Topical Review

The 2021 room-temperature superconductivity roadmap

Boeri Lilia[1,29,*], Richard Hennig[2,29,*], Peter Hirschfeld[3,29,*],
Gianni Profeta[4,29,*], Antonio Sanna[5,29,*], Eva Zurek[6,29,*],
Warren E Pickett[7], Maximilian Amsler[8,9], Ranga Dias[10],
Mikhail I Eremets[11], Christoph Heil[12], Russell J Hemley[13],
Hanyu Liu[14], Yanming Ma[14], Carlo Pierleoni[3],
Aleksey N Kolmogorov[15], Nikita Rybin[15], Dmitry Novoselov[17],
Vladimir Anisimov[17], Artem R Oganov[16], Chris J Pickard[18],
Tiange Bi[6], Ryotaro Arita[19,20], Ion Errea[21], Camilla Pellegrini[22],
Ryan Requist[5,23], E K U Gross[5,23], Elena Roxana Margine[15],
Stephen R Xie[3], Yundi Quan[3], Ajinkya Hire[3], Laura Fanfarillo[3,24],
G R Stewart[3], J J Hamlin[3], Valentin Stanev[25], Renato S Gonnelli[26],
Erik Piatti[26], Davide Romanin[27], Dario Daghero[26] and
Roser Valenti[28]

[1] Physics Department, Sapienza University and Enrico Fermi Research Center, Rome, Italy
[2] Deparment of Material Science and Engineering and Quantum Theory Project, University of Florida, Gainesville 32611, United States of America
[3] Department of Physics, University of Florida, Gainesville, FL 32611, United States of America
[4] University of L'Aquila, Italy
[5] Max Planck Institute of Microstructure Physics, Halle, Germany
[6] University at Buffalo, SUNY, United States of America
[7] University of California Davis, United States of America
[8] Department of Chemistry and Biochemistry, University of Bern, Freiestrasse 3, CH-3012 Bern, Switzerland
[9] Department of Materials Science and Engineering, Cornell University, Ithaca, NY 14853, United States of America
[10] University of Rochester, United States of America
[11] Max Planck Institute for Chemistry, Mainz, Germany
[12] Graz University of Technology, Austria
[13] University of Illinois at Chicago, United States of America
[14] Jilin University, People's Republic of China
[15] Binghamton University, United States of America
[16] Skolkovo Institute of Science and Technology, Russia
[17] Ural Federal University, Yekaterinburg, Russia
[18] Cambridge University, United Kingdom
[19] University of Tokyo, Japan
[20] RIKEN, Japan
[21] University of the Basque Country, Spain
[22] University of Perugia, Italy
[23] Hebrew University of Jerusalem, Israel
[24] Scuola Internazionale Superiore di Studi Avanzati (SISSA), Via Bonomea 265, 34136 Trieste, Italy
[25] University of Maryland, United States of America

[29] Guest editors of the roadmap.
* Author to whom any correspondence should be addressed.

[26] Politecnico di Torino, Italy
[27] Sorbonne Université, France
[28] Goethe University Frankfurt, Germany

E-mail: lilia.boeri@uniroma1.it, rhennig@mse.ufl.edu, pjh@phys.ufl.edu, gianni.profeta@aquila.infn.it, sanna@mpi-halle.mpg.de and ezurek@buffalo.edu

Abstract

Designing materials with advanced functionalities is the main focus of contemporary solid-state physics and chemistry. Research efforts worldwide are funneled into a few high-end goals, one of the oldest, and most fascinating of which is the search for an ambient temperature superconductor (A-SC). The reason is clear: superconductivity at ambient conditions implies being able to handle, measure and access a single, coherent, macroscopic quantum mechanical state without the limitations associated with cryogenics and pressurization. This would not only open exciting avenues for fundamental research, but also pave the road for a wide range of technological applications, affecting strategic areas such as energy conservation and climate change. In this roadmap we have collected contributions from many of the main actors working on superconductivity, and asked them to share their personal viewpoint on the field. The hope is that this article will serve not only as an instantaneous picture of the status of research, but also as a true roadmap defining the main long-term theoretical and experimental challenges that lie ahead. Interestingly, although the current research in superconductor design is dominated by conventional (phonon-mediated) superconductors, there seems to be a widespread consensus that achieving A-SC may require different pairing mechanisms.

