Discovery of Novel Superconducting Materials with Deep Learning

Colin Burdine
E. P. Blair

IEEE Quantum Week
September 19, 2023

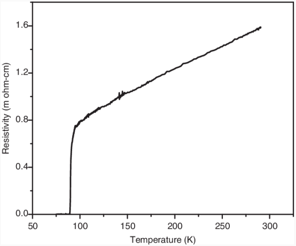
Baylor University

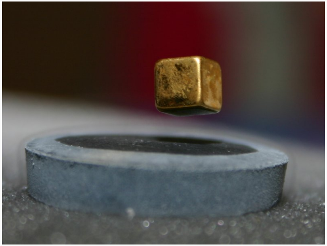
Background

Superconductors

- Superconductors are materials that exhibit two key properties at low temperatures:
  1. The resistivity ρ drops to 0Ω (Current flows without resistance).
  2. Magnetic fields are expelled in the bulk. (Meissner Effect)

Example: $YBa_2Cu_3O_{7-\delta}$ [VBRS14]



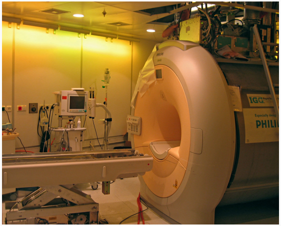


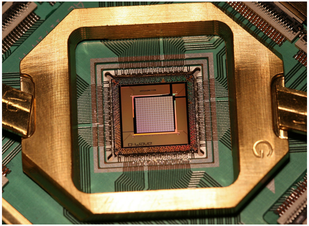
- A superconductor's critical temperature (T_c) is the highest temperature at which it exhibits these two properties.

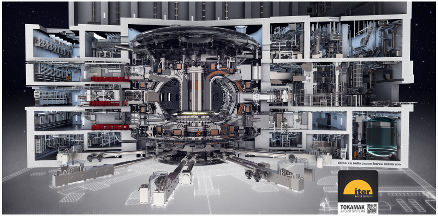
https://commons.wikimedia.org/wiki/File:Meissner_effect_p1390048.jpg

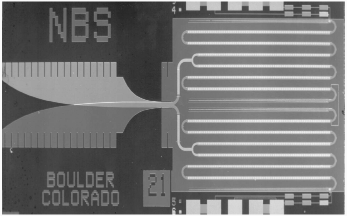
Superconductors

Industrial Applications of Superconductors









https://www.flickr.com/photos/14646075@N03/2833410223
https://www.dwavesys.com/solutions-and-products/systems/
https://commons.wikimedia.org/wiki/File:Transrapid-emsland.jpg
https://www.flickr.com/people/37940997@N05
https://nvlpubs.nist.gov/nistpubs/sp958-lide/315-318.pdf

Superconductors

- Superconductivity occurs due to anomalous quantum mechanical pairing of electrons [Coo60].
- Pairing allows for condensation of conducting electrons into a single energy state at the Fermi level, E_F:

$$\underbrace{|\uparrow\rangle, |\downarrow\rangle}_{\text{Spin-1/2 electrons}} \longrightarrow \underbrace{\frac{(|\uparrow\downarrow\rangle - |\downarrow\uparrow\rangle)}{\sqrt{2}}}_{\text{Spin-0 Singlet Cooper pair}}$$

$$\underbrace{|\uparrow\rangle, |\uparrow\rangle}_{\text{Spin-1/2 electrons}} \longrightarrow \underbrace{|\downarrow\downarrow\rangle, \quad \frac{(|\uparrow\downarrow\rangle + |\downarrow\uparrow\rangle)}{\sqrt{2}}, \quad |\uparrow\uparrow\rangle}_{\text{Spin-1 Triplet Cooper pairs}} \quad [\text{m} = \text{-}1,\ 0,\ +1]$$

- Pairing in conventional (i.e. BCS) superconductors is mediated by phonons [BCS57].
- Pairing in unconventional superconductors has not yet been fully explained [ZLI+21].

What makes a good Superconductor?

- High T_c is desirable in most industrial applications, since He-based cryogenics are required for most conventional superconductors.
  - The known limit of conventional superconductors is 44 K at ambient pressure (MgB_2) and 250K+ (Hydrides) at high pressure.
  - Cost effective cooling begins at 77 K (Liquid N_2).

