In this contribution it is shown that modest calculations combining first principles evaluations of the molecular properties with electrostatic interaction schemes to account for crystal environment are reliable for predicting and interpreting the experimentally-measured electric linear and second-order nonlinear optical susceptibilities within the experimental error bars. This is illustrated by considering two molecular crystals, namely: 2-methyl-4-nitroaniline (MNA) and 4-(N,N-dimethylamino)-3-acetamidonitrobenzene (DAN) [1]. A good agreement between theory and experiment (see figure below for DAN) is achieved providing the electric field effects originating from the electric dipoles of the surrounding molecules are accounted for. The presentation will also i) highlight the key role of the geometry on the $\chi(1)$ and $\chi(2)$ responses, ii) demonstrate the impact of electron correlation on the molecular and crystal properties, iii) assess the performance of exchange-correlation functionals, and iv) address the amplitude of the zero-point vibrational energy contributions [2]. A second illustration will deal with the $\chi(1)$ and $\chi(2)$ responses of two anil crystals, [N-(4-hydroxy)-salicylidene-amino-4-(methylbenzoate) and N-(3,5-di-tert-butylsalicylidene)-4-aminopyridine, which can switch between a enol (E) and a keto (K) form [3].