

First-principles evidence for conventional superconductivity in a quasicrystal approximant

Pedro N. Ferreira^{1,*}, Roman Lucrezi², Sangmin Lee³, Lucy Nathwani³, Matthew Julian⁴, Rohit P. Prasankumar⁴, Warren E. Pickett⁵, Chris J. Pickard^{6,7}, Philip Kim³, and Christoph Heil^{1,†}

¹*Institute of Theoretical and Computational Physics, Graz University of Technology, NAWI Graz, 8010 Graz, Austria*

²*Department of Chemistry, Stockholm University, SE-10691 Stockholm, Sweden*

³*Department of Physics, Harvard University, Cambridge, Massachusetts 02138, USA*

⁴*Enterprise Science Fund, Intellectual Ventures, Bellevue, Washington 98005, USA*

⁵*Department of Physics and Astronomy, University of California Davis, Davis, California 95616, USA*

⁶*Department of Materials Science and Metallurgy, University of Cambridge, 27 Charles Babbage Road, Cambridge CB3 0FS, United Kingdom*

⁷*Advanced Institute for Materials Research, Tohoku University, Sendai 980-8577, Japan*



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Quasicrystals (QCs) host long-range order without translational symmetry, a regime in which the very foundations of BCS theory are not straightforwardly applicable, yet experiments on QCs and their approximant crystals (ACs) point to conventional, *s*-wave, electron–phonon-coupled superconductivity. Here, we test the predictive power of the electron–phonon framework in a representative decagonal AC from first principles. Using state-of-the-art *ab initio* methods, we compute the superconducting properties of the recently discovered AC $\text{Al}_{13}\text{Os}_4$ and quantitatively reproduce its bulk T_c . This provides an *ab initio* determination of T_c for an AC and establishes that the electron–phonon framework is predictive in these systems. Using the generalized quasi-chemical approximation for alloy modeling in the decagonal Al–Os family, we predict tunable superconductivity in $\text{Al}_{13}\text{Os}_{4-x}\text{Re}_x$ and $\text{Al}_{13}\text{Os}_{4-x}\text{Ir}_x$; in particular, $\text{Al}_{13}\text{Re}_4$ is dynamically stable and estimated to have a T_c about 30% above that of $\text{Al}_{13}\text{Os}_4$. Finally, we discuss the role of ACs as high-fidelity proxies for their parent QCs. Although long-range quasiperiodicity may introduce subtle electronic features, our findings indicate that the key ingredients for superconductivity are already encoded in the local structural motifs preserved by the AC. This places the Al–Os and Al–Re families among the most promising candidates for the highest- T_c quasicrystalline superconductivity.

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I. INTRODUCTION

Quasicrystals (QCs) represent an extraordinary class of solids that have challenged the classical laws of crystallography. They are characterized by their aperiodic yet long-range ordered atomic arrangements, exhibiting noncrystallographic rotational symmetries [1–3]. This conceptual breakthrough was first reported in 1984 in rapidly solidified Al alloys containing 10–14 at.% Mn [4]. Since then, QCs have attracted sustained interest both for applications—ranging from photonics [5–8] and catalysis [9,10] to functional coatings [11,12]—and as platforms for magnetism [13], topology [14,15], and strongly correlated matter [16–18].

Despite indications that QCs are not exceedingly rare in the universe [19], superconducting QCs remain exceptionally

scarce. The first indications of superconductivity (SC) in a QC were reported in 1987, observed initially in the icosahedral phase of $\text{Mg}_{71}\text{Zn}_{21}\text{Al}_{12}$ [20, 21], in the Frank-Kasper phases of Al–Cu–Li and Al–Cu–Mg alloys [22], and in the rapidly quenched Ti–Zr–Al alloys [23].

Nevertheless, none of these initial reports provided comprehensive evidence for SC by simultaneously demonstrating zero resistivity, the Meissner effect, a specific heat jump at the superconducting transition, and explicit quasiperiodic symmetry within the same sample. This milestone was achieved conclusively only in 2018, with the characterization of a bulk superconducting state at a critical temperature (T_c) of 0.05 K in the so-called Bergmann-type Al–Zn–Mg QC alloys [24].

Earlier theoretical studies had predicted that Cooper pairs in structures such as the Penrose lattice would exhibit exotic, unconventional superconducting pairing symmetries characterized by finite center-of-mass momentum, fundamentally driven by the lack of translational symmetry [25]. Nevertheless, none of the predicted exotic phenomena have been experimentally observed thus far [24,26,27]. In fact, experimental measurements in both the Al–Zn–Mg QCs and their corresponding ACs indicate *s*-wave superconducting pairing consistent with a conventional BCS weak-coupling electron–phonon (el–ph) mechanism [24].

*Contact author: nunesferreira@tugraz.at

†Contact author: christoph.heil@tugraz.at

Despite the often anticipated unconventional superconducting mechanisms in QCs, there is also support for more conventional scenarios. For instance, in Ref. [28], the authors employed the inhomogeneous Bogoliubov–de Gennes (BdG) mean-field theory to explore SC within a two-dimensional Ammann–Beenker tiling. Predictions from this model closely align with experimental observations for the Al–Zn–Mg QC, indicating that BdG solutions for QCs can indeed reproduce conventional, BCS-like SC [28]. Furthermore, the authors of Ref. [28] provided theoretical evidence that ACs successfully capture the superconducting behavior of QCs, demonstrating that the unique electronic wave function characteristics of an infinite QC may not significantly influence its superconducting properties [28]. In fact, as QCs lack translational symmetry, ACs provide the most practical way to simulate and gain insights into their parent QCs and have therefore been investigated via *ab initio* calculations to shed light on, for instance, their thermodynamic [29], electronic [30], magnetic [31], and—beginning with the present work—superconducting properties.

Here, we address from first principles the question of whether SC in ACs can originate from a conventional electron–phonon coupling mechanism. To this end, we perform density functional perturbation theory (DFPT) calculations on the recently reported $\text{Al}_{13}\text{Os}_4$ AC superconductor [32].

$\text{Al}_{13}\text{Os}_4$ is a hitherto unexplored AC [33], isotypic to the well-known $\text{Al}_{13}\text{Fe}_4$ [34,35] and homeotypic to $\text{Al}_{13-x}(\text{Co}_{1-y}\text{Ni}_y)_4$ [36], associated with the decagonal phase of the Al–Os alloy [37]. In Ref. [32], SC in $\text{Al}_{13}\text{Os}_4$ was established via resistivity, specific heat, and magnetization measurements, as well as muon-spin rotation spectroscopy, revealing a record T_c of approximately 5 K for QCs and ACs, accompanied by unambiguous BCS-like signatures.

