

# AstroChemical Newsletter #84

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## Abstracts

### Gas-phase spectroscopy of photostable PAH ions from the mid- to far-infrared

Sandra D. Wiersma, Alessandra Candian, Joost M. Bakker, Annemieke Petrigani

We present gas-phase InfraRed Multiple Photon Dissociation (IRMPD) spectroscopy of cationic phenanthrene, pyrene, and perylene over the 100–1700  $\text{cm}^{-1}$  (6–95  $\mu\text{m}$ ) spectral range. This range covers both local vibrational modes involving C–C and C–H bonds in the mid-IR, and large-amplitude skeletal modes in the far-IR. The experiments were done using the 7T Fourier-Transform Ion Cyclotron Resonance (FTICR) mass spectrometer integrated in the Free-Electron Laser for Intra-Cavity Experiments (FELICE), and findings were complemented with Density Functional Theory (DFT) calculated harmonic and anharmonic spectra, matching the experimental spectra well. The experimental configuration that enables this sensitive spectroscopy of the strongly bound, photoresistant Polycyclic Aromatic Hydrocarbons (PAHs) over a wide range can provide such high photon densities that even combination modes with calculated intensities as low as 0.01  $\text{km mol}^{-1}$  near 400  $\text{cm}^{-1}$  (25  $\mu\text{m}$ ) can be detected. Experimental frequencies from this work and all currently available IRMPD spectra for PAH cations were compared to theoretical frequencies from the NASA Ames PAH IR Spectroscopic Database to verify predicted trends for far-IR vibrational modes depending on PAH shape and size, and only a relatively small redshift (6–11  $\text{cm}^{-1}$ ) was found between experiment and theory. The absence of spectral congestion and the drastic reduction in bandwidth with respect to the mid-IR make the far-IR fingerprints viable candidates for theoretical benchmarking, which can aid in the search for individual large PAHs in the interstellar medium.

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Full-text URL: <https://arxiv.org/abs/2111.08592>

### Energetic Electron Irradiations of Amorphous and Crystalline Sulphur-Bearing Astrochemical Ices

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Laboratory experiments have confirmed that the radiolytic decay rate of astrochemical ice analogues is dependent upon the solid phase of the target ice, with some crystalline molecular ices being more radio-resistant than their amorphous counterparts. The degree of radio-resistance exhibited by crystalline ice phases is dependent upon the nature, strength, and extent of the intermolecular interactions that characterise their solid structure. For example, it has been shown that crystalline  $\text{CH}_3\text{OH}$  decays at a significantly slower rate when irradiated by 2 keV electrons at 20 K than does the amorphous phase due to the stabilising effect imparted by the presence of an extensive array of strong hydrogen bonds. These results have important consequences for the astrochemistry of interstellar ices and outer Solar System bodies, as they imply that the chemical products arising from the irradiation of amorphous ices (which may include prebiotic molecules relevant to biology) should be more abundant than those arising from similar irradiations of crystalline phases. In this present study, we have extended our work on this subject by performing comparative energetic electron irradiations of the amorphous and crystalline phases of the sulphur-bearing molecules  $\text{H}_2\text{S}$  and  $\text{SO}_2$  at 20 K. We have found evidence for phase-dependent chemistry in both these species, with the radiation-induced exponential decay of amorphous  $\text{H}_2\text{S}$  being more rapid than that of the crystalline phase, similar to the effect that has been previously observed for  $\text{CH}_3\text{OH}$ . For  $\text{SO}_2$ , two fluence regimes are apparent: a low-fluence regime in which the crystalline ice exhibits a rapid exponential decay while the amorphous ice possibly resists decay, and a high-fluence regime in which both phases undergo slow exponential-like decays. We have discussed our results in the contexts of interstellar and Solar System ice astrochemistry and the formation of sulphur allotropes and residues in these settings.

