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ABSTRACT

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SINGLE-MOLECULE INFRARED SPECTROSCOPY WITH SCANNING TUNNELING MICROSCOPY

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ABSTRACT

Probing vibrations at the single-molecule level is essential for achieving bond-specific chemical control in realistic heterogeneous environments. Here, we introduce a new measurement scheme that integrates frequency-tunable infrared excitation with scanning tunneling microscopy to characterize vibration-mediated nuclear motions of individual molecules. The technique is first validated by monitoring infrared-induced rotation of the ethynyl radical and is subsequently applied to mapping the conformational dynamics of pyrrolidine. The resulting broadband spectra capture fundamental vibrational modes as well as rich overtone and combination bands that are inaccessible by conventional spectroscopic methods, as confirmed through isotopic substitution experiments. Density functional theory calculations reveal that delocalized vibrational modes coupled with pyrrolidine ring puckering drive the observed structural transitions, indicating altered selection rules compared to traditional infrared spectroscopy. This experimental platform enables the probing of molecular vibrations and transformations with atomic-scale precision.

ABSTRACT TOPIC

- Chemical Physics

KEYWORDS

Single-molecule spectroscopy, Infrared excitation, Scanning tunneling microscopy, Molecular vibrations, Conformational dynamics

Memory Kernel Coupling Theory for Spin-Phonon Interactions in Molecular Qubits

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ABSTRACT

Dynamical observables are often described by time correlation functions (TCFs). However, efficiently computing TCFs for complex quantum systems remains a significant challenge, as it typically requires solving the full dynamical evolution of the system. In this work, we propose the memory kernel coupling theory (MKCT)—a general theoretical framework for calculating TCFs. This approach builds upon the memory kernel formalism established by Mori and avoid the calculation of projected dynamics by further decomposing the memory kernel into auxiliary kernel functions. As a result, only steady state moments are required to obtain the TCFs, which greatly simplify the calculation of quantum dynamics in complex systems. This general framework is numerically validated on typical open quantum systems, namely the spin-boson model and the Anderson impurity model. Furthermore, we apply it to systems with nonlinear environment coupling, obtaining accurate dynamical and spectral properties. Finally, we employ MKCT to study the spin-phonon relaxation process in a molecular qubit. The numerical results successfully explain novel spin relaxation phenomena observed in experiments.

REFERENCE STYLE

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KEYWORDS

Spin-phonon interactions, Molecular qubits, memory kernel

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AI acceleration of AIMD simulation of electrochemical interfaces

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ABSTRACT

Ab initio molecular dynamics (AIMD) has been proven to be a powerful tool to study complex chemical systems such as electrochemical interfaces [1]. Insisting on rigorous treatment of electrochemical interfaces both quantum and statistical mechanically, not only has AIMD helped resolve the microscopic structures of electric double layers under bias potential [2], often in collaboration with in situ spectroscopic characterization, but also demonstrated that water adsorption on metal electrodes like Pt has significant impact on dielectric properties of the interfaces, leading to negative capacitive response and thus bell-shaped differential capacitances of Helmholtz layers [3].

The high computational cost of AIMD however limits its application to small model systems consisting of hundreds of atoms at timescale of tens of ps. While, AI-accelerated AIMD (AI2MD) significantly increases size and timescale, promising in situ modeling of realistic electrochemical systems. A prerequisite is that machine learning potentials (MLPs) accurately capture long-range electrostatics and dielectric responses. In this talk, I present our electrochemical MLP (ec-MLP) using a hybrid scheme of Wannier localization and polarizable electrode methods to account for interface polarization [4]. The ec-MLP has been validated against AIMD of electrified Pt/water interfaces, reproducing the bell-shaped differential capacitive curve.

The ultimate test is of course being able to connect the simulation models to experiments under the same electrochemical conditions. The bridge in between is through spectroscopies. Envisioning the important role of spectroscopies, we have been developing AI accelerated methods for computing vibrational (IR, Raman and SFG) [5] and NMR [6,7] spectroscopies of electrochemical systems, so as to eventually be able to compare against experiments at the same level of complexity (e.g. composition, heterogeneity, disorder, as well as external condition). The hope is to combine in situ/operando characterizations to resolve the structures of highly complex materials, interfaces and even interphases in electrochemical devices.

