Journal of the Physical Society of Japan **87**, 041012 (2018)
https://doi.org/10.7566/JPSJ.87.041012

Ab initio Eliashberg Theory: Making Genuine Predictions of Superconducting Features

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(Received April 20, 2017; accepted July 6, 2017; published online February 14, 2018)

We present an application of Eliashberg theory of superconductivity to study a set of novel superconducting systems with a wide range of structural and chemical properties. The set includes three intercalated group-IV honeycomb layered structures, SH_3 at 200 GPa (the superconductor with the highest measured critical temperature), the similar system SeH_3 at 150 GPa, and a lithium doped mono-layer of black phosphorus. The theoretical approach we adopt is a recently developed, fully ab initio Eliashberg approach that takes into account the Coulomb interaction in a full energy-resolved fashion avoiding any free parameters like μ^*. This method provides reasonable estimations of superconducting properties, including T_C and the excitation spectra of superconductors.

1. Introduction

Prediction of functional materials is a fast-growing field within physics, chemistry and material science. It has lead to significant predictions for semiconductors,[1] magnetic materials,[2,3] thermoelectrics,[4,5] batteries[6–8] and energy storage materials,[9] to name but a few.

This is now possible because of decades of intense theoretical development. Improvement in Kohn–Sham density-functional theory (DFT)[10–14] and its computational implementations,[15–23] alongside the development of material design methods such as evolutionary algorithms,[24] particle swarms,[25] random sampling[26] and hopping based algorithms,[27,28] that allows for an unbiased and efficient exploration[27,28] of the potential energy surface.

A striking success of these methods was the recent prediction of superconductivity in SH_3 at high pressure[29] that has since received a full experimental confirmation.[30]

Until a few years ago, it was believed that predicting novel superconductors and superconducting critical temperatures (T_C) would be impossible, due to the complexity of the superconducting state. Theoretical methods were used mostly to characterize known superconductors and to improve on their experimental understanding.[31] This changed in the last decade, where many predictions of superconductors and their properties have been made[29,32–40] and several have been confirmed experimentally. Among these are important cases such as superconductivity of graphene[36] and SH_3.

So far, ab initio superconductivity prediction are limited to phonon-driven superconductors, while important classes such as cuprates and iron based superconductors, are still out of reach due to the lack understanding of the microscopic pairing mechanism[41] and treatment[42] of the Coulomb interactions. In this respect, Superconducting Density Functional Theory[43] (SCDFT), has been the leading theory which includes Coulomb interactions solely by first principles.[44–48] While Eliashberg theory,[31,49–52] which is a Green's function method, has never been employed with a complete treatment of Coulomb forces for real materials.[31,53]

We have recently formulated[54] an extension of the Eliashberg approach to include full scale Coulomb interactions from first principles. This theory is reviewed in Sect. 2. Here we will use it to study superconductivity for a set of recently predicted superconductors (Sect. 3). Although all the materials in the set are exclusively phonon driven, our approach includes Coulomb forces without the use of any empirical parameter such as μ^* and perform genuine ab initio predictions. Our method is formally similar to the recent work of Sano and coworkers,[55] with the difference that we solve the full Eliashberg set instead of its linearised form. Also in this work we will not consider the effect of Fermi level shifts.

2. Eliashberg Theory of Superconductivity with a Full Static Coulomb Interaction

Eliashberg theory of superconductivity[49,50,56,57] is one of the most powerful and versatile approaches to superconductivity. It is derived from a Green's function based perturbative approach that starts from a Kohn–Sham system and assumes an approximation for the self energy in a GW like form. Here the interaction W is the sum of a fully dressed phononic propagator and a screened Coulomb interaction.[56] However for computational convenience the Coulomb part is usually simplified by introducing an adjustable Coulomb strength parameter,[50,58] called μ^*. This is de rigueur even for the most recent and advanced implementations and applications.[51,59–62] This approximation allows significant savings in the computational cost of the method since the energy integration (integration over the Kohn–Sham states) can be reduced to a narrow energy window near the Fermi level. Nevertheless, this approximation remains extremely successful because in the McMillan parameterization of the Eliashberg approach[56,63,64] experience shows[31] that the μ^* required to fit the experimental T_C has a narrow range of values (normally between 0.1 and 0.15).

