Quantum Dynamics

Introduction

The conditions in astronomical environments differ substantially from those found in most terrestrial laboratories. Simulating interstellar processes through experiment and theory thus often explores extreme regimes in both our experimental capabilities and theoretical understanding. This is reflected in the activities in our institute, which cover a wide range in temperature and density.

Astrochemical studies with stored molecular ions at the Test Storage Ring and the new Cryogenic Storage Ring aim at a detailed understanding of the formation and destruction of molecules in the cold interstellar medium, and we are currently developing a novel spectroscopic technique for complex interstellar molecules, based on the ultra-sensitive detection of mid-infrared radiation. On the other hand, experiments at the various electron beam ion traps (EBIT) at MPIK study the spectral properties and photo-induced processes of highly charged ions (HCI), which are prevalent in very hot interstellar objects, covering the range from visible light to X-rays. These efforts are complemented by advanced theoretical calculations of processes in intense laser fields, which aid the interpretation of laboratory data and shed light on the origin of interstellar phenomena like ultra-relativistic jets generating positron-electron plasmas, at the far end of the temperature scale.

Photodissociation of CH⁺ inside the Cryogenic Storage Ring

The methylidyne cation CH⁺ was the first molecular ion detected in interstellar space and its high abundance in many interstellar environments continues to puzzle observational astronomers. No efficient formation pathway for CH⁺ at low temperatures is known, and various scenarios introducing non-thermal effects, ranging from turbulent mixing to thermodynamic shocks and Alfvén waves, have been proposed.

A possible pathway to create CH⁺ in space relies on radiative attachment of C⁺ and H. This reaction has not been studied in the laboratory yet, therefore, theory has to resort to the reverse process, the photodissociation of CH⁺, to benchmark the potentials that are involved and understand the relevant resonances.

We have studied the near-threshold photodissociation spectrum of CH⁺ in the new Cryogenic Storage Ring (CSR) [1], at nominal temperatures of 6 K. Unlike earlier studies, we focused on the lowest rotational quantum states J=0-2, to verify the resonances originating from those states and also to monitor the rotational cooling of molecular ions inside the CSR.

To this end we have produced CH⁺ ions in a caesium sputter ion source, mass-selected them, accelerated the ion beam to 60 keV kinetic energy and injected it into the CSR. The ions were stored for several minutes and then exposed to ultraviolet photons from a pulsed optical parametric oscillator (OPO) laser system. The photons excited the ions from the 1Σ⁺ electronic ground state to a vibrational band inside an excited 1Π state, just above the dissociation threshold. From here the ions pre-dissociate into C⁺ and H. The neutral H atoms were detected by a single-particle detector downstream of the laser interaction zone. This two-step dissociation process leads to Feshbach resonances in a narrow energy window around the dissociation threshold. We used state-of-the-art close-coupled calculations to

2.4 Astrophysics with Ions and Strong Light Fields

The image shows the light collector mirror assembly of a mid-infrared detector module developed to detect emission of large gas phase molecular ions in laboratory experiments. Background: Messier 82: Composite of Chandra, HST and Spitzer images (NASA/JPL-Caltech/STScI/CXC/UCF/ESA/AURA/Johns Hopkins University).
Astrophysics with Super Intense Laser Pulses

Astrophysical spectra recorded by space observatories provide the only means to determine the element composition, temperature, density, and velocity of distant celestial objects such as stars, X-ray binaries, black-hole accretion disks, or active galactic nuclei. A large body of reliable atomic data is needed, either from theory or experiment, for the extraction of the expected object properties from astrophysical spectra. The X-ray lines of highly charged ions, in particular, Fe$^{33+}$, are among the brightest in astrophysical spectra, and were observed within the last decade with the orbiting laboratories Chandra and XMM-Newton. The X-ray spectrum of Fe$^{33+}$ is, however, poorly reproduced by astrophysical models: the observed line-strength ratio of the X-ray lines 3C and 3D is in stark contrast with its predicted value, preventing a reliable analysis of observatory data. The sources of astrophysical discrepancy were narrowed down after the first X-ray laser spectroscopic experiment, employing a purely photonic excitation of the ions with the LCLS X-ray free-electron laser (XFEL). A disagreement of 30% between all theoretical predictions and the experimental line-strength ratio was stated, hinting to a shortcoming of atomic structure theory [S. Bernitt et al., Nature 492, 225 (2012)].