- However, T_c isn't everything:
  - In quantum information devices, coherence time and fidelity of superconducting states is more desirable than high operating T_c.
  - Many unconventional superconductors have superconducting states that are robust under magnetic interference or other perturbations.
  - Unconventional superconductors may provide platforms for the development of the next generation of superconducting quantum devices.

Example: UTe_2

- UTe_2 is an unconventional heavy fermion superconductor in which magnetic and superconducting spin triplet phases coexist [ABF$^+$22].

nature > articles > article

Article | Published: 25 March 2020

Chiral superconductivity in heavy-fermion metal UTe_2

Lin Jiao orcid.org/0000-0002-8343-0190[1], Sean Howard orcid.org/0000-0002-6132-4688[1], Sheng Ran orcid.org/0000-0003-1557-267X[2,3], Zhenyu Wang orcid.org/0000-0002-4857-7234[1], Jorge Olivares Rodriguez[1], Manfred Sigrist[4], Ziqiang Wang[5], Nicholas P. Butch orcid.org/0000-0002-6083-8388[2,3] & Vidya Madhavan[1]

Nature **579**, 523–527 (2020) | Cite this article

Subjects

Superconducting properties and materials   Topological matter

- UTe_2's chiral *p*-wave states may give rise to topologically robust Majorana edge states [JHR$^+$20].

Prediction of T_c

- Prediction of T_c via *ab initio* methods is only possible for conventional (BCS) superconductors.

McMillan Equation [McM68]

$$T_c = \frac{\vartheta}{1.45} \exp\left(-\frac{1.04(1+\lambda)}{\lambda - \mu^*(1+0.62\lambda)}\right)$$

(ϑ: characteristic phonon frequency, μ^*: Coulomb pseudopotential, λ: electron-phonon coupling)

- Typically, λ is computed from the Eliashberg spectrum, which is obtainable through density functional theory (DFT) methods.
- For large or disordered systems, estimating λ comes at a significant computational cost.

(at least, on a classical computer...).

Machine Learning and Superconductivity

- There has been great interest in applying machine learning to superconductor discovery (e.g. see: [SOK+18][KKN+21][XQH+22])
- Few of these approaches directly incorporate atomic structure. E.g. [CG22]:

nature > npj computational materials > articles > article

Article | Open Access | Published: 22 November 2022

Designing high-T_C superconductors with BCS-inspired screening, density functional theory, and deep-learning

Kamal Choudhary orcid.org/0000-0001-9737-8074[1,2,3] & Kevin Garrity[1,2]

npj Computational Materials **8**, Article number: 244 (2022) | Cite this article

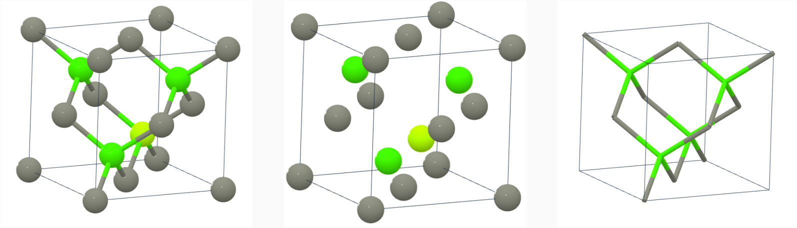
Subjects

Atomistic models   Superconducting properties and materials

Deep Graph Neural Networks

- Graphs are data structures consisting of networks of nodes connected by edges.
- Atomic structures are naturally interpreted as periodic graphs:

Crystal (graph) Atoms (nodes) Bonds (edges)



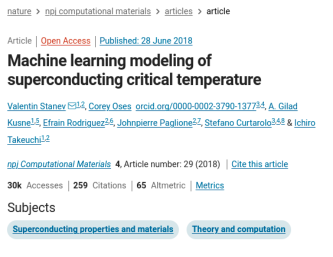
- In this work, we applied these graph neural networks to identify superconductors and predict T_c from graph representations of crystal structure using experimental data.

Methods

Data Sources

- We combined two large experimental measurement datasets (*Supercon v1* [SOK+18] and *Supercon v2* [FdCS+23]).