Despite this apparent universality, such an approximation makes the Eliashberg approach semi-empirical, preventing true ab initio investigations of all the physics related to the Coulomb interaction, among which are isotope coefficients and strong plasmonic effects.[46,47,65–67]

Moreover, the success of μ^* is limited to the McMillan formula. In the full solution of the Eliashberg equations, the

value of μ^* depends strongly on the chosen energy cutoffs that, in turn, are bound to the phonon energy scale. This makes the choice of μ^* far less trivial. If the dos is strongly varying close to the Fermi level the assumption of constant μ^* can not be accepted, recently this was investigated by using and extended Eliashberg theory in which the dos variation was considered while however not accounting for the momentum dependence of the dielectric screening an investigation in which the effect of the dos while keeping.

Anisotropic or multiband superconductors, such as MgB_2 where Coulomb interactions are strongly band dependent,[68,69] are also cases for which the μ^* rule is inadequate. Probably the most important case where the Coulomb renormalization has to be included from first principles is for *unconventional* superconductivity, where the phase of the order parameter changes on the Fermi surface.[41,70,71] This renormalization cannot follow the Morel–Anderson theory[50,58] that is limited to order parameters with the full crystal symmetry. In fact, SCDFT calculations including full Coulomb interactions demonstrate a quite intricate interplay between phonons, spin fluctuations and screened Coulomb repulsion,[48,72] and there is compelling evidence that a low-energy effective theory, like conventional Eliashberg, can not fully explain the phenomenology of unconventional superconductors and particularly their multigap structure.[72–74]

Motivated by these facts, we have recently developed[54] a more general Eliashberg approach that, within an isotropic approximation in momentum space, allows the Eliashberg equations to be solved including the full Coulombic interaction computed from first principles. So far this kind of approach has never been performed on new materials but only tested on classic superconductors which are already well understood.

In its final form this fully ab initio Eliashberg approach consists in the solution to the following set of coupled self-consistent equations:

$$Z_n(\xi) = 1 - \frac{T}{\omega_n}\int d\xi'\, N(\xi') \sum_{\omega_{n'}} V^{ph}_{n-n'} \frac{Z_{n'}(\xi')\omega_{n'}}{\Theta_{n'}(\xi')},$$

$$\phi_n(\xi) = -T\int d\xi'\, N(\xi') \sum_{\omega_{n'}} [V^{ph}_{n-n'} + V^c_0(\xi\xi')] \frac{\phi_{n'}(\xi')}{\Theta_{n'}(\xi')},$$

$$\chi_n(\xi) = \frac{T}{\omega_n}\int d\xi'\, N(\xi') \sum_{\omega_{n'}} V^{ph}_{n-n'} \frac{\chi_{n'}(\xi') + \xi'}{\Theta_{n'}(\xi')},$$

$$\Theta_n(\xi) = [Z_n(\xi)\omega_n]^2 + \xi^2 + \phi_n^2(\xi), \tag{1}$$

where Z is the mass renormalization function responsible for all the polaronic physics,[52,75] ϕ is the gap function (the superconducting gap being $\Delta = \phi/Z$), $N(\xi)$ is the density of states (DOS), T the temperature, ω_n are Fermionic Matsubara energies and χ a Fermi energy shift that, since one should preserve the particle number,[60] is negligible in the isotropic approximation. This set of equations is solved on the imaginary frequency axis. To obtain the physical gap function, we perform an analytic continuation to the real axis which is done by means of the Padé's approximants.[52,76]

The interactions are given by a phononic contribution: $V^{ph}_{n-n'}$ and a Coulomb contribution $V^c_0(\xi\xi')$. $V^{ph}_{n-n'}$ is well-understood in Eliashberg theory, and is related to the Eliashberg spectral function, $\alpha^2F(\omega)$, in the following way:

$$V^{ph}_n = -\frac{1}{N_F}\int d\omega\, \frac{2\omega\alpha^2F(\omega)}{\nu_n^2 + \omega^2}, \tag{2}$$

where N_F is the DOS at the Fermi level $[N(\xi = 0)]$ and ν_n is a bosonic Matsubara frequency. Moments of the $\alpha^2F(\omega)$ function are of particular physical importance because they are used in the McMillan–Allen–Dynes formula,[64]

$$\lambda = 2\int \frac{\alpha^2F(\omega)}{\omega}\, d\omega, \tag{3}$$

$$\omega_{\log} = \exp\left[\frac{2}{\lambda}\int \alpha^2F(\omega)\frac{\ln(\omega)}{\omega}\, d\omega\right],$$

$$\omega_2 = \frac{2}{\lambda}\int \alpha^2F(\omega)\omega\, d\omega,$$

where λ is the usual electron–phonon coupling strength. While $\omega_{\log}$ and ω_2 are two characteristic phonon energies that express the scale at which the phonon pairing is most effective.