With this work, we show that the bulk superconducting response of $\text{Al}_{13}\text{Os}_4$ is well described within the conventional el–ph framework. This is, to our knowledge, an *ab initio* prediction of T_c for an AC using Migdal–Eliashberg theory, and it reproduces the experimental superconducting energy scale in a system where measurements already indicate a fully gapped, weak-coupling *s*-wave state [32]. Moreover, we predict that the substitutional solid solutions $\text{Al}_{13}\text{Os}_{4-x}\text{Re}_x$ and $\text{Al}_{13}\text{Os}_{4-x}\text{Ir}_x$ provide routes to improve superconducting properties; in particular, $\text{Al}_{13}\text{Re}_4$ is dynamically stable and exhibits a higher T_c and larger Δ_0 compared with $\text{Al}_{13}\text{Os}_4$. We therefore propose that the quasicrystalline counterparts of $\text{Al}_{13}\text{Os}_4$ and $\text{Al}_{13}\text{Re}_4$ may harbor the high- T_c values among QCs.

II. RESULTS

Crystal structure. $\text{Al}_{13}\text{Os}_4$ has a monoclinic structure (space group $C2/m$) and belongs to the family of decagonal ACs commonly designated as $\text{Al}_{13}\text{TM}_4$ (TM = transition metal) [38]. It has drawn particular interest because, among known superconducting ACs, it exhibits the highest T_c reported so far, and its atomic arrangement closely resembles that of $\text{Al}_{13}\text{Fe}_4$, a compound that approximates both decagonal and icosahedral QCs [34]. $\text{Al}_{13}\text{Os}_4$ is isostructural to the Y-phase of the Al–Co–Ni intermetallic, $\text{Al}_{13-x}(\text{Co}_{1-y}\text{Ni}_y)_4$,

and both of these compounds are ACs of the QC $\text{Al}_{70}\text{Ni}_{15}\text{Co}_{15}$ [36,39]. $\text{Al}_{13}\text{Os}_4$ is also related to the metastable decagonal quasicrystal phase of the Al–Os alloy [37,38].

Fully *ab initio* structural optimization of the oblique rhombic-prism primitive cell of $\text{Al}_{13}\text{Os}_4$ [17 atoms in the primitive cell, Fig. 1(a)] yields lattice parameters $a = 9.11 \text{ \AA}$ and $c = 7.82 \text{ \AA}$ and angles $\alpha = 115.04^\circ$ and $\gamma = 26.91^\circ$, all within 1% of the experimental values. We also fully relaxed the primitive cell while accounting for spin–orbit coupling (SOC) and nonlocal van der Waals (vdW) interactions [40,41]. The resulting lattice parameters differ by less than 0.2% from those obtained without these effects, indicating that SOC and vdW interactions do not play a significant role in the structural properties of $\text{Al}_{13}\text{Os}_4$.

$\text{Al}_{13}\text{Os}_4$ is layered along the *b* axis with a periodicity of $\sim 4 \text{ \AA}$. The layers are structurally identical, but the layer at $y = 1/2$ is shifted by half a unit cell in the *a*-axis direction [Figs. 1(b) and 1(c)] [32]. Within these layers, structural motifs can be tiled in two ways: smaller tiles formed by connecting predominantly Al atoms, and larger tiles formed by connecting predominantly Os atoms [32,36]. Figures 1(b) and 1(c) below show these tilings on each layer. The smaller tiles provide only a partial tessellation of the unit cell, whereas the larger tiles generate a gapless tiling of the entire cell. The larger tiling scheme resembles a distorted Penrose tiling composed of one pentagon and two types of rhombi: a narrow rhombus with an acute angle of $\sim 35^\circ$, and a wider rhombus with an angle of $\sim 80^\circ$.

Variations of both the smaller and larger tiling schemes, shown in Fig. 1(d), can be used to tile $\text{Al}_{70}\text{Ni}_{15}\text{Co}_{15}$. The larger tiling in $\text{Al}_{70}\text{Ni}_{15}\text{Co}_{15}$ is a 2D Penrose tiling composed of one pentagon and one rhombus with an acute angle of $\sim 35^\circ$. The tile shapes in $\text{Al}_{13}\text{Os}_4$ are slightly distorted relative to those in the ideal quasicrystal [36]. $\text{Al}_{13}\text{Os}_4$ is, therefore, a promising model system for studying the emergence of quasicrystalline symmetry in Al–TM systems and for understanding the structures of other $\text{Al}_{13}\text{TM}_4$ ACs.

Electrons. To gain insights into the microscopic behavior of $\text{Al}_{13}\text{Os}_4$, we start with a discussion of its electronic properties. The electronic band structure along high-symmetry directions of the BZ is shown in Fig. 2(a), with the states projected onto Al-*p* (blue) and Os-*d* (red) orbital characters.

The electronic structure of $\text{Al}_{13}\text{Os}_4$ is, in fact, quite complex: six distinct bands cross the Fermi level along every high-symmetry path of the BZ, as shown in Fig. 2(c). The Fermi energy ε_F lies near a local maximum in the DOS. The total DOS at the Fermi level, N_{ε_F} , is 5.67 states/eV/formula unit, in line with the calculated value of 5.38 states/eV/f.u. reported in Ref. [32]. Decomposing N_{ε_F} by orbital character, 50% of the states at ε_F originate from Al-*p* orbitals and 34% from Os-*d* orbitals. The nearly equal energy dependence of the Al-*p* and Os-*d* DOS within $\pm 0.5 \text{ eV}$ of ε_F indicates strong hybridization, corroborated by the orbital-projected bands in Fig. 2(a).

To obtain further insights into the bonding characteristics of $\text{Al}_{13}\text{Os}_4$, we computed the electron localization function (ELF); isosurfaces at selected values are shown in Fig. 2(b). These isosurfaces place $\text{Al}_{13}\text{Os}_4$ closer to covalent metals than to simple metals: unlike simple metals, where interstitial networks typically percolate near $\text{ELF} \approx 0.5$ [42,43], clear

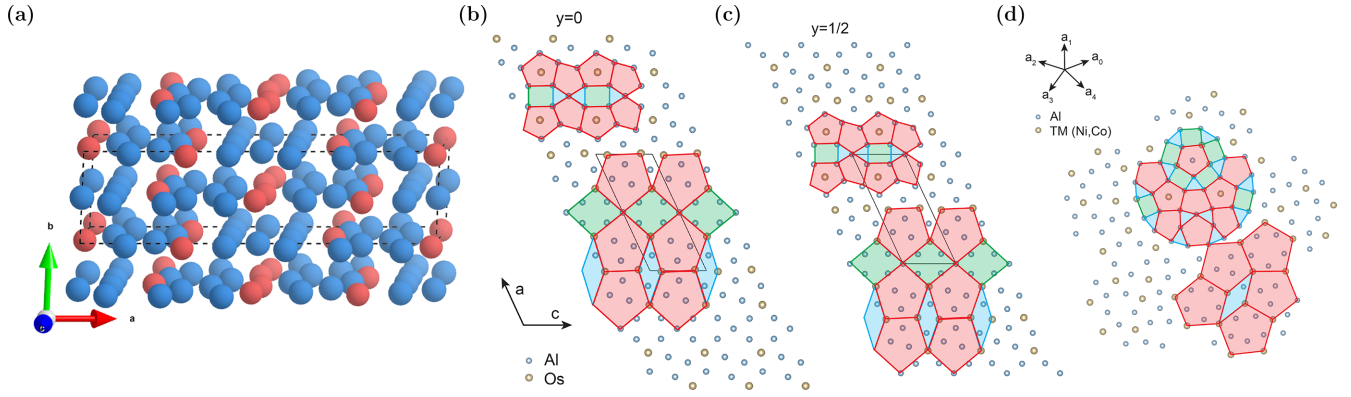


FIG. 1. Crystal structure of $\text{Al}_{13}\text{Os}_4$. (a) Primitive cell (1 formula unit) of $\text{Al}_{13}\text{Os}_4$. Aluminum is represented by blue spheres and osmium by red spheres. (b) The $y = 0$ layer of $\text{Al}_{13}\text{Os}_4$ with one unit cell indicated by thin black lines. (c) The $y = 1/2$ layer of $\text{Al}_{13}\text{Os}_4$. (d) Variations of the smaller and larger tilings can be used in the quasicrystal $\text{Al}_{70}\text{Ni}_{15}\text{Co}_{15}$ as marked. Projections of the quasicrystal's five basis axes are also shown.

