

FINAL REPORT

1. DOE AWARD DE-SC0005180, Princeton University

2. CMCSN: Structure and dynamics of water and aqueous solutions in materials science, PI: Roberto Car, Princeton University, CO-PIs: Giulia Galli, University of California Davis, John J. Rehr, University of Washington

3. Award period: 09/01/2010 - 08/31/2013 extended at no cost for two additional years up to 08/31/2015

4. Accomplishments

This award has contributed to build a network of scientists interested in the structure and dynamics of water. Such network extends well beyond the PI and the co-PIs and includes both theoreticians and experimentalists. Scientific interactions within this community have been fostered by three workshops supported by the grant. The first workshop was held at Princeton University on December 6-8, 2010. The second workshop was held at the Talaris Conference Center in Seattle on February 10-12, 2012. The third workshop was held at UC Davis on June 19-22, 2013. Each workshop had 40-50 participants and about 20 speakers. The workshops have been very successful and stimulated ongoing discussions within the water community. This debate is lasting beyond the time frame set by the grant. The following events are just a few examples: (i) the month long activity on "Water: the most anomalous liquid" organized at NORDITA (Stockholm) in October-November 2014 by A. Nilsson and L. Petterson who participated in all the three CMCSN sponsored workshops; (ii) the workshop on "ice nucleation" organized by R. Car, P. Debenedetti and F. Stillinger at the Princeton Center for Theoretical Science in April 23-24 2015; (iii) the 10 days workshop on water organized by R. Car and F. Mallamace at the E. Majorana Centre in Erice (Sicily) in July 2016, an activity that will morph into a regular summer school of the E. Majorana Centre in the years to come under the directorship of R. Car, F. Mallamace (U. Messina), A. Nilsson (U. Stockholm) and L. Xu (Beijing U.). All these activities were stimulated by the scientific discussions within the network initiated by this CMCSN grant.

In addition to building a collaborative network, the CMCSN grant partially supported graduate students and postdoctoral associates in the groups of the PI and co-PIs at Princeton, UC Davis and U of Washington. Their research activity led to important progress in the simulation of the structure and dynamics of water at different thermodynamic conditions. Important phenomena such as ice nucleation, pressure induced phase transitions and ionic solvation were studied. Several investigations impacted experimental spectroscopic studies, including x-ray diffraction, neutron scattering, x-ray absorption, optical spectra, infrared spectra, and dielectric relaxation spectroscopy. New interpretations of experimental data were made possible by the theoretical developments originating from the research supported by this grant.

The scientific results led to several publications on scientific journals, the full list of which is given below.

Publications supported by the grant

2011

1. L. Lin, J.A. Morrone, R. Car, M. Parrinello, Momentum distribution, vibrational dynamics, and potential of mean force in ice, *Phys. Rev. B* **83**, 220302(R) (2011)
2. L. Lin, J.A. Morrone, R. Car, Correlated Tunneling in Hydrogen Bonds, *J. Stat. Phys.* **145**, 365 (2011)
3. Cui Zhang, Leonardo Spanu and Giulia Galli, Entropy of Liquid Water from Ab-initio Molecular Dynamics, *J. Phys. Chem. B* **115**, 14190–14195 (2011)
4. Tianshu Li, Davide Donadio, Giovanna Russo and Giulia Galli, Homogeneous ice nucleation from supercooled water, *Phys. Chem. Chem. Phys.* **13**, 19807-19813 (2011)

2012

5. D. Flammini, A. Pietropaolo, R. Senesi, C. Andreani, F. McBride, A. Hodgson, M.A. Adams, L. Lin, R. Car, Spherical momentum distribution of the protons in hexagonal ice from modeling of inelastic neutron scattering data, *J. Chem. Phys.* **136**, 024504 (2012)
6. L. Kong, X. Wu, and R. Car, Roles of quantum nuclei and inhomogeneous screening in the x-ray absorption spectra of water and ice, *Phys. Rev. B* **86**, 134203 (2012)
7. Tuan Anh Pham, Patrick Huang, Eric Schwegler and Giulia Galli, First Principles Study of the Infrared Spectra of the Ice Ih (0001) Surface, *J. Phys. Chem. A* **116**, 9255 (2012)
8. J. Vinson, J. Kas, F. Vila, J. J. Rehr and E. L. Shirley, Theoretical optical and x-ray spectra of liquid and solid H₂O, *Phys. Rev. B* **85**, 045101 (2012)

2013

9. Roberto Senesi, Davide Flammini, Alexander Kolesnikov, Eamonn Murray, Giulia Galli, and Carla Andreani, The quantum nature of the OH stretching mode in ice and water probed by neutron scattering experiments, *J. Chem. Phys.* **139**, 074504 (2013)
10. Cui Zhang, François Gygi and Giulia Galli, Strongly Anisotropic Dielectric Relaxation of Water at the Nanoscale, *J. Phys. Chem. Lett.* **4**, 2477 (2013)

11. Cui Zhang, Tuan Anh Pham, François Gygi and Giulia Galli, Electronic structure of the solvated chloride anion from first principles molecular dynamics, *J. Chem. Phys-Comm.* **138**, 181102 (2013)
12. Tianshu Li, Davide Donadio and Giulia Galli, , Ice nucleation at the nanoscale probes no man's land, *Nature Commun.* **4** 1887 (2013)
13. Ding Pan, Leonardo Spanu, Brandon Harrison, Dimitri A. Sverjensky and Giulia Galli, Dielectric properties of water under extreme conditions and transport of carbonates in the deep Earth, *Proc. Natl. Acad. Sci. USA* **110**, 6646 (2013)

2014

14. R. A. DiStasio, Jr., B. Santra, Z. Li, X. Wu, and R. Car, The individual and collective effects of exact exchange and dispersion interactions on the ab-initio structure of liquid water, *J. Chem. Phys.* **141**, 084502 (2014)
15. Ding Pan, Quan Wan, and Giulia Galli, The refractive index and electronic gap of water and ice increase with increasing pressure, *Nature Commun.* **5**, 3919 (2014)

2015

16. B. Santra, R. A. DiStasio Jr., F. Martelli, R. Car, Local structure analysis in ab-initio liquid water, *Mol. Phys.* **113**, 2829 (2015)
17. J. Sun, B.K. Clark, S. Torquato, R. Car, The phase diagram of high-pressure superionic ice, *Nature Comm.* **6**, 8156 (2015)
18. K. Gilmore, John Vinson, E.L. Shirley, D. Prendergast, C.D. Pemmaraju, J.J. Kas, F.D. Vila, and J.J. Rehr, Efficient implementation of core-excitation Bethe-Salpeter equation calculations, *Comput. Phys. Comm.* **197**, 109 (2015)