Preferential adsorption of para and ortho water molecules on charged nanoparticles in planetary ice clouds

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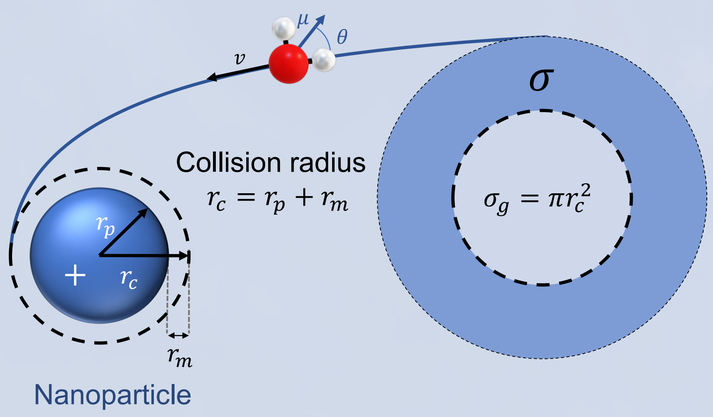
Motivation

In the Earth mesopause and other extraterrestrial settings, **nanometer-sized singly charged particles** form by condensation of evaporated meteorite material. They exhibit an **enhanced water adsorption cross section** due to the strong **charge-dipole-interaction** [1]. The enhancement factor depends on the average dipole orientation of the water molecules in their Stark-shifted rotational states.

$$\rightarrow \quad \sigma = \Gamma \cdot \sigma_g$$

Γ – enhancement factor
σ_g – geometrical cross section

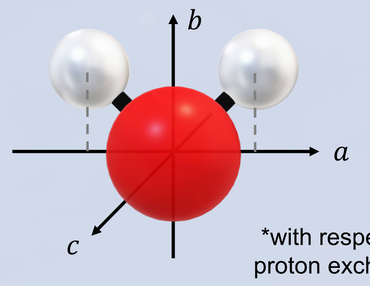


In water, rotational states are strongly linked to the nuclear spin. The spins of the two indistinguishable protons in water can be either parallel (ortho) or antiparallel (para) [2]:

Para, $I = 0$

1 antisymmetric* nuclear wave function

$(\uparrow\downarrow - \uparrow\downarrow)$



*with respect to proton exchange

Ortho, $I = 1$

3 symmetric* nuclear wave functions

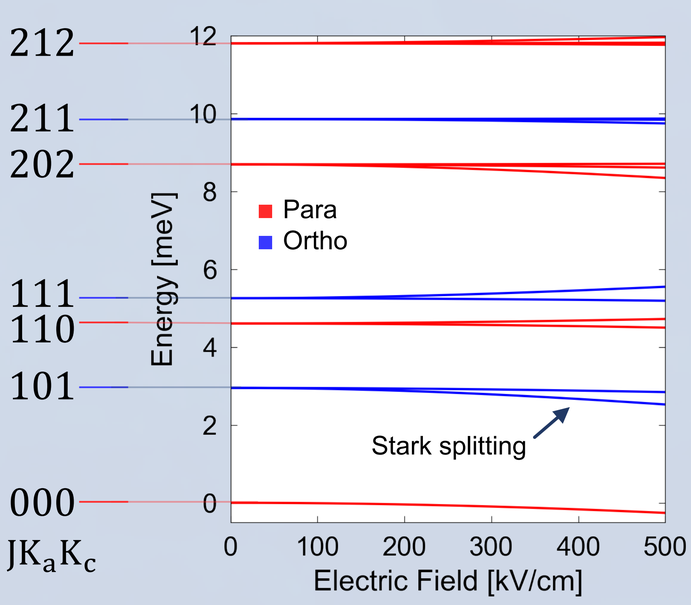
$\downarrow\downarrow$, $\uparrow\uparrow$, $(\uparrow\downarrow + \uparrow\downarrow)$

Symmetry constraints (Pauli principle) → Ortho and para water occupy different rotational states → Different average dipole orientation of molecular ensembles

