The Role of Solid Phase in the Radiation Astrochemistry of Ices

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Introduction

- We have performed comparative 2 keV electron irradiations of the crystalline and amorphous phases of a number of pure astrophysical ice analogues.
- The radiation-induced molecular decay rates of crystalline ices were found to be slower than those of the amorphous ones.
- The discrepancy in the decay rates between the amorphous and crystalline phases of an ice was found to be related to the nature, strength, and extent of the intermolecular bonding of the structure.
- Such results have implications for the chemical productivities of astrophysical ices undergoing cycles of radiation-induced amorphization and thermally-induced crystallization.

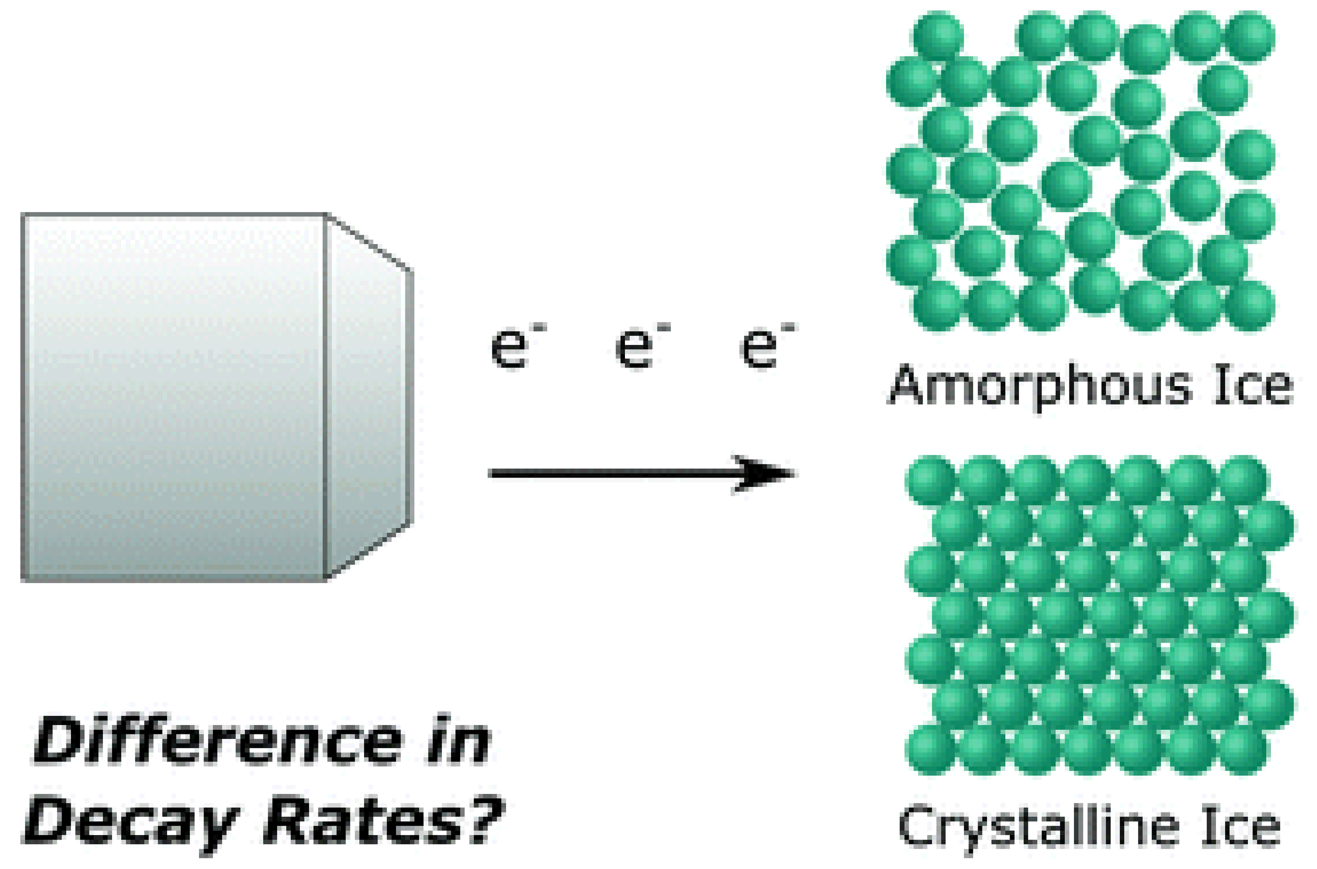


Fig. 1: Cartoon illustration of the conceptual basis of this study.