In memoriam, to Neil Ashcroft, who inspired us all.

Keywords: superconductor, superconductivity, hydrides, electron–phonon interaction, , crystal structure prediction, novel superconductors

(Some figures may appear in colour only in the online journal)
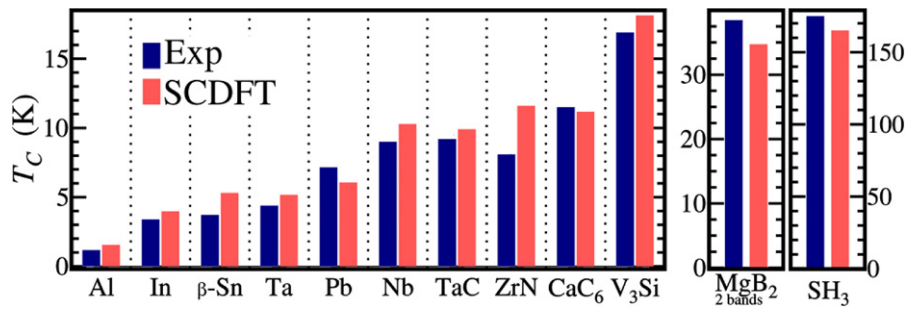
Contents

Figure 23. SCDFT predicted critical temperatures for a set of conventional electron phonon superconductors and comparison with experimental values. Details can be found in reference [142].

high-T_c structures thermodynamically stable at much lower pressures. Therefore, if we were able to perform crystal structure searches in the quantum energy landscape, we would probably find new high-T_c compounds at much lower pressures and put experimental colleagues on the right track for finally reaching high-temperature and ambient pressure superconductivity.

Acknowledgments

16. Extending the predictive power of DFT for superconductors: toward nonadiabatic electron–phonon coupling

Camilla Pellegrini[1], *Antonio Sanna*[2], *Ryan Requist*[2] and *E K U Gross*[2]

[1]Hebrew University of Jerusalem, Israel
[2]Max'Planck of Microstructure Physics, Halle Germany

16.1. Status

Over the past 30 years DFT has enjoyed enormous popularity as an accurate yet computationally feasible method for the *ab initio* calculation of normal-state electronic properties. The discovery of high-temperature superconductors motivated an extension of DFT to superconductivity in order to predict properties such as the superconducting critical temperature (T_c) and the gap at zero temperature [140]. Within SCDFT, a superconducting material is fully described in terms of two quantities: the ordinary electron density and the superconducting order parameter. This additional density is determined by solving a BCS-like gap equation, where the screened Coulomb repulsion and the phonon-mediated attraction between the electrons are incorporated via the xc potentials, which are universal functionals of the two densities. Compared to traditional many-body Eliashberg theory, SCDFT yields two major advantages: the numerical simplicity of the gap equation, and the absence of any empirical parameter like the Coulomb pseudopotential.

The basic Hohenberg–Kohn and Kohn–Sham theorems of SCDFT were proved by Oliveira, Gross and Kohn in

the late 1980s. A first approximation to the xc functionals was derived on the basis of many-body perturbation theory by employing the optimized effective potential method in the exchange-only approximation [141]. However, in addition to neglecting vertex corrections, as justified by Migdal's theorem, the phononic part of the functional also neglected the dressing of the electronic Green's function, which made it of questionable accuracy for treating phonon-mediated superconductivity. Recently, a new functional that retains the accuracy of Migdal's approximation has been derived from the parametrization of the Eliashberg self-energy for a simple Einstein phonon spectrum [142]. This functional has enabled accurate density functional calculations of experimental superconducting gaps without resorting to expensive GW calculations. For all the materials tested so far, the T_c's and gaps predicted by the new functional turn out to be in excellent agreement with the experimental data, while the numerical effort is significantly lower than solving the full Eliashberg equations (figure 23).

