Simultaneous determination of structures, vibrations, and frontier orbital energies from a self-optimizing range-separated hybrid functional

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We present a new self-optimizing tuned range-separated hybrid functional approach within density functional theory (DFT), which leads to accurate predictions for both frontier orbital eigenvalues *and* geometries and vibrational frequencies. We demonstrate the efficacy of the approach by using it to compute the energetic and electronic properties for small organic molecules within the G2/97 test set, and comparing our results to gas-phase experimental data and calculations with a conventional hybrid functional. We find that our approach greatly improves the agreement between highest occupied molecular orbital energy and experimental ionization potentials. At the same time, it maintains a high level of accuracy in predicting experimental bond-lengths and angles, and is only slightly less accurate than a conventional hybrid functional for vibrational properties. We discuss prospects for this approach, as well as caveats and limitations.