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KEYWORDS

AIMD, Machine learning potentials, Electrochemical interfaces, In situ spectroscopy, Electric double layer

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Momentum-dependent nonlinear optical spectroscopy of electrical double layer

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ABSTRACT

Electrical double layer (EDL) controls the energy transfer and chemical reaction pathway at many aqueous interfaces, yet to characterize the interface-specific hydrogen- (H-)bonding network and ionic structure therein remains a challenge. Nonlinear optics is known as a perceptive probe to the interfacial properties, whereas the structural identification of an EDL is difficult owing to interference of various nonlinear optical responses. In this talk, I shall report our recent development of a sum-frequency (SF) spectroscopic scheme with varying photon momenta as an all–optic solution for retrieving the vibrational spectra of the bonded water layer and the ion diffuse layer, and hence microscopic structural and charging information about an interface. Application of the method to a model surfactant–water interface reveals a hidden weakly-donor-H-bonded water species, suggesting an asymmetric hydration-shell structure of fully solvated surfactant headgroups. In another application to a zwitterionic phosphatidylcholine lipid monolayer–water interface, we find a highly polarized bonded water layer structure associating to the phosphatidylcholine headgroup, while the diffuse layer contribution is experimentally proven to be negligible. Our all-optic method offers an in situ microscopic probe of electrochemical and biological interfaces and the route toward future imaging and ultrafast dynamics studies. At the end of this talk, I shall present an extension of this project to an ongoing study on hydroxide ion depletion at the air/water interface.

REFERENCE STYLE

Momentum-Dependent Sum-Frequency Vibrational Spectroscopy of Bonded Interface Layer at Charged Water Interfaces, *Science Advances*, 9, eadg2823, 2023, Y. Hsiao, T.-H. Chou, A. Patra, and Y.-C. Wen.

KEYWORDS

Sum-frequency vibrational spectroscopy, water interface, bonded interface layer, electrical double layer, diffuse layer.

Anion Binding in Solution Investigated by Ultrafast Vibrational Spectroscopy

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ABSTRACT

In this talk, the ultrafast structural dynamics and nonlinear spectroscopic study of ion binding and recognition in solution will be presented. By constructing host-guest interaction systems and studying the dynamics between anions and receptors using linear and nonlinear vibrational spectroscopy techniques, it was found that enthalpy change is the main driving force for anion recognition. Additionally, the reorientational time constant of anions and the activation energy of rotational relaxation dynamics processes during the ion recognition were reported. Through the combination of theoretical analysis and experimental results, it was revealed that reorientational dynamics is controlled by solvent viscosity and the anion recognition achieved through rearrangement of hydrogen bond structures. This study, based on the framework of classical thermodynamics and ultrafast dynamics, provides insightful guidance for future supramolecular chemistry and is of great significance for better understanding ion transport and regulation in life processes.

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KEYWORDS

Anion Recognition, Structural Dynamics, Ultrafast IR Spectroscopy

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Open system quantum dynamics approaches for chemical dynamics: Methodological developments and applications

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ABSTRACT

Open system quantum dynamics provides a rigorous route to describing chemical processes in condensed phases, where strong system-bath coupling, non-Markovian effects, and quantum coherence often coexist. This talk first reviews recent advances from our group in hierarchical equations of motion (HEOM) and tensor network methods that substantially expand the scope of accurate reduced dynamics simulations. These methods are applied to several representative chemical problems. In strongly light-matter coupled molecular systems, we show how collective coupling, vibrational structure, and dissipation shape polariton and plasmon-exciton relaxation and spectroscopic signals. For atoms and molecules scattering on metal surfaces, HEOM captures transient charge transfer and nonadiabatic dynamics beyond simplified electronic friction models. Together, these results illustrate how open system quantum dynamics can connect microscopic Hamiltonians with experimentally measurable dynamics and spectra across a broad range of chemical dynamics problems.

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KEYWORDS

Hierarchical equations of motion, Chemical dynamics, Strong light-matter interaction, Nonadiabatic dynamics

Beyond Pairwise Interactions in Ternary Heteromolecular Clusters

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ABSTRACT

Ternary heteromolecular clusters represent the smallest nontrivial molecular systems in which intermolecular interactions extend beyond a simple pairwise picture, making them ideal platforms for probing many-body effects, particularly nonadditive three-body contributions, as well as the competition and cooperativity of noncovalent interactions. In this presentation, I will discuss our recent experimental and theoretical studies of ternary clusters using Fourier-transform microwave spectroscopy in combination with high-level quantum-chemical calculations. Our results show that water and CO_2 can play active and sometimes unexpected roles in molecular nucleation, microsolvation, CO_2 capture, and solvent-controlled reaction pathways. In particular, stepwise hydration can alter the interaction pattern of amine-CO_2 complexes and may facilitate bicarbonate formation, while solvent molecules can redirect the geometry of pre-reactive intermediates and affect reaction selectivity. These findings demonstrate that ternary clusters provide a sensitive molecular-level platform for understanding how beyond-pairwise interactions govern structure, stability, and reactivity in chemically relevant environments.

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KEYWORDS

Rotational Spectroscopy, Fourier transform microwave spectroscopy, Non-covalent interactions, Ternary heteromoclecular cluster

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Development of *Ab Initio* Anharmonic Algorithms

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ABSTRACT

One of the central challenges in Chemical Physics is that the vibrational Hamiltonian of a given molecular system is not known and the construction of the vibrational Hamiltonian requires performing computational expensive electronic structure calculations. The second challenge is known as "curse of dimensionality" that severally limits the size of the quantum systems that can be solved. Our group has been working on developing "*Ab Initio* Anharmonic Algorithms" (A^3) to simulate and to gain further insight of vibrational coupling in the proton transfer processes.