16.2. Current and future challenges

SCDFT is presently a powerful tool, but its predictive power is limited to conventional phonon-driven superconductors. Although this proves sufficient to describe room temperature superconductivity in the high pressure regime, at atmospheric pressure pairing mechanisms of purely electronic origin are believed to be essential for achieving high T_c ($\gg 100$ K).

One mechanism of utmost importance is the spin-fluctuation mechanism, which is commonly assumed to be responsible for high-T_c superconductivity in Fe and Cu based superconductors [143, 144]. Electronic charge fluctuations, including plasmons [145] and orbital fluctuations [144], may also favor superconductivity by providing additional contributions to the attractive pairing. Along these lines there has already been some progress in the construction of a plasmonic xc kernel [145] and a preliminary functional approximation including spin-fluctuations [143].

Even though, despite many years of systematic theoretical search, no phononic superconductor with $T_c > 39$ K has been found, there might be situations in which the electron–phonon coupling leads to superconductivity beyond the predictive capability of conventional theory. One aspect that has been long overlooked, especially in SCDFT, is the role of nonadiabatic effects in the dynamics of electrons and
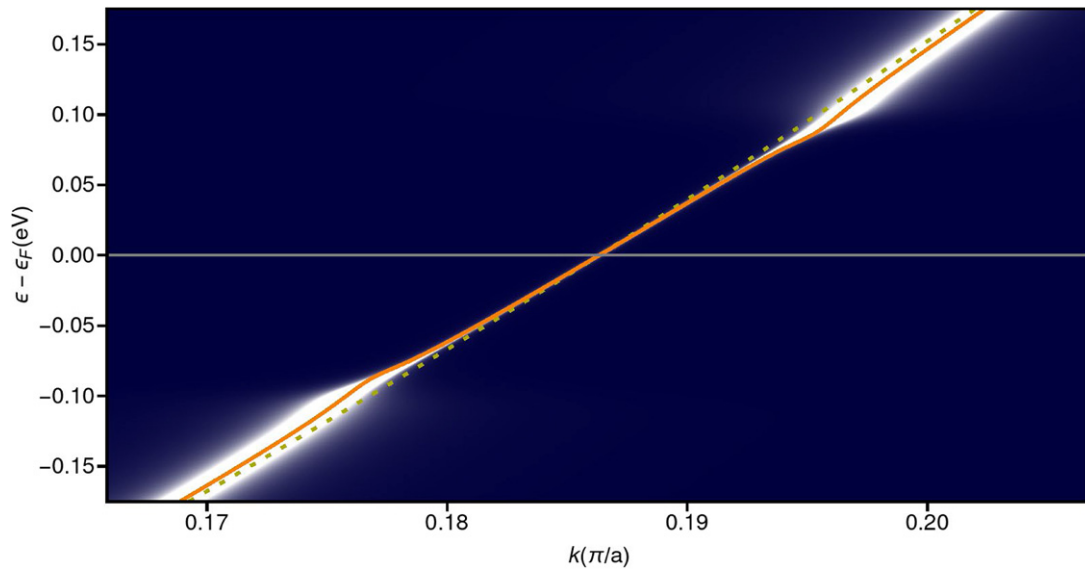
Figure 24. The Kohn–Sham band structure (orange) in exact factorization-based DFT captures the phonon-induced wiggle in the electronic spectral function (background blue-white color scale) of the Fröhlich model with a parabolic electronic band (dashed yellow) linearly coupled to a single Einstein phonon with energy 0.1 eV.

phonons. There is indeed experimental evidence that a number of phononic superconductors are not described by the conventional many-body theory of superconductivity based on Migdal's theorem, which breaks down when the energies of electrons and phonons are comparable. In this regard, a generalization of the functional construction scheme [142] within SCDFT, by effectively including vertex corrections to the electron–phonon self-energy, seems to be particularly appealing. Evaluating these higher order contributions in a direct many-body approach is in fact extremely costly due to cumbersome Matsubara frequency summations. On the other hand, in SCDFT these sums can be integrated analytically by following a procedure similar to that used in reference [142].