Experimental Methodology

- Experiments were performed using the Ice Chamber for Astrophysics-Astrochemistry at the Institute for Nuclear Research (Atomki) in Debrecen, Hungary.[1,2]
- The base pressure of the chamber is ~10^{-9} mbar.
- The amorphous and various crystalline phases of CH_3OH, N_2O, and H_2O were deposited *via* background deposition at different temperatures.[3-5]
- Deposited ices were irradiated using 2 keV electrons at 20 K.
- Phyisco-chemical changes accompanying irradiation could be monitored *in situ* using FT-IR transmission absorption spectroscopy.

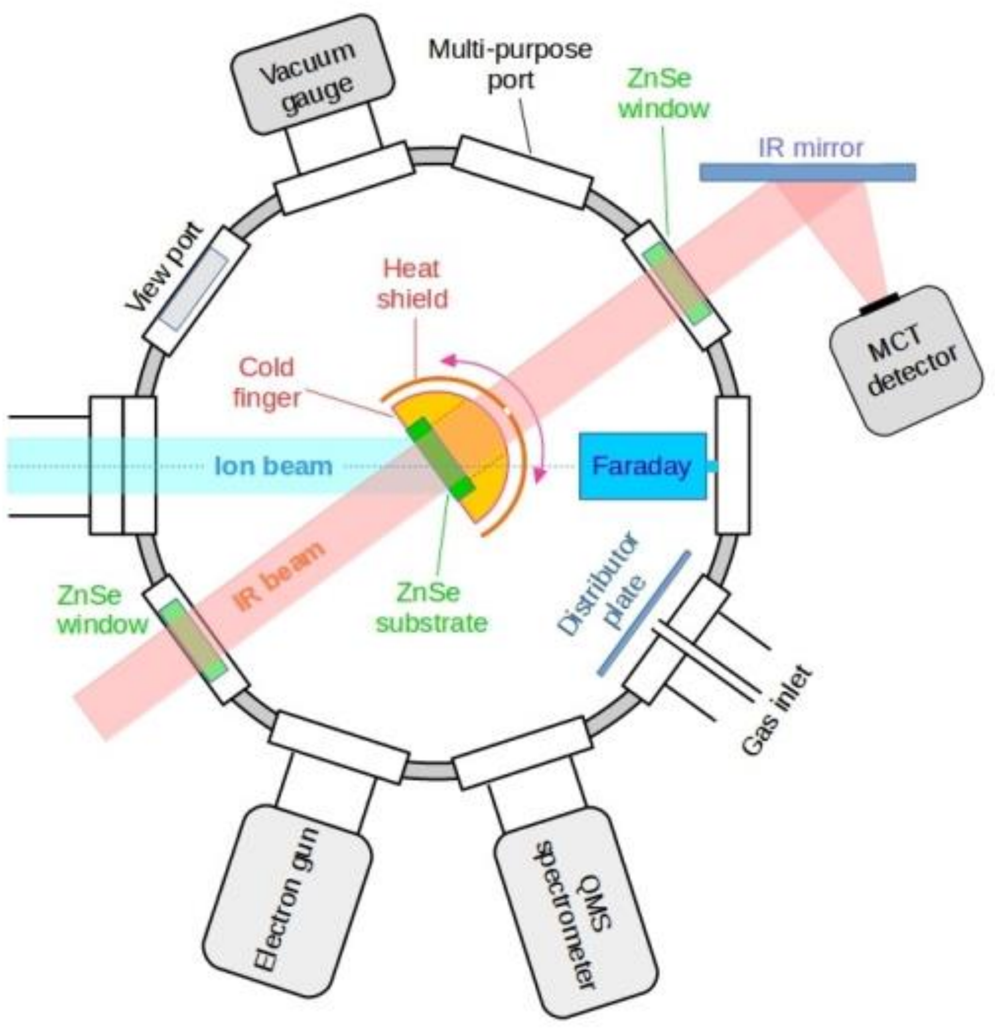


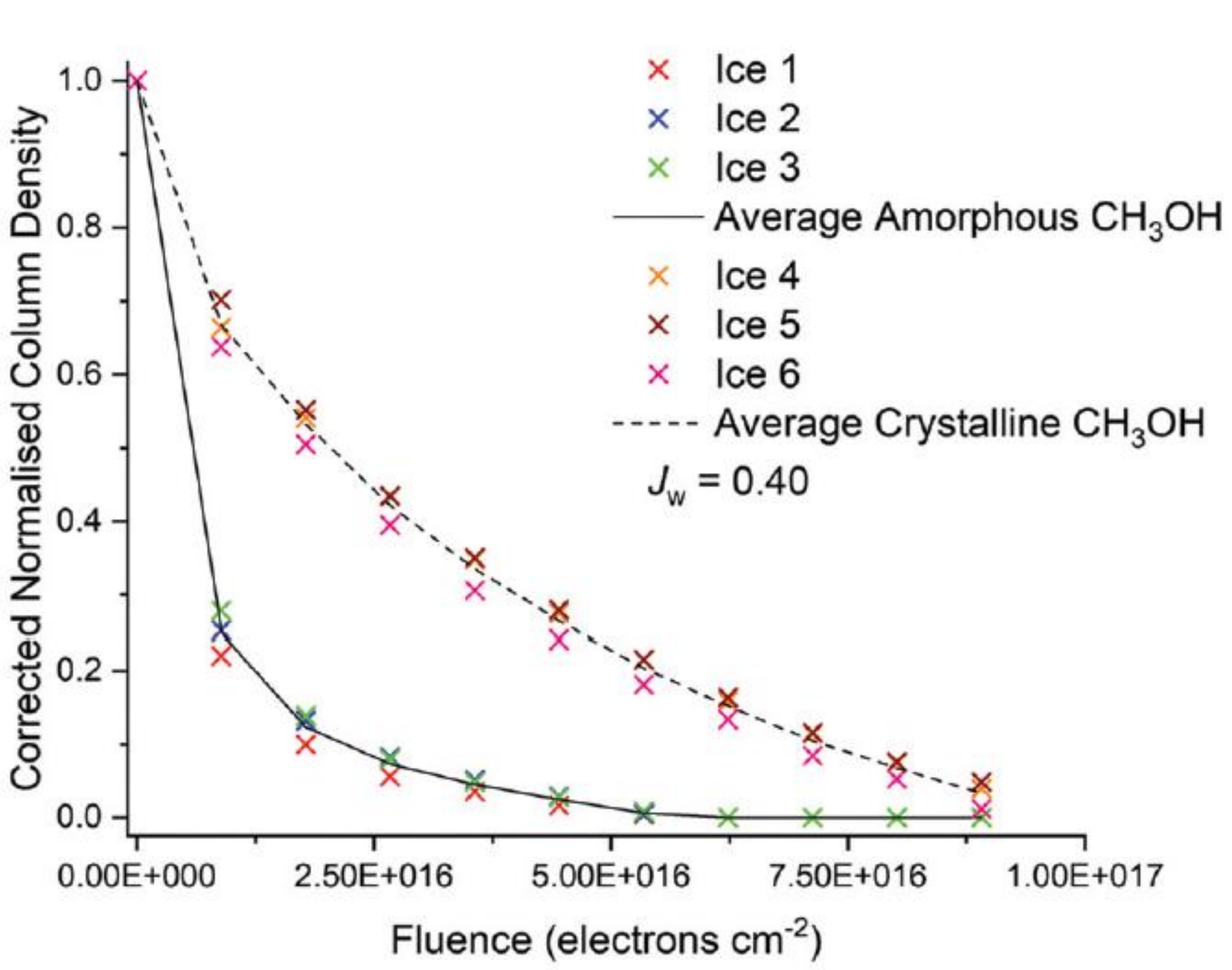
Fig. 2: *Left*: Top-view schematic diagram of the ICA set-up. Note that electron irradiations are carried out at 36° to the target ice sample normal. *Above:* The substrate holder and ZnSe deposition substrates.

Results and Interpretation

- The radiation-induced decay of the crystalline phase of an ice was always found to be slower than that of its amorphous phase.
- The decay of amorphous CH_3OH was significantly more rapid than that of the crystalline phase.
- Similar results were observed for the studied H_2O ice phases.
- The decay of amorphous N_2O was only moderately more rapid than that of the crystalline phase.

- The H-bonding network is crystalline CH_3OH is significantly more extensive than that in amorphous CH_3OH, and so adds a considerable degree of radio-resistance.
- Dipole interactions in the crystalline N_2O ice stabilize it against radiation-induced decay compared to the amorphous phase, but since these interactions are weak this stabilization is not considerable.

- Astrophysical ices may therefore be more chemically productive in the amorphous phase; where radiation processes dominate over thermal ones, or where the latter are absent (e.g., in pre-stellar cores).