To fully describe the quantum dynamics of a solvated hydronium will require treating all 33 degrees of freedom (DoF). Even if one assumes that only the 12 DoF of the central H_3O^+ are important remain as a numerical challenging task. Our A^3 codes work with Cartesian coordinates make the construction of kinetic energy operator (KEO) a trivial task. Furthermore, we have developed a set of code to construct potential energy operator (PEO) with different resolutions that can balance computational costs and accuracy. Very recently, we are working to integrate A^3 with Cytnx to transfer PEO into Matrix Product Operator (MPO) form and to use DMRG tools to solve time independent Schrödinger equations with up to 12 DoF of the central H_3O^+.

REFERENCE STYLE

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KEYWORDS

Chemical Physics, Vibrational Spectroscopy, Quantum Resonance

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New developments in density-matrix based quantum embedding approaches to strongly correlated systems

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ABSTRACT

Developing electronic structure theory for strongly correlated systems (SCSs) is regarded as one of the greatest challenges in molecular quantum chemistry and condensed matter theory. A lot of efforts have been devoted to the development of various quantum embedding techniques to treat complex SCSs accurately and efficiently. Density-matrix embedding theory (DMET), formulated based on the Schmidt decomposition of Slater determinants, provides a systematic framework to combine low- and high-level quantum chemistry methods to treat strongly correlated systems. In this talk, I will present our recent efforts of further developing and applying DMET-based quantum embedding approaches to strongly correlated systems including 3d and 4f single-ion magnets (SIMs) and lanthanide luminescent materials [1-4].

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KEYWORDS

Quantum embedding theory, Density-matrix embedding theory, strongly correlated systems, single-ion magnets, lanthanide luminescent materials

Quantum Dynamical Simulations of Two-Dimensional Spectroscopy via the Schrödinger–Langevin Equation

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ABSTRACT

Two-dimensional spectroscopy (2DS) provides a powerful window into ultrafast electronic and vibrational dynamics, yet its interpretation remains challenging due to the intricate interplay between coherence, dissipation, and nonlinear light–matter interactions. In this work, we present a unified theoretical framework for simulating multidimensional spectroscopic signals based on the quantum Schrödinger–Langevin equation (QSLE). It combines explicit wavepacket dynamics in the Schrödinger picture with a Langevin friction operator to incorporate environmental effects. By treating system–field interactions explicitly while retaining a tractable description of dissipation, this framework enables efficient and physically transparent simulations of coupled electronic–nuclear motion in complex environments. We demonstrate the generality of the method through applications to 2D electronic, 2D electronic–vibrational, and 2D infrared spectroscopy. The QSLE-based simulations reveal rich spectral features arising from nontrivial quantum effects, including Berry phase contributions, vibronic coherence, and Fermi resonances. These results highlight the capability of the QSLE approach to bridge quantum dynamics and nonlinear spectroscopy, providing a versatile platform for interpreting multidimensional spectra and uncovering signatures of quantum coherence in molecular systems.

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2. Frozen-mode small polaron quantum master equation with variational bound for excitation energy transfer in molecular aggregates, *J. Chem. Phys.*, **150**, 224110, 2019, H.-H. Teh, B.-Y. Jin, and Y.-C. Cheng.

KEYWORDS

Two-dimensional spectroscopy, Schrödinger–Langevin equation, Dissipative systems, Electronic–vibrational coupling

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Unified Theoretical Framework of Metal-Support Interaction for Designing Ultrastable and Active Catalysts

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ABSTRACT

The rational design of supported nanocatalysts requires precise control over metal-support interaction (MSI) to balance stability and activity. We have established a unified theoretical framework that transforms MSI from qualitative observations into a quantitative design tool. This framework begins with a Sabatier principle for sintering, which reveals a universal volcanic dependence of sintering kinetics on MSI strength. This principle identifies an "optimal interaction" window that simultaneously suppresses Ostwald ripening and particle migration, enabling catalysts to reach or exceed the Tammann temperature. To provide a rigorous physical basis for this principle, we utilized interpretable machine learning to disentangle MSI into short-range metal-metal and metal-oxygen interactions. This quantitative approach yields a predictive formula for interfacial adhesion and a universal criterion for suboxide encapsulation across diverse oxide supports. Finally, we extended this framework to nitride-supported systems, demonstrating that strong metal-metal interactions drive spontaneous support restructuring and encapsulation. In ammonia synthesis, this structural evolution constructs unique interfacial perimeter sites that effectively decouple nitrogen activation from hydrogen poisoning. Collectively, this integrated work provides a robust strategy for the rational engineering of next-generation ultrastable and high-performance industrial catalysts.

