

Computational prediction of strontium carbide phases under compression

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Exploring the chemistry of materials at high pressures enables the discovery of previously unknown, but frequently exotic, compounds. We systematically searched for all thermodynamically stable Sr-C compounds under pressure (up to 100 GPa) using the *ab initio* evolutionary crystal structure prediction method. Our search led to the discovery of hitherto unknown phases of SrC₃, Sr₂C₅, Sr₂C₃, Sr₂C, Sr₃C₂, and SrC. The discovered crystal structures feature a variety of different carbon environments ranging from isolated C anions and C dimers to exotic polyatomic carbon anions, including chains, stripes, and infinite ribbons consisting of pentagonal C₅ and hexagonal C₆ rings. The dynamical stability of all predicted compounds is confirmed by phonon calculations. Bader analysis unravels very diverse chemistry in these compounds, and bonding patterns in some of them can be described using the Zintl-Klemm rule.

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I. INTRODUCTION

Chemical reactions that defy conventional chemical intuition can happen under extreme conditions, resulting in the emergence of rich phase diagrams and materials possessing intriguing properties since pressure is known to greatly affect the chemistry of elements and cause the formation of exotic compounds [1,2]. Well-known examples of such reactions involve the formation of sodium chlorides [3], a stable compound of helium and sodium [4], platinum group metal nitrides [5], and superconducting metal hydrides [6]. Nowadays, experimental high-pressure techniques allow one to explore materials chemistry up to terapascal pressure [7] and hence explore theoretical predictions of exotic (from the chemical perspective) compounds. State-of-the-art computational techniques are reliable in predicting which compounds might be formed under certain thermodynamic conditions [8,9]. For example, evolutionary crystal structure prediction has led to the *in silico* discovery of transparent dense sodium, unexpected forms of sodium chloride, and various superconducting metal hydrides, which were later confirmed experimentally [3,10,11].

Among all compounds previously investigated at high pressure, particular attention was given to carbon-containing materials. Carbon itself has the ability to form *sp*-, *sp*²-, and *sp*³-hybridized bonds and therefore exists in various allotropic forms, including known structures of graphite, diamond, graphene, fullerenes, and nanotubes [12–19]. High

pressure alters the bonding patterns of carbides and leads to the formation of compounds with unusual carbon environments [20–22]. A large number of metal carbides contain C₂ dimers (acetylide group) [23–28]; less common is the C₃ unit (allylenide group) [29–32]. For the binary systems Mg-C [33], Ca-C [34,35], Y-C [36–38], and La-C [20], *ab initio* structure searches predicted the formation of unusual metal carbides with exotic C₄, C₅ units, C₆ rings, and graphene carbon sheets.

Despite the thorough exploration of the chemical space of metal carbides in the past, recent experiments revealed several novel compounds in the Y-C and Ca-C systems [35,37]. Motivated by these findings, we decided to construct a pressure-composition phase diagram of the Sr-C system. Compressed strontium shows unique structural and electronic properties [39,40], but to the best of our knowledge, strontium carbides were not investigated under compression. Using a variable-composition structure prediction methodology, we explored the pressure-composition phase diagram of the Sr-C system in order to fully understand the structural diversity and evolution of the C–C bonding types under high pressure in the Sr-C system. This resulted in several predicted stable stoichiometries (SrC₃, Sr₂C₅, Sr₂C₃, Sr₂C, Sr₃C₂, and SrC) with a diverse set of carbon anions: isolated C atoms in Sr₂C, dimers in Sr₃C₂ and SrC, linear trimers in Sr₂C₃, chains in SrC, stripes with five-membered (C pentagons) rings in SrC₂. There are also infinite ribbons consisting of five-membered (C pentagons) rings in SrC₃, ribbons consisting of C pentagons in SrC₂, and ribbons made of C pentagons and C hexagons in Sr₂C₅. Here, we discuss the structures and some properties of these compounds.