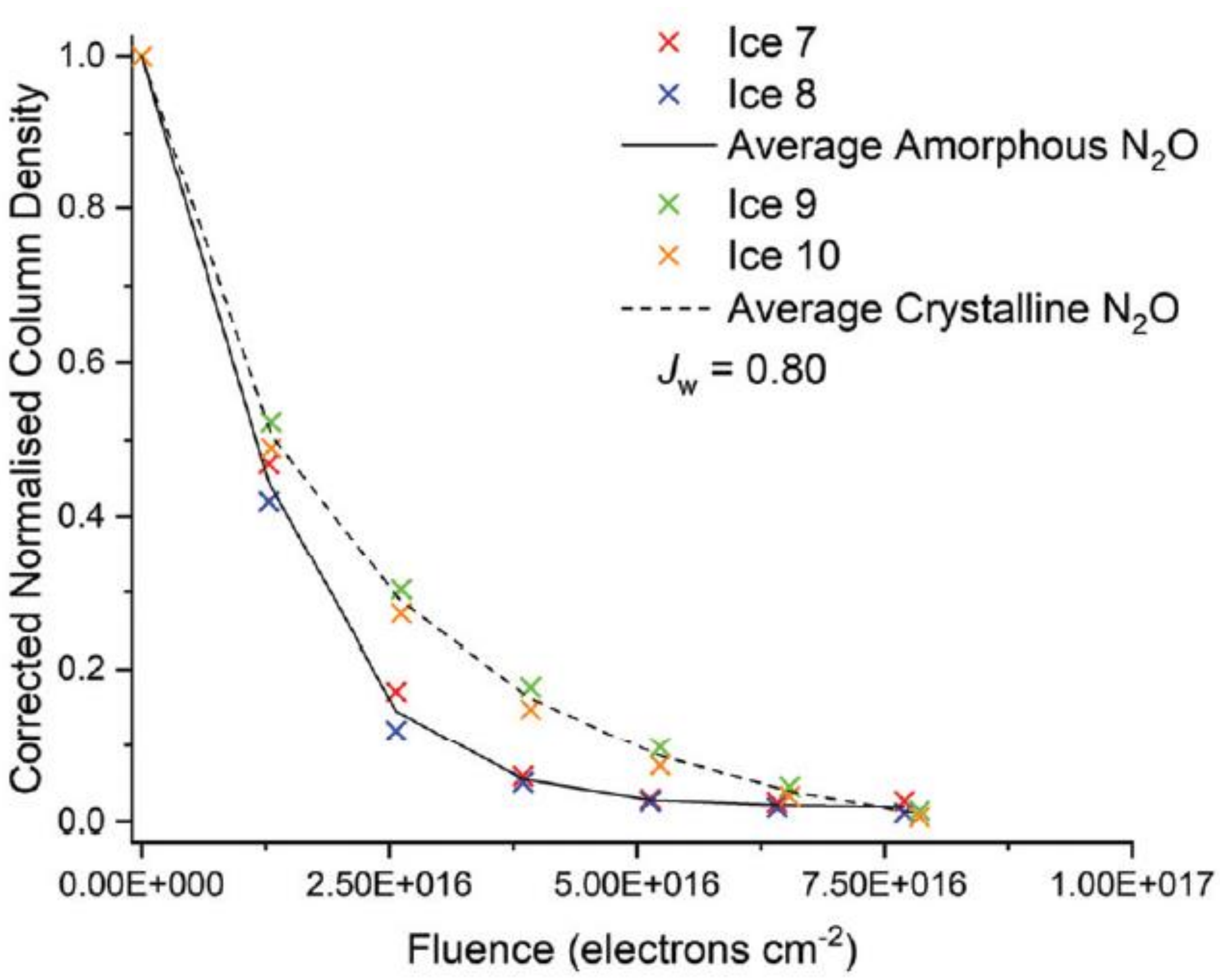


Fig. 3: Differences in electron-induced radiation decay rates between the amorphous and crystalline phases of CH_3OH (*left*) and N_2O (*right*).

References and Acknowledgements

1. P. Herczku *et al.* (2021). *Rev. Sci. Instrum.* **92**, 084501.
2. D.V. Mifsud *et al.* (2021). *Eur. Phys. J. D: Atom. Mol. Opt. Plasma Phys.* **75**, 182.
3. D.V. Mifsud *et al.* (2022). *Phys. Chem. Chem. Phys.* **24**, 10974.
4. D.V. Mifsud *et al.* (2022). *Eur. Phys. J. D: Atom. Mol. Opt. Plasma Phys.* **76**, 87.
5. D.V. Mifsud *et al.* (2022). *Front. Chem.* submitted.

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