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First-principles photoemission spectroscopy in DNA and RNA nucleobases from Koopmans-compliant functionals NGOC LINH NGUYEN, Theory and Simulations of Materials (THEOS), and National Centre for Computational Design and Discovery of Novel Materials (MARVEL), EPFL, GIOVANNI BORGHI, ANDREA FERRETTI, Centro S3, CNR-Istituto Nanoscienze, 41125 Modena, Italy, NICOLA MARZARI, Theory and Simulations of Materials (THEOS), and National Centre for Computational Design and Discovery of Novel Materials (MARVEL), EPFL — The determination of spectral properties of the DNA and RNA nucleobases from first principles can provide theoretical interpretation for experimental data, but requires complex electronic-structure formulations that fall outside the domain of applicability of common approaches such as density-functional theory. In this work, we show that Koopmans-compliant functionals [1], constructed to enforce piecewise linearity in energy functionals with respect to fractional occupation-i.e., with respect to charged excitations-can predict not only frontier ionization potentials and electron affinities of the nucleobases with accuracy comparable or superior with that of many-body perturbation theory and high-accuracy quantum chemistry methods, but also the molecular photoemission spectra are shown to be in excellent agreement with experimental ultraviolet photoemsision spectroscopy data. The results highlight the role of Koopmans-compliant functionals as accurate and inexpensive quasiparticle approximations to the spectral potential, which transform DFT into a novel dynamical formalism where electronic properties, and not only total energies, can be correctly accounted for. Reference [1] N.L. Nguyen et al., PRL (2015).

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