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### VUV Photoabsorption of Thermally Processed Carbon Disulfide and Ammonia Ice Mixtures - Implications for Icy Objects in the Solar System

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Many icy bodies in the solar system have been found to contain a rich mixture of simple molecules on their surfaces. Similarly, comets are now known to be a reservoir of molecules ranging from water to amides. The processing of

planetary/cometary ices leads to the synthesis of more complex molecules some of which may be the harbingers of life. Carbon disulphide (CS<sub>2</sub>) and ammonia (NH<sub>3</sub>) are known to be present on many icy satellites and comets. Reactions involving CS<sub>2</sub> and NH<sub>3</sub> may lead to the formation of larger molecules that are stable under space conditions. In this paper we present temperature dependent VUV spectra of pure CS<sub>2</sub> in the ice phase, and of CS<sub>2</sub> and NH<sub>3</sub> ices deposited as (i) layered, and (ii) mixed ices at 10 K and warmed to higher temperatures until their sublimation. Pure CS<sub>2</sub> ice is found to have a broad absorption in the VUV region, which is unique for a small molecule in the ice phase. In layered and mixed ices, the molecules tend to affect the phase change and sublimation temperature of each other and also leave behind a form of CS<sub>2</sub>-NH<sub>3</sub> complex after thermal annealing. This study of CS<sub>2</sub>-NH<sub>3</sub> ice systems in layered and mixed configurations would support the detection of these species/complexes in mixed molecular ices analogous to that on planetary and cometary surfaces.

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## Discovery of non-metastable ammonia masers in Sagittarius B2

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We report the discovery of widespread maser emission in non-metastable inversion transitions of NH<sub>3</sub> toward various parts of the Sagittarius B2 molecular cloud/star forming region complex: We detect masers in the J,K= (6,3), (7,4), (8,5), (9,6), and (10,7) transitions toward Sgr B2(M) and Sgr B2(N), an NH<sub>3</sub> (6,3) maser in Sgr B2(NS), and NH<sub>3</sub> (7,4), (9,6), and (10,7) masers in Sgr B2(S). With the high angular resolution data of the Karl G. Jansky Very Large Array (JVLA) in A-configuration we identify 18 maser spots. Nine maser spots arise from Sgr B2(N), one from Sgr B2(NS), five from Sgr B2(M), and three in Sgr B2(S). Compared to our Effelsberg single dish data, the JVLA data indicate no missing flux. The detected maser spots are not resolved by our JVLA observations. Lower limits to the brightness temperature are > 3000~K and reach up to several 1e5 ~K, manifesting the lines' maser nature. In view of the masers' velocity differences with respect to adjacent hot molecular cores and/or UCHII regions, it is argued that all the measured ammonia maser lines may be associated with shocks caused either by outflows or by the expansion of UCHII regions. Overall, Sgr B2 is unique in that it allows us to measure many NH<sub>3</sub> masers simultaneously, which may be essential to elucidate their so far poorly understood origin and excitation.

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## Surveying the inner structure of massive young stellar objects using L-band spectroscopy

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We present results from a high spectral resolution (6kms<sup>-1</sup>) survey of five massive protostars in the wavelength range of 2.95 and 3.25 μm, conducted with iSHELL at the InfraRed Telescope Facility (IRTF). Our targets are Mon R2 IRS 2, Mon R2 IRS 3, AFGL 2136, Orion BN and S140 IRS 1. Two of our five targets (Mon R2 IRS 3 and AFGL 2136) show transitions from organic species, with MonR2 IRS 3 showing HCN lines in emission, and AFGL 2136 showing HCN and C<sub>2</sub>H<sub>2</sub> lines in absorption. The velocity of the emission lines of HCN of MonR2 IRS 3A are consistent with CO emission features in lines up to J = 26, as both are red-shifted with respect to the systemic velocity. Carbon monoxide lines also show blue-shifted absorption. This P-Cygni line profile, commonly observed towards massive young stellar objects, is likely due to an expanding shell, which is supported by sub-millimetre velocity maps of HCN. Alternatively HCN emission may arise from the upper layers of a disk photosphere, as has been suggested for the massive protostar AFGL 2591. Absorption lines in AFGL 2136 may either originate in foreground cloud or in the disk photosphere. For a foreground cloud, the data require that the foreground gas only covers the source partially (0.3) at 13 μm. In contrast, absorption lines at 3 and 7 μm require a covering factor of >0.9. Analysing the 13 μm HCN absorption lines in terms of absorption by gas in the photosphere of a disk, results in physical conditions that are consistent over all three vibrational modes. C<sub>2</sub>H<sub>2</sub> absorption lines reveal an increasing temperature and abundance with decreasing wavelength, indicative of a radial abundance gradient. We conclude that the disk model is the best interpretation of the absorption lines of AFGL 2136.