REFERENCE STYLE

Sabatier principle of metal-support interaction for design of ultrastable metal nanocatalysts, *Science*, 374, 1360–1365, 2021, S. Hu, W.-X. Li.

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Strong Metal–Metal Interaction-Induced Encapsulation of Cobalt by Lanthanum Nitride for Efficient Ammonia Synthesis, *J. Am. Chem. Soc.*, 2026, J. Luo, W.-X. Li. et al., https://doi.org/10.1021/jacs.6c01112

KEYWORDS

MSI theory, Sabatier principle, Machine learning, Encapsulation, Ammonia synthesis

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OBSERVING MOLECULAR TRANSPORT THROUGH LIVING CELL MEMBRANES BY NONLINEAR OPTICAL SPECTROCOPY

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ABSTRACT

The nonlinear optical phenomenon Second Harmonic Generation (SHG), operated in scattering and microscopy modes, can be developed for observing, with time and spatial resolutions, the adsorption and transport of small molecules at the membranes of liposomes and biological cells such as human and bacterial cells. The physical principles and theoretical foundation of this nonlinear optical probe of surfaces of colloidal particles including biological cells have been well-established. Its application to biological cells provides an unprecedented opportunity to understand a variety of membrane related phenomena and biological mechanisms.

We have shown that SHG can be used to observe, in real time and with spatial resolution, adsorption and transport of molecules at living cell membranes. With the unique ability afforded by SHG to detect molecules at membranes, we have embarked on a series of studies to gain understanding of molecule-membrane interactions in liposomes and living bacterial and mammalian cells. The following topics will be discussed: membrane phase transition, membrane asymmetry, thermosensitive liposomes, ion channel and membrane diffusive transport, molecular structural influence on membrane transport, efflux rates, mechanosensitive channels, antibiotics interaction with membranes, Gram bacterial differentiation mechanism, and bacteria membrane homeoviscous adaptation.

The other 2nd order nonlinear optical phenomenon Sum Frequency Generation, with the characteristic of molecular selectivity, can in principle be also applied to the study of molecule-membrane interactions in living biological cells. The experiments will be more challenging but the information acquired will be more revealing.

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KEYWORDS

Second Harmonic Generation, Molecule-Membrane Interactions, Liposome Membranes, Bacteria Membranes

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What Leads to Aggregation-Induced Emission?

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ABSTRACT

Molecular aggregation typically leads to fluorescence quenching, yet a small class of systems exhibits the opposite behavior. Using ultrafast UV–vis and infrared multi-pulse mixing schemes together with nonlinear spectroscopic techniques, we systematically probe the evolution dynamics of molecular electronic excited states. This approach elucidates the mechanism of aggregation-induced emission (AIE) in TPE molecules and reveals that their fluorescence does not obey Vavilov's rule. Building on this mechanistic insight, we further demonstrate control of photochemical reactions through mode-selective vibrational excitation.

REFERENCE STYLE

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KEYWORDS

AIE, Conical intersection

Matter Growth in the Interstellar Medium: A Chemical Physics Approach to Molecular Complexity in the Era of AI

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ABSTRACT [*major heading style: Bold 12 pt, UPPERCASE*]

Understanding how matter grows from simple atoms to complex molecules is a fundamental problem in chemical physics, especially under the extreme low-temperature, low-density, and strongly non-equilibrium conditions of the interstellar medium (ISM). In star-forming environments, molecular complexity emerges through the coupled action of microscopic reaction dynamics, grain-surface diffusion and chemistry, and macroscopic physical evolution, making the ISM a unique natural laboratory for chemical physics beyond terrestrial conditions.

In this talk, I will present recent progress toward a unified chemical physics based framework for molecular growth in the ISM. On the modeling side, multiphase gas-grain simulations incorporating nonthermal processes show that complex organic molecules can form efficiently even in cold (~10 K) environments, highlighting the key roles of non-equilibrium surface chemistry and stochastic effects. In parallel, improved treatments of grain-surface diffusion and reaction kinetics provide quantitative constraints on microscopic parameters, helping bridge laboratory-scale physics and astrophysical conditions.

To address the increasing complexity of astrochemical reaction networks, we further develop physics-informed and data-driven approaches for reaction prediction, including deep learning based methods that enable efficient exploration of large reaction spaces beyond traditional expert-curated networks. Complementarily, automated spectral analysis makes it possible to identify molecular lines with high recall from large observational datasets, directly connecting theoretical models with observable signatures.

By integrating reaction prediction, chemical evolution, and spectral inference, this framework moves toward a closed-loop description of molecular growth under interstellar conditions. It reduces degeneracies in interpreting line-rich spectra and provides a scalable route for connecting microscopic chemical physics with macroscopic observables. More broadly, these results show how modern modeling and AI can work together to advance the physical understanding of non-equilibrium

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matter evolution, molecular complexity, and the chemical initial conditions of star and planet formation, including pathways potentially relevant to prebiotic chemistry.