Supercon v1



nature > npj computational materials > articles > article

Article | Open Access | Published: 28 June 2018

Machine learning modeling of superconducting critical temperature

Valentin Stanev[1,2], Corey Oses orcid.org/0000-0002-3790-1377[3,4], A. Gilad Kusne[1,5], Efrain Rodriguez[2,6], Johnpierre Paglione[2,7], Stefano Curtarolo[3,4,8] & Ichiro Takeuchi[1,2]

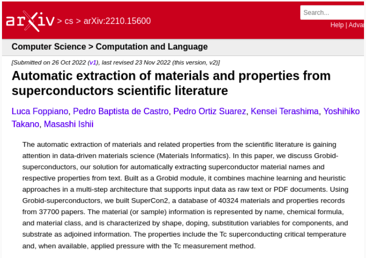
npj Computational Materials **4**, Article number: 29 (2018) | Cite this article

30k Accesses | **259** Citations | **65** Altmetric | Metrics

Subjects

Superconducting properties and materials | Theory and computation

Supercon v2



arXiv > cs > arXiv:2210.15600

Search...

Help | Adva

Computer Science > Computation and Language

[Submitted on 26 Oct 2022 (v1), last revised 23 Nov 2022 (this version, v2)]

Automatic extraction of materials and properties from superconductors scientific literature

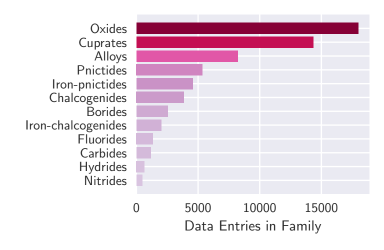
Luca Foppiano, Pedro Baptista de Castro, Pedro Ortiz Suarez, Kensei Terashima, Yoshihiko Takano, Masashi Ishii

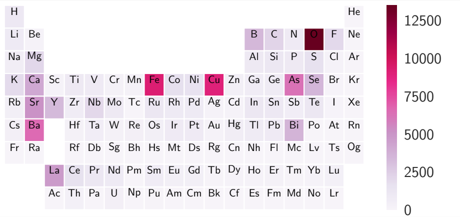
The automatic extraction of materials and related properties from the scientific literature is gaining attention in data-driven materials science (Materials Informatics). In this paper, we discuss Grobid-superconductors, our solution for automatically extracting superconductor material names and respective properties from text. Built as a Grobid module, it combines machine learning and heuristic approaches in a multi-step architecture that supports input data as raw text or PDF documents. Using Grobid-superconductors, we built SuperCon2, a database of 40324 materials and properties records from 37700 papers. The material (or sample) information is represented by name, chemical formula, and material class, and is characterized by shape, doping, substitution variables for components, and substrate as adjoined information. The properties include the Tc superconducting critical temperature and, when available, applied pressure with the Tc measurement method.

- These datasets were still need of significant cleaning:
  - Corrupted data and missing paper references
  - Algebraic chemical formulas without substituted values.
  - Extraction of wrong physical properties (e.g. Curie Temperature)

Data Sources

Dataset Distribution

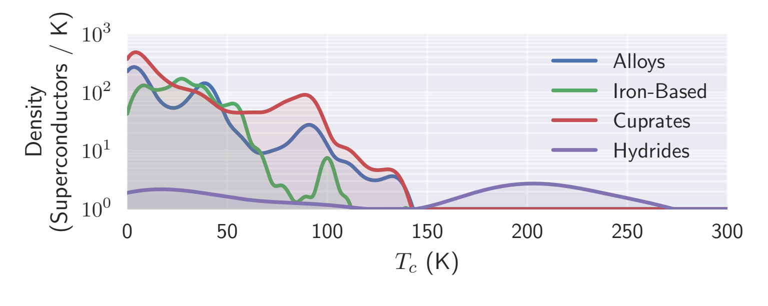




- These datasets had T_c measurements, chemical formulas, and other important metadata, but no atomic structure.
- To obtain crystal structure, we cross-referenced chemical formula data and paper metadata with the Materials Project database.