bond basins are already visible at 0.75. Consistent with this picture, two Os sites are more ionic, and the remaining two Os sites hybridize with the Al network, which partially explains the smaller Os contribution at ε_F .

We also investigated the effects of SOC and nonlocal vdW interactions on the electronic properties of $\text{Al}_{13}\text{Os}_4$ [44]. vdW interactions produce no substantive changes. SOC lifts key degeneracies and opens a continuous pseudogap between the

valence and conduction bands; nevertheless, the overall FS topography remains essentially unchanged.

Hybridization at the Fermi level can give rise to multiband behavior that may influence the superconducting properties [45]. To sharpen this picture, we examined the FS topography: we resolve two small hole-like FS sheets with predominantly Al- p character (α and β); a third hole-like sheet with pronounced Al- p /Os- d hybridization (γ); and three additional,

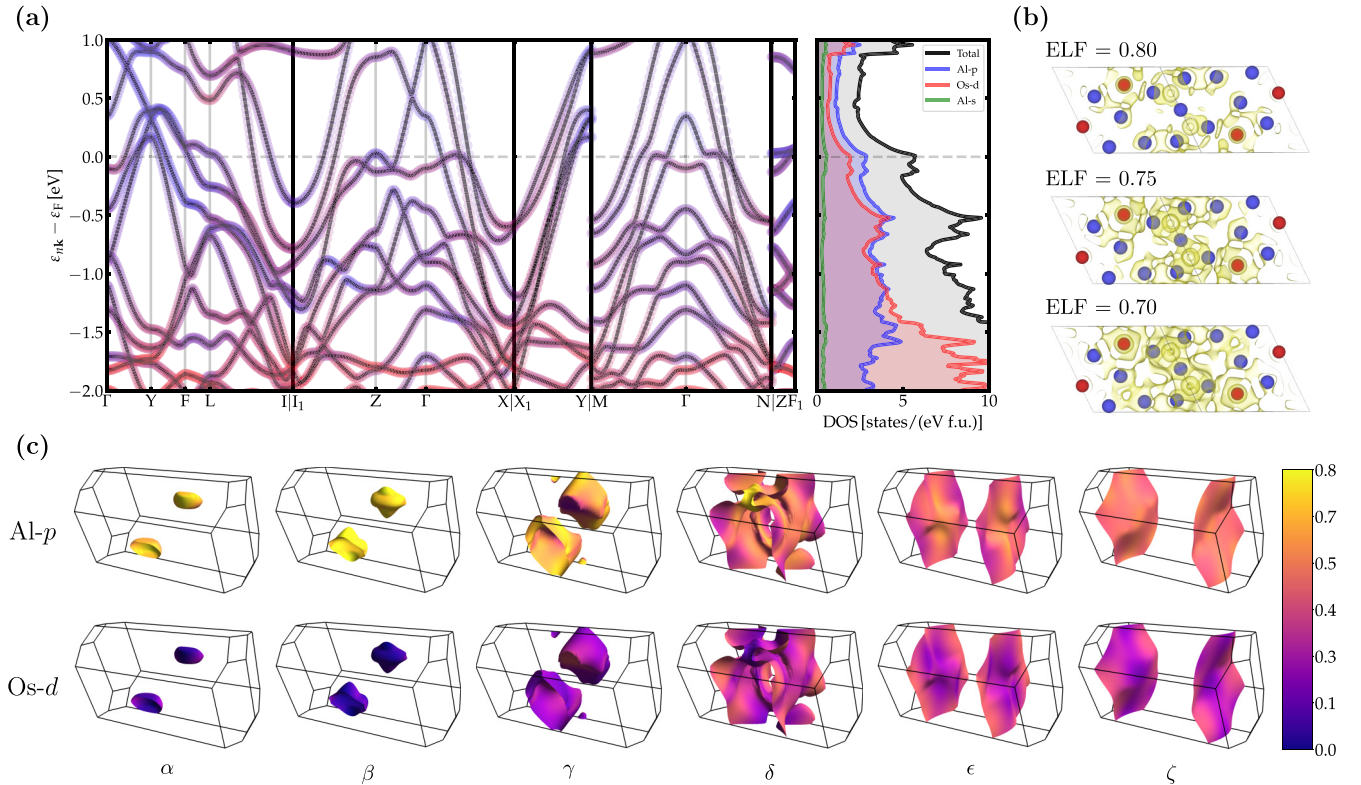


FIG. 2. Electronic properties of $\text{Al}_{13}\text{Os}_4$. (a) Orbital-projected band structure along the selected high-symmetry path (thin black lines). Each marker's hue is a combination of the colors assigned to the contributing orbitals, with coefficients given by their fractional projections at that state; the marker opacity encodes the total projected weight. Vertical guidelines indicate the high-symmetry points. Total electronic density of states (black, light fill) and orbital-resolved partial DOS for Al- p (blue), Os- d (red), and Al- s (green) are also displayed. (b) Electron localization function (ELF) isosurfaces at selected ELF values. (c) Fermi-surface sheets projected onto the Al- p and Os- d character.