II. COMPUTATIONAL METHODOLOGY

Crystal structure prediction techniques allow one to determine pressure-dependent phase diagrams for a given

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chemical space and a set of thermodynamic conditions. Such methodology, called a variable-composition search, is not limited to the prediction of the ground state structure of a particular stoichiometry and explores the whole chemical space of the system in a single calculation [41]. Recently, by exploring pressure-composition phase diagrams of Cu-F, Ag-F, and Nd-F, we showed that a variable-composition evolutionary crystal structure search allows one to find new stable compounds even after fixed-composition searches are done [11,42,43].

Here, we used two crystal structure prediction codes which both implement the idea of evolutionary crystal structure prediction. One is the field-leading crystal structure prediction package USPEX [44–46]. The other is a python-based implementation of the *ab initio* evolutionary algorithm which has the functionality to perform variable-composition search. We call this implementation SPUTNIK (structure prediction using theoretical kristallography), and details of the implementation will be published elsewhere, but we have to mention that workflow management in SPUTNIK is done using the fireworks functionality [47]. The theoretical basis under the SPUTNIK’s implementation is identical to USPEX’s as presented in [41,45,48].

The evolutionary searches were combined with structure relaxation and energy calculations using density-functional theory (DFT) within the Perdew-Burke-Ernzerhof [49] exchange-correlation functional. In the case of SPUTNIK, DFT calculations were performed using the plane-wave quantum ESPRESSO (qe) package [50,51], while in the case of USPEX, we used VASP (Vienna Ab initio Simulations Package) [52] with the projector augmented wave method [53]. We used standard VASP pseudopotentials with four and ten valence electrons for C and Sr, respectively (pseudopotentials C and Sr_sv in VASP notation). In the case of qe we used pseudopotentials from the gbrv library [54] for Sr and pslibrary [55] (version 1.0) for C following the suggestions of the benchmark performed in [56]. For the variable-composition structure search, the first generation of 160 structures was composed of randomly generated unit cells containing up to 18 atoms (see the details of the random structure generation procedure in the associated papers [46,57]). Seventy percent of the next generation was obtained by applying variation operators (heredity, soft mutation, lattice mutation) to 70% of the lowest-energy structures in the previous generation, while the remaining 30% of the generation was generated randomly. The percentage of structures produced by each of the variation operators was dynamically adjusted on the fly based on the performance of each operator [58]. During the structure search, each generation of structures was relaxed through a series of steps with increasingly more stringent calculation parameters (plane-wave cutoff, k -point density, electronic density convergence threshold). In the last step, we performed single-point calculations using a plane-wave energy cutoff of 60 Ry (in the case of VASP the kinetic energy cutoff was set to 600 eV) and Γ -centered k meshes with a reciprocal space resolution of $2\pi \times 0.05 \text{ \AA}^{-1}$ for the Brillouin zone sampling. In these calculations, no constraints on stoichiometries were imposed as long as the unit cell had no more than 18 atoms, and calculations were performed at 10, 25, 50, 75, and 100 GPa.

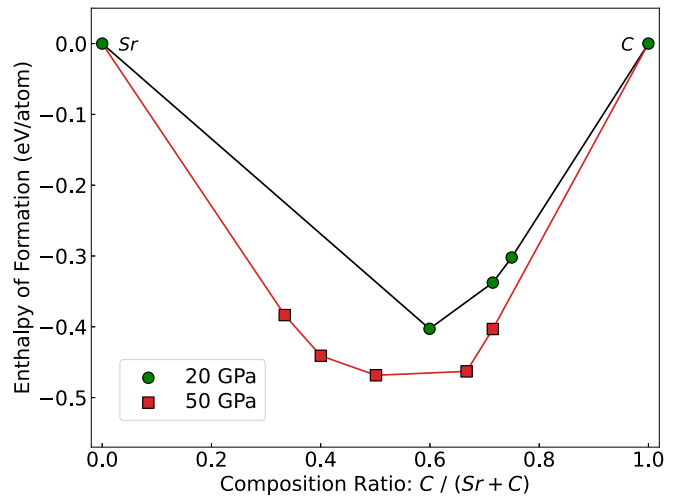


FIG. 1. Convex hull of the Sr-C system at 20 GPa (green disks) and 50 GPa (red squares). The enthalpy of formation H is defined as $\Delta H(\text{Sr}_x\text{C}_y) = [H(\text{Sr}_x\text{C}_y) - xH(\text{Sr}) - yH(\text{C})]/(x + y)$.