Data Sources

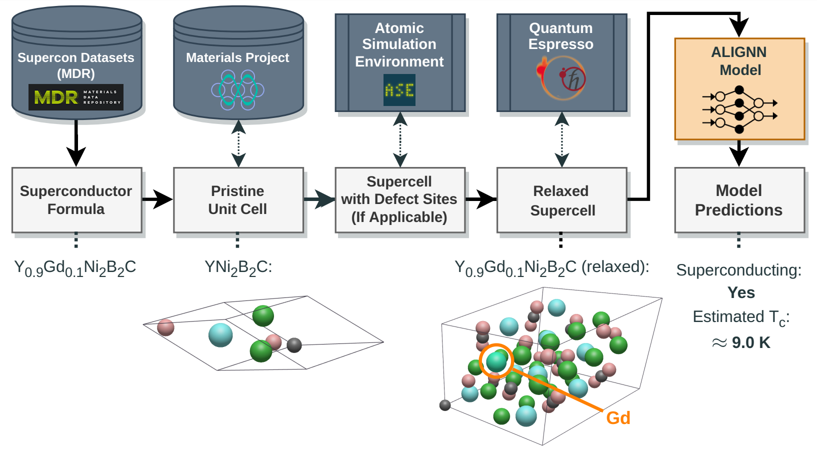
Distribution of Experimental T_c measurements



- 44 K: Limit of conventional ambient superconductivity.
- 100 K: Limit of FeSe thin films on $SrTiO_3$ substrate []
- $\approx$ 150 K: Limit of the cuprates (HgTlBaCaCuO @ 164 K)
- 250+ K: Hydrides (at high pressures of 100-300 GPa)

Generating Atomic Structures

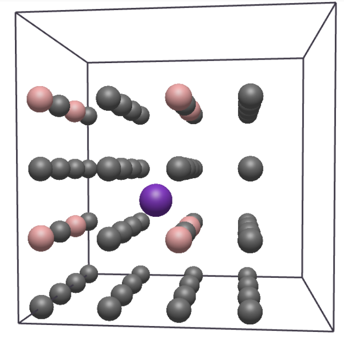
Data Generation Pipeline

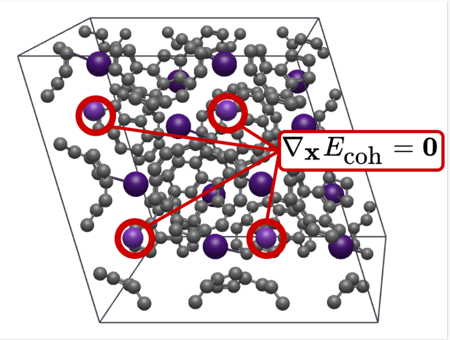


Data Cleaning Challenges

- A significant challenge was handling the placement of defects (vacancies, interstitials, etc. via the semi-classical embedded atom method [DFB93])

Defect Placement Examples: Fake Lattice (left), KCs_3C_{60} (right).

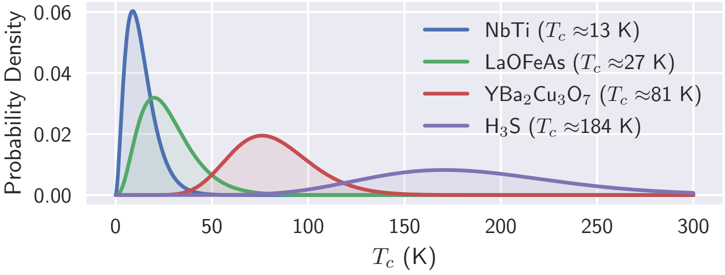




Data Cleaning Challenges

- For each superconducting material, many differing measurements were reported due to various factors (applied pressure, choice of substrate, film thickness, etc.)
- We constructed "empirical" distributions of reported T_c values, and had the model predict these distributions:

Examples of Empirical T_c Distributions:

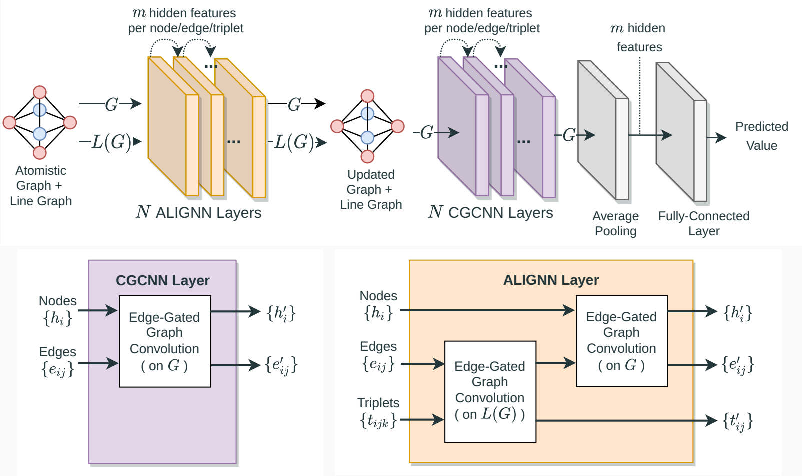


ALIGNN Model

- We used the Atomistic Line Graph Neural Network (ALIGNN) model for both classifying superconductors and predicting T_c [CD21].
- ALIGNN is one of the top performing deep learning models for structure-based material property predictions on the Materials Project's Matbench benchmarking leaderboard [DWG+20].
- Pros:
  - Is naturally invariant under $E(3)$ (Euclidean) symmetries and space group symmetries.
  - Learns representations of bonds and bond features directly from the atomic structure (No DFT required).
  - Time complexity is $\mathcal{O}(n)$ (by comparizon DFT is $\mathcal{O}(n^3)$).
  - Incorporates both bond lengths and bond angles.
- Cons:
  - Lacks interpretability.
  - Prone to overfitting and requires lots of data to perform well.

ALIGNN Model

ALIGNN Model Architecture [XG18][CD21]



ALIGNN Training

- First, we trained the ALIGNN model as a superconducting/non-superconducting binary classifier.
    - We used randomly sampled materials from the Materials Project database to serve as the non-superconducting class.
    - Non-superconducting materials were drawn according to the same distribution of elements as the superconductors class to mitigate sampling bias.
- We trained another ALIGNN regression model to predict the parameters of the empirical T_c distributions for identified superconductors.
    - To compute the loss between predicted and actual empirical T_c distributions, we used an approximation to the the squared $p = 2$ Wasserstein metric:

$$W_2(p_1(x), p_2(x))^2 = \inf_{\gamma(x,y)} \mathbb{E}_{\gamma(x,y)} |x - y|^2$$

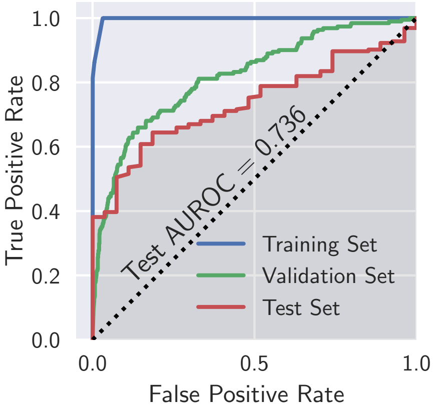
Results

Model Evaluation

Classifying Superconductors (Confusion Matrix and ROC Curve)

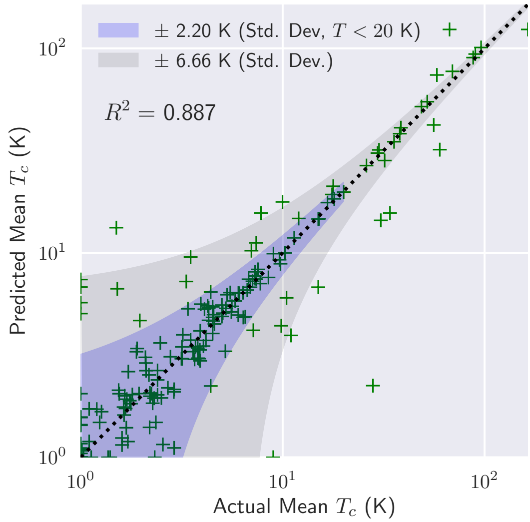
		Predicted Superconductor	
		Yes	**No**
Contained in Dataset	**Yes**	**Total: 102** $TaSe_3$, AlCu, W_2C, Nb_3, RhGe, Ce, $TlNiSe_2$, $SrIr_2$	**Total: 92** $LaPd_2Al_3$, Ru_2Zr, In_3Sn, La_3Sn, $PdZr_2$
	No	**Total: 95** $YbLuB_{24}$, Ti_3Ir*, $CePb_3$*, $SrAu_5$, $SrGe_3$*	**Total: 917** Sc_3Nb, Yb_5Sn_3, $Ba(SmSe_2)_2$, $YAsO_4$, Th_2Zn