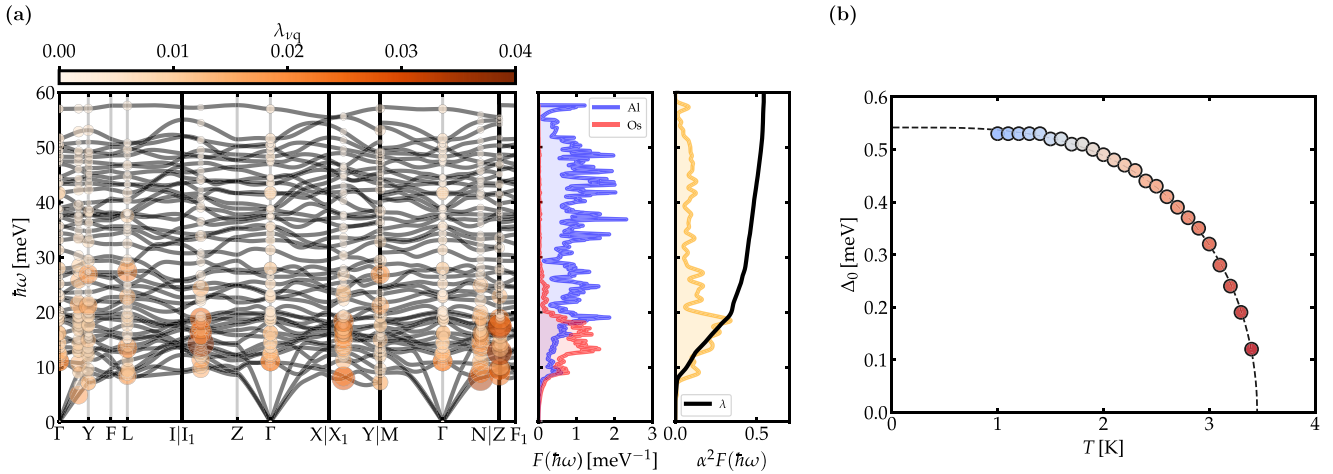


FIG. 3. El-ph interactions and superconducting properties of $\text{Al}_{13}\text{Os}_4$. (a) (Left) Phonon dispersion along a high-symmetry path in the BZ. Superimposed circles indicate the mode-resolved el-ph coupling λ_{vq} at selected q points; colors and marker size encode λ_{vq} . (Middle) Atom-projected phonon density of states $F(\hbar\omega)$ in meV^{-1} for Al (blue) and Os (red). (Right) Eliashberg spectral function $\alpha^2F(\hbar\omega)$ (orange line) and the cumulative total electron-phonon coupling parameter λ . (b) Superconducting gap Δ_0 as a function of temperature obtained by solving the isotropic Migdal-Eliashberg equations within the full-bandwidth approximation.

more extended sheets (δ , ϵ , ζ) that exhibit strong Al-Os hybridization, as shown in Fig. 2(c).

In general, disconnected FS sheets with different electronic character lead to two or more well-separated superconducting gaps [46–50]. As follows from multiband gap equations [51,52], appreciable interband pairing combined with an approximately sheet-independent pairing interaction favors a common gap over the FS, whereas well-separated gaps typically emerge when the effective couplings differ substantially among the sheets. Such gap separation occurs naturally when distinct FS sheets have different orbital character, resulting in strongly sheet-dependent pairing interactions and weak interband impurity scattering, as in the σ and π states of MgB_2 [53,54]. In $\text{Al}_{13}\text{Os}_4$, the dominant sheets share substantial Al- p /Os- d hybridization, so the electronic structure provides no mechanism for a MgB_2 -like separation of the order parameter. The hybridized bands are also likely to dominate the superconducting quasiparticle excitations because of their appreciable DOS, potentially suppressing any additional, smaller gap [55].

Consistent with this scenario, Ref. [32] reports that the temperature dependence of the superconducting relaxation rate in muon spin rotation/relaxation measurements σ_{SC} is well described by an isotropic conventional s -wave model. The low-temperature plateau in σ_{SC} down to the lowest measured T is, indeed, consistent with a single, isotropic gap [56–61].

Nevertheless, there are cases where the temperature-dependent TF- μ SR signal fails to distinguish single-gap from multigap order parameters [62]. Hence, we believe that the single versus multigap nature of SC in $\text{Al}_{13}\text{Os}_4$ is not completely resolved on theoretical or experimental grounds, and further investigations along these lines would be highly valuable. A definitive theoretical resolution would require solving the fully anisotropic multiband ME equations [63–65]—an exceptionally demanding computation that lies beyond the scope of the present work.

Phonons and el-ph coupling. We now turn to the phonons and their coupling to electrons. We confirm that $\text{Al}_{13}\text{Os}_4$ is dynamically stable in the harmonic picture. The phonon dispersion and phonon density of states, shown in Fig. 3(a), indicate that the phonon spectrum extends up to 57.6 meV, where low-energy branches are hybrid Al-Os vibrations, while middle- and high-energy branches are derived from Al.

We also computed the el-ph coupling and found that λ is broadly distributed across the BZ, as indicated by the size and color of the markers in Fig. 3(a). Integrating the Eliashberg spectral function up to 25 meV yields $\lambda \approx 0.43$, i.e., about 80% of the total el-ph coupling is provided by the hybrid Al-Os phonon branches, while the mostly Al-derived optical branches above 25 meV contribute the remaining ≈ 0.11 . Notably, the low-frequency branches along X_1 –Y and Γ –N host strongly coupled chiral (circularly polarized) and in-plane bending Al modes, with a smaller contribution from in-plane Os stretching. We also identify strongly coupled modes at the Z high-symmetry point, comprising a mixture of in-plane Al stretching and bending together with out-of-phase, out-of-plane Al/Os vibrations.

The total el-ph coupling is $\lambda = 0.54$, placing $\text{Al}_{13}\text{Os}_4$ in the weak-coupling regime. For comparison, the authors of Ref. [32] estimated λ from the measured T_c and the Debye frequency using the McMillan semiempirical model [66], obtaining $\lambda = 0.63$. This independent estimate corroborates weakly coupled BCS SC and is in excellent agreement with our fully *ab initio* determination.

Superconductivity. To investigate the superconducting properties, we solved the isotropic Migdal-Eliashberg equations [67] within the full-bandwidth approximation [64] using the ISOME code [68] to obtain a reliable estimate of T_c including retardation effects and the el-ph scattering processes beyond the Fermi surface.

Within this framework, we obtain $T_c = 3.5$ K and a zero-temperature superconducting gap $\Delta_0(0) = 0.54$ meV using $\mu_{\text{AD}}^* = 0.1$ for the Morel-Anderson Coulomb

pseudopotential [69,70]. Experimentally, Ref. [32] reports onsets of the superconducting transition at 5.47 K in resistivity, 5.45 K in zero-field-cooled/field-cooled magnetization at 1 mT, 5.44 K in specific heat, and close to 5 K in the TF- μ SR relaxation rate; the magnetization offset likewise occurs near 5 K. The experimentally fitted superconducting gap is 0.79 meV. This represents remarkable agreement between theory and experiment for such a complex system. Indeed, the agreement is particularly compelling given the level of theory employed. Our calculations deliberately use an isotropic ME framework; achieving still higher quantitative accuracy would require a fully anisotropic treatment that accounts for the momentum and band dependence of both the el-ph and screened el-el interactions on the same footing, at substantially higher computational cost. Taken together, reproducing the correct scale of T_c and obtaining a gap magnitude consistent with experiment—alongside experimental indications of an isotropic s -wave pairing [32]—show that superconductivity in $\text{Al}_{13}\text{Os}_4$ is quantitatively consistent with a conventional electron-phonon mechanism. While this does not uniquely exclude more complex pairing scenarios, it places strong constraints on them.