The $V^c_0(\xi\xi')$ term is the novelty in our the approach. This type of static interaction has been almost exclusively used in SCDFT[43–45,77] and proven to be quite accurate, allowing calculation of T_C and other superconducting properties for broad classes of superconductors[68,78–82] and making significant predictions.[83–89] This term is defined as an iso-energy surface average of the screened Coulomb matrix elements:

$$V^c_{kk',n} = \frac{4\pi}{\Omega}\sum_{\mathbf{GG}'}\sum_{\mathbf{q}} \epsilon^{-1}_{\mathbf{GG}'}(\mathbf{q}, i\nu_n) \times \frac{\rho_{kk'}(\mathbf{qG})\rho^*_{kk'}(\mathbf{qG}')}{|\mathbf{q}+\mathbf{G}||\mathbf{q}+\mathbf{G}'|}, \tag{4}$$

where Ω is the unit cell volume, k is a joint index representing the momentum and the band index of the Kohn–Sham Bloch orbitals, $\mathbf{G}$ are reciprocal lattice vectors, $\mathbf{q}$ are vectors within the first reciprocal unit cell, $\rho_{kk'}(\mathbf{qG}) = \langle k|e^{i(\mathbf{q}+\mathbf{G})\mathbf{r}}|k'\rangle$ is the polarization matrix and $\epsilon^{-1}_{\mathbf{GG}'}(\mathbf{q}, i\nu_n)$ is the inverse dielectric function that, in this work, is evaluated within the random phase approximation (RPA). The iso-energy average is performed according to the following definition:

$$V^c_0(\xi\xi') := \sum_{k,k'} V^c_{kk',0}\frac{\delta(\xi - \xi_k)}{N(\xi)}\frac{\delta(\xi' - \xi_{k'})}{N(\xi')} \tag{5}$$

similar to the definitions adopted[69,82,90] in SCDFT.

In multiband or strongly anisotropic superconductors one may require the full anisotropic Coulomb coupling $V^c_{kk',0}$ and the fully k-resolved electron–phonon matrix elements. In this work, however, we will only consider materials that are sufficiently well described within the isotropic approximation.

3. Superconducting Properties of Selected Set of Novel Superconductors

The set of materials chosen for this study have recently been proposed using theoretical methods and expected to be useful. We will analyze their superconducting properties by means of our *full* Eliashberg approach. In the present section we review their structural properties and discuss their coupling with particular attention to the Coulomb coupling that is the novelty introduced in our approach as compared to conventional Eliashberg theory. Among these selected materials only SH_3 has been synthesized and confirmed experimentally.[30]

