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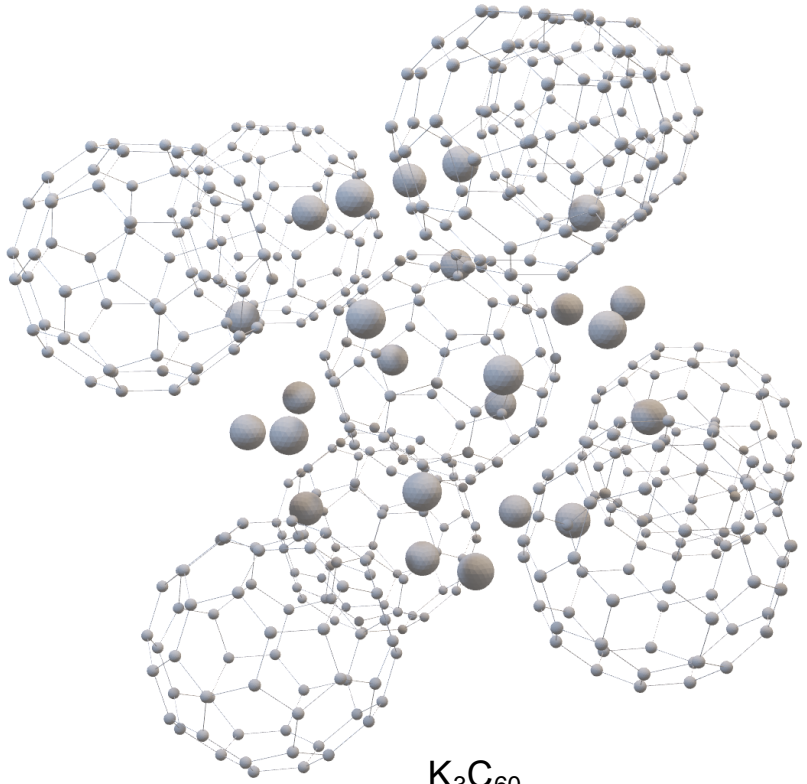
Electron–Phonon Coupling and Superconductivity in FHI-aims

Connor Box



K_3C_{60}

Model credit: www.chemtube3d.com

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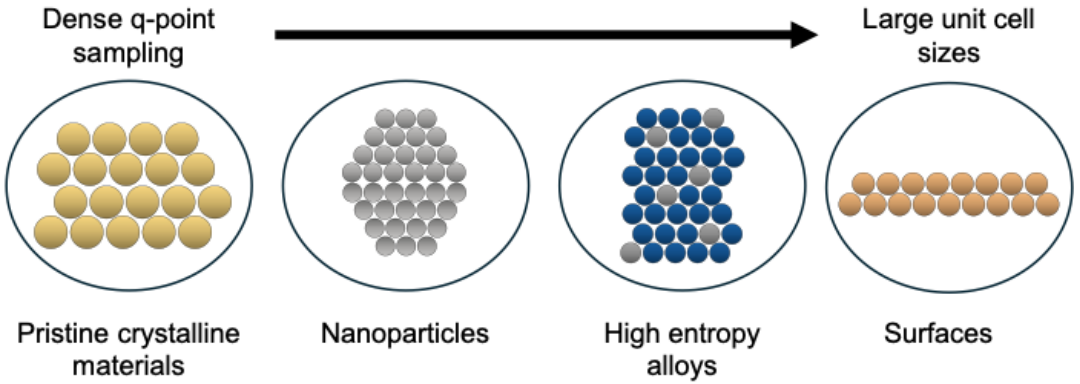
Superconductivity in Real space

- Predicting superconductivity requires access to phonons, electron–phonon coupling, and superconducting observables (α^2F, λ, T_c)
- Existing workflows are dominated by pseudo-potential plane-wave implementations, while FHI-aims provides an all-electron, numeric atom-centered orbital framework in real space.
- Many superconducting systems of interest do not reduce to a small unit cell.



"It would be amazing if we could calculate electron-phonon coupling / superconductivity for hundreds of atoms" Nov 2024

Pedro Nunes Ferreira

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What was already implemented in FHI-aims?

Reinhard Maurer

Time-dependent perturbation theory
electronic friction

Kohn-Sham eigenvalues

$$\Lambda_{a\kappa,a'\kappa'} = 2\pi\hbar \sum_{mn} \int \frac{\mathrm{d}\mathbf{k}}{\Omega_{BZ}} \tilde{g}^{a\kappa}_{mn}(\mathbf{k}) \left(\tilde{g}^{a'\kappa'}_{mn}\right)^{*}(\mathbf{k}) \left(f_{n\mathbf{k}} - f_{m\mathbf{k}}\right) \frac{\delta\left(\epsilon_{m\mathbf{k}} - \epsilon_{n\mathbf{k}}\right)}{\epsilon_{m\mathbf{k}} - \epsilon_{n\mathbf{k}}}.$$