The underestimation of the critical temperature by about 30% lies within the typical accuracy range of first-principles superconductivity calculations for low- T_c materials [71]. This is because, in a weak-to-intermediate-coupling system such as $\text{Al}_{13}\text{Os}_4$, small changes in the effective el-ph coupling or screened Coulomb repulsion strongly affect T_c . Using the Allen-Dynes expression, the experimental $T_c \approx 5$ K is recovered by slightly increasing the coupling to $\lambda \simeq 0.60$ at fixed $\mu_{\text{AD}}^* = 0.10$. Varying μ_{AD}^* from 0.07 to 0.13 changes T_c from approximately 5.0 K to 2.3 K within the isotropic ME framework. Note, however, that the reduced gap ratio $2\Delta_0/k_B T_c$ is 3.54 in our calculations and 3.4–3.7 experimentally, both close to the BCS weak-coupling value of 3.53. Moreover, gap and coupling anisotropy can increase the predicted T_c , bringing it closer to the experimental value [63].

We would like to emphasize that our calculations address bulk superconductivity. Reported spin-polarized surface states in $\text{Al}_{13}\text{Os}_4$ [32] could, in principle, host additional pairing phenomena confined to the surface. Such effects would not necessarily be reflected in the bulk thermodynamic T_c reproduced here.

We next address an important theoretical consideration. While we have demonstrated that *ab initio* calculations within the conventional el-ph framework accurately reproduce the experimental T_c of ACs, QCs differ in their lack of translational invariance. This absence undermines conventional prerequisites of BCS theory, including well-defined momentum space and Fermi surfaces, and in principle permits Cooper pairs with finite center-of-mass momentum. However, theoretical studies indicate that the exotic electronic wave functions of infinite QCs do not qualitatively alter their superconducting character [28], and experimental evidence consistently points to a conventional pairing mechanism.

From a practical standpoint, standard DF(P)T cannot be directly applied to infinite QCs for T_c predictions because of its reliance on translational periodicity. ACs therefore serve as computationally tractable proxies that capture the

essential local structural and electronic features of QCs. The key question is whether T_c values predicted for ACs using DF(P)T and ME theory reliably approximate those of the corresponding quasicrystalline phases. If AC-based T_c predictions prove reliable, they can be leveraged in first-principles calculations to identify candidate QC phases with enhanced superconducting properties.

We argue that Ref. [25], although advocating unconventional pairing in QCs, also offers useful insights into why ACs can still provide reasonable predictions for their QC counterparts. Using real-space dynamical mean-field theory, the authors of Ref. [25] studied the attractive Hubbard model on a Penrose lattice. In the weak-coupling regime, they find a finite Fourier-transformed pair amplitude of the order parameter, $\text{OP}_{\mathbf{k}\mathbf{k}'}$, beyond $\mathbf{k}' = -\mathbf{k}$ and distinct high-intensity spots along the $\mathbf{k}' = -\mathbf{k}$ line. Nevertheless, most of $\text{OP}_{\mathbf{k}\mathbf{k}'}$ remains concentrated along $\mathbf{k}' = -\mathbf{k}$, with high-intensity spots reminiscent of the BCS regime. This indicates that, even without translational invariance, a large fraction of the pair amplitude remains concentrated near $\mathbf{k}' = -\mathbf{k}$, reminiscent of the dirty-limit robustness of conventional s -wave pairing [72]. This motivates the expectation that the bulk T_c of a QC and that of a closely related AC will be comparable when their local chemistry, dominant phonon spectrum, and low-energy electronic states are similar.

In a follow-up work [73], the same authors showed that, within the Penrose-tiling model, the specific-heat jump is only about 10%–20% smaller than the BCS value. For comparison, multiband effects in s -wave BCS superconductors can reduce the specific-heat anomaly by more than 20% [52,74,75], further supporting our argument.

We propose that the T_c of an approximant crystal provides a physically motivated baseline for the T_c scale of its parent quasicrystal. This is because the quantities that govern conventional pairing—bond chemistry and stiffness, characteristic phonon frequencies, and the dominant el-ph matrix elements—are controlled primarily by the local atomic environment, which is shared by the AC and QC. Quasiperiodicity can, however, modify quantities that depend sensitively on the reciprocal-space structure, such as the fine DOS near ε_F , the pseudogap structure, and therefore the precise values of N_{ε_F} . This distinction is consistent with model studies, where quasiperiodic superconductors remain broadly BCS-like in the weak-coupling regime but can show gap inhomogeneity and modest thermodynamic deviations [25,28,73], and with experiments establishing bulk superconductivity in Al–Zn–Mg QCs and approximants [24] as well as in the van der Waals QC $\text{Ta}_{1.6}\text{Te}$ [26,27]. Thus, the AC should be viewed as a proxy for the pairing mechanism and local pairing scale, while the exact T_c of the parent QC may differ as quasiperiodicity can modify the DOS near ε_F .

Therefore, we strongly encourage experimental efforts to synthesize and characterize this material. Moreover, this AC strategy opens an unexplored route to predict new QCs *in silico*, as we will demonstrate for $\text{Al}_{13}\text{Re}_4$.

Designing new superconducting ACs. In the first report of the $\text{Al}_{13}\text{Os}_4$ decagonal phase in 1987 [37], the author also described the synthesis of an analogous decagonal phase in the Al–Rh system and speculated that a similar phase might exist in Al–Ir. However, to the best of our knowledge, no

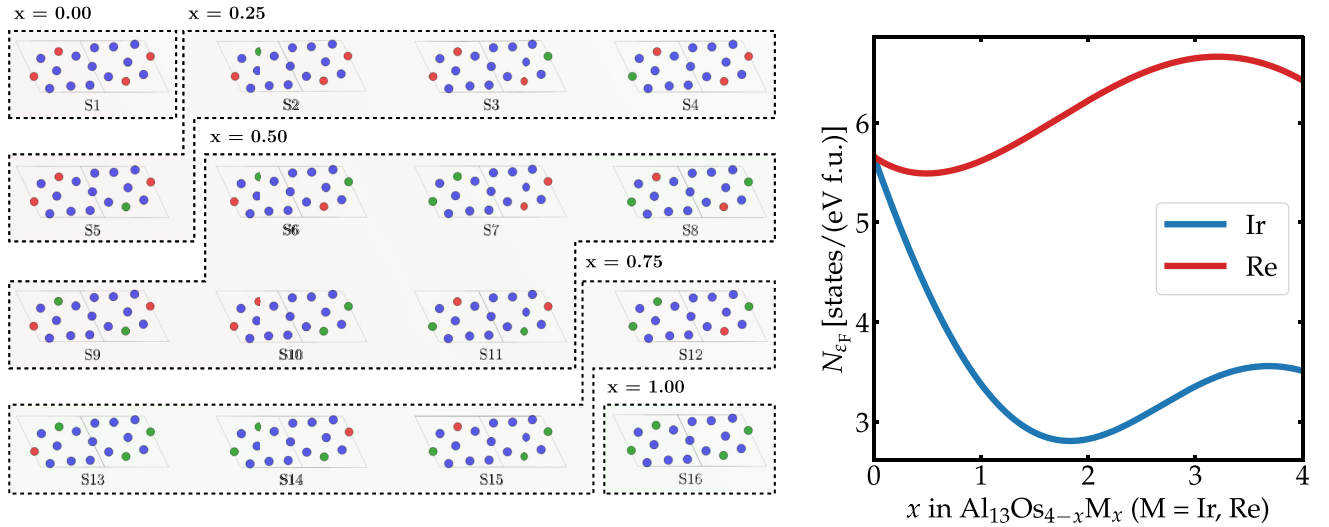


FIG. 4. (left) All supercell configurations obtained by substituting Os with Re or Ir in $\text{Al}_{13}\text{Os}_4$. (right) Total electronic DOS of $\text{Al}_{13}\text{Os}_{4-x}\text{Ir}_x$ and of $\text{Al}_{13}\text{Os}_{4-x}\text{Re}_x$ as a function of composition x , computed within the GQCA framework.