*** Materials that are actual superconductors, but were not contained in the dataset.**



Model Evaluation

Predicting Empirical T_c Distributions



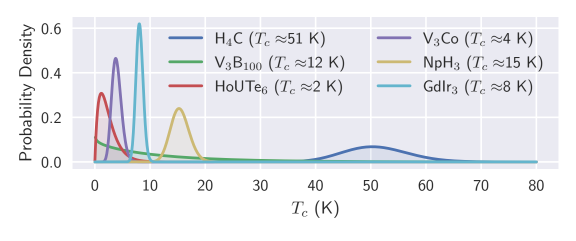
Identified Superconductors

- Using the model, we screened over 40,000 metals, oxides, metallic compounds, and hydrides from the Materials Project database [JOH+13].
- We identified over 600 candidate superconductors not contained in our dataset:

Materials Project ID	Formula	Stable	Experimentally Observed	Predicted Mean T_c	Closest Superconductor in Dataset
mp-672238	$CeCuSb_2$	Yes	Yes	1.88 K	Cu_2Sb
mp-1025564	$LuAl_2Pd_5$	Yes	No	8.12 K	Pd
mp-10898	$ScAlNi_2$	Yes	Yes	1.25 K	Ni_3Al
mp-573601	Th_7Ru_3	Yes	Yes	0.95 K	Th
mp-28280	$K_5V_3O_{10}$	Yes	Yes	4.93 K	KO_3
mp-1224184	$HfZrB_4$	Yes	No	2.57 K	HfB_2
mp-1228895	$AlGaSb_2$	No	No	4.15 K	AlSb
mp-1222266	Lu_3S_4	Yes	No	3.63 K	LuS
mp-1079796	Ti_3Pd	Yes	No	4.24 K	Ti
mp-1218331	Sr_3CaSi_8	No	No	1.88 K	$Sr(Si)_2$
mp-1021328	H_4C	Yes	No	50.97 K	H_2
mp-11494	$LuPb_3$	No	Yes	3.80 K	Pb
mp-1226890	Ce_4H_{11}	Yes	No	331.83 K	CeH_9
mp-22266	GdB_6	Yes	Yes	2.22 K	B
mp-1184695	Ho_3Er	No	No	6.22 K	Ho
...	...	...	...	...	...

Predicting T_c of Candidates

- Using the model trained on the T_c of known superconductors, we estimated the empirical T_c distribution of all predicted candidates.



- The T_c of identified candidates range from 0 to 335 K.
- The non-hydride, non-cuprate superconductors range from 0 to approximately 50 K.
- Several of the candidates contain magnetic rare-earth elements.

Ongoing Work

- Currently, we are collaborating with Dr. Julia Y. Chan and her lab at Baylor University to synthesize and measure the properties of the most promising intermetallic and oxide candidates.
- So far, we have confirmed two of the model's predictions:
    1. La_2Sn_3 (Observed T_c: 2.5 K, Predicted: 2.8 K)
    2. $Lu_3Ir_4Ge_{13}$ (Observed T_c: 2.8 K, Predicted: 1.8 K)
- However, some of the proposed candidates have proved difficult to synthesize:
    - CeBi (oxidizes very rapidly)
    - CeTe (flammable and toxic)
- More candidates are under development.

Conclusion

Conclusion

- We trained an atomic structure-based graph neural network model (ALIGNN) to identify and predict superconductor T_c with a standard deviation error of 6.66 K ($2.20K$ for low-T_c).
- After screening the Materials Project database, we found more than 600 candidate superconductors.

Ongoing and Future Work

- We are currently collaborating with an inorganic chemistry lab at Baylor University to synthesize some of the more promising candidates.
- Additional work needs to be done incorporating electronic features (e.g. band structure) and incorporating structural features (e.g. substrates, film thickness) into the model.

Acknowledgments

- The author would like to extend special thanks to:
  - Dr. E. P. Blair
  - Dr. Julia Y. Chan (and her students)

Advisor

Collaborators

- The IEEE Quantum Week 2023 chairs, organizers, presenters, and attendees for making this an amazing conference experience.