We performed a large-scale configuration-interaction calculation of line strengths involving higher-order correlations, suggest explaining the discrepancy [6]. In addition, quantum electromagnetic (QED) corrections were reliably estimated. We furthermore investigate the light-matter interaction of the system in a dynamic way, showing that, for high pulse intensities available at LCLS, the 3C and 3D transitions can show a complete population inversion. Thus, nonlinear dynamic effects are induced, significantly influencing the line strengths, which could not be explained by the previous weak-field modeling. Such effects possibly resolve the discrepancy, motivating the use of light-matter interaction models valid for strong fields also in the analysis of astrophysical spectra. For accurately modeling strong-field effects on line shapes, an improved experimental determination of the XFEL parameters is called for. At sufficiently high intensities, the weak-field atomic theory is also not applicable in corresponding astrophysical scenarios, e.g., in the early stages of the Universe. In the experiment described in [7], this unique state of matter has been generated in a terrestrial laboratory, opening the possibility of scrutinizing such extreme astrophysical phenomena and regimes under controlled conditions. An ultra-relativistic electron beam, produced in an all-optical setup via laser wake-field acceleration, hits a Pb solid target. Due to the complex interaction of the electron beam with the nuclei and the electrons in the target, an ultra-relativistic electron-positron bunch was observed on the rear side of the target, with a fraction up to 50-50 of electrons and positrons depending on the target thickness. The bunch density was found to be sufficiently high that its skin-depths resulting from the length, duration, and bandwidth of the simulated XFEL pulse [7]. The gray shaded area shows the experimentally observed ratio together with its error bar, taken from [6].
target. As a result, only two fundamental QED processes have been found to play a substantial role: 1) bremsstrahlung of electrons and positrons, and 2) electron-positron production by photons, both occurring in the presence of the screened electromagnetic field of the target nuclei. Analytical estimations and numerical integrations of the corresponding kinetic equations agree extremely well with the experimental results on the relative population of electrons and positrons in the generated bunch.

Natalia S. Oreshkina, Zoltán Harman, Autunno Di Piazza, Christoph H. Keitel

Laser-Induced Vibrational Emission: A New Type of Spectroscopy for Complex Molecular Ions

As more and more molecules are identified in interstellar space, two spectral phenomena remain mysterious, even after decades of investigation. The first one, termed the Difuse Interstellar Bands (DIBs), stands for more than 500 absorption features that are seen in many interstellar environments, stretching from the visible to the near-infrared. Only very recently one of these features could be attributed to the \( \text{C}_60^+ \) molecule. The other ubiquitous feature is the so-called Undetected Infrared Emission (UIE) that is seen in the mid-infrared (IR) range throughout the universe. It is not clear at present whether these features have a common origin or stem from the same class of molecules, however, there are strong hints that make complex organic ions likely candidates. To shed light on the interstellar observations, spectroscopic data of complex ions in the gas phase are required. However, owing to the difficulties to achieve high enough densities, experimental data are sparse.

We are currently developing a new spectroscopic technique (in collaboration with researchers of the MPI for astronomy and the Karlsruhe Institute for Technology) based on the direct detection of emission from gas phase molecular ions using highly sensitive blocked-impurity-band (BIB) mid-IR detectors operated in a cryogenic ion beam trap. While the technique should be fairly universal, the implementation is extremely challenging, and the goal for the proof-of-principle experiment is to achieve the first true gas phase electronic spectra of \( \text{C}_60^+ \). To this end, we will store \( \text{C}_60^+ \) inside the cryogenic beam trap at nominal temperatures around 10 K. The stored ions will be exposed to pulsed laser radiation in the near-infrared. As direct fluorescence is a very unlikely process in large molecules, the absorbed energy will be re-distributed among the internal degrees of freedom and – after a delay of typically several ten microseconds – will be re-emitted at vibrational frequencies in the mid-IR. The emitted photons will be detected by the BIB detector assembly and serves as a signal that absorption took place. By monitoring the emission while scanning the frequency of the excitation laser, a gas phase absorption spectrum of the stored molecular species is recorded. After the feasibility of this approach has been demonstrated, it is foreseen to add spectral sensitivity to the mid-IR detector module and thereby get spectroscopic information on both the absorption and emission features. This would permit us to follow the heating and cooling processes that large gas phase molecules undergo in the interstellar medium in real time, and thus this technique has the potential to yield unprecedented and valuable information on the entire class of complex organic molecular ions.

Sandu Kumar, Sebastian George, Jürgen Göck, Holger Kreckel

Laboratory Astrophysics with Highly Charged Ions

The conditions in CryPTEs, developed at the MPlK for investigations of HCl, are close to those of interstellar clouds in terms of low radiation temperature (7 K) and residual gas density (below 10\(^{-14}\) mbar). Thus, the device is also ideally suited to study molecular ions. In commissioning experiments in collaboration with Aarhus University, we succeeded in producing molecular ions (MgH\(^+\)) at the lowest hitherto reported ro-vibrational temperature.

We demonstrated another novel method for controlling at will their internal tempera-