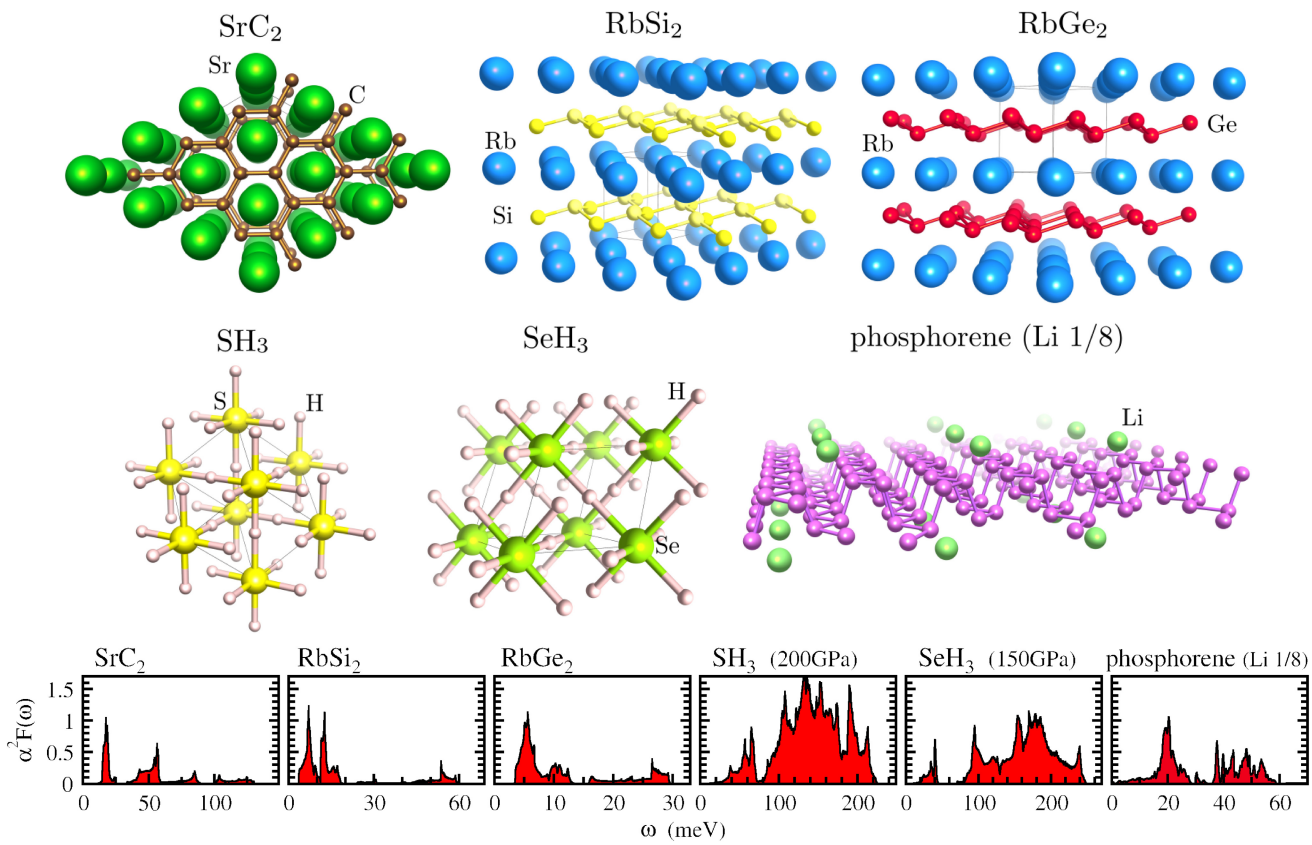

Fig. 1. (Color online) Crystal structure representation of the different materials studied in this work. Electron–phonon spectral functions (bottom panels) for the set of systems under investigation.

Table I. Couplings and predicted critical temperatures. λ, $\omega_{\log}$, and ω_2 are characteristic integrals over the electron phonon coupling defined by Eq. (3). μ is the Coulomb coupling at the Fermi level, defined as $V_0^c(0,0)$ [Eq. (5)] times N_F. μ^*(fit) is the value of the μ^* parameter that used in combination with the McMillan–Allen–Dynes formula[56,63,64] leads to the full Eliashberg T_C, presented here. $2\Delta/\mathrm{kb}T_C$ is the ratio between gap at $T = 0$ and $\omega = 0$ with the T_C. For BCS[114] superconductors this value should be 3.5.

	Phononic			Electronic			T_C (K)	$2\Delta/\mathrm{kb}T_C$
	λ	$\omega_{\log}$ (meV)	ω_2 (meV)	μ	N_F (st./eV/cell/spin)	μ^* (fit)		
SrC_2	0.68	26.5	40.8	0.16	0.40	0.11	9.45	3.64
$RbSi_2$	1.26	8.7	14.1	0.19	0.53	0.11	10.0	4.16
$RbGe_2$	1.12	6.6	9.4	0.12	0.67	0.09	7.4	4.08
SH_3	2.28	109.5	130.9	0.26	0.3	0.125	226.9	4.79
SeH_3	1.38	107.2	142.6	0.23	0.26	0.12	136.1	4.12
phos. (Li 1/8)	1.08	19.3	27.9	0.37	3.01	0.11	14.5	3.87

