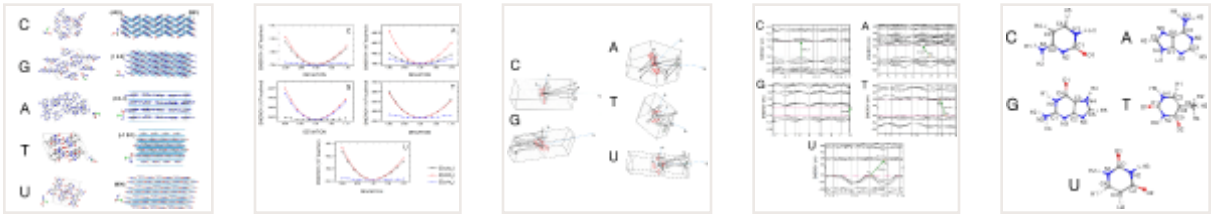
Improved description of the structural and optoelectronic properties of DNA/RNA nucleobase anhydrous crystals: Experiment and dispersion-corrected density functional theory calculations

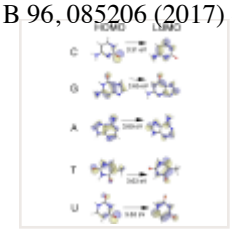
M. B. da Silva, T. S. Francisco, F. F. Maia, Jr., E. W. S. Caetano, U. L. Fulco, E. L. Albuquerque, and V. N. Freire

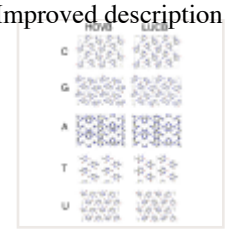
Phys. Rev. B **96**, 085206 – Published 17 August 2017

ABSTRACT

The development of low cost and environmentally friendly organic electronic/optoelectronic devices has attracted a lot of interest. The integration of DNA and RNA nucleobases to improve the performance of organic light-emitting diodes has been proposed recently [Gomez *et al.*, Sci. Rep. **4,** 7105 (2014) (http://dx.doi.org/10.1038/srep07105)], notwithstanding limited experimental and theoretical information on the optoelectronic properties of DNA/RNA thin films. As a contribution to an improved understanding of DNA/RNA-based devices in the solid state, we have performed in this paper dispersion corrected density functional theory (DFT) and time-dependent DFT (TDDFT) calculations to obtain the optimized geometries, Kohn-Sham band structures and orbitals, charge distribution, optical absorption, Frenkel exciton binding energies, and complex dielectric functions of the five DNA/RNA nucleobase anhydrous crystals, namely cytosine, guanine, adenine, thymine, and uracil. Optical absorption measurements on DNA/RNA nucleobase powders were also performed for comparison with the simulations. An improvement on the local density approximation (LDA) description of the lattice parameter estimates was achieved considering the generalized gradient approach (GGA) with a semiempirical dispersion correction scheme in comparison with structural x-ray data found in the literature. Energy gap correction using the Δ-sol methodology provided a good agreement between theory and experimental estimates from our optical absorption data, greatly surpassing the quality of previous simulations. Effective masses for the carriers were also found, indicating that the guanine crystal as well as the cytosine one (although with some drawbacks) has potential applications in optoelectronics as a direct gap semiconductor, with the other nucleobases presenting either a semiconductor or an insulator character depending on the carrier type. The complex dielectric function exhibits a high degree of anisotropy for different states of light polarization relative to the molecular stacking planes, while the Frenkel exciton binding energy estimation for the adenine crystal is very close to the optical absorption experimental data.







6 More

Received 8 February 2017

DOI: https://doi.org/10.1103/PhysRevB.96.085206

Physics Subject Headings (PhySH)

Research Areas

Band gap (http://physh.aps.org/concepts/16575caeb51f473c975cdcbf05d2de80)

Density functional theory (http://physh.aps.org/concepts/05d1f0ed0bf54c6d9e38a9cf3a67eb5f)

Density of states (http://physh.aps.org/concepts/aaff251998914d4ea0671ea36b6b08d4)

Electronic structure (http://physh.aps.org/concepts/9e019803f9d84802b1acd2c035d9508f)

First-principles calculations (http://physh.aps.org/concepts/55bba34b0d31428fa4304f502667434a)