Once structure prediction with SPUTNIK and USPEX was done, we calculated the pressure-composition phase diagram with a resolution of 10 GPa; then we confirmed the dynamical stability of all novel structures with phonon calculations using the supercell approach and the finite displacement method [59], as implemented in the phonopy package [60]. The electron localization function (ELF) was calculated for all the novel phases using VASP and visualized using vesta [61]. Bader charge analysis was performed on the total charge density [62–65]. Structural information for all discovered compounds, band structures, and phonon dispersion curves are presented in the Supplemental Material (SM) [66].

III. RESULTS

To determine stable phases in a variable-composition system, it is convenient to use the convex hull construction. Phases located on the thermodynamic convex hull are stable with respect to decomposition into elemental Sr and C or other Sr-C compounds. All the values of the chemical potentials, delimiting the fields of stability of compounds, are derived directly from the convex hull. Consequently, the convex hull construction is the cornerstone of the variable-composition structure prediction, which determines the stability of compounds at particular thermodynamic conditions [41].

Convex hulls of the Sr-C system at 20 and 50 GPa are presented in Fig. 1. The pressure-composition phase diagram of the Sr-C system is shown in Fig. 2, which was obtained by calculating the enthalpies of the most stable structures for each composition at a given pressure (steps of 5 GPa were used). According to this diagram, we found ten hitherto unknown phases in the Sr-C system. These compounds have the following stoichiometries: SrC_3 , Sr_2C_5 , SrC_2 , Sr_2C_3 , SrC , Sr_3C_2 , and Sr_2C (for some stoichiometries there are a couple of different structures). These structures are shown in Fig. 3. For all predicted structures, the calculated phonon dispersion relations confirmed their dynamical stability (see SM Fig. 8).

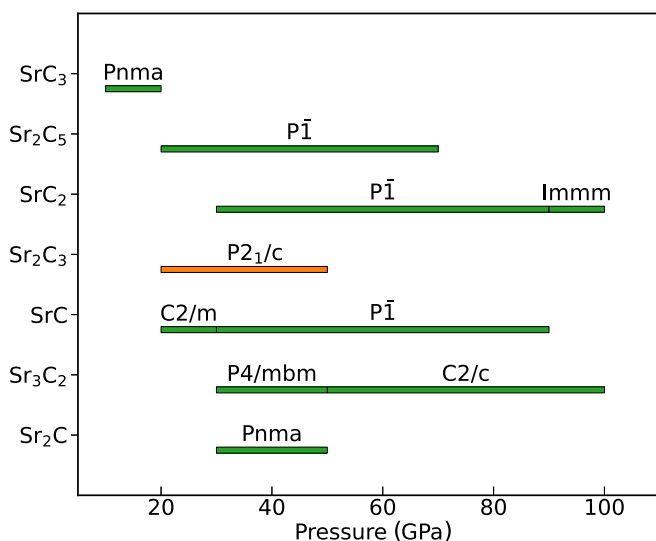


FIG. 2. Pressure-composition phase diagram for the Sr-C system. Sr_2C_3 is the only semiconducting system and is highlighted in orange, while all other systems are metallic (highlighted in green).

The identified compounds have a variety of different carbon motifs, as demonstrated in Fig. 4. In the following, we consider the predicted phases in order of increasing carbon content. In order to analyze these structures, we recall that the length of the C–C bond depends on the bond order, and at 1 atm these lengths are 1.20 Å for the triple C–C bond, 1.33 Å for the double C–C bond, and 1.54 Å for the single C–C bond. Combining this knowledge with the results of Bader analysis, we unravel a very diverse chemistry.

According to our calculations Sr_2C crystallizes in the orthorhombic *Pnma* space group. This structure is stable from 30 to 50 GPa, is metallic, and contains isolated carbon atoms—the simplest carbon motif among all compounds obtained, as shown in Fig. 3. The Sr atom is surrounded by four carbon atoms arranged in a tetrahedral environment. Such stoichiometry was previously found in other metal-carbon compounds, for example, Ca_2C and Mg_2C [34,67]. However, there are no reported theoretical or experimental data on Sr_2C . This metallic phase [see SM Fig. 9(a)] can be represented as a methanide with an idealized charge distribution of $(\text{Sr}^{2+})_2\text{C}^{4-}$ and is in line with the Zintl concept.