3.1 Intercalated group-IV honeycomb structures

The first set of systems studied here are the intermetallic layered materials (structurally similar to MgB_2) namely SrC_2, $RbGe_2$, and $RbSi_2$ discussed in Ref. 85. SrC_2 is an intercalated graphite corresponding to the maximum intercalation limit achievable in layered graphite (carbon layers sp^2). $RbGe_2$ and $RbSi_2$ show a buckled honeycomb of the Si and Ge plane, due to the sp^3 bonding. Their electron–phonon coupling strength is weak to moderate, with relatively low phononic energies (see Fig. 1 and Table I). These systems are of particular interest for this work, because from an electronic point of view, they are similar to doped insulators with covalent states crossing the Fermi level. It is evident from the panels in Fig. 2, that these materials feature structured screened matrix elements. There is a relatively weak interaction between deep valence states and states at the Fermi level. The Coulomb function in Eq. (5) therefore has an almost block-diagonal structure. Meaning that states far from the Fermi level (especially in the valence energy window) will be weakly coupled with those at the Fermi level thus have little contribution to the Coulomb renormalization[50,58] mechanism. The electron screening for these systems is high and the matrix elements at the Fermi level are rather small, leading to values of μ (the average matrix element at the Fermi level times N_F), which are extremely low: respectively 0.16, 0.19, and 0.12 (see Table I).

The critical temperatures in these systems are around 10 K which are significant for graphite-like materials.[36] This predictions are close to those obtained with SCDFT.[85] Although SCDFT and the present approach are formally constructed from the same couplings and formally include the interactions in a similar approximation, this agreement

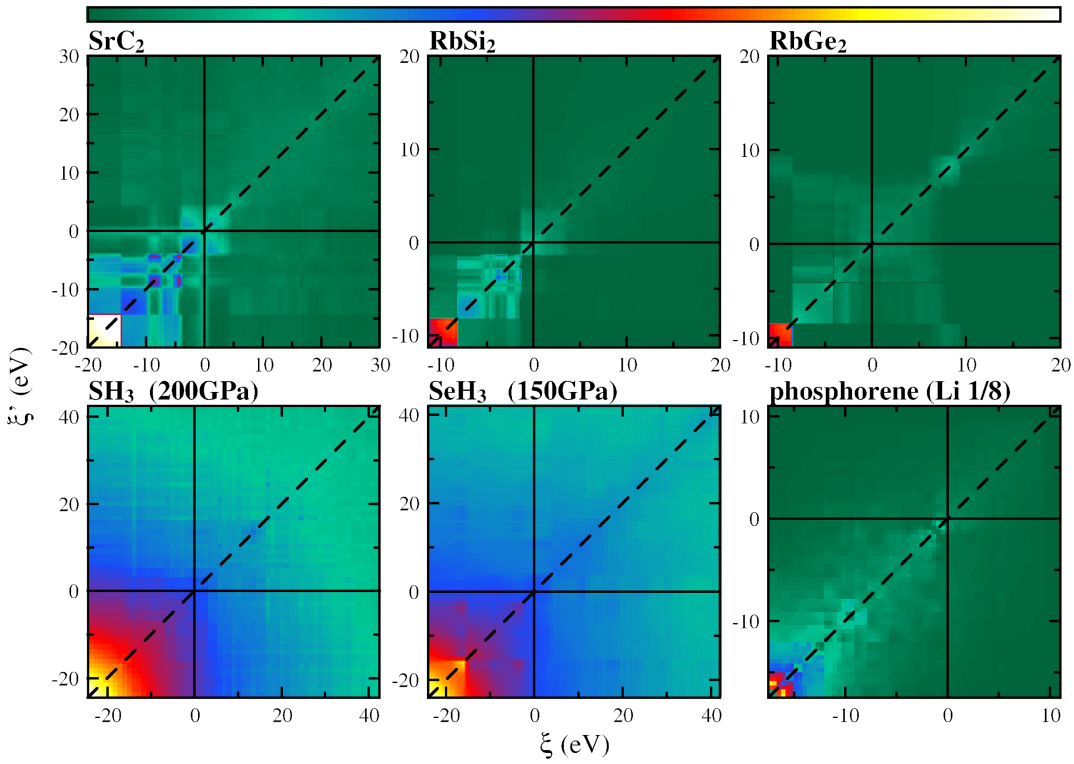

Fig. 2. (Color online) Screened Coulomb interaction averaged matrix elements $V_0^c(\xi\xi')$ [as defined by Eq. (5)] for the set of studied materials. The value indicated by the color scale ranges from zeros (dark green) and a maximum (white) corresponding to 2.2 (st./eV/cell/spin)$^{-1}$.

between the theories predicted critical temperatures is not a foregone conclusion, the form of the equations that are solved is completely different, the numerical Matsubara integration of Eq. (1) is numerically non-trivial, and SCDFT requires extra approximations for the functional.[45)]