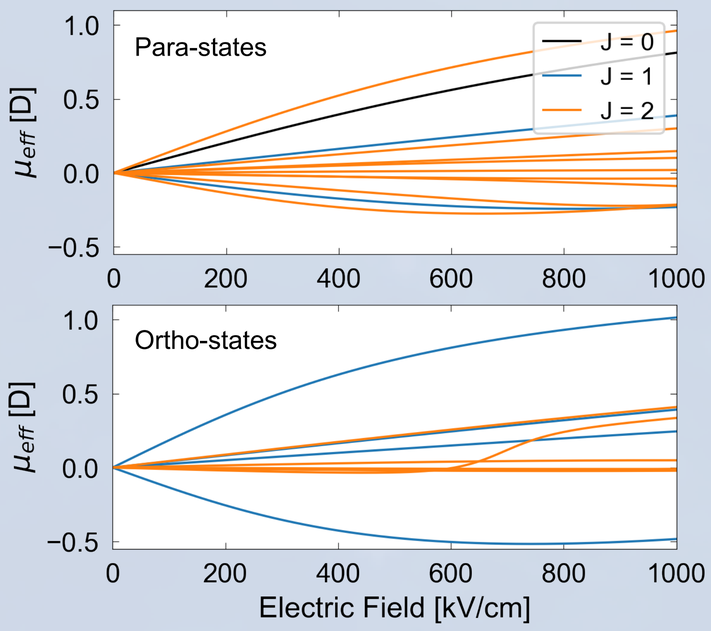
How does the adsorption enhancement factor of the water molecule depend on its nuclear spin state?

Method

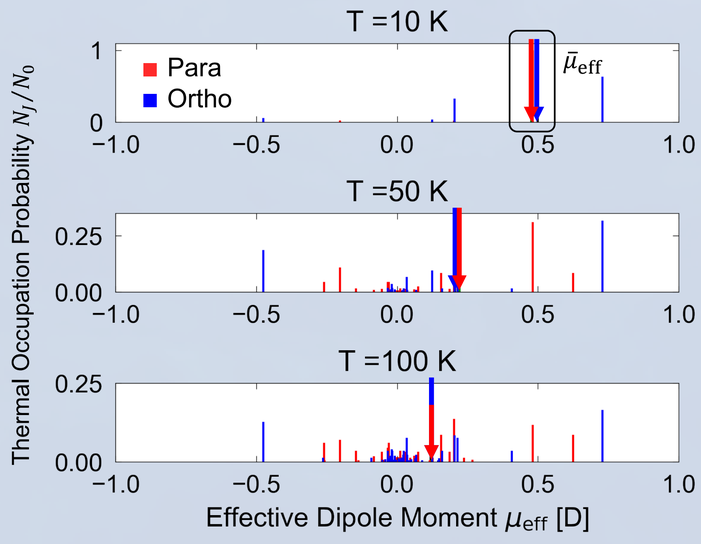
Calculation of the **rotational states** of water and division into **ortho- and para** states using the program CMIStark [4]



Calculation of the **effective dipole moment** of each energy state [4]



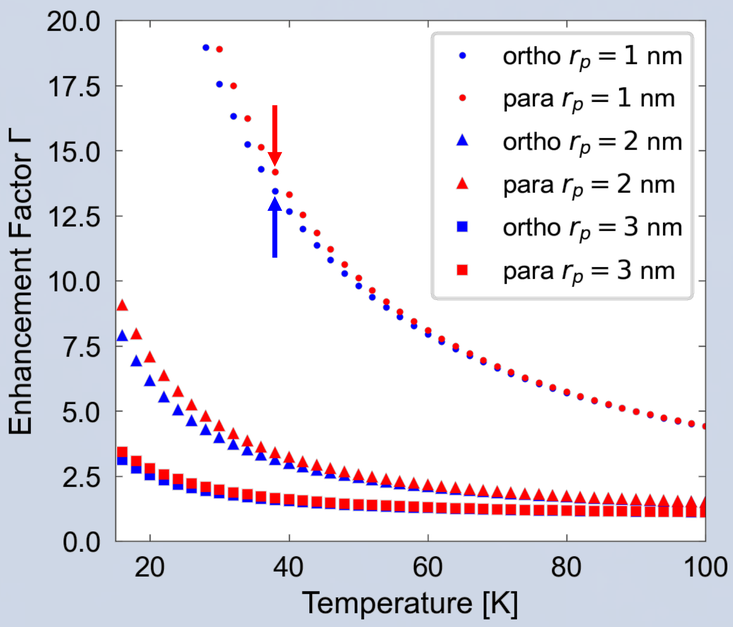
Calculation of the **average effective dipole moment** of a molecular ensemble (weighted by thermal occupation) consisting of only para- or only ortho-water, depending on temperature and field strength [3]