REFERENCE STYLE

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KEYWORDS

Interstellar molecules, Matter growth, Chemical physics, Molecular complexity, Artificial intelligence

High-Resolution Reactive Scattering Experiments Using Velocity Map Ion Imaging

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ABSTRACT

The crossed molecular beam technique, combined with modern laser-based detection, has become a pivotal tool for unraveling the quantum dynamics of chemical reactions. Recent advances in high-resolution imaging and ionization methods have enabled unprecedented access to state-to-state dynamics, offering new insights into fundamental molecular processes [1-2].

In this talk, we present comprehensive ion imaging results for benchmark systems including H + HD/D_2 [3-4], N + D_2 and H + O_2. The observed differential cross sections and quantum-state-resolved product distributions provide stringent tests for theoretical models and contribute to a deeper mechanistic understanding of reaction dynamics at the quantum level.

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KEYWORDS

Reactive scattering, Differential cross section, High resolution, Velocity map imaging, Quantum effects

Tunable Coherent Energy Transfer Networks in Quasi-2D Perovskites Revealed by Two-Dimensional Electronic Spectroscopy

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ABSTRACT

Quasi-2D hybrid organic-inorganic perovskites (HOIPs) combine the excellent stability of 2D HOIPs and high carrier mobility of 3D HOIPs, making them promising candidates for advanced optoelectronic devices. However, the multiphase coexistence in solution-processed quasi-2D HOIP films leads to complex energy transfer pathways, which remain poorly understood due to the limited time and spectral resolution of conventional ultrafast spectroscopic techniques. In this work, we investigated model quasi-2D HOIP spin-coated thin films [$(PEA)_2(MA)_2Pb_3I_{10}$ and $(PMA)_2(MA)_2Pb_3I_{10}$, hereafter abbreviated as PEA and PMA, respectively] with similar structures but different organic layer spacing. Firstly, we characterized them using steady-state spectroscopy and found that both perovskite films exhibit a multiple-phase coexisting phenomenon. Then, we conducted femtosecond transient absorption spectroscopy (Fs-TAS) experiments and observed energy funneling in two samples. Subsequently, we performed two-dimensional electronic spectroscopy (2DES) tests on the two samples. Clear cross-peaks indicated coupling between different phases in quasi-2D HOIPs. By extracting the dynamics of the cross-peaks, we observed bidirectional energy transfer dynamics in adjacent and non-adjacent phases of the two samples within 1 ps, which deviates from typical Förster resonance energy transfer (FRET). It reveals a complex energy transfer network in quasi-2D HOIPs and we attributed it to coherent energy transfer (CET) caused by the interphase delocalization of excitons. By comparing the energy transfer dynamics of PEA and PMA, we found that PMA, due to its shorter organic layer spacing, exhibited enhanced interphase coupling and thus faster CET. Our research provides a potential approach to optimize their optoelectronic performance via cation engineering, which will benefit the design of high-performance perovskite-based devices.

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KEYWORDS
Two-dimensional electronic spectroscopy, Hybrid perovskite, Coherent energy transfer, Exciton delocalization, Cation engineering

Quantum Interferences in Photodissociation of Small Free Radicals

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ABSTRACT

The near-ultraviolet state-to-state predissociation dynamics of SH($A^2\Sigma^+$, v', N') are probed as a function of rovibrational levels of $A^2\Sigma^+$, from v'=0 to 7 up to the S(^{1}D) threshold. The S(^3P_J) product spin-orbit branching fractions change drastically as the $A^2\Sigma^+$ v' level tunes across the crossing points of the repulsive states $^4\Sigma^-$, $^2\Sigma^-$, and $^4\Pi$, manifesting quantum interferences among the three repulsive states. Furthermore, near the H + S(^{1}D) threshold, the bound $A^2\Sigma^+$ v'=5 or 6 level and the continuum $^2\Sigma^-$ state can both be optically excited with good Franck-Condon factors from $X^2\Pi$ $v''\geq3$. Since these two excited states are coupled via spin-orbit interactions and lead to the same S(^3P_J) products, quantum interferences (predissociation of discreet level of $A^2\Sigma^+$ *vs*. direct dissociation of $^2\Sigma^-$ continuum) would result due to the Fano-Feshbach resonance, and the S(^3P_J) product spin-orbit branching ratios and angular distributions vary drastically across the discreet-continuum resonance. The S(^3P_J) product distributions could provide deeper insights into the quantum interference phenomenon.