Within SCDFT, the frequency dependent gap function is not easily accessible while, in the present Eliashberg formalism, this quantity is straightforward to compute. In Fig. 3 we present the real and imaginary part of the frequency-dependent gap function together with the temperature dependence of the superconducting gap at zero frequency. From these functions it is simple to estimate important experimental observables such as coherence length, STM spectra or ARPES spectral functions. These are key quantities required to achieve a complete characterization of a superconducting material beyond the mere critical temperature.[31)]

3.2 *SH_3 and SeH_3*

Sulfur hydride (SH_3) is the superconductor that set the record for the highest critical transition temperature ever measured (200 K) at a pressure of 200 GPa. This system was first predicted by means of an evolutionary method (genetic algorithm) by Duan and coworkers.[29)] Shortly afterward Drozdov, Eremets, and coworkers[30)] succeed on synthesizing SH_3 and measured the electrical resistivity at high pressure. The superconducting mechanism has been thoroughly investigated and unambiguously identified as being phononic.[66,83,91–100)]

Shortly after this discovery it was predicted[83,85,101)] that SeH_3 also should form at high pressure with the same cubic crystal lattice as SH_3 and have strong superconducting

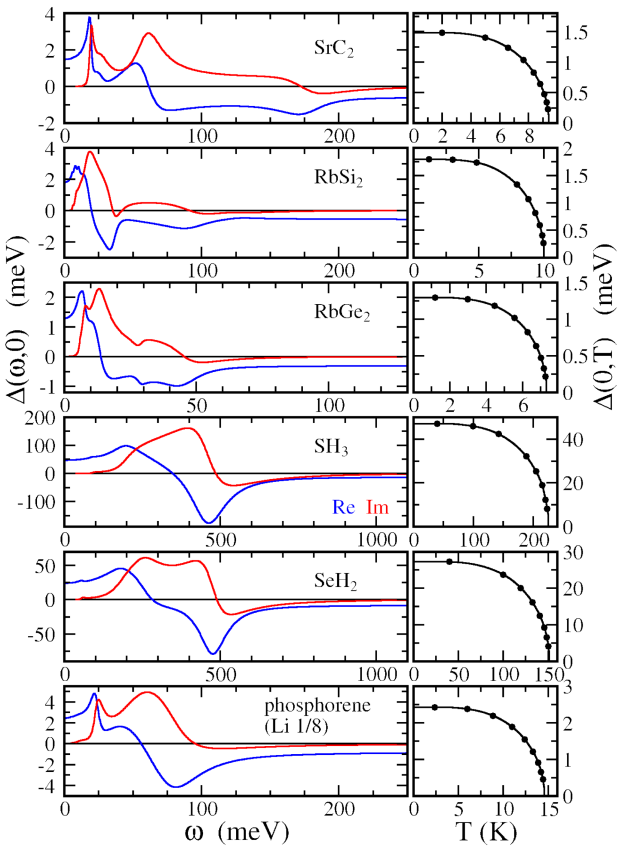

Fig. 3. (Color online) Superconducting gap $\Delta(\omega, T)$ as a function of frequency and at low temperature (left) and as a function of temperature and zero frequency (right). On the left we plot the real and imaginary part of Δ in blue and red, respectively.

properties. The crystal structure is shown in Fig. 1. Hydrogen is bonded to S or Se forming a metallic band of hybrid character that is the key ingredient for the existence of such spectacular superconducting coupling. As pointed out by Ashcroft,[102] hydrogen is the best candidate for high-temperature phononic superconductivity due to its low mass that results in large phonon energies (see the scale of the α^2F function in Fig. 1). In addition hydrogen, having only one electron, presents a weakly screened ionic potential[88,103,104] that leads to large values of the matrix elements. This can also be understood in SH_3 and SeH_3 by looking at the configurational space,[83,105] where it is clear that all low-enthalpy structures show hydrogen in molecular form, apart from the lowest where *S–H* bonds are formed. Indeed, much higher pressures would be necessary to reach the metallization of pure molecular hydrogen.[106–108]

From the point of view of the Coulomb interaction point of view, these high pressure systems show an electron-gas like behaviour[109] with screened matrix elements that decrease monotonically and smoothly with the quasi-particle energy. While the μ itself is quite large, Coulomb renormalization leads to an effective μ^* of 0.12, a rather conventional value for standard *sp* systems in spite of the unusual properties of these materials and the extreme pressures.