Metallic Sr_3C_2 crystallizes in the tetragonal *P4/mbm* space group at 30 GPa and transforms into a structure with a monoclinic *C2/c* space group at 50 GPa. The structure of the identified Sr_3C_2 is of the U_3Si_2 type, a common arrangement found in silicides [68], borides [69], and intermetallic compounds [70]. Both structures are metallic and contain carbon dimers as presented in Figs. 3(b) and 3(c). Calcium carbides with this stoichiometry and space groups were previously predicted [34]. In the *P4/mbm* Sr_3C_2 phase, the structure contains C_2 groups (the C–C distance is 1.39 Å at 30 GPa) carrying an ideal charge of -6 , receiving two electrons from three Sr atoms each. This leaves three electrons per C atom to establish C–C double bonds in the metallic framework. A carbon atom with three additional electrons becomes isoelectronic with a fluorine atom, similar to the configuration found in the singly bonded F_2 molecule (where the length of the F–F

bond is 1.42 Å). The Sr atoms are located in the same plane as the C_2 dumbbells, collectively forming the Cairo pentagonal tiling, which consists of $(\text{Sr})_2\text{C}_3$ pentagons [see Fig. 5(a)]. As shown in Fig. 5(a), Sr atoms are fourfold coordinated by C atoms, with a Sr–C distance of 2.63 Å at 30 GPa. A similar coordination environment was previously reported in dysprosium carbide, *P4/mbm* Dy_3C_2 [71]. The ELF indicates a strong covalent bond between the carbon atoms in the dimers and an ionic Sr–C interaction [see Fig. 5(c)]. Above 50 GPa *P4/mbm* Sr_3C_2 transforms into *C2/c* Sr_3C_2 , and the Cairo pentagonal tiling becomes distorted. This distortion is accompanied by an increase in the C–C bond length of the C dimers to 1.46 Å. Electron density maps in the plane containing the C_2 dimers reveal slightly higher ELF values between carbon atoms in the dimers of the *P4/mbm* Sr_2C_3 phase compared to the *C2/c* Sr_2C_3 phase (see SM Fig. 13). This suggests a higher bond order in the *P4/mbm* Sr_2C_3 dimer.

Metallic SrC exhibits two thermodynamically stable phases below 100 GPa. The low-pressure monoclinic *C2/m* phase on further compression transitions into a low-symmetry triclinic $\bar{P}1$ SrC structure. At 30 GPa the structural formula of *C2/m* is $\text{Sr}_2(\text{C}_2)$, featuring a doubly bonded C_2 group with a C–C bond length of 1.34 Å. The Sr atom is coordinated with seven carbon atoms arranged in a heptagonal environment. This C_2 group carries an ideal charge of -4 (Bader charge of -2.175), perfectly balancing the charge of two strontium atoms, and *C2/m* SrC is classified as an ethylenide. The $\bar{P}1$ SrC structure is particularly intriguing due to its novel zigzag C chain with C–C bond lengths ranging from 1.45 to 1.50 Å, implying bond orders between 1 and 2. The bond angles are 125.6° and 121.3° , which are remarkably close to the 120° expected for sp^2 -hybridized carbon. The ELF maps clearly indicate strong covalent bonding between the carbon atoms and ionic bonding between Sr and C atoms (see SM Fig. 12).

