# Is the HDL-LDL equilibrium a thermally distorted version of HDA-LDA?

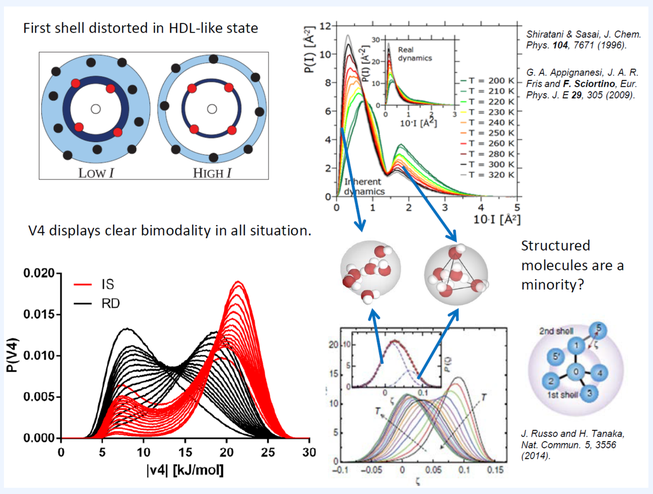

## Montes de Oca Joan[1], Verde Alejandro, Accordino Sebastian , Alarcón Laureano, Appignanesi Gustavo

[1]INQUISUR, Univerisdad Nacional del Sur, Bahía Blanca, ARG.,



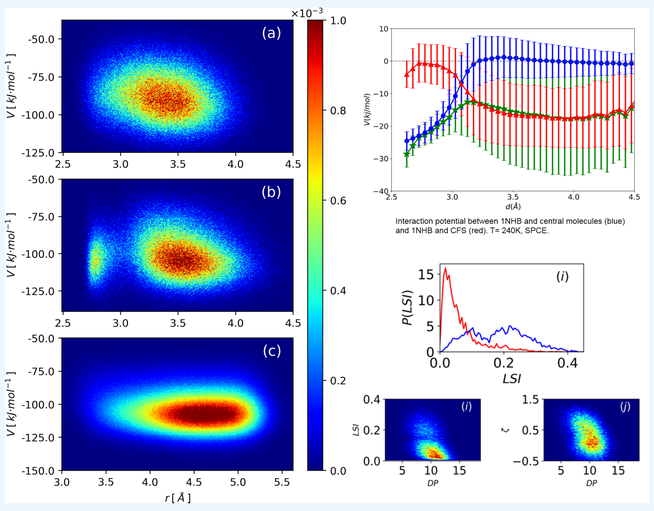
## abstract

Several indicators of local structure have been devised to evidence the two hypothetical types of coexisting liquid water molecules: high and low local density ones (HDL and LDL, respectively). In general, all of them have focused on detecting the more-ordered lower density species assuming its structural similarity with ice. However, we will show that the HDL-like molecules are not as unstructured as it was originally supposed. We will also evidence that locally denser molecules are extremely subject to high dynamic propensity. Moreover, we shall develop a new structure indicator (producing neat bimodal behavior) that will re-emphasize the relevance of the unstructured fraction of liquid water. We will show that truly HDL-like molecules are much scarcer than formerly supposed and easily confused with inherently structured but thermally distorted molecules. Finally, we show new support for the theory of HDL-LDL coexistence.