The $\tilde{A}^2A''$-$\tilde{X}^2A'$ predissociation dynamics of the HCO radical is examined at a state-to-state level in a combined experimental and theoretical study. The complete CO (v, J) product state distributions from various vibrational levels of the $\tilde{A}^2A''$ state are obtained from the H-atom product translational energy distributions. The CO product rotational distributions are highly inverted, and furthermore, intensity oscillations at high J states are observed. The quantum reaction dynamics show that the oscillations in the CO product state distributions are attributed to an interference effect stemming from unique features on the ground-state potential energy surface (PES) of HCO, following the decay of the excited $\tilde{A}^2A''$ state to the ground $\tilde{X}^2A'$ state via Renner-Teller coupling at linearity. This interference effect is exquisitely sensitive to the PES and thus can be used to probe dynamically important regions of the reactive PES.

KEYWORDS

Photodissociation, quantum interferences, SH radical, HCO radical.

Ultrafast Spectroscopy at Microscale: Principles and Applications

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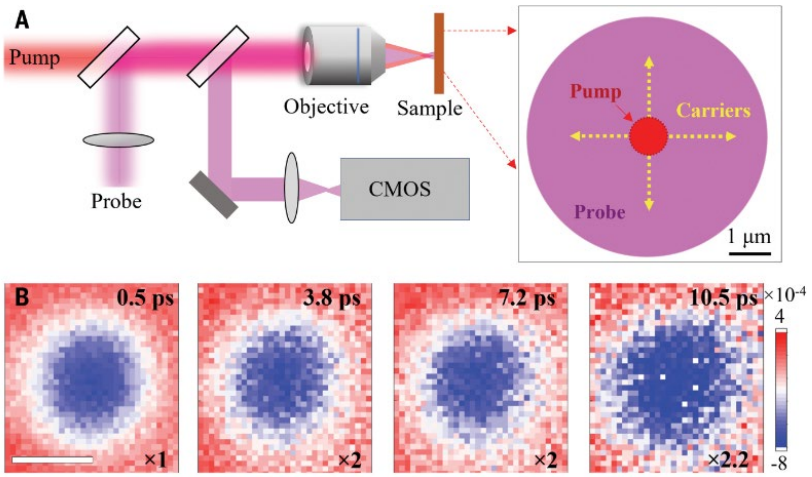
ABSTRACT

Ultrafast spectroscopy imaging is an important technical means for the characterization of charge carriers, which are widely used in the fields of materials, information, physics and chemistry. In recent years, we have been committed to developing spectral measurement systems with spatial, temporal, and momentum resolution capabilities, studying the carrier characteristics in semiconductor material systems, and laying an important foundation for a deeper understanding of their photoelectric properties and related devices. In this talk, I mainly introduce some progress based on micro/nanoscale ultrafast spectroscopy imaging in the study of carrier mobility, electron phonon coupling, as well as the spectral characteristics and related carrier dynamics of nanoscale semiconductor materials induced by the edges.



Keywords: Ultrafast spectroscopy, carrier diffusion, BAs, Spatial resolution, semiconductor materials

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Nonadiabatic Field:
A Conceptually Novel Approach for Nonadiabatic Transition Dynamics

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Nonadiabatic transition dynamics lies at the core of many electron/hole transfer, photoactivated, and vacuum field-coupled processes. About a century after Ehrenfest proposed "Phasenraum" and the Ehrenfest theorem, we report a conceptually novel trajectory-based nonadiabatic dynamics approach, nonadiabatic field (NaF), based on a generalized exact coordinate–momentum phase space formulation of quantum mechanics, which includes constraint phase space, whose mathematical structure is diffeomorphic to the complex Stiefel manifolds, for electronic degrees of freedom (DOFs) and infinite Wigner phase space for nuclear DOFs. It does not employ the conventional Born–Oppenheimer or Ehrenfest trajectories in the nonadiabatic coupling region. Instead, in NaF the equations of motion of the independent trajectories involve a nonadiabatic nuclear force term in addition to an adiabatic nuclear force term of a single electronic state. A few benchmark tests for gas phase and condensed phase systems indicate that NaF offers a practical tool to capture the correct correlation of electronic and nuclear dynamics for processes where the states remain coupled all the time, as well as for the asymptotic region where the coupling of electronic states vanishes. NaF has been integrated in the PSiNad (phase space-integrated non-adiabatic dynamics) simulation package, where both benchmark models and *ab initio* electronic structure methods can be used to study nonadiabatic transition processes and related spectroscopy.

