Fast and Accurate Quantum Monte Carlo for Molecular Crystals

Andrea Zen * (1,2), Jan Gerit Brandenburg (1,2), Jiri Klimes (3), Alexandre Tkatchenko(4), Dario Alfe (1,5), Angelos Michaelides (1,2)

(1) Thomas Young Centre and London Centre for Nanotechnology, 17–19 Gordon Street, London, WC1H 0AH, U.K., (2) Department of Physics and Astronomy, University College London, 20 Gordon Street, London, WC1H 0AJ, U.K., (3) J. Heyrovsky Institute of Physical Chemistry, Academy of Sciences of the Czech Republic, Dolejskova 3, CZ-18223 Prague 8, Czech Republic and Department of Chemical Physics and Optics, Faculty of Mathematics and Physics, Charles University, Ke Karlovu 3, CZ-12116 Prague 2, Czech Republic, (4) Physics and Materials Science Research Unit, University of Luxembourg, L-1511 Luxembourg, (5) Department of Earth Sciences, University College London, Gower Street, London WC1E 6BT, U.K.

*** presenting author Email: a.zen@ucl.ac.uk**

Abstract:
Computer simulation plays a central role in modern day materials science. The utility of a given computational approach depends largely on the balance it provides between accuracy and computational cost. Molecular crystals are a class of materials of great technological importance which are challenging for even the most sophisticated ab initio electronic structure theories to accurately describe. This is partly because they are held together by a balance of weak intermolecular forces but also because the primitive cells of molecular crystals are often substantially larger than those of atomic solids. Here, we demonstrate that diffusion quantum Monte Carlo (DMC) delivers sub-chemical accuracy for a diverse set of molecular crystals at a surprisingly moderate computational cost. As such, we anticipate that DMC can play an important role in understanding and predicting the properties of a large number of molecular crystals, including those built from relatively large molecules which are far beyond reach of other high accuracy methods.