Electron-phonon coupling

Fermi occupations

$$\tilde{g}^{a\kappa}_{mn}(\mathbf{k}) = \sum_{ij} \left(C^{j}_{m\mathbf{k}}\right)^{*} C^{i}_{n\mathbf{k}} \left(\mathbf{H}^{a\kappa,(1)}_{ij}(\mathbf{k}) - \epsilon_{n\mathbf{k}} \mathbf{S}^{a\kappa,L}_{ij}(\mathbf{k}) - \epsilon_{m\mathbf{k}} \mathbf{S}^{a\kappa,R}_{ij}(\mathbf{k})\right)$$

Response matrices

Response matrices calculated with **finite difference**
or **density functional perturbation theory (DFPT)**

`calculate_friction` `numerical_friction`

`calculate_friction` `DFPT`

R. J. Maurer, M. Askerka, V. S. Batista, and J. C. Tully, PRB, **94**, 115432, (2016)
C. L. Box, W. G. Stark, R. J. Maurer, Electron. Struc. 5 , 035005, (2023)

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Phonon linewidths

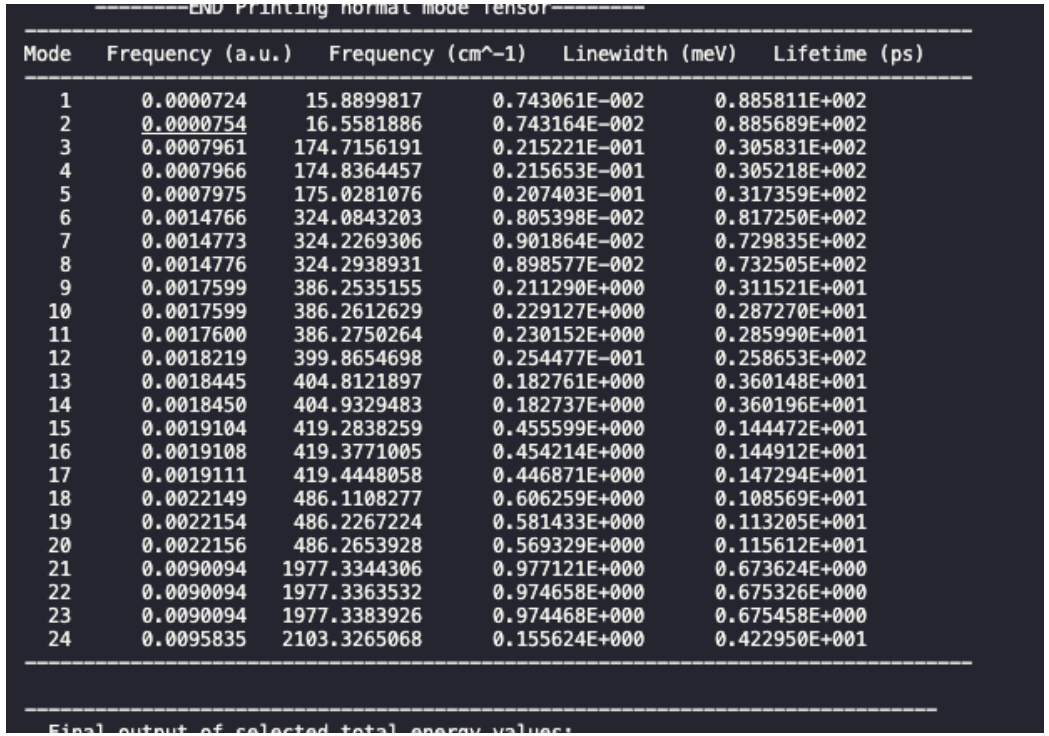
$$\gamma_{\mathbf{q}\nu} = \hbar\Gamma_{\mathbf{q}\nu} = \hbar\left[\tilde{\mathbf{u}}_{\mathbf{q}\nu}\mathbf{\Lambda}^{\mathbf{q}}\left(\hbar\omega_{\mathbf{q}\nu}\right)\tilde{\mathbf{u}}^{T}_{\mathbf{q}\nu}\right]$$

Keyword allows the calculation of γ

Normal modes ($\boldsymbol{u}$) are required for this transformation.

Usage: `friction_output_vibrations`
Purpose: Calculates the normal modes of vibration and Hessian during finite difference (if requested)
Default is .false.
`boolean`: .true. or .false.

Set an appropriate `sc_accuracy_forces` and integration grid.



--------END Printing normal mode Tensor--------

Mode	Frequency (a.u.)	Frequency (cm^-1)	Linewidth (meV)	Lifetime (ps)
1	0.0000724	15.8899817	0.743061E-002	0.885811E+002
2	0.0000754	16.5581886	0.743164E-002	0.885689E+002
3	0.0007961	174.7156191	0.215221E-001	0.305831E+002
4	0.0007966	174.8364457	0.215653E-001	0.305218E+002
5	0.0007975	175.0281076	0.207403E-001	0.317359E+002
6	0.0014766	324.0843203	0.805398E-002	0.817250E+002
7	0.0014773	324.2269306	0.901864E-002	0.729835E+002
8	0.0014776	324.2938931	0.898577E-002	0.732505E+002
9	0.0017599	386.2535155	0.211290E+000	0.311521E+001
10	0.0017599	386.2612629	0.229127E+000	0.287270E+001
11	0.0017600	386.2750264	0.230152E+000	0.285990E+001
12	0.0018219	399.8654698	0.254477E-001	0.258653E+002
13	0.0018445	404.8121897	0.182761E+000	0.360148E+001
14	0.0018450	404.9329483	0.182737E+000	0.360196E+001
15	0.0019104	419.2838259	0.455599E+000	0.144472E+001
16	0.0019108	419.3771005	0.454214E+000	0.144912E+001
17	0.0019111	419.4448058	0.446871E+000	0.147294E+001
18	0.0022149	486.1108277	0.606259E+000	0.108569E+001
19	0.0022154	486.2267224	0.581433E+000	0.113205E+001
20	0.0022156	486.2653928	0.569329E+000	0.115612E+001
21	0.0090094	1977.3344306	0.977121E+000	0.673624E+000
22	0.0090094	1977.3363532	0.974658E+000	0.675326E+000
23	0.0090094	1977.3383926	0.974468E+000	0.675458E+000
24	0.0095835	2103.3265068	0.155624E+000	0.422950E+001