Questions

Superconductor Candidates List

https://cburdine.github.io/files/qce23.html

GitHub repository: https://github.com/cburdine/sc-screening

For more detailed questions, email: `colin_burdine1@baylor.edu`

References i

- D. Aoki, J.-P. Brison, J. Flouquet, K. Ishida, G. Knebel, Y. Tokunaga, and Y. Yanase, *Unconventional superconductivity in UTe2*, J. Phys.: Condens. Matter **34** (2022), no. 24, 243002 (en).
- J. Bardeen, L. N. Cooper, and J. R. Schrieffer, *Theory of Superconductivity*, Phys. Rev. **108** (1957), no. 5, 1175–1204.
- Kamal Choudhary and Brian DeCost, *Atomistic Line Graph Neural Network for improved materials property predictions*, npj Comput Mater **7** (2021), no. 1, 1–8 (en).
- Kamal Choudhary and Kevin Garrity, *Designing high-TC superconductors with BCS-inspired screening, density functional theory, and deep-learning*, npj Comput Mater **8** (2022), no. 1, 1–12 (en).

References ii

- Leon N. Cooper, *Theory of Superconductivity*, American Journal of Physics **28** (1960), no. 2, 91–101.
- Murray S. Daw, Stephen M. Foiles, and Michael I. Baskes, *The embedded-atom method: a review of theory and applications*, Materials Science Reports **9** (1993), no. 7, 251–310 (en).
- Alexander Dunn, Qi Wang, Alex Ganose, Daniel Dopp, and Anubhav Jain, *Benchmarking materials property prediction methods: the Matbench test set and Automatminer reference algorithm*, npj Comput Mater **6** (2020), no. 1, 1–10 (en).
- Luca Foppiano, Pedro Baptista de Castro, Pedro Ortiz Suarez, Kensei Terashima, Yoshihiko Takano, and Masashi Ishii, *Automatic extraction of materials and properties from superconductors scientific literature*, Science and Technology of Advanced Materials: Methods **3** (2023), no. 1, 2153633.

- Lin Jiao, Sean Howard, Sheng Ran, Zhenyu Wang, Jorge Olivares Rodriguez, Manfred Sigrist, Ziqiang Wang, Nicholas P. Butch, and Vidya Madhavan, *Chiral superconductivity in heavy-fermion metal UTe2*, Nature **579** (2020), no. 7800, 523–527 (en).
- Anubhav Jain, Shyue Ping Ong, Geoffroy Hautier, Wei Chen, William Davidson Richards, Stephen Dacek, Shreyas Cholia, Dan Gunter, David Skinner, Gerbrand Ceder, and Kristin A. Persson, *Commentary: The Materials Project: A materials genome approach to accelerating materials innovation*, APL Materials **1** (2013), no. 1, 011002.
- Tomohiko Konno, Hodaka Kurokawa, Fuyuki Nabeshima, Yuki Sakishita, Ryo Ogawa, Iwao Hosako, and Atsutaka Maeda, *Deep learning model for finding new superconductors*, Phys. Rev. B **103** (2021), no. 1, 014509.

- W. L. McMillan, *Transition temperature of strong-coupled superconductors*, Physical Review **167** (1968), no. 2, 331–344 (en).
- Valentin Stanev, Corey Oses, A. Gilad Kusne, Efrain Rodriguez, Johnpierre Paglione, Stefano Curtarolo, and Ichiro Takeuchi, *Machine learning modeling of superconducting critical temperature*, npj Comput Mater **4** (2018), no. 1, 1–14 (en).
- C. Vijayakumar, M. Brightson, S.L. Rayar, and P. Selvarajan, *Development of yba2cu3o7-d superconducting thick film on ba2 smsbo6 substrate material synthesised by combustion technique*, Journal of Experimental Nanoscience **9** (2014), no. 5, 525–532 (en).
- Tian Xie and Jeffrey C. Grossman, *Crystal Graph Convolutional Neural Networks for an Accurate and Interpretable Prediction of Material Properties*, Phys. Rev. Lett. **120** (2018), no. 14, 145301.

S. R. Xie, Y. Quan, A. C. Hire, B. Deng, J. M. DeStefano, I. Salinas, U. S. Shah, L. Fanfarillo, J. Lim, J. Kim, G. R. Stewart, J. J. Hamlin, P. J. Hirschfeld, and R. G. Hennig, *Machine learning of superconducting critical temperature from Eliashberg theory*, npj Comput Mater **8** (2022), no. 1, 1–8 (en).

Xingjiang Zhou, Wei-Sheng Lee, Masatoshi Imada, Nandini Trivedi, Philip Phillips, Hae-Young Kee, Päivi Törmä, and Mikhail Eremets, *High-temperature superconductivity*, Nat Rev Phys **3** (2021), no. 7, 462–465 (en).