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CHIRALITY AT AIR/LIQUID INTERFACE AND IN 2D MATERIALS DETECTED BY SHG POLARIZATION AND IMAGING

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ABSTRACT

In this study, we developed and integrated a system for interfacial chirality detection based on second harmonic generation (SHG) techniques, enabling high-sensitivity measurements ranging from molecular orientation analysis and chiral characterization to real-time monitoring of biofilm dynamic processes. The system comprises three home-built modules: a second harmonic generation linear dichroism (SHG-LD) spectroscopy system for studying interfacial molecular orientation and chiral response (for the first time, we observed consistent and directionally reproducible chiral signals from achiral dye molecules at the air/water interface, revealing the mechanism of chirality induction through symmetry breaking at the interface); a high spatial resolution second harmonic generation circular dichroism (SHG-CD) microscopy imaging system for direct chiral imaging of micro/nano-scale chiral materials; and a time-resolved second harmonic generation scattering technique for real-time, quantitative characterization of dynamic processes at soft matter interfaces. The series of SHG spectroscopy and imaging methods developed in this study provide powerful technical tools for in-situ, high-sensitivity investigations into the origins of interfacial chirality, chiral structures of low-dimensional materials, and physicochemical processes at biofilm interfaces.

KEYWORDS

Molecular structure, Molecular dynamics, Second harmonic generation, Interfacial chirality, Biofilm interface

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QUANTITATIVE PHASE RECONSTRUCTION AND $\chi^{(2)}/\chi^{(3)}$ SEPARATION AT CHARGED AQUEOUS INTERFACES USING HIGH-RESOLUTION BROADBAND SFG-VS

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ABSTRACT

Interfacial hydrogen-bonding structures are inherently complex, making their spectra difficult to resolve using conventional SFG-VS. Here, we employ a high-resolution broadband sum frequency generation vibrational spectroscopy (HR-BB-SFG-VS) system to investigate charged surfactant air/water interfaces by measuring SFG intensity spectra of anionic (SDS) and cationic (CTAB, DTAB) surfactants over the 2700-3800 cm^{-1} region, covering both C-H and O-H stretching vibrations. Building on our previous study of the air/pure water interface, where phase-resolved spectra were quantitatively reconstructed from intensity data and validated against independent phase measurements, we extend this approach to charged interfaces by introducing free phase parameters into the lineshape fitting. This method not only enables high-quality fitting of the intensity spectra, but also allows extraction of the relative phase information of individual vibrational modes and reconstruction of the corresponding real and imaginary spectra. The results indicate that this approach can reliably retrieve relative phase information of interfacial vibrational modes without direct phase-resolved measurements, providing an effective route to analyze interfacial hydrogen-bonding structures and their nonlinear optical origins at charged interfaces.

Furthermore, by tuning the ionic strength to vary the Debye length, we aim to distinguish the relative contributions of $\chi^{(2)}/\chi^{(3)}$, and to better understand the hydrogen-bonding water structures at charged interfaces.

KEYWORDS

Charged interfaces, Surfacants, Hydrogen bond, SFG

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Femtosecond optics and X-ray lasers and its application in ultrafast dynamics research

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ABSTRACT

Advanced experimental methods with high spatial-temporal resolutions are essential for ultrafast dynamics at microscopic scales, primarily focusing on the movements of individual electrons or nuclei. The rapid development of Xray free-electron lasers (XFELs) has created a new foundation for ultrafast dynamics research by offering fully coherent ultrabright femtosecond X-ray pulses. The cutting-edge technology developed around the XFELs has enabled revolutionary breakthroughs in multiple disciplines in the past decade. X-ray emission/absorption spectroscopy, X-ray scattering, and X-ray crystallography have quickly adapted to the XFEL facilities, investigating high-resolution structure and dynamics. The development of the XFEL-based time-resolved methods is particularly significant for light-sensitive macromolecules. Novel insights into the structural dynamics are being gathered for a comprehensive understanding of the detailed process and its underlying molecular mechanism.

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KEYWORDS

ultrafast dynamics, Xray free-electron lasers, X-ray emission spectroscopy, X-ray absorption spectroscopy

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Interface THz Nonlinear Optical Spectroscopy

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ABSTRACT

Quantum confinement and proximity effects at material interfaces give rise to a range of novel physical and chemical phenomena, substantially advancing our ability to understand and control material properties. The development of in situ and interface-sensitive characterization techniques is therefore critical for uncovering the microscopic mechanisms governing interfacial systems and for improving device performance. This presentation will outline recent advances in interface nonlinear optical spectroscopic techniques, with a particular focus on their extension into the (multi-)terahertz spectral regime. Applications of these methods in probing interfacial microstructures and dynamic processes will be discussed, highlighting their potential for resolving complex interfacial phenomena with high specificity.

KEYWORDS

Nonlinear optics, interface, Terahertz spectroscopy

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Physics-Regularized Deep Learning for EIS-Based Degradation Diagnosis in Silicon–Carbon Composite Anodes

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ABSTRACT

Silicon–carbon composite anodes provide high capacity but undergo coupled degradation involving lithium inventory loss, active-material isolation, interfacial growth, and impedance increase. Electrochemical impedance spectroscopy (EIS) offers a non-destructive probe of these processes, yet translating spectra into interpretable degradation states remains challenging because direct ground-truth labels for internal degradation modes are rarely available.