However, this approach would only involve the electronic degrees of freedom and is expected to increase the superconducting T_c for low Fermi velocity [146]. Nevertheless, nonadiabatic corrections could also significantly affect the electron phonon coupling strength and the lattice dynamics, either stabilizing or suppressing the superconducting phase, depending on the material properties. A way to systematically include nonadiabatic electron–phonon effects would be to use a DFT/many-body formalism which does not depend on the Born–Oppenheimer (BO) approximation.

16.3. Advances in science and technology to meet challenges

Standard DFT relies, by its very nature, on the BO approximation: the ground-state density is calculated from the electrostatic potential of clamped nuclei. Since first-principles calculations of electron–phonon coupling (used, e.g., in SCDFT and Eliashberg theory [147]) start from DFT calculations of the electronic structure, the BO approximation is implicitly assumed. Recently, a generalized DFT framework for the treatment of electrons and phonons has been developed [148] starting from the exact factorization of the electron–nuclear wave

function into electronic and nuclear components [149]. The basic electronic variable is the exact (rather than BO) electronic density, which depends parametrically on the set of nuclear coordinates. The equations to be solved are generalized KS equations with a nonadiabatic Hartree xc potential and a nuclear Schrödinger equation with a beyond-BO PES. Exact phonons are defined by the harmonic expansion of the latter equation. Results for the Fröhlich model show that already the simplest approximation, in which nonadiabatic contributions to the PES are added to standard BO-DFT functionals like LDA or GGA, can reproduce the leading-order phonon-induced band structure renormalization, including the renormalization of the Fermi velocity (see figure 24).

Extending exact factorization-based DFT to superconductivity would establish a beyond-BO SCDFT which would allow for (i) a seamless description of electron–phonon effects in the normal state and (ii) the inclusion of nonadiabatic effects on the superconducting side. The latter effects might be relevant for systems like molecular superconductors or doped insulators such as Cs_3C_{60}, BaK_xBiO_3 and BEDT-TTF.

An alternative way of capturing non-adiabaticity in the superconducting state would be to set up a connection to the recent many-body Green's function theory beyond the BO approximation proposed in reference [150]. A first SCDFT nonadiabatic functional could be derived starting with the Eliashberg (GW) self-energy diagram, where standard Green's functions are replaced by nonadiabatic propagators [150], and then following a similar procedure as in reference [142].

16.4. Concluding remarks

SCDFT has attained a level of maturity where it allows for accurate and computationally efficient calculations of T_c's and gap functions of phononic superconductors. However, superconductors displaying strong nonadiabatic effects, as well as
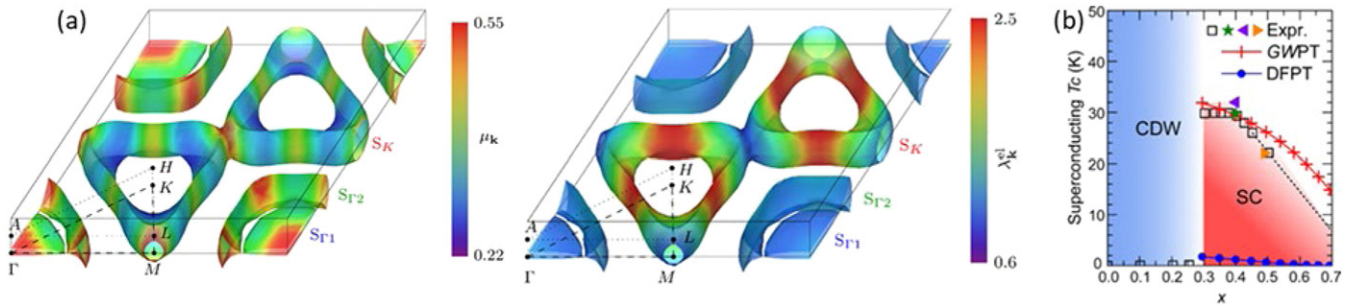
Figure 25. Momentum resolved screened Coulomb interaction and e–ph interaction strength on the FS of 2H-NbS$_2$. Reprinted figure with permission from [155], Copyright (2017) by the American Physical Society. (b) Superconducting phase diagram of Ba$_{1-x}$K$_x$BiO$_3$. Calculated T_c with $\mu^* = 0.16$ are plotted as red crosses for GWPT and blue dots for DFPT. Experimental data are plotted as squares, star, left-pointing triangle, and right-pointing triangle. Reprinted figure with permission from [158], Copyright (2019) by the American Physical Society.

