Autoionizing excited states of N_2 using complex-basis function spin-flip coupled cluster theory

Cite as: J. Chem. Phys. 164, 014118 (2026); doi: 10.1063/5.0300522
Submitted: 2 September 2025 • Accepted: 17 December 2025 •
Published Online: 7 January 2026

View Online Export Citation CrossMark

Abhisek Ghosal and George C. Schatz[a)]

AFFILIATIONS

Department of Chemistry, Northwestern University, Evanston, Illinois 60208, USA

[a)]Author to whom correspondence should be addressed: g-schatz@northwestern.edu

ABSTRACT

Collision-induced autoionizing excited states play an important role in plasma formation through associative ionization, where excited states lie in resonance with the continuum. In this work, we compute the autoionization widths of various doubly excited states of the N_2 molecule using equation-of-motion coupled-cluster theory combined with complex basis functions. This study represents the first application of spin-flip methods to doubly excited autoionizing states, enabled by a newly developed computational protocol based on Kaufmann basis functions. We apply this protocol to N_2 and determine the widths of the $^3\Sigma_g^+$, $3\text{–}4^3\Pi_u$, and $2^3\Delta_g$ states, which are potential contributors to the associative ionization process. Our results establish the complex basis function-based spin-flip method as a reliable and systematically improvable approach for resonance width calculations, opening avenues for its application to a broader class of autoionizing states in molecular systems.