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Full-text URL: <https://www.aanda.org/articles/aa/abs/2022/10/aa43003-21/aa43003-21.html>

## The Formation of Monosubstituted Cyclopropenylidene Derivatives in the Interstellar Medium via Neutral-Neutral Reaction Pathways

Athena R. Flint and Ryan C. Fortenberry

Five substituted cyclopropenylidene derivatives (c-C<sub>3</sub>HX, X = CN, OH, F, NH<sub>2</sub>), all currently undetected in the interstellar medium (ISM), are found herein to have mechanistically viable, gas-phase formation pathways through neutral-neutral additions of X onto c-C<sub>3</sub>H<sub>2</sub>. The detection and predicted formation mechanism of c-C<sub>3</sub>HC<sub>2</sub>H introduces a need for the chemistry of c-C<sub>3</sub>H<sub>2</sub> and any possible derivatives to be more fully explored. Chemically accurate CCSD(T)-F12/cc-pVTZ-F12 calculations provide exothermicities of additions of various radical species to c-C<sub>3</sub>H<sub>2</sub>, alongside energies of submerged

intermediates that are crossed to result in product formation. Of the novel reaction mechanisms proposed, the addition of the cyano radical is the most exothermic at -16.10 kcal/mol. All five products are found to or are expected to have at least one means of associating barrierlessly to form a submerged intermediate, a requirement for the cold chemistry of the ISM. The energetically-allowed additions arise as a result of the strong electrophilicity of the radical species as well as the product stability gained through substituent-ring conjugation.

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## Chemistry and dynamics of the prestellar core L1544

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We aim to quantify the effect of chemistry on the infall velocity in the prestellar core L1544. Previous observational studies have found evidence for double-peaked line profiles for the rotational transitions of several molecules, which cannot be accounted for with presently available models for the physical structure of the source, without ad hoc up-scaling of the infall velocity. We ran one-dimensional hydrodynamical simulations of the collapse of a core with L1544-like properties (in terms of mass and outer radius) using a state-of-the-art chemical model with a very large chemical network combined with an extensive description of molecular line cooling, determined via radiative transfer simulations, with the aim of determining whether these expansions of the simulation setup (as compared to previous models) can lead to a higher infall velocity. After running a series of simulations where the simulation is sequentially simplified, we found that the infall velocity is almost independent of the size of the chemical network or the approach to line cooling. We conclude that chemical evolution does not have a large impact on the infall velocity, and that the higher infall velocities that are implied by observations may be the result of the core being more dynamically evolved than what is now thought, or alternatively the average density in the simulated core is too low. However, chemistry does have a large influence on the lifetime of the core, which varies by about a factor of two across the simulations and grows longer when the chemical network is simplified. Therefore, although the model is subject to several sources of uncertainties, the present results clearly indicate that the use of a small chemical network leads to an incorrect estimate of the core lifetime, which is naturally a critical parameter for the development of chemical complexity in the pre-collapse phase.

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## ALCHEMI Finds a "Shocking" Carbon Footprint in the Starburst Galaxy NGC 253

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The centers of starburst galaxies may be characterized by a specific gas and ice chemistry due to their gas dynamics and the presence of various ice desorption mechanisms. This may result in a peculiar observable composition. We analyse the abundances of CO<sub>2</sub>, a reliable tracer of ice chemistry, from data collected as part of the Atacama Large Millimeter/submillimeter Array large program ALCHEMI, a wide-frequency spectral scan toward the starburst galaxy NGC 253 with an angular resolution of 1."6. We constrain the CO<sub>2</sub> abundances in the gas phase using its protonated form HOCO<sup>+</sup>. The distribution of HOCO<sup>+</sup> is similar to that of methanol, which suggests that HOCO<sup>+</sup> is indeed produced from the protonation of CO<sub>2</sub> sublimated from ice. The HOCO<sup>+</sup> fractional abundances are found to be (1–2)e–9 at the outer part of the central molecular zone (CMZ), while they are lower (~1e–10) near the kinematic center. This peak fractional abundance at the outer CMZ is comparable to that in the Milky Way CMZ, and orders of magnitude higher than that in Galactic disk, star-forming regions. From the range of HOCO<sup>+</sup>/CO<sub>2</sub> ratios suggested from chemical models, the gas-phase CO<sub>2</sub> fractional abundance is estimated to be (1–20)e–7 at the outer CMZ, and orders of magnitude lower near the center. We estimate the CO<sub>2</sub> ice fractional abundances at the outer CMZ to be (2–5)e–6 from the literature. A comparison between the ice and gas CO<sub>2</sub> abundances suggests an efficient sublimation mechanism. This sublimation is attributed to large-scale shocks at the orbital intersections of the bar and CMZ.

