Abstract Submitted
for the MAR17 Meeting of
The American Physical Society

Anharmonic and Quantum Fluctuations in Molecular Crystals from Ab Initio Simulations MARIANA ROSSI, Fritz Haber Institut der Max Planck Gesellschaft, PIERO GASPAROTTO, MICHELE CERIOTTI, École Polytechnique Fédérale de Lausanne — Molecular crystals often exist in multiple competing polymorphs which are challenging to be predicted computationally, but show significantly different physicochemical properties. This challenge is not due only to the combinatorial search space, but also to the complex interplay of subtle effects determine the relative stability of different structures. Here we estimate all contributions to the free energies of these systems with density-functional theory, including the oft-neglected anharmonic contributions and nuclear quantum effects, by using a series of different flavors of thermodynamic integration. As an example, for the two most stable forms of paracetamol we find that anharmonic contributions, different descriptions of van der Waals interactions, and nuclear quantum effects **all** matter to quantitatively determine the stability of different phases [1]. Our studies indicate that anharmonic free energies could play an important role for molecular crystals composed by large molecules and opens the way for a systematic inclusion of these effects in order to obtain a predictive screening of structures. [1] Rossi, Gasparotto, Ceriotti, *Phys. Rev. Lett.* **117**, 115702 (2016).

Mariana Rossi
Fritz Haber Institut der Max Planck Gesellschaft

Date submitted: 19 Oct 2016 Electronic form version 1.4