Final output of selected total energy values:

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Eliashberg theory

`calculate_superconductivity` `numerical`

$$\alpha^2 F(\omega) = \frac{1}{2\pi N_{\epsilon_F} \hbar\omega} \sum_\nu \delta(\omega - \omega_\nu)\, \gamma_\nu, \qquad \mathbf{q} = 0$$

Density states at Fermi level, evaluated using smeared function e.g Gaussian

Phonon linewidths from friction code


Pedro Nunes Ferreira


Eva Kogler


Reinhard Maurer


Christoph Heil

$$\lambda = 2\int d\omega \frac{\alpha^2 F(\omega)}{\omega}.$$

$$T_c = \frac{f_1 f_2 \omega_{\log}}{1.20} \exp\left[-\frac{1.04(1+\lambda)}{\lambda - \mu^*(1+0.62\lambda)}\right]$$

P. B. Allen and R. C. Dynes, Phys. Rev. B 12, 905 (1975)

Correction factors (simply evaluated)

Coulomb pseudopotential (user input)

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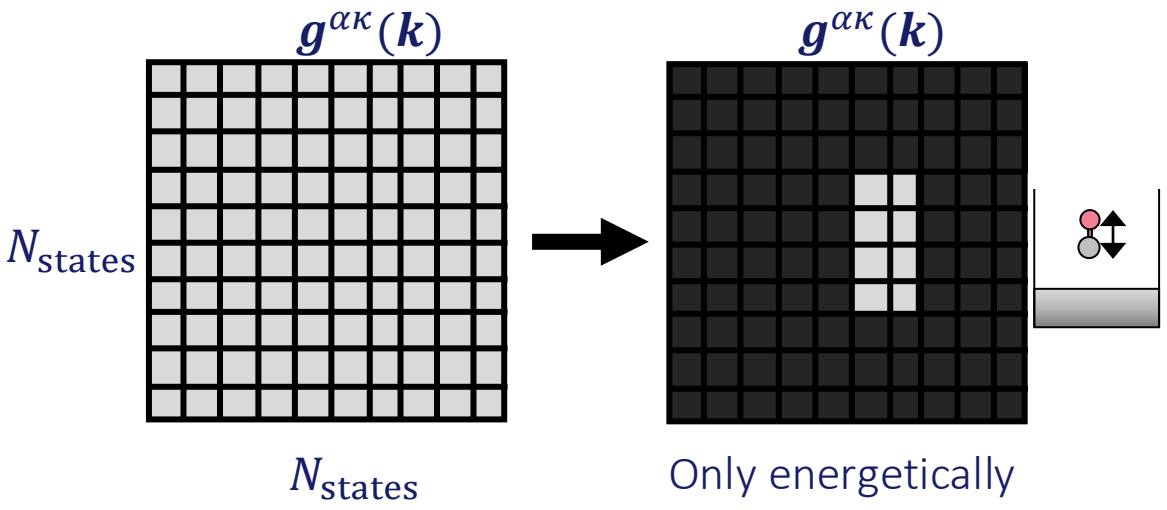
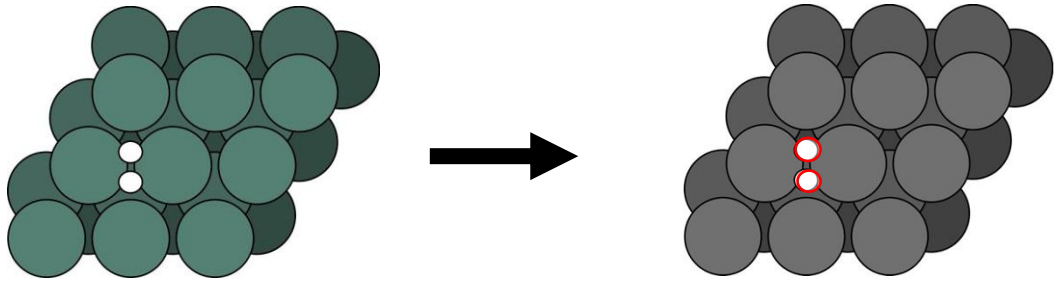
Code choices and efficiency