Al–Ir QC or their AC phases have been realized so far. Notably, iridium carries one additional $5d$ electron relative to osmium, while rhenium has one less. Thus, substitution on the transition-metal sublattice provides a way to tune the electronic properties of the system. Realizing such phases could open unexplored avenues for synthesizing high-quality, homogeneous quasicrystalline solid solutions with tunable superconducting properties, potentially surpassing those observed in the $\text{Al}_{13}\text{Os}_4$ AC.

Motivated by this, we investigated the thermodynamic stability and electronic properties of $\text{Al}_{13}\text{Os}_{4-x}\text{Re}_x$ and $\text{Al}_{13}\text{Os}_{4-x}\text{Ir}_x$ within the generalized quasichemical approximation (GQCA) [76,77].

Modeling $\text{Al}_{13}\text{Os}_4$ alloys. To model the alloy within GQCA, we generated all possible supercell configurations by substituting osmium atoms with rhenium or iridium, resulting in 16 distinct configurations, as illustrated in Fig. 4(a). Among these, 13 configurations are symmetry inequivalent. For each inequivalent configuration, we performed full *ab initio* relaxations of the lattice parameters and atomic positions while constraining the $C2/m$ base-centered monoclinic Bravais lattice symmetry. The total energies of the relaxed supercells were then used to evaluate the Gibbs mixing free energy as a function of composition for $\text{Al}_{13}\text{Os}_{4-x}\text{Ir}_x$ and $\text{Al}_{13}\text{Os}_{4-x}\text{Re}_x$ from 300 K to 1400 K (see Figs. 2 and 3 within the Supplemental Material [44]).

There exist clusters (supercells) with positive excess energies ($\Delta_f > 0$), which yield an asymmetric mixing enthalpy, $\Delta H(x, T)$, for $\text{Al}_{13}\text{Os}_{4-x}\text{Re}_x$ (Fig. 3 within the Supplemental Material [44]). This asymmetry suggests a departure from an ideal random-cluster distribution. As temperature increases, the configurational entropy dominates over enthalpy, and the Gibbs mixing free energy $\Delta G(x, T)$ becomes symmetric about $x = 0.5$. Moreover, no common-tangent construction connects distinct compositions in ΔG in either systems, which, together with the positive curvature, rules out binodal and spinodal decompositions above ambient temperature. From a thermodynamic point of view, therefore,

we expect that—subject to dynamical stability and kinetic constraints—high-quality, homogeneous solid solutions of $\text{Al}_{13}\text{Os}_{4-x}\text{Ir}_x$ and $\text{Al}_{13}\text{Os}_{4-x}\text{Re}_x$ should be experimentally accessible.

We note that this analysis addresses the configurational mixing thermodynamics within the $\text{Al}_{13}\text{TM}_4$ approximant structure, i.e., the stability of the pseudobinary substitutional solid solution with respect to its end members. Vibrational contributions to the mixing free energy are expected to be small for the present substitutions since Os, Re, and Ir are neighboring heavy elements with similar masses and bonding environments, and the phonon spectra of the end members closely resemble each other (Figs. 3 and 5). A global convex-hull construction including all competing Al–Os–Re phases was not performed and would require a separate extensive search over competing structures. We therefore restrict our conclusion to the configurational mixing stability within the $\text{Al}_{13}\text{TM}_4$ substitutional line, where no binodal or spinodal phase separation is found. This distinction is relevant because decagonal QCs and several of their ACs, including the Al–Os decagonal phase [37,38], are often metastable and can be obtained through nonequilibrium synthesis routes such as rapid solidification.

We now examine their potential for SC. In conventional phonon-mediated superconductors, T_c increases with both the electron–phonon coupling parameter λ and the logarithmic average phonon frequency $\omega_{\log}$. This trend is captured by the modified semiempirical McMillan expression [66,78,79],

$$T_c = \frac{\omega_{\log}}{1.2} \exp \left[-\frac{1.04(1 + \lambda)}{\lambda - \mu^*(1 + 0.62\lambda)} \right]. \quad (1)$$

As λ is proportional to N_{ϵ_F} , a well-established route to enhance T_c is to increase λ by maximizing N_{ϵ_F} through electronic gating or doping. However, stronger coupling is typically accompanied by a decrease of the characteristic phonon scale $\omega_{\log}$, so our task is to identify the optimal balance between large λ and high $\omega_{\log}$.