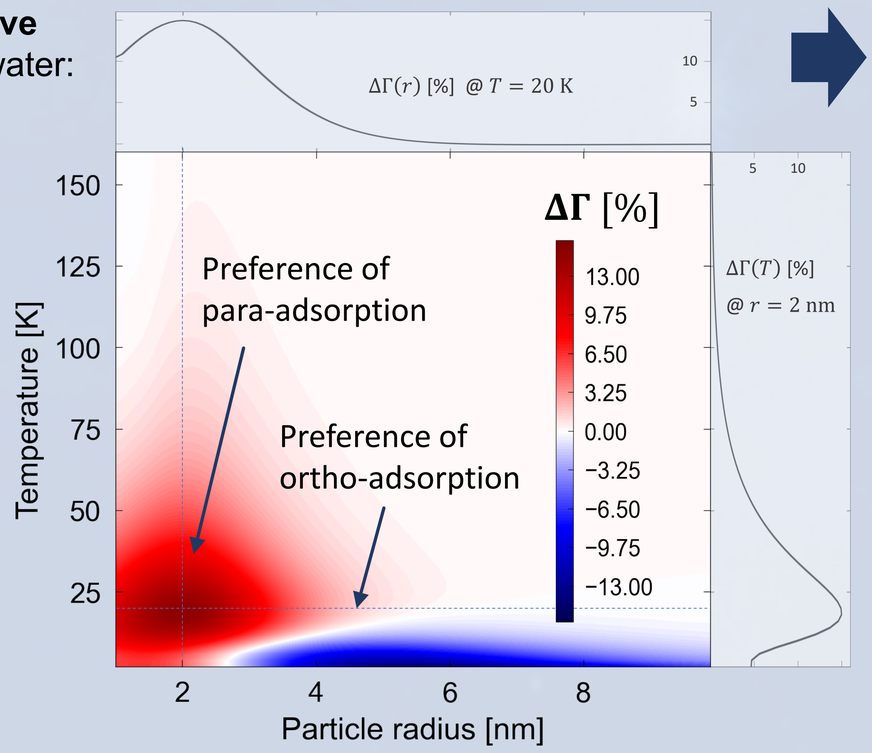


Having calculated the average effective dipole moment of a molecular ensemble of ortho- and para molecules, each, we can obtain their respective enhancement factor [1]:

Results: Collision Cross Section Enhancement

Resulting **enhancement factor Γ** (left) and its **relative deviation $\Delta\Gamma$** (right) of para water compared ortho water:





Under certain conditions, there are significant deviations in the adsorption behavior of water molecules with different total nuclear spin.

Whether ortho or para water is adsorbed preferably depends on the temperature and the radius of the involved nanoparticle.

Outlook:
What happens to the nuclear spin state of the water molecule during and after the adsorption?

References:

1) M. Nachbar et al., DPG Annual Conference UP6.1 (2021)
2) D. Horke et al., Ang. Chem. **126**, 12159 (2014)
3) R. Moro et al., Phys. Rev. A **75**, 013415 (2007)
4) Y. P. Chang et al., Comput Phys Commun **185**, 339–349 (2014)

Background image: Orion Nebula James Webb Telescope NASA