Only energetically accessible states stored

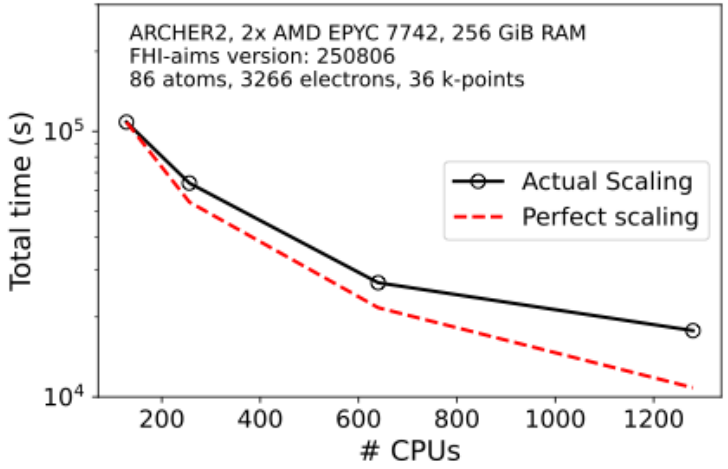


Select atoms to calculate el-ph coupling

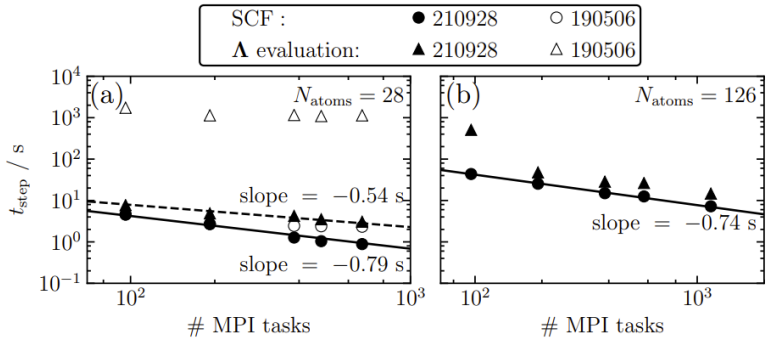
C. L. Box, W. G. Stark, R. J. Maurer, Electron. Struc. 5 , 035005, (2023)

FHI-aims
The ab initio materials simulation package

Local atomic orbital basis



Strong scaling for many cores



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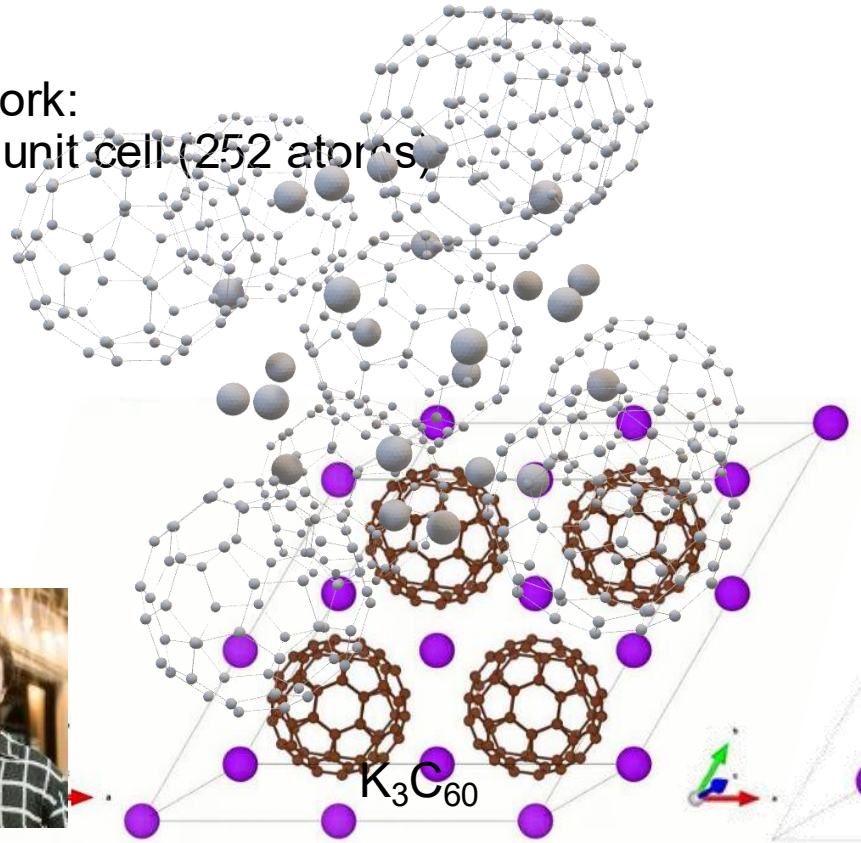
Case study: K_3C_{60}

- Previous theoretical work ignored the disorder in the orientation of the C_{60} units.
- This work:
  2x2x1 unit cell (252 atoms)