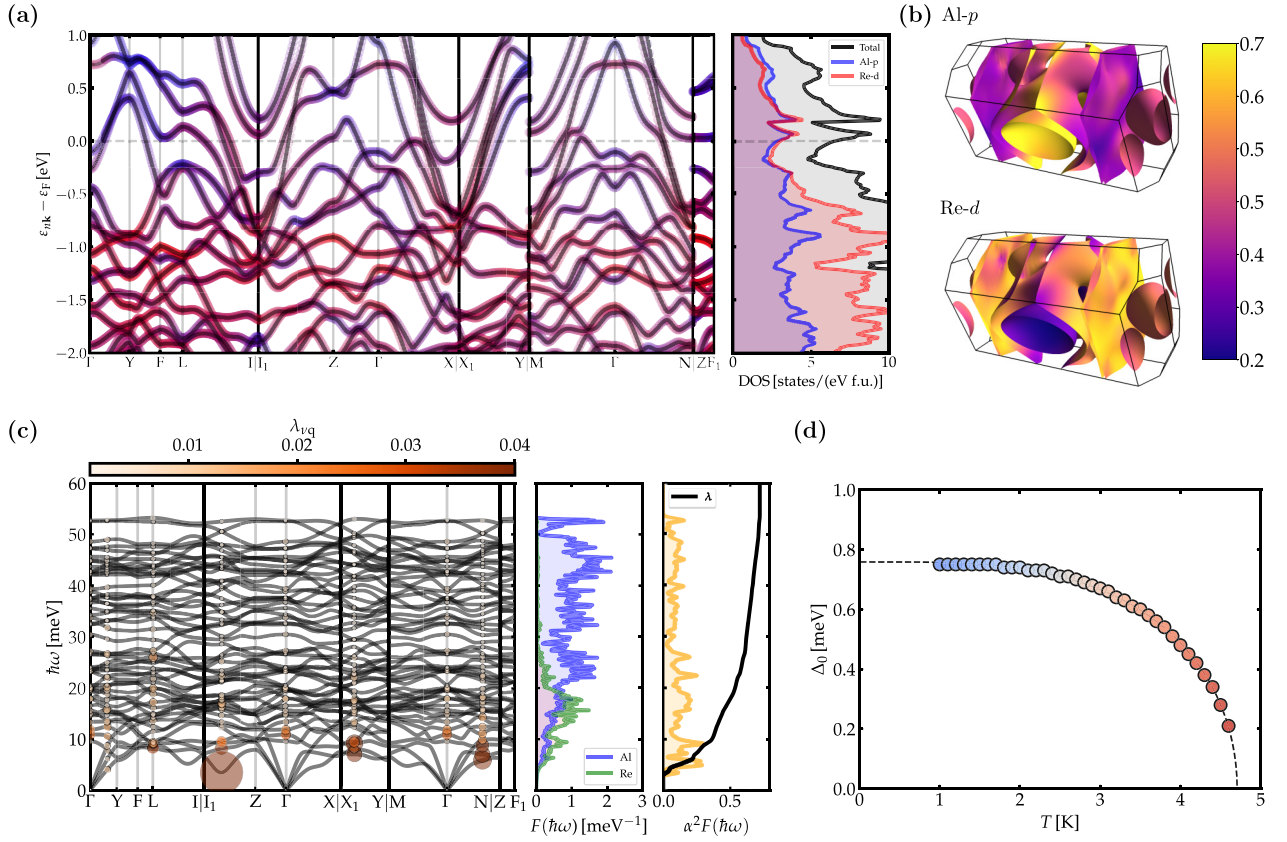


FIG. 5. Electronic, el-ph and superconducting properties of $\text{Al}_{13}\text{Re}_4$. (a) Orbital-projected band structure, total electronic density of states (black, light fill), and orbital-resolved partial DOS for Al- p (blue) and Re- d (red). Each marker's hue in the electronic dispersion is a combination of the colors assigned to the contributing orbitals, with coefficients given by their fractional projections at that state; the marker opacity encodes the total projected weight. (b) Fermi surface projected onto the Al- p and Re- d states. (c) (Left) Phonon dispersion along the same high-symmetry path. Superimposed circles indicate the mode-resolved el-ph coupling λ_{vq} at selected q points; colors and marker size correlate with the values of λ_{vq} . (Middle) Atom-projected phonon density of states $F(\hbar\omega)$ for Al (blue) and Re (green). (Right) Eliashberg spectral function $\alpha^2F(\hbar\omega)$ (orange line) and the cumulative total el-ph coupling parameter λ . (d) Superconducting gap Δ_0 as a function of temperature obtained by solving the isotropic Migdal-Eliashberg equations within the full-bandwidth approximation.

With this in mind, we investigate whether T_c in the $\text{Al}_{13}\text{Os}_4$ AC can be fine-tuned by electron or hole doping via Ir or Re alloying. We then computed the total and partial DOS at ε_F for the solid solutions $\text{Al}_{13}\text{Os}_{4-x}\text{Ir}_x$ and $\text{Al}_{13}\text{Os}_{4-x}\text{Re}_x$, using the GQCA ensemble average defined in Eq. (6). The results are shown in Fig. 4.

Ir doping suppresses the total DOS at ε_F , which would be detrimental to SC. Furthermore, we found that within the harmonic approximation $\text{Al}_{13}\text{Ir}_4$ is dynamically unstable, exhibiting imaginary phonon modes around various high-symmetry points of the BZ (see Fig. 4 within the Supplemental Material [44]). We further verified that these instabilities persist under hydrostatic pressure up to 10 GPa. Thus, $\text{Al}_{13}\text{Ir}_4$ is unlikely to form, and the solid solution $\text{Al}_{13}\text{Os}_{4-x}\text{Ir}_x$ is expected to be feasible only over a narrow composition range near the Os-rich end.

In contrast, the $\text{Al}_{13}\text{Os}_{4-x}\text{Re}_x$ solid solution provides a promising route to tune SC in Al-TM ACs. Our calculations show that N_{ε_F} increases with Re substitution, reaching 6.57 states/eV/f.u. in $\text{Al}_{13}\text{Re}_4$, where Al- p and Re- d states contribute nearly equally at ε_F . $\text{Al}_{13}\text{Re}_4$ is also predicted to be dynamically stable. Since both end members are stable and the mixing thermodynamics are favorable, $\text{Al}_{13}\text{Os}_{4-x}\text{Re}_x$ is

a plausible candidate for a homogeneous, continuous solid solution. In light of the enhanced N_{ε_F} , which boosts λ , this system is expected to exhibit a higher T_c than pristine $\text{Al}_{13}\text{Os}_4$.

Superconductivity in $\text{Al}_{13}\text{Re}_4$. To assess this scenario, Fig. 5 summarizes the electronic structure, phonons, el-ph coupling, and superconducting properties of $\text{Al}_{13}\text{Re}_4$. In this compound, five bands cross ε_F and the FS now includes an enlarged Al- p -derived sheet alongside hybridized sheets. This configuration naturally accommodates an anisotropic distribution of the superconducting gap over the FS or quasiparticle excitations with well-separated gap energies on different sheets. Consequently, $\text{Al}_{13}\text{Os}_{4-x}\text{Re}_x$ may exhibit a composition-tuned crossover from a single- ($\text{Al}_{13}\text{Os}_4$) to a multigap ($\text{Al}_{13}\text{Re}_4$) superconducting state.

The phonon spectrum of $\text{Al}_{13}\text{Re}_4$ closely resembles that of $\text{Al}_{13}\text{Os}_4$, and λ_{qv} is likewise well distributed throughout the BZ, except for a single soft branch along I_1-Z composed of mixed in-plane and out-of-plane vibrations that couples strongly with the electrons at ε_F . Since it occupies only a small fraction of $\mathbf{q}$ -space, this branch does not produce a pronounced peak in the Eliashberg spectral function, and the overall electron-phonon coupling retains a distribution

comparable to its Os analog. Assuming isotropic s -wave pairing, we predict $T_c = 4.7$ K, about 30% higher than that of $\text{Al}_{13}\text{Os}_4$.

Building on experimental evidence that the T_c of QCs closely follows that of their crystalline ACs, the Al–Re QC corresponding to $\text{Al}_{13}\text{Re}_4$ is therefore expected to exhibit a high T_c among QCs reported to date. Notably, introducing small amounts of osmium could further enhance the T_c of $\text{Al}_{13}\text{Re}_4$ by maximizing N_{EF} .

III. SUMMARY

In summary, our work demonstrates that fully *ab initio* calculations accurately capture the superconducting state of the newly discovered $\text{Al}_{13}\text{Os}_4$ AC. We reproduce its bulk T_c and Δ_0 , providing a first theoretical determination of T_c for an AC. The close agreement with experiment offers strong evidence that the SC phase originates from a conventional el–ph coupling mechanism.

