultrashort bursts of short-wavelength radiation. These pulses provide a way to capture ultrafast electronic and nuclear motion in matters with an exquisite temporal resolution down to the attosecond timescale. However, due to the low conversion efficiency of the HHG process at high photon energies, experiments have so far been limited to a photon energy range below 100 eV. We are currently developing a bright soft X-ray source making use of two-color HHG [1], where the first laser has a wavelength centered around 800 nm and the other laser is in the mid-infrared part of the spectrum. We will present the design of our table-top soft X-ray source that includes a new high gas pressure cell that will be used to generate water window harmonics using this two-color laser scheme. The source will be used together with a velocity map imaging spectrometer to perform time-resolved photoion and photoelectron spectroscopy and a soft X-ray spectrometer enabling transient absorption spectroscopy.

[1] B. Schütte, Optics Express, 23, 26 (2015)

MO 16.7 Thu 17:00 P OGs

Molecular processes with intense laser pulses — •Denhi Martínez[1,2], Alfonso Guerrero[2], Ignacio Álvarez[2], Thomas Pfeifer[1], Robert Moshammer[1], and Carmen Cisneros[2] — [1]Max-Planck-Institut für Kernphysik, Saupfercheckweg 1, 69117 Heidelberg — [2]Instituto de Ciencias Físicas, Universidad Nacional Autónoma de México, Av. Universidad s/n, Chamilpa, 62210 Cuernavaca, Morelos.

Through the interaction between strong laser fields and atoms or molecules, it is possible to elucidate the dynamical evolution in those systems. In the present work are shown some results for the dissociation and multiphoton ionisation processes of the nitromethane (CH_3NO_2) with V-UV radiation from laser pulses having nanoseconds pulse duration and at an intensity ranging from 10^9 to 10^{10} W/cm^2 . The observed ionic fragments were identified using high resolution mass spectrometry (Reflectron and TOF technique) and provide information on the fragmentation channel. We also present how to extend this research using a femtosecond laser providing higher intensity ranges and enabling therefore to study tunneling ionization. Moreover, femtosecond laser pulses, due to their timescale, allow to obtain a clearer picture of the electron dynamics the system We combine this pump probe spectroscopy technique with a reaction microscope to determine the initial momenta of ions and electrons, based on the position and time of flight of their detection.

MO 16.8 Thu 17:00 P OGs

A molecular movie of Interatomic Coulombic Decay in NeKr — •Florian Trinter[1], Tsveta Miteva[2], Miriam Weller[1], Sebastian Albrecht[1], Alexander Hartung[1], Martin Richter[1], Joshua Williams[1], Averell Gatton[3], Bishwanath Gaire[3], Thorsten Weber[3], James Sartor[4], Allen Landers[4], Ben Berry[5], Vasili Stumpf[2], Kirill Gokhberg[2], Reinhard Dörner[1], and Till Jahnke[1] — [1]Institut für Kernphysik, Goethe-Universität, 60438 Frankfurt am Main, Germany — [2]Theoretische Chemie, Physikalisch-Chemisches Institut, Universität Heidelberg, 69120 Heidelberg, Germany — [3]Lawrence Berkeley National Laboratory, Chemical Sciences Division, Berkely, California 94720, USA — [4]Department of Physics, Auburn University, Auburn, Alabama 36849, USA — [5]J. R. MacDonald Laboratory, Department of Physics, Kansas State University, Manhattan, Kansas 66506, USA

During the last 15 years a novel decay mechanims of excited atoms has been discovered and investigated. This so called "Interatomic Coulombic Decay" (ICD) involves the chemical environment of the electronically excited atom or molecule: the excitation energy is transferred to a neighbor of the initially excited particle usually ionizing that neighbor. It turned out that ICD is a very common decay route in nature as it occurs across van der Waals and hydrogen bonds. The time evolution of ICD is predicted to be highly complex, as its efficiency strongly depends on the distance of the atoms involved and this distance typically changes during the decay. Here we present a direct measurement of the temporal evolution of ICD using a novel experimental approach.

MO 16.9 Thu 17:00 P OGs

Fluorescence measurement of gas phase molecules by a single photon detector — •Sharmila Sait, Alexander Kastner, Philipp Schmidt, Arne Senftleben, Thomas Baumert, Andre Knie, Arno Ehresmann, and Martin Pitzer — Institute for Physics and Center for Interdisciplinary Nanostructure Science and Technology (CINSaT), University of Kassel

Being one of the oldest techniques in molecular physics, fluorescence spectroscopy is broadly used and still advancing in a wide range of application. Time-resolved detection of single photons has over the last two decades become an established technique to study dynamics and solvent effects of biomolecules in solution. Nevertheless, obtaining a complete picture, i.e. spectral and polarization information, still requires scans or arrays of photomultipliers [1]. This poster highlights a novel, time efficient and portable setup for measuring the behavior of isolated molecules in gas phase. The synchrotron radiation is focused on the sample. Emitted fluorescence is passed through a Wollaston prism, diffraction grating and detected by a time and position sensitive single photon detector [2]. Consequently, polarization, wavelength and lifetime of fluorescence of each photon are recorded in a single measurement.