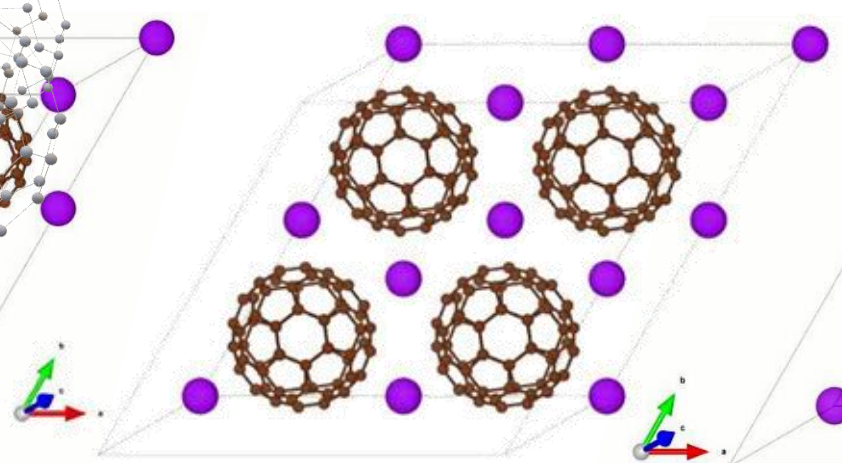
Mode coupling and electronic structure calculations. Electronic structure, lattice dynamics, and nonlinear phonon couplings after excitation of the $T_{1u}(4)$ phonon were obtained using density functional theory (DFT) calculations with a plane-wave basis set and projector augmented wave pseudopotentials (VASP software package)[35]. These calculations were performed within the local density approximation (LDA) with a 900 eV cut-off for the plane-wave basis set. A $6 \times 6 \times 6$ k-point grid and a 0.1 eV Gaussian smearing were used for the Brillouin-zone integration during the self-consistency cycles. Lattice parameters and atomic positions were obtained by energy minimization, ignoring the C_{60} orientational disorder. The calculated lattice constant is 13.888 Å, in good agreement with the experimental value of 14.240 Å.

Mitrano, M., Cantaluppi, A., Nicoletti, D. *et al. Nature* **530**, 461–464 (2016).



K_3C_{60}

Pedro Nunes Ferreira



AAAA

BBBB

ABAB

Model credit: www.crystalmaker.com

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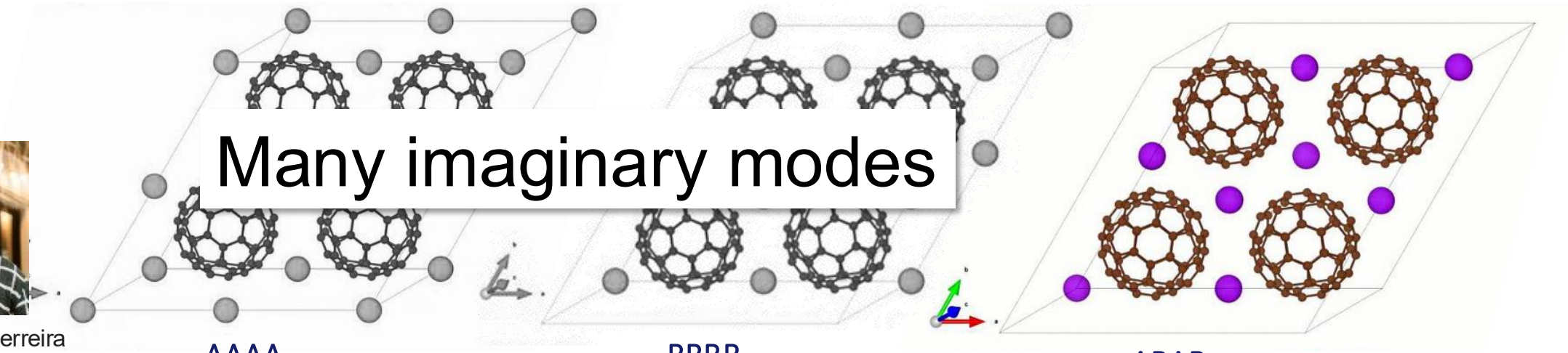
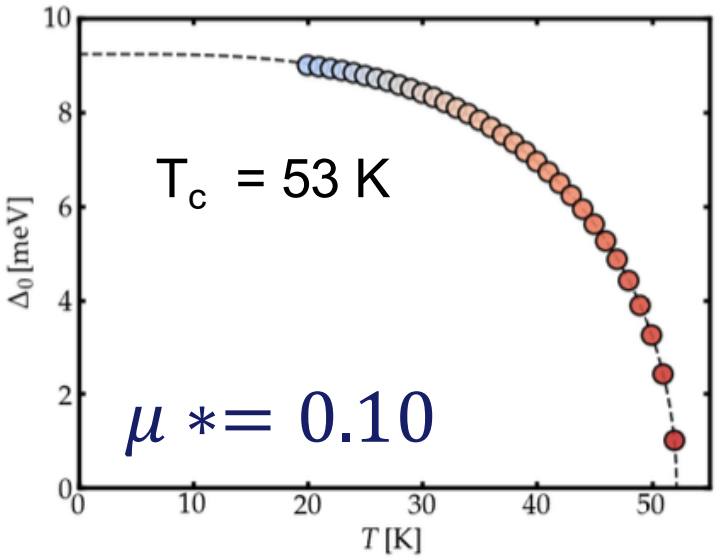
Pedro Nunes Ferreira