## HDL-like molecules

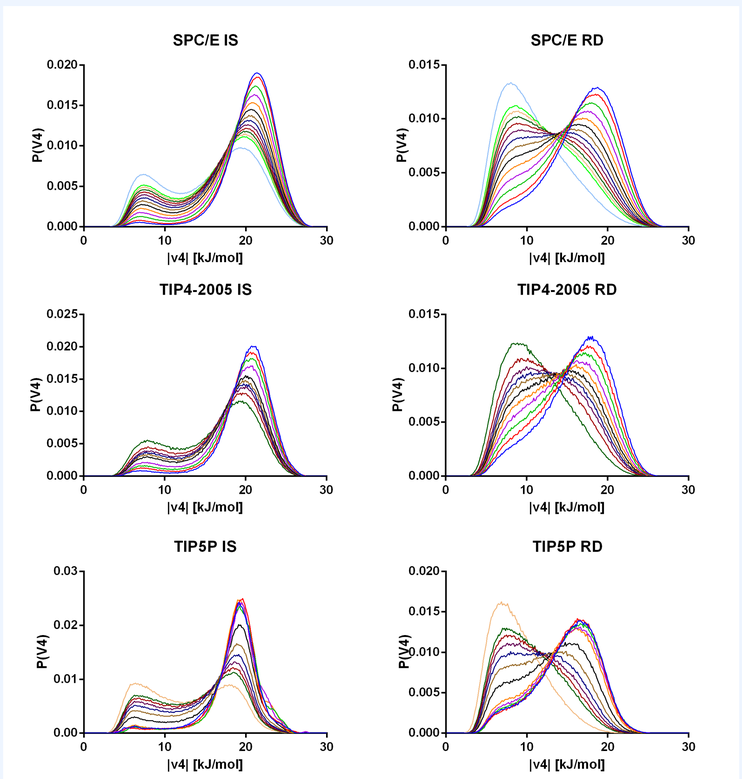
We have shown that HDL-like molecules, independently on the index employed to classify them, not only display clear structural preferences[1] but also play an important role in the dynamic of the system[2].




## V4 index

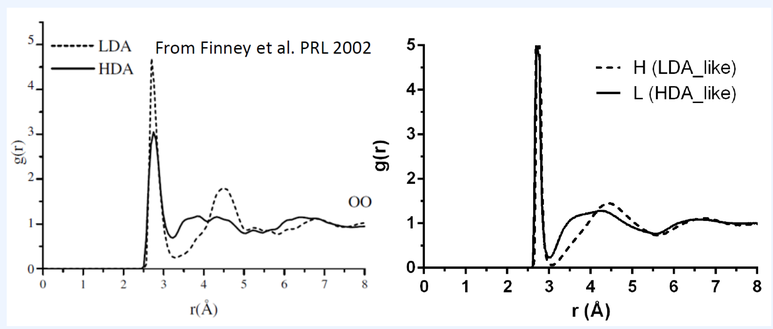
V4 measures the interaction of a central molecule with its fourth neighbor (ordered by potential energy with a central molecule).

Senses disorder on the first shell (from well HB to non-HB attraction).

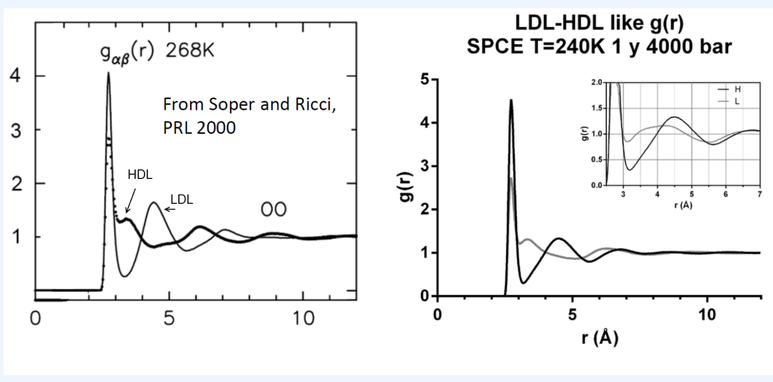



## O-O RDFs

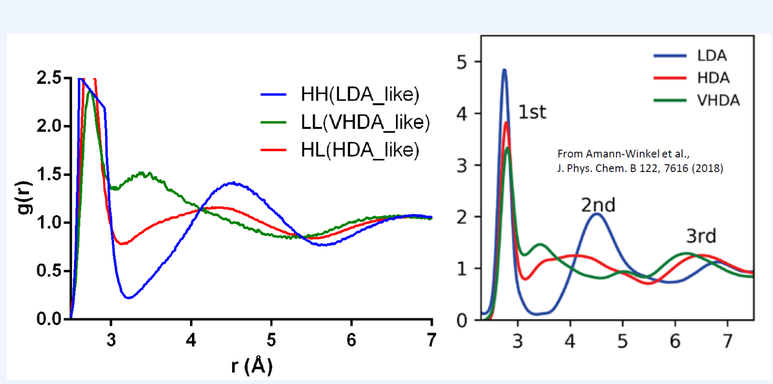
Inheren structutres:



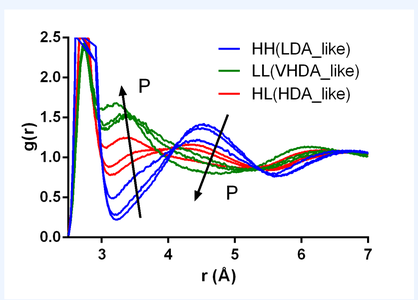

Real MD



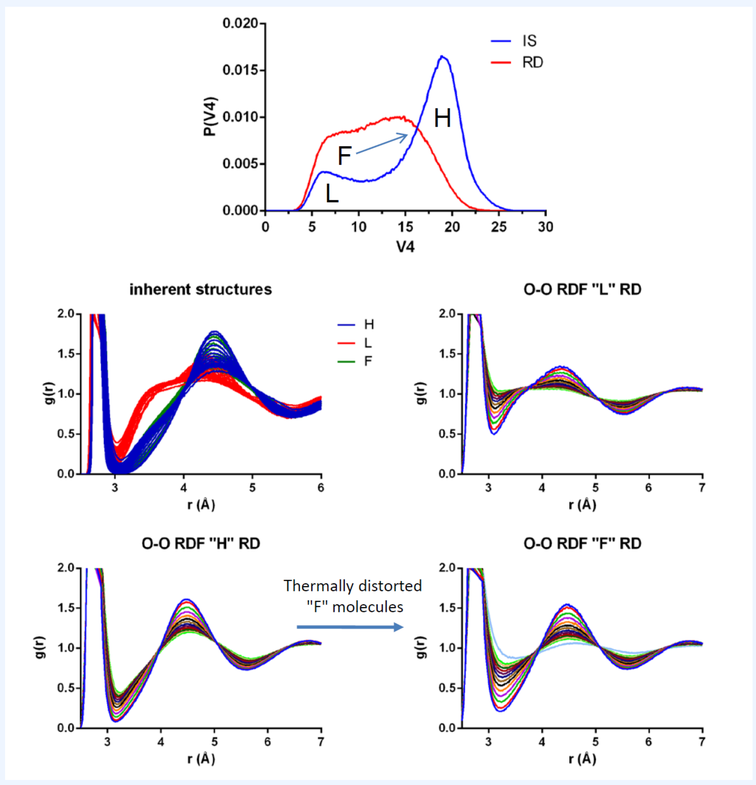

Partial O-O RDFs:



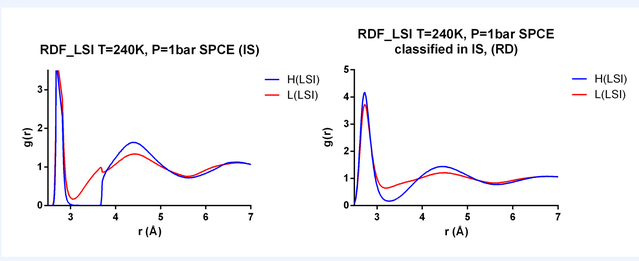

Effect of pressure:




Effect of temperature:




classified by LSI:




## Conclusions

- New index, V4: neat bimodality, generalizable to non-bulk contexts.
- We discriminate between “inherently unstructured”, “L” molecules, and “thermally distorted configurations” but still belonging to a structured IS.
- L molecules are found to be central:

1. To rationalize structuring: Essential for the emergence of the salient features of the radial distribution functions. HDL/LDL RDFs are correctly reproduced by L/H molecules, while the minimization makes them resemble the HDA/LDA RDFs. HDL/LDL equilibrium as a thermally distorted version of HDA/LDA.
2. For dynamics: might be essential to HB dynamics, and have been proven to show enhanced dynamic propensity.

## References


1. Montes de Oca, J. M., Accordino, S. R., Verde, A. R., Alarcón, L. M. & Appignanesi, G. A. Phys. Rev. E, 99 (2019) 062601.

2. Verde, A. R., Montes de Oca, J. M., Accordino, S. R., Alarcón, L. M. & Appignanesi, G. A. J. Chem. Phys., 150 (24):244504 (2019)