Molecular Quantum Optics: Tomographic state reconstruction in ensembles and single molecules
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Through electronically resonant four-wave mixing measurements, it is possible to reconstruct the complete quantum state of ensembles evolving in coherence, in the form of the Wigner Distribution Function (WDF). This, we illustrate in ordinary matter, for a diatomic molecule doped in ice. Although the system is coupled to its environment, the WDF shows a persistent negative hole, which quantifies its “cattiness”. The moving pictures of the WDF gives the most complete quantum description of a bond in motion, both system wavefunctions and system-bath entanglement in phase space, are observable.

Fig. 1: Measurements on bromine doped ice (a), allow the tomographic quantum state reconstruction [\(\left|0\right>+\left|1\right>\) state shown in (b)], the indirectly prepared states show a persistent a negative hole in their Wigner distribution function, characteristic of (microscopic) “cat”-states (c).

The manipulation of vibrational coherences on single molecules, through Coherent anti-Stokes Raman scattering (CARS), is a more direct implementation of quantum molecular optics. The first of such measurements, carried out on molecules equipped with plasmonic antennas, and under ambient conditions, illustrates the information content of such measurements. Single molecules do not diphase, however they acquire characteristic phase noise, which defines the state through measurement.