Optoelectronics (http://physh.aps.org/concepts/3d71d9d8-fab6-4338-be83-7624e12f1438)

Structural properties (http://physh.aps.org/concepts/15f520a35c6c4d9e897581d399140562)

Physical Systems

Crystal structures (http://physh.aps.org/concepts/6af5be88489f4a509abf998e8912254a)

Crystalline systems (http://physh.aps.org/concepts/1128a9f370c1409b84b244063aa12108)

Nucleic acids (http://physh.aps.org/concepts/870d415a-ceb3-4e49-a475-798d07177a83)

Organic compounds (http://physh.aps.org/concepts/101b99f734cb4ea98f26b5d649d98640)

Semiconducting systems (http://physh.aps.org/concepts/27c8f20be22444b3bd139233ce79dcb8)

Atomic, Molecular & Optical Biological Physics Condensed Matter & Materials Physics

AUTHORS & AFFILIATIONS

M. B. da Silva (/search/field/author/M%20B%20da%20Silva)[1], T. S. Francisco (/search/field/author/T%20S%20Francisco)[2], F. F. Maia, Jr. (/search/field/author/F%20F%20Maia%20Jr)[3], E. W. S.

Caetano (/search/field/author/E%20W%20S%20Caetano)[4], U. L. Fulco (/search/field/author/U%20L%20Fulco)[5], E. L. Albuquerque (/search/field/author/E%20L%20Albuquerque)[5], and V. N. Freire (/search/field/author/V%20N%20Freire)[1]

[1]Departamento de Física, Universidade Federal do Ceará, Caixa Postal 6030, 60455-760 Fortaleza, CE, Brazil
[2]Universidade Estadual Vale do Acaraú, Av. da Universidade, 850–Campus da Betânia, 62040-370, Sobral, CE, Brazil
[3]Universidade Federal Rural do Semi-Árido, Av. Francisco Mota, 572, 59625-900, Mossoró, RN, Brazil
[4]Instituto Federal de Educação, Ciência e Tecnologia do Ceará, 60040-531 Fortaleza, CE, Brazil
[5]Departamento de Biofísica e Farmacologia, Universidade Federal do Rio Grande do Norte, 59072-970 Natal, RN, Brazil

ARTICLE TEXT (SUBSCRIPTION REQUIRED)
CLICK TO EXPAND

SUPPLEMENTAL MATERIAL (SUBSCRIPTION REQUIRED)
CLICK TO EXPAND

REFERENCES (SUBSCRIPTION REQUIRED)
CLICK TO EXPAND

Issue

Vol. 96, Iss. 8 — 15 August 2017 (/prb/issues/96/8)

Access Options

AUTHORS

General Information (/prb/authors)

Submit a Manuscript (https://authors.aps.org/Submissions/)

Publication Rights (/pub_rights.html)

Open Access (/open_access.html)

Policies & Practices (/prb/authors/editorial-policies-practices)

Tips for Authors (/authors/tips-authors-physical-review-physical-review-letters)

Professional Conduct (/authors/professional-conduct-ethics)

REFEREES

General Information (/prb/referees)

Submit a Report (http://referees.aps.org/)

Update Your Information (http://referees.aps.org/)

Policies & Practices (/prb/authors/editorial-policies-practices)

Referee FAQ (/referees/faq.html)

Guidelines for Referees (/prb/referees/advice-referees-physical-review)

Outstanding Referees (/OutstandingReferees)

LIBRARIANS

General Information (https://librarians.aps.org/)

Subscriptions (https://librarians.aps.org/subscriptions)

Online License Agreement (https://librarians.aps.org/sitelicense.pdf)

Usage Statistics (http://counter.aps.org/)

Your Account (https://librarians.aps.org/account)

STUDENTS

Physics (https://physics.aps.org)

PhysicsCentral (http://www.physicscentral.com/)

Student Membership (https://www.aps.org/membership/student.cfm)

APS MEMBERS

Subscriptions (https://www.aps.org/membership/aps-publications.cfm)

Article Packs (https://journals.aps.org/article-packs)

Membership (https://www.aps.org/membership/index.cfm)

FAQ (https://www.aps.org/membership/faq.cfm)

APS News (https://www.aps.org/publications/apsnews/index.cfm)

Meetings & Events (https://www.aps.org/meetings/index.cfm)