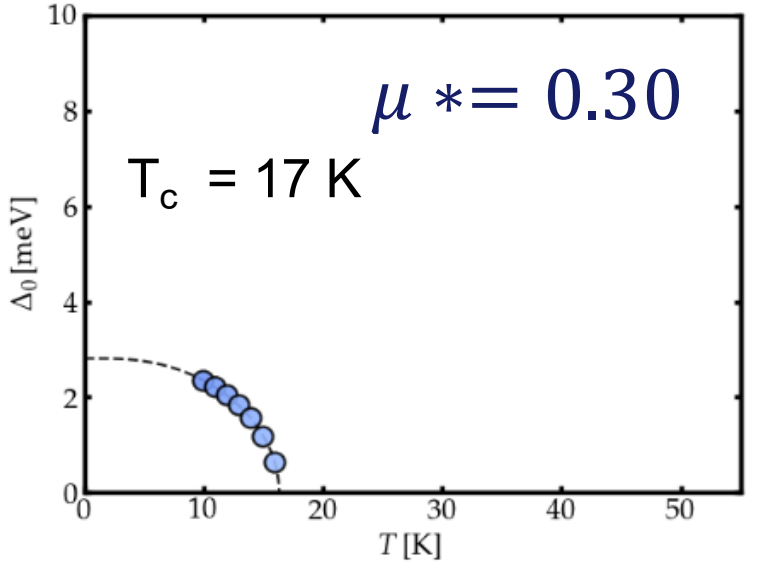


Case study: K_3C_{60}

Produced using IsoME.jl [1]

- Previous theoretical work ignore the disorder in the orientation of the C_{60} units.
- This work:
  2x2x1 unit cell (252 atoms)
- Experimental T_c = 19 K [1]
- A mu* value of 0.3 is suggested in literature for C_{60} systems: O. Gunnarsson and G. Zwicknagl, Phys. Rev. Lett. 69, 957 (1992)





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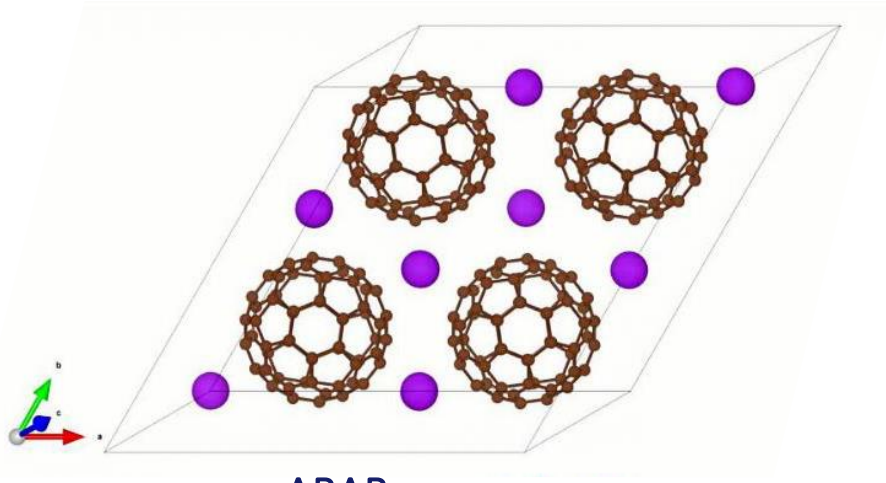
We reproduce experimental T_C with a physically justified mu* value



ABAB

[1] E. Kogler et al, Comp. Phys. Comm, (2025)

Important user choices

- **Electronic smearing** that enters friction/linewidth expression
  - Anecdotally, we see we require much smaller smearing widths (σ) than plane wave codes for convergence w.r.t N_{k}
  - We are looking into a sensible automatic choice workflow [1] :
    1. Calculate $N(\epsilon_F)$ with DOS Tetrahedron method (very well converged)
    2. Calculate $N(\epsilon_F)$ with different σ values
    3. Choose smallest σ that reproduces DOS tetrahedron method

- **Supercell size** for periodic systems
  - $q = 0$ is not a free lunch.
  - Convergence of phonons with respect to supercell size is material dependent.
  - Interpolation to dense **q**-grids is possible (as a post-processing script right now)
  - T_c often convergences faster than $\alpha^2 F(\omega)$ - relevant for high-throughput

[1] Similar logic to T. Koretsune, R. Arita Comp. Phys. Comm. (2017)