Guided by this el–ph framework, we identify $\text{Al}_{13}\text{Os}_{4-x}\text{Re}_x$ as a promising platform for enhanced superconducting properties; notably, the end member $\text{Al}_{13}\text{Re}_4$ is dynamically stable and is predicted to raise T_c by $\approx 30\%$ over $\text{Al}_{13}\text{Os}_4$. We further argue that T_c in ACs provides a practical baseline for their quasicrystalline relatives, highlighting the QCs related to $\text{Al}_{13}\text{Os}_4$ and $\text{Al}_{13}\text{Re}_4$ as promising candidates for the high- T_c quasicrystalline superconductivity.

Together, these results demonstrate the computational feasibility of applying state-of-the-art Eliashberg theory to complex, large-unit-cell approximants. By successfully navigating the high computational cost associated with these systems, we provide a template for using ACs as predictive proxies. This work, therefore, provides a basis for a systematic search for new superconducting QCs by leveraging the translational symmetry of their approximant counterparts to probe the local physics and el–ph pairing of the quasiperiodic state.

IV. METHODS

DFT. We performed density functional theory (DFT) [80,81] calculations using the QUANTUM ESPRESSO (QE) package [82,83]. Scalar-relativistic optimized norm-conserving Vanderbilt (ONCV) pseudopotentials [84,85] were taken from the PSEUDODOJO library [86]. Exchange–correlation effects were treated within the generalized gradient approximation (GGA) using the Perdew–Burke–Ernzerhof (PBE) parametrization [87]. Self-consistent-field (SCF) cycles were converged to a threshold of 10^{-10} Ry, with a plane-wave kinetic-energy cutoff of 80 Ry. Brillouin-zone sampling employed a Γ -centered Monkhorst–Pack $\mathbf{k}$ -point grid [88] of $8 \times 8 \times 12$ and a Methfessel–Paxton smearing [89] of 0.04 Ry. This setup yields total-energy convergence well below 5 meV/atom. Lattice parameters and atomic positions were fully relaxed until changes in total energy and residual forces were smaller than 10^{-7} Ry and 10^{-6} Ry/ a_0 , respectively.

Phonons and el–ph coupling. Phonons and el–ph coupling were computed within density-functional perturbation theory (DFPT) [90].

To improve the convergence of the superconducting properties with respect to the Gaussian smearing width and grid sizes, we introduce a correction factor to $\alpha^2 F(\hbar\omega)$, as described in Ref. [91],

$$\alpha^2 F_r(\hbar\omega) = \frac{N_{\text{EF}}^{\text{tetra}}}{N_{\text{EF}}^{\text{smear}}} \alpha^2 F(\hbar\omega). \quad (2)$$

Here, $N_{\text{EF}}^{\text{tetra}}$ is a high-quality, well-converged density of states at the Fermi level computed with the tetrahedron method on a fine $\mathbf{k}$ -point grid, $N_{\text{EF}}^{\text{smear}}$ is the DOS at the Fermi level obtained with the Gaussian smearing used in the el–ph calculations, and $\alpha^2 F_r(\hbar\omega)$ is the rescaled Eliashberg spectral function. We find this rescaling to be important for improving the convergence of the el–ph coupling [91].

To obtain the total el–ph coupling parameter λ , we integrated $\alpha^2 F_r(\hbar\omega)$ over the phonon spectrum,

$$\lambda = 2 \int d\omega \frac{\alpha^2 F_r(\hbar\omega)}{\omega}. \quad (3)$$

The electron–phonon matrix elements were computed on homogeneous $2 \times 2 \times 3$ (six irreducible q points) and $3 \times 3 \times 4$ (14 irreducible q points) q -point grids, which were subsequently combined to obtain the results shown in Fig. 3 on a regular grid containing 19 irreducible q points in the BZ. We verified, however, that the electron–phonon coupling and superconducting properties of $\text{Al}_{13}\text{Os}_4$ are already well converged even on a $2 \times 2 \times 2$ uniform Γ -centered q -point grid with a reduced k -point sampling for the electronic states.

The phonon self-consistency threshold was set to 10^{-16} Ry. Electron–phonon matrix elements were integrated over a denser $24 \times 24 \times 36$ $\mathbf{k}$ -grid using 50 double-delta smearing values in the range 0.001–0.05 Ry; unless noted otherwise, the reported results correspond to 0.05 Ry together with a phonon smearing of 0.25 THz for the $\mathbf{q}$ -grid integration.

The isotropic Migdal–Eliashberg equations within the full-bandwidth approximation [64] were solved using the ISOME code [68]. The Matsubara frequency cutoff was set to 7000 meV. In variable DOS computations, the chemical potential was updated continuously. An energy cutoff of 5000 meV was applied to all quantities, except for the energy shift and μ update, where a lower cutoff of 2000 meV was used.

GQCA. Disorder and alloying were treated within the generalized quasichemical approximation [76,77,92]. In the GQCA formalism, an alloy is represented as an ensemble of nonequivalent supercells spanning the entire compositional range. These supercells are assumed to be both energetically and statistically independent of their local atomic environments, while remaining spatially homogeneous on a macroscopic scale. Within this approximation, the probability of occurrence of a given supercell at temperature T and composition x is given by

$$p_j = \frac{g_j \eta^{n_j} e^{-\Delta_j/k_B T}}{\sum_{j=1}^J g_j \eta^{n_j} e^{-\Delta_j/k_B T}}, \quad (4)$$

where

$$\eta = \frac{x e^{\lambda_L/k_B T}}{1 - x}. \quad (5)$$

Here, g_j denotes the supercell degeneracy, Δ_j is the excess energy per permutable site relative to the end members of the solid solution, and n_j is the number of dopant atoms in supercell j . Once the probabilities $p_j(x, T)$ are determined by solving the n th order polynomial arising from free-energy minimization, any composition- and temperature-dependent property $P(x, T)$ accessible from first-principles can be obtained as the ensemble average

$$P(x, T) = \sum_{j=1}^J p_j(x, T) P_j, \quad (6)$$

where P_j is the property associated with supercell j . A comprehensive description of the GQCA formalism and derivation of the equations are provided in Ref. [77].

Supercells were generated with the SUPERCELL program [93].

Visualization. Fermi surfaces were visualized using FERMI SURFER [94]. Crystal structures were visualized with VESTA [95]. The high-symmetry path in the first Brillouin zone was generated with XCRYSDEN [96].

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P.N.F. performed the DFT, DFPT, and GQCA calculations and wrote the main draft. R.L. did additional calculations. S.L. and L.N. analyzed the QC tilings. W.E.P. and C.J.P. participated in the discussions. M.J., R.P.P., P.K., and C.H. supervised this project. All authors participated in the discussions and revised the manuscript.

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

DATA AVAILABILITY

The data that support the findings of this article are openly available [97].

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- $$\frac{1}{\mu_{\text{ME}}^*} = \frac{1}{\mu_{\text{AD}}^*} + \ln\left(\frac{\omega_{\text{ph}}}{\omega_{\text{c}}}\right),$$
- where ω_{ph} is the characteristic cutoff frequency for the phonon-induced interaction and ω_{c} is the Matsubara frequency cutoff.
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