unconventional pairing, are presently out of reach. In our opinion, these materials are the most promising candidates for achieving high-temperature superconductivity at ambient pressure as they belong to a largely unexplored basin of search. Future developments of SCDFT should thus aim at their description. With regard to nonadiabatic effects we propose that SCDFT, as well as the conventional theory of superconductivity, may benefit from a change in paradigm where the starting point is a unified treatment of electrons and nuclei based on the exact-factorization approach.

Acknowledgments

17. Modeling superconducting properties with the Eliashberg formalism

Elena Roxana Margine

Binghamton University-SUNY, United States of America

17.1. Status

Advances in electronic structure methods alongside developments in software packages and computing hardware have made it possible to model a host of complex materials properties from first principles. In particular, the research in conventional superconductivity has come a long way from making rough estimates of the critical temperature (T_c) to probing pairing mechanisms and resolving superconducting energy gaps. Nevertheless, fully *ab initio* prediction of superconducting properties remains challenging, and the vast majority of investigations rely on the semi-empirical McMillan or Allen–Dynes formulas for estimations of the critical temperature.

A major part of the success achieved in the characterization of the superconducting state in materials with conventional pairing can be attributed to the development of the

many-body Eliashberg theory [151], a generalization of the Bardeen–Cooper–Schrieffer microscopic theory of superconductivity. This state-of-the-art theoretical method rests on the Green's function formalism and gives direct access to the frequency-dependent superconducting energy gap as a solution to a set of self-consistent coupled diagrammatic equations for the electronic and phononic propagators. The approach has been particularly useful for solving Eliashberg equations in the anisotropic, i.e. band and crystal momentum-dependent, form. With the capability of naturally resolving multiple gaps on the FS, it has been used to analyze the two-gap structure in MgB$_2$ [151] and other intriguing superconductors.

The accurate anisotropic description is achievable only with extremely dense electron and phonon meshes to properly sample the electron–phonon (e–ph) scattering processes in the vicinity of the FS. The development of a first-principles interpolation technique based on Wannier functions was instrumental for overcoming this computational challenge [152]. The approach takes advantage of the spatial localization of the e–ph matrix elements in the Wannier basis and enables their efficient interpolation from a given uniform coarse grid to a desired arbitrary dense Brillouin zone (BZ) grid at a significantly reduced computational cost. So far, the only open-source implementation of the anisotropic Eliashberg formalism with Wannier-based interpolation is available in the EPW code [153] (https://epw-code.org/) distributed as part of the Quantum ESPRESSO package (https://github.com/QEF/q-e/releases). A numerical procedure based on symmetric Helmholtz Fermi-surface harmonics (HFSH) has also been proposed as a robust approach to treat the anisotropy of the e–ph interaction in the Eliashberg equations [154]. The basic idea is to replace BZ integrals in the k space with inexpensive sums over a few HFSH coefficients.

While these advances have enabled accurate modeling of the e–ph anisotropy from first principles, the Coulomb correlations effects in the Eliashberg equations are still treated predominantly with a single empirical parameter μ^* that acts only near the Fermi level. A description of the e–ph and electron–electron (e–e) interactions on an equal footing will