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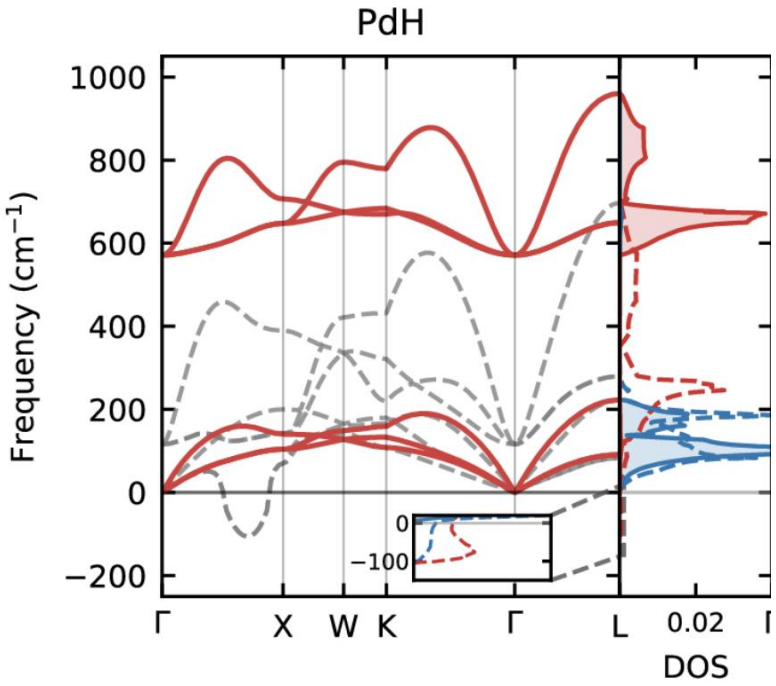
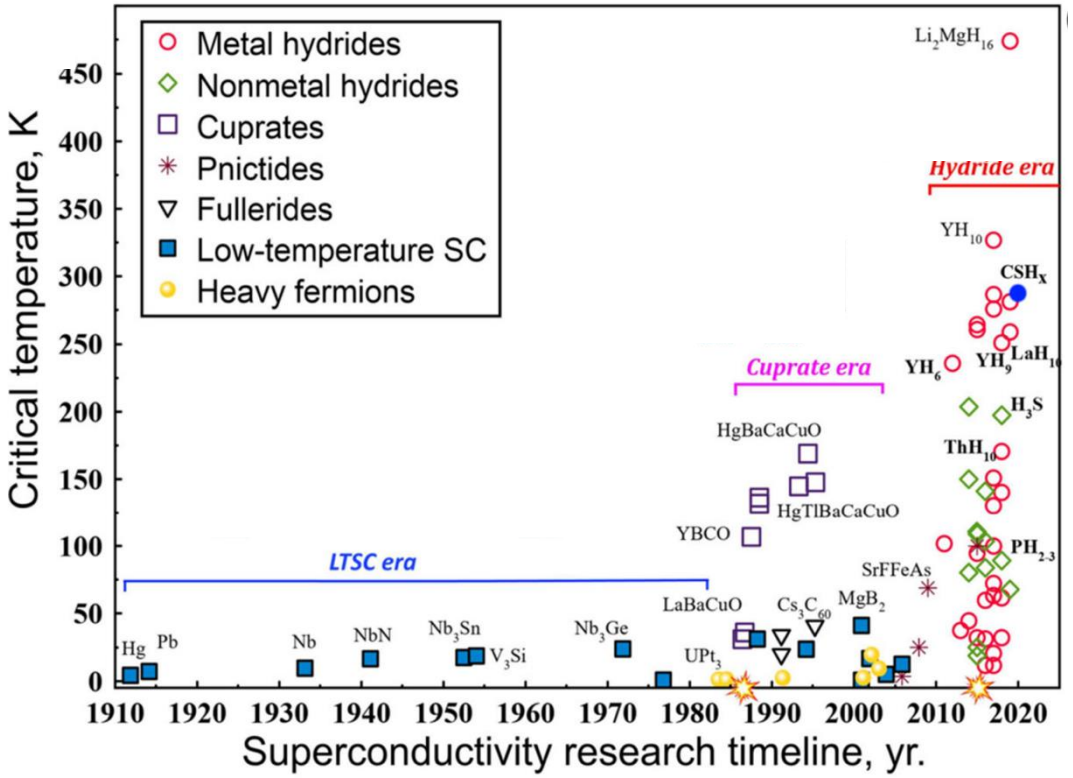
Future directions: beyond harmonic superconductivity



Anharmonic phonons from SSCHA + MLIP

Harmonic el-ph elements from Quantum ESPRESSO.

B. Yin et al, arXiv:2603.25373 (2026)



Nuclear electronic orbital (NEO)-DFT in FHI-aims.

L.E. Smith, P. Settembri, A. Cucciari, L. Boeri, G. Profeta, S. Hammes-Schiffer, *Proc. Natl. Acad. Sci. U.S.A.* 123 (21), (2026).

I A Troyan et al, Physics-Uspekhi, 65 (7) 748 -761 (2022)

If you are interested in using the code – tutorials!

Tutorial 2: Nb 2x2x2 Superconductivity Example

This tutorial demonstrates a complete superconductivity calculation in FHI-aims using a bcc Nb `2x2x2` supercell example.

The reference calculation used here is:

`/Users/u1865573/work/2025/tutorial/superconductivity_calc/nb_222`

1. Input files

Prepared input files are provided in:

`Tutorial-2/input_files/`

Files:

- `control.in` : superconductivity settings and numerical parameters
- `geometry.in` : Nb `2x2x2` supercell with friction flags on all atoms

Reference output files from the Nb `2x2x2` run are also provided in:

`Tutorial-2/output_files/`

Available in the Aims club tutorials

(Not on index page yet)

Tutorial 3: Interpolating Eliashberg Function and Tc on Dense q Grids

This tutorial demonstrates an interpolation workflow that maps supercell (`q=0`) results back to primitive-cell `q` points and evaluates `alpha^2F(omega)` and `T_c` on a much denser `q` mesh.