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## Network Analysis Reveals Spatial Clustering and Annotation of Complex Chemical Spaces: Application to Astrochemistry

Alexander Ruf and Grégoire Danger

How are molecules linked to each other in complex systems? In a proof-of-concept study, we have developed the method mol2net (<https://zenodo.org/record/7025094>) to generate and analyze the molecular network of complex astrochemical data (from high-resolution Orbitrap MS1 analysis of H<sub>2</sub>O:CH<sub>3</sub>OH:NH<sub>3</sub> interstellar ice analogs) in a data-driven and unsupervised manner, without any prior knowledge about chemical reactions. The molecular network is clustered according to the initial NH<sub>3</sub> content and unlocked HCN, NH<sub>3</sub>, and H<sub>2</sub>O as spatially resolved key transformations. In comparison with the PubChem database, four subsets were annotated: (i) saturated C-backbone molecules without N, (ii) saturated N-backbone molecules,

(iii) unsaturated C-backbone molecules without N, and (iv) unsaturated N-backbone molecules. These findings were validated with previous results (e.g., identifying the two major graph components as previously described N-poor and N-rich molecular groups) but with additional information about subclustering, key transformations, and molecular structures, and thus, the structural characterization of large complex organic molecules in interstellar ice analogs has been significantly refined.

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## The transition from soluble to insoluble organic matter in interstellar ice analogs and meteorites

Grégoire Danger , Alexander Ruf , Thomas Javelle , Julien Maillard , Vassilissa Vinogradoff , Carlos Afonso , Isabelle Schmitz-Afonso , Laurent Remusat , Zelimir Gabelica , and Philippe Schmitt-Kopplin

Context. Carbonaceous chondrites are sources of information on the origin of the Solar System. Their organic content is conventionally classified as soluble (SOM) and insoluble organic matter (IOM), where the latter represents the majority. Aims. In this work, our objectives are to identify possible relations between soluble and insoluble organic matter generated in laboratory experiments and to extrapolate the laboratory analog findings to soluble and insoluble organic matter of meteorites to test their connection. Methods. Using laboratory experiments, processes possibly linking IOM analog (IOMA) to SOM analog (SOMA) precursors are investigated by assuming that dense molecular ices are one of the sources of organic matter in the Solar System. Each organic fraction is analyzed by laser desorption coupled to a Fourier transform ion cyclotron resonance mass spectrometer on a comprehensive basis. Results. SOMA and IOMA significantly differ in their chemical fingerprints, and particularly in their aromaticity, O/C, and N/C elemental ratios. Using an innovative molecular network, the SOMA-IOMA transition was tested, revealing connection between both classes. This new network suggests that IOMA is formed in two steps: a first generation IOMA based on precursors from SOMA, while a second IOMA generation is formed by altering the first IOMA generation. Finally, using the same analytical technique, the molecular content of IOMA and that of the Paris IOM are compared, showing their molecular similarities for the first time. The molecular network application to the Paris SOM and IOM demonstrates that a possible connection related to photochemical ice processing is present, but that the overall history of IOM formation in meteorites is much more complex and might have been affected by additional factors (e.g., aqueous alteration). Conclusions. Our approach provides a new way to analyze the organic fraction of extraterrestrial material, giving new insights into the evolution of organic matter in the Solar System

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## Cluster Beam Study of (MgSiO<sub>3</sub>)<sup>+</sup>-Based Monomeric Silicate Species and Their Interaction with Oxygen: Implications for Interstellar Astrochemistry

Joan Mariñoso Guiu, Bianca-Andreea Ghejan, Thorsten M. Bernhardt, Joost M. Bakker, Sandra M. Lang, and Stefan T. Bromley