[1] Becker, et al., Microscopy Research and Technique 70, 403-409 (2007)

[2] A. Czasch et al., Nuclear Instruments and Methods in Physics Research A 580 (2007) 1066-1070

MO 16.10 Thu 17:00 P OGs

Implementation of a mechanical shutter into a He droplet apparatus using a hard disk drive actuator — •Katharina Schneider — Institute of Physics, University of Freiburg, Germany

Controlled modulation or switching of light intensity is indispensable to many laser-based experiments. There are various ways to provide fast shuttering, varying in speed, reliability, expenses and extinction ratio.

Mechanical shutter excel in usability, expenses and transmission rate. Referring to [1], the voice-coil system of a computer hard disc drive can be modified to make a fast mechanical shutter, with switching times comparable to commercial devices. The voice-coil is driven by a custom circuit, using high-current pulses to ensure rapid shutter action. The direction of current is controlled by a TTL signal, enabling varying shutter frequencies and an asymmetric performance.

We setup such a shutter in a vacuum chamber to block a He droplet beam in front of a doping cell. This allows us to measure the remaining effusive signal in femtosecond HENDI (helium nanodroplet isolation) spectroscopy experiments. By subtracting this background from the measurement signal with the shutter open, the real doped droplet signal can be obtained.

[1] L. P. Maguire, S. Szilagyi, and R. E. Scholten, Rev. Sci. Instrum. 75, 3077 (2004)

MO 16.11 Thu 17:00 P OGs

Natural chemical conversion and catalytic ortho/para conversion of the non-radioactive hydrogen isotopologues — •Bennet Krasch, Robin Grössle, Sebastian Mirz, and Florian Altenbrand — Karlsruher Institute of Technology

Besides the chemical conversion processes an additional one exists for the homonuclear hydrogen isotopologues according to their relative nuclear spin orientation. There are two different species the so-called ortho hydrogen for parallel nuclear spins and the para hydrogen for antiparallel nuclear spins. These two species differ slightly in their thermodynamic properties as boiling point or specific heat capacity. The time constants for the natural chemical conversion is in the order of days to weeks in the gas phase. The ortho/para conversion time constant is in the order of thousand hours in the gas phase and ten hours in the liquid. At the Tritium Laboratory Karlsruhe (TLK) an analysis system has been developed to measure the isotopologue concentration in the liquid phase of a cryogenic distillation process, based on infrared absorption spectroscopy. Since the ortho/para ratio has an influence on the IR spectra, it is indispensable to calibrate the system with catalytically produced ortho/para mixtures. Currently at the TLK an experiment has been installed to measure the time constants of catalytic conversion at a temperature range 70K-400K to determine the efficiency of the iron oxide used catalyst. A Laser Raman setup is used for a real-time analysis. This contribution presents first results of these measurements of the catalyst efficiency.

MO 16.12 Thu 17:00 P OGs

Progress on a Zeeman Slower for Molecules — •Maurice Petzold, Paul Kaebert, Philipp Gersema, Mirco Siercke, and Silke Ospelkaus — IQO, Leibniz Universität Hannover, Hannover, Germany

Recently there has been great progress in laser cooling of molecules with quasi-diagonal Franck-Condon factors such as SrF CaF and YO. In particular, optical molasses, 2D- and 3D-magneto-optical traps have all been demonstrated. This is a tremendous step towards the creation of ultracold or even quantum degenerate samples of molecules via direct cooling techniques, extending the range of accessible molecules in the ultracold regime. Currently the molecule number in these traps is severely limited to $\sim 10^3$ due to the lack of high flux sources of molecules with velocities slow enough to be captured by magnetic or magneto-optical traps. Zeeman slowing, which is commonly used for atoms, is often thought to be impossible to implement for molecules due to the nature of the optical transitions used. Here we present a scheme of how to realize a Zeeman Slower for molecules. We simulate molecular trajectories emitted by a buffer gas cell and show that the slowing device provides significantly more flux of cold and slow molecules than other techniques used to date. The scheme is continuous and provides compression of the 1D-velocity distribution. We further report on progress on our demonstration experiment, which employs the proposed Zeeman slower scheme to Potassium atoms on the D1-line, an atomic system which exhibits many of the features and difficulties of molecular Zeeman slowing.