Here, we develop a physics-regularized deep learning framework for EIS-based degradation diagnosis of silicon–carbon composite anodes. The framework predicts a set of physically interpretable diagnostic indices, including SOH, resistance growth, and degradation-mode indicators associated with LLI, LAM, and contact loss. The framework combines spectral encoding, reduced electrochemical latent variables, equivalent-circuit-parameter distillation, impedance reconstruction, and monotonicity regularization to encourage physically plausible aging representations. Using a public EIS dataset from commercial silicon–graphite cells, the model is benchmarked against standard neural-network baselines. The results suggest that electrochemical regularization improves stability and interpretability under limited-data conditions. This work positions EIS-based learning as a fast diagnostic surrogate for generating degradation hypotheses, not a replacement for direct experimental validation.

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KEYWORDS
ASim 2026, Abstract template, Paper instruction

Energy storage, Lithium-ion batteries, Battery diagnostics, Machine learning, Electrochemical physics

Observer only, no abstract

3D laser nanoprinting of inorganic nanomaterials

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ABSTRACT

3D printing of inorganic materials with nanoscale resolution offers a different materials processing pathway to explore devices with emergent functionalities. However, existing technologies typically involve photocurable resins that reduce material purity and degrade properties. To address this challenge, we developed two novel photochemical bonding approaches for polymer-independent 3D printing of inorganic materials. Firstly, we developed a photoexcitation-induced chemical bonding (PEB) strategy to directly assemble and print semiconductor quantum dots into 3D nanostructures. Under laser irradiation, the holes excited inside semiconductor quantum dots are transferred to the nanocrystal surface and improve their chemical reactivity, leading to interparticle chemical bonding. Without any additives, quantum dots can be directly printed into arbitrary 3D architectures ranging from linear, curved to volumetric structures at nanoscale resolution (~77 nm). Secondly, we further developed a general photochemical bonding approach for 3D printing more than 10 semiconductors, metal oxides, metals, and their mixtures. Mimicking matter organization at atomic scale, inorganic NCs capped with native ligands were used as artificial atoms, and a small amount of photosensitive bisazide molecules was added to NC dispersion, behaving as artificial chemical bonds. Under laser irradiation, bisazide molecules generate reactive nitrene radicals to bond nanocrystals together via C-H insertion. This technique adapts to a broad spectrum of inorganic materials, including II-VI/III-V semiconductors, metal halide perovskites, metals, and oxides. Printed structures have a large mass fraction (~90%), high mechanical strength (>1 GPa) and broadband chiroptical response.

The bonding photochemistry and nanocrystal building blocks at the nanoscale create “plenty of room” for the material toolbox of 3D printing. The printed structures preserve the intrinsic properties encoded in the nanocrystals and also show additional functionalities stemming from their 3D geometries. We believe that these techniques will unlock new ways to fabricate inorganic devices for mechanical, optical, electrical applications, and even for advanced chip manufacturing.

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KEYWORDS
3D printing, photochemical bonding, inorganic nanomaterials, quantum dots

Chiral ligand-directed nanoscale symmetry breaking forward and inverse design

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ABSTRACT

The rational design of chiral ligand-protected nanoclusters is limited by a fundamental gap: molecular chirality is easy to label, but difficult to translate into predictable nanoscale symmetry breaking. Here we develop CHILI platform, a closed-loop computational discovery framework that connects large molecular libraries, ligand–gold assembly, molecular dynamics sampling, continuous chirality analysis, interpretable feature learning, and inverse ligand evolution. Rather than serving only as a screening workflow, CHILI enables the extraction of physical design rules from high-throughput ligand-induced deformation of Au clusters. We find that strong geometric chirality is not encoded by molecular stereochemistry alone, but by how the ligand shell imposes an asymmetric boundary condition on the metallic core. Two symmetry-breaking channels emerge. In the first, anchor-local motifs near the sulfur binding site inject torque or curly density into the cluster surface, producing a directional bias during structural relaxation. In the second, ligands with weak single-molecule torque can still generate high chirality when biased tri-ring or multi-ring frameworks assemble into a collective chiral envelope, where rigid offset units, spatial superposition, and distal mass distribution select one handed deformation. These results explain why static descriptors define only a necessary region for high chirality, while assembly and conformational sampling determine whether this bias is amplified or lost. The extracted rules, including short-range anchor dominance, the Golden 8 Atomic Principle, and the Heavy Arm Rule, are then embedded into inverse design through protected anchoring, local molecular editing, envelope screening, stability filtering, and reward-based evolution. This work establishes a physics-informed route from molecular libraries to programmable nanoscale chirality, transforming chiral ligand design from empirical stereochemical selection into mechanism-guided engineering of interfacial symmetry breaking.

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KEYWORDS

Chiral nanoclusters, Ligand design, Gold nanoparticles, Symmetry breaking, Machine learning