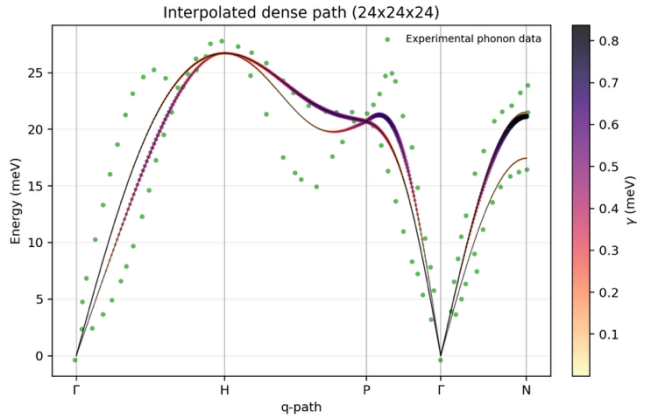
1. Motivation

The direct FHI-aims superconductivity implementation operates in a `q=0` framework.
Large supercells fold additional primitive-cell `q` points to supercell `q=0` , but the resulting sampling is still sparse.
Interpolation helps recover a denser `q` representation and more converged spectral properties. For Nb in particular, a denser electronic `k` -grid is also required to converge the Eliashberg spectrum and derived `T_c` . In practice, moving from a `2x2x2` to a `3x3x3` supercell, together with denser electronic `k` -point sampling, should better recover both the experimental phonon dispersion and the Eliashberg spectrum for Nb. This is the main reason to treat the present `2x2x2` example as a workflow demonstration rather than a fully converged productio

2. Input data used here

This example uses the Nb `2x2x2` outputs copied into:

- `Tutorial-3/input_files/geometry.in`
- `Tutorial-3/input_files/friction_hessian.out`
- `Tutorial-3/input_files/friction_normal_mode_`
- `Tutorial-3/input_files/eliashberg_function.`
- `Tutorial-3/input_files/superconductivity_tc.`
- `Tutorial-3/input_files/exp2.csv` (experimenta
- `Tutorial-3/input_files/exp_phonon_dispersio`



Conclusion

- Superconductivity in FHI-aims is:
  - Not a replacement for existing plane-wave implementations
  - But rather a complementary tool

- Features:
  - Hessian (inc acoustic sum rule)
  - Phonon DOS and atom-resolved Phonon DOS
  - Eliashberg function and atom-resolved Eliashberg function
  - Phonon linewidths
  - Electron-phonon coupling matrix elements using ELSI
  - Electron-phonon coupling constant
  - T_c

C. L. Box, P. N. Ferreira, E. Kogler, C. Heil, R. J. Maurer, in preparation, (2026)

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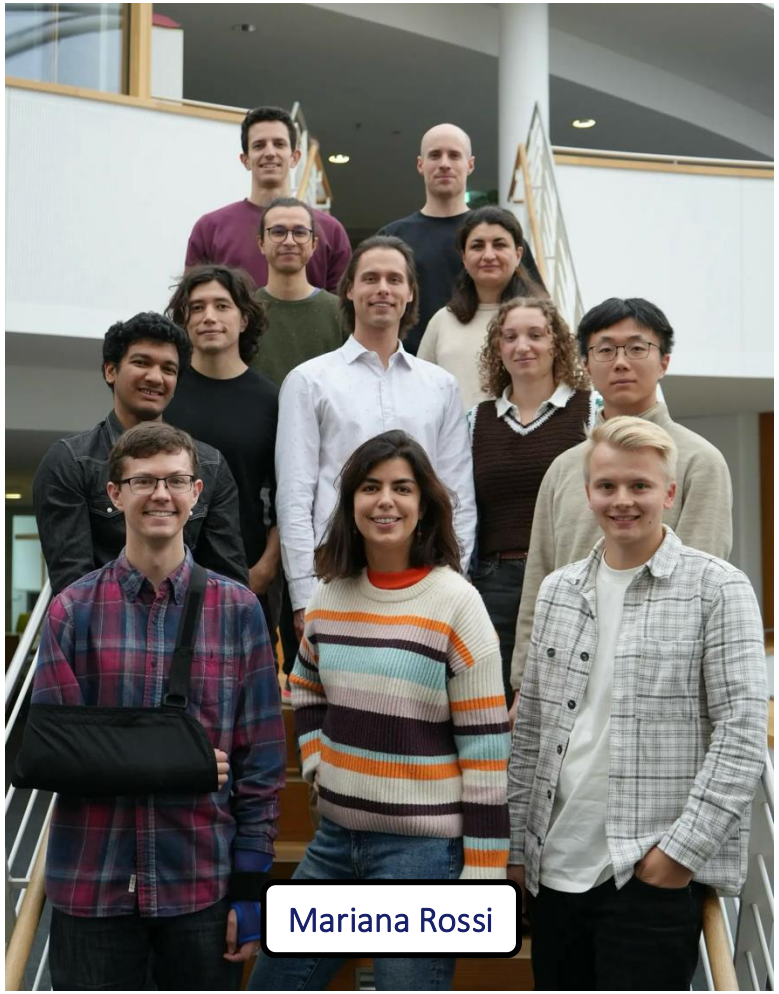

SABIA

General thanks to FHI-aims community & MS1P team

Engineering and Physical Sciences Research Council

HEC MCC UK

hpc MIDLANDS+

Centre for Scientific Computing

archer2

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