MO 16.13 Thu 17:00 P OGs

Cold collisions and slow reactions in a 22-pole trap — •Eric Endres, Daniel Hauser, Olga Lakhmanskaya, Steffen Spieler, Gerhard Egger, Malcolm Simpson, Fabio Carelli, Franco Gianturco, and Roland Wester — Institute for Ion Physics and Applied Physics, University of Innsbruck, Austria

Today the study of ion-molecule reactions moves towards cold chemistry which implies well controlled ion ensembles in a cold environment. A widely used tool to store and prepare ions by buffer gas cooling is a 22-pole rf ion trap [1].

For most applications it is important to know the translational and internal temperatures of the trapped ions. Here, we present detailed rotational state thermometry measurements over an extended temperature range for hydroxyl anions in different buffergases and show incomplete rotational cooling of OH^-. Furthermore we present a method to manipulate and measure molecular quantum states by non-resonant photodetachment. Based on this we provide quantum scattering rate coefficients under full quantum state control for the rotationally inelastic collision of hydroxyl anions with helium [2]. Additionally, preliminary results of the influence of temperature on the inelastic collision rate are shown. Finally the exothermic proton transfer reaction $D^- + H_2 \rightarrow HD + H^-$ with a barrier of about 0.33 eV [3] is analysed. Here we investigate whether this reaction may occur at low temperature via tunneling through this barrier.

[1] R. Wester, J.Phys.(2009); [2] D. Hauser Nat.Phys.(2015); [3] E. Haufler, J.Phys.Chem.(1997)

MO 16.14 Thu 17:00 P OGs

A novel spectroscopy technique for complex molecular ions in the gas phase — •Jürgen Göck[1], Klaus Blaum[1], Christian Breitenfeldt[1,2], Ulrich Gröozinger[3], Sebastian George[1], Thomas Henning[3], Holger Kreckel[1], Sunil Kumar S[1], Christian Meyer[1], Gaël Rouillé[3], Dmitry Strelnikov[4], and Andreas Wolf[1] — [1]Max-Planck-Institut für Kernphysik, Heidelberg, Germany — [2]Institut für Physik, Ernst-Moritz-Arndt-Universität Greifswald, Greifswald, Germany — [3]Max-Planck-Institut für Astronomie, Heidelberg, Germany — [4]Institut für Physikalische Chemie II, Karlsruher Institut für Technologie, Karlsruhe, Germany

The origin of the diffuse interstellar bands (DIBs)[1] and the unidentified infrared emission bands (UIEs) are long-standing questions in the field of astrophysics. DIBs are interstellar absorption features in the visible to near-infrared range, whereas the UIEs can be found in the emission spectra of astronomical objects. Complex molecular ions are candidates in particular since the first and only DIB carrier identified so far is the C_{60}^+ ion[2]. At the Max-Planck-Institut für Kernphysik we are developing a new technique for gas-phase spectroscopy of molecular ions that combines a cryogenic ion trap, which emulates the extreme conditions in the interstellar medium (10 K and 2000 particles/cm^3), with extremely sensitive mid-infrared detectors. The experimental aim is to identify unknown carrier species of the spectral lines. The measurement scheme and the current status of the experimental setup will be presented. [1] Herbig, G. H., Annu. Rev. Astron. Astrophys. 33, 19-73 (1995). [2] Campbell, E. K. et al., Nature 523, 322-323 (2015)

MO 16.15 Thu 17:00 P OGs

Stochastic Wavepacket Approach to Modelling Penning Ionisation — •Alexander Blech[1], Daniel M. Reich[1], Edvardas Narevicius[2], and Christiane P. Koch[1] — [1]Universität Kassel, Deutschland — [2]Weizmann Institute of Science, Rehovot, Israel

Cold Penning ionisation reactions of metastable helium display sharp tunneling resonances. In this regime the reactants can be described by a modicum of partial waves with one dominant one, when the collision energy matches a resonance [1,2].

Recently, the products of the Penning ionisation reaction have been detected using VMI. For Ar colliding with He*, an additional ring is observed which is absent for Kr. This situation can be attributed to the spin-orbit interaction. However, the angular dependence is more isotropic than one would expect. We therefore model the ionisation process using a stochastic wavepacket approach and calculate the cross section in the exit channels.

[1] Henson et al., Science 338, 234 (2012).
[2] Klein et al., Nat. Phys., doi:10.1038/nphys3904, (2016).