Silicates are ubiquitously found as small dust grains throughout the universe. These particles are frequently subject to high-energy processes and subsequent condensation in the interstellar medium (ISM), where they are broken up into many ultrasmall silicate fragments. These abundant molecular-sized silicates likely play an important role in astrochemistry. By approximately mimicking silicate dust grain processing occurring in the diffuse ISM by ablation/cooling of a Mg/Si source material in the presence of O<sub>2</sub>, we observed the creation of stable clusters based on discrete pyroxene monomers (MgSiO<sub>3</sub><sup>+</sup>), which traditionally have only been considered possible as constituents of bulk silicate materials. Our study suggests that such pyroxene monomer-based clusters could be highly abundant in the ISM from the processing of larger silicate dust grains. A detailed analysis, by infrared multiple-photon dissociation (IR-MPD) spectroscopy and density functional theory (DFT) calculations, reveals the structures and properties of these monomeric silicate species. We find that the clusters interact strongly with oxygen, with some stable cluster isomers having a silicate monomeric core bound to an ozone-like moiety. The general high tendency of these monomeric silicate species to strongly adsorb O<sub>2</sub> molecules also suggests that they could be relevant to the observed and unexplained depletion of oxygen in the ISM. We further find clusters where a Mg atom is bound to the MgSiO<sub>3</sub> monomer core. These species can be considered as the simplest initial step in monomer-initiated nucleation, indicating that small ionized pyroxenic clusters could also assist in the reformation of larger silicate dust grains in the ISM.

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## Phosphine in the Venusian Atmosphere: A Strict Upper Limit from SOFIA GREAT Observations

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The presence of phosphine (PH<sub>3</sub>) in the atmosphere of Venus was reported by Greaves et al. (2021a), based on observations of the J=1-0 transition at 267 GHz using ground-based, millimeter-wave spectroscopy. This unexpected discovery presents a challenge for our understanding of Venus's atmosphere, and has led to a reappraisal of the possible

sources and sinks of atmospheric phosphorous-bearing gases. Here we present results from a search for PH<sub>3</sub> on Venus using the GREAT instrument aboard the SOFIA aircraft, over three flights conducted in November 2021. Multiple PH<sub>3</sub> transitions were targeted at frequencies centered on 533 GHz and 1067 GHz, but no evidence for atmospheric PH<sub>3</sub> was detected. Through radiative transfer modeling, we derived a disk-averaged upper limit on the PH<sub>3</sub> abundance of 0.8 ppb in the altitude range 75-110 km, which is more stringent than previous ground-based studies.

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## OH mid-infrared emission as a diagnostic of H<sub>2</sub>O UV photodissociation. II. Application to interstellar PDRs.

M. Zannese, B. Tabone, E. Habart, F. Le Petit, E. van Dishoeck, E. Bron

Water photodissociation in the 114 - 144 nm UV range forms excited OH which emits at mid-infrared wavelengths via highly excited rotational lines. These lines have only been detected with Spitzer in several proto-planetary disks and shocks. Previous studies have shown they are a unique diagnostic for water photodissociation. Thanks to its high sensitivity and angular resolution, the James Webb Space Telescope (JWST) could be able to detect them in other environments such as interstellar Photo-Dissociation Regions (PDRs). In order to predict the emerging spectrum of OH, we use the Meudon PDR Code which compute the thermal and chemical structure of PDRs. The influence of thermal pressure and UV field strength on the integrated intensities, as well as their detectability with the JWST are studied in details. OH mid-IR emission is predicted to originate very close to the H<sub>0</sub>/H<sub>2</sub> transition and is directly proportional to the column density of water photodissociated in that layer. Because neutral gas-phase formation of water requires relatively high temperatures ( $T_k \geq 300$  K), the resulting OH mid-IR lines are primarily correlated with the temperature at this position, and are therefore brighter in regions with high pressure. This implies that these lines are predicted to be only detectable in strongly irradiated PDRs with high thermal pressure. In the latter case, OH mid-IR lines are less dependent on the strength of the incident UV field. The detection in PDRs like the Orion bar, which should be possible, is also investigated. To conclude, OH mid-IR lines observable by JWST are a promising diagnostics for dense and strongly irradiated PDRs.

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## Announcements

### The Olympian Symposium 2023: Star formation in the era of JWST

29 May - 2 June, 2023

Paralia Katerini, Mt. Olympus, Greece

More details at [olympiansymposium.org](https://olympiansymposium.org)