3.3 *Lithium doped phosphorene*

Phosphorene[110] is a monolayer of black phosphorus,[111] the layered allotrope of elemental phosphorus. Phosphorene is a small gap semiconductor that, upon Li deposition it was predicted to become metallic and superconducting.[87,112,113] In this work we consider Li doping on both faces of the monolayer, a configuration that is likely more difficult to realise experimentally. However it to achieve a higher level of doping and consequently larger critical temperatures. This is important because superconductivity is induced the a charge transfer from the dopant to the valence states of Phosphorene. The highest the doping level the larger will be the Fermi DOS and consequently the electron–phonon coupling.

In fact superconductivity is induced by the intrinsic superconducting coupling of P atoms (from the black-P) lattice as soon as electrons are injected in the anti-bonding states of the system. Hence, superconductivity is largely independent from the actual dopant species and instead depends on the doping level that these atoms provide. Since this system is a pseudo-2D material it is expected to have an intrinsic Coulomb repulsion stronger than that in typical 3D superconductors. Nevertheless the effective μ^* is, once again of the order of 0.11 confirming the empirical rule.

From the physical point of view it is quite interesting to observe that doping induces a very strong superconducting coupling in the black phosphorus system. This result is similar to that obtained for doped graphene,[36] however with an important difference: in graphene the superconducting states are partly due to the dopant atom while in doped phosphorene all states at the Fermi level are phosphorus states the actual dopant is irrelevant and merely transfers charge to the substrate.

3.4 *Computational details*

All calculations were performed at the level of Kohn–Sham[11] DFT,[10] within the local density approximation LDA[115] for SH_3 and SeH_3 and GGA-PBE[116] for SrC_2, $RbSi_2$, $RbGe_2$, and phosphorene. The electron–phonon coupling was computed with Quantum Espresso[17] by means of density functional perturbation theory.[117–120] Electronic screening and screened Coulomb interaction matrix elements were computed with the ELK code.[18] The computational cost of the solution of the Eliashberg Eq. (1), is only little more demanding than a similar SCDFT calculation and overall negligible as compared to the computational cost involved in computing the Eliashberg spectral functions. This is due to the isotropic approximation we assumed. On the other hand, the calculation of the screened Coulomb interactions in Fig. 2 are demanding due to the large number of Kohn–Sham states required to reach an energy scale sufficiently large to converge the ξ' integral in the ϕ part of Eq. (1).

4. Conclusions and Outlook

In this work, we present a thorough investigation of several important novel superconductors ranging from high pressure systems, layered materials and doped monolayers, by means of a fully ab initio Eliashberg approach. This method provides reasonable predictions of superconducting properties, including T_C and the excitation spectrum of the superconductor without any adjustable parameter. Overall results are consistent with predictions performed within superconducting density functional theory, so far the only alternative parameter-free method for studying superconducting materials. This scheme establishes Eliashberg theory as a method for discovery new superconductors without the need of material-dependent parameters. However it is important to observe that the calculations presented here, rather than invalidate the μ^* approximation, are actually confirming it. In fact, once the Coulomb renormalization is computed ab-initio the value of μ^* that gives the computed T_C (at least within the McMillan theory) is very close to 0.11 (see Table I). On the basis of empirical observation this value is broadly accepted for *sp* systems. It is fascinating to observe that in spite of having very different Coulomb interactions (Fig. 2 and Table I) and phononic characteristic energies (Fig. 1 and Table I) these different materials, yield almost the same effective coupling strength.

It will be interesting to test our fully parameter free Eliashberg method on even more complex physical systems, where the μ^* approximation is known to fail. This approach can be easily extended to include anisotropy effects or unconventional order parameters by separating bands.[72,121] A further extension would be to consider more complex Coulomb pairing forces such as those dynamical effects arising from plasmons or spin fluctuations.

Acknowledgments J.A.F.-L. acknowledges the computational resources under the project s752 from the Swiss National Supercomputing Center (CSCS) in Lugano.

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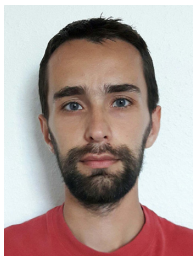

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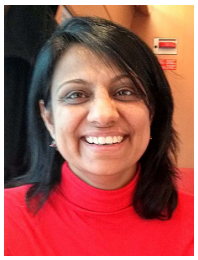

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