MO 16.16 Thu 17:00 P OGs

Three-body bound states induced by a p-wave resonant two-body interaction in 1D — •Lucas Happ[1], Maxim A Efremov[1,2], and Wolfgang P Schleich[1,3] — [1]Institut für Quantenphysik and Center for Integrated Quantum Science and Technology (IQ^{ST}), Universität Ulm, D-89081 Ulm, Germany — [2]A.M. Prokhorov General Physics Institute, Russian Academy of Sciences, 119991 Moscow, Russia — [3]Institute for Quantum Science and Engineering (IQSE), Department of Physics and Astronomy, Texas A&M University, College Station, TX 77843

The emergence of an infinite number of bound states in a system of three bosons, given the s-wave two-body binding energy is at the dissipation threshold, was first predicted by V. Efimov in 1970, and finally verified experimentally during the last decade. Since then, studies are exploring this effect in one- and two-dimensional systems, for different types of two-body interactions (long- and short-range) as well as symmetries of the underlying two-body resonance.

In this poster we present a system containing one light particle and two identical heavy ones, provided i) all particles move on a line, and ii) the heavy-light interaction potential has a p-wave (asymmetric) resonance. Within the Born-Oppenheimer approach we find the corresponding energy spectrum and the number of three-body bound states.

MO 16.17 Thu 17:00 P OGs

Correlated electron-nuclear dissociation dynamics: Classical versus quantum motion — •Julian Albert, Thomas Schaupp, and Volker Engel — Institut für Physikalische und Theoretische Chemie, Universität Würzburg, Emil-Fischer-Str. 42, 97074 Würzburg

We investigate the coupled electron-nuclear dynamics in a model system which undergoes dissociation. In choosing different initial conditions, the cases of adiabatic and non-adiabatic dissociation are realized. We treat the coupled electronic and nuclear motion in the complete configuration space so that classically, no surface hopping procedures have to be incorporated in the case that more than a single adiabatic electronic state is populated during the fragmentation. Concerning the highly anharmonic interaction potential, where it is expected that classical mechanics substantially deviates from quantum mechanics, it is found that the densities and fragmentation yields obtained from the two treatments are in astonishingly good agreement.

MO 16.18 Thu 17:00 P OGs

Transient wavepacket dynamics in excited-state potentials — Markus Gühr and •Carsten Henkel — Universität Potsdam

Time dependent wavepacket dynamics driven by short laser pulses can be simulated in two different ways. First, the static eigenstates of the system can be superposed with a spectral phase given by the excitation pulse. Second, a given static wavepacket can be propagated in the system's potential by solving the time-dependent Schrödinger equation.

Using molecular vibrational wavepackets, we illustrate that the two methods may deliver slightly different results at small propagation times and give an intuitive explanation based on causality. We point towards methods to verify the short-scale dynamics experimentally.

MO 16.19 Thu 17:00 P OGs

bound state properties of two dipoles in harmonic waveguides — •gaoren wang[1], panogiotis giannakeas[2], and peter schmelcher[1] — [1]Zentrum für Optische Quantentechnologien,Universität Hamburg — [2]Department of Physics and Astronomy, Purdue University

The bound states of two dipoles in a harmonic waveguide are investigated based on the local frame transformation (LFT) approach. Both the identical Bosonic and Fermionic dipoles are considered. In the weak dipole interaction regime, the length scale separation, which is the key prerequisite for the application of LFT approach, is verified by examing the wavefunction. Comparing the bound state energies obtained from LFT approach with the numerically accurate ones, it is found that, away from the threshold, the LFT approach with single partial wave approximation is accurate enough. Close to the threshold, higher partial wave states are strongly coupled, and more partial wave states should be incorprated in the LFT approach to obtain accurate results.

MO 16.20 Thu 17:00 P OGs

Generalization of the Davydov Ansatz — •Michael Werther[1,2] and Frank Grossmann[1] — [1]TU Dresden, Institut für Theoretische Physik, Zellescher Weg 17, 01069 Dresden — [2]Max Planck Institute for the Physics of Complex Systems, Nöthnitzer Straße 38, 01187 Dresden

The Davydov Ansatz, originally introduced to study transport of bio-

logical energy in proteins by Davydov and co-workers[1], is an efficient numerical tool for approximate solution of the Schrödinger-equation for different Hamiltonians. In recent works the D1-Ansatz has been successfully applied to the spin boson model as well as the Holstein molecular crystal model.[2] For strong coupling the D1-Ansatz wave function can only insufficiently describe the dynamics of the system, according to its fixed phase space width. Here we propose an extension by adding a further degree of freedom which enables squeezing in phase space.[3] The Dirac-Frenkel variational principle is applied to establish equations of motion for the parameters. First numerical results, executed for the prototype system with one oscillator, show a considerable improvement especially for strong coupling. This allows for an improved description of systems with many oscillators. As an outlook we present how the Davydov Ansatz can be used to include temperature effects into these models for strong coupling.

[1] A.S. Davydov, N.I. Kislukha, Phys. Status Solidi B 59 (1973) 465.

[2] K.-W. Sun et.al., J. Chem. Phys. 142 (2015) 212448

[3] F. Grossmann et.al., Chem. Phys. (2016), in press