At pressures of about 20 GPa Sr_2C_3 becomes stable and crystallizes in the *P21/c* space group. Its structure contains C_3 units (allylenide groups). This structure is the only semiconducting compound found in this study (see SM Fig. 9). Usually, C_3 trimers are linear $[\text{C}=\text{C}=\text{C}]^{4-}$ groups with a C–C bond length of 1.34–1.35 Å (e.g., in Mg_2C_3), which is close to that in gaseous allene (1.335 Å) [72]. The structure of *P21/c* Sr_2C_3 at 20 GPa consists of Sr_2 layers connected by nearly linear symmetric C_3 groups featuring double C–C bonds, with C–C distances measuring 1.31 Å. The Sr atom is surrounded by seven carbon atoms arranged in a heptagonal environment. In this configuration, the ideal charge of the C_3 group is -4 (Bader charge of -2.62), which precisely balances the total charge of the two Sr atoms in the formula, with one Sr atom having a $+1.31$ Bader charge. The central carbon atom in the C_3 group should be neutral, and the terminal carbons should have -2 charges as in gaseous allene. The terminal carbon atoms with -2 charges are isoelectronic with oxygen atoms. Therefore, the linear $[\text{C}=\text{C}=\text{C}]^{4-}$ group can structurally behave as $[\text{O}=\text{C}=\text{O}]$, according to the Zintl-Klemm rule. However, the central atom of the C_3 unit in Sr_2C_3 exhibits a higher negative Bader charge of -1.098 , while the terminal carbon atoms show lower Bader charges of -0.761 (see SM Table 1). This discrepancy is attributed to the significant ionic interactions between the Sr atom and the central carbon atom of the C_3 unit. This can be seen in the

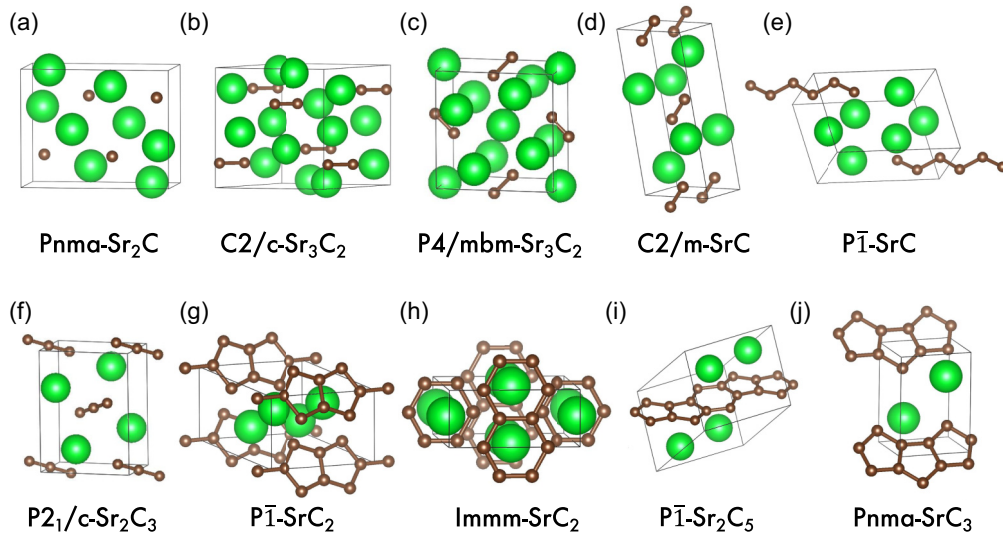


FIG. 3. Crystal structures of the compounds predicted in this work. The green and brown spheres represent strontium and carbon atoms, respectively.

calculated two-dimensional (2D) ELF of these structures (see SM Fig. 10).

Metallic SrC_2 becomes stable at 30 GPa and crystallizes in the low-symmetry $P\bar{1}$ space group. This structure contains exotic carbon stripes with fused five-membered C rings; the structural motif is presented in Fig. 4(e). In this structure, two layers of Sr atoms are stacked between two layers of these stripes. The C–C distances within the C rings are in the range from 1.41 to 1.48 Å at 30 GPa, indicating bonds that

are intermediate between single and double. Upon compression, at 90 GPa the phase transition $P\bar{1} \text{SrC}_2 \rightarrow Immm \text{SrC}_2$ occurs. The $Immm \text{SrC}_2$ structure contains extended carbon ribbons consisting of fused hexagonal C rings, as presented in Fig. 4(f). These carbon ribbons are planar, with C–C distances of 1.43 and 1.44 Å at 90 GPa. The $Immm \text{SrC}_2$ phase has the same structure as the previously observed high-pressure $Immm \text{CaC}_2$ [34] and the predicted high-pressure $Immm \text{YC}_2$ [28]. In the low-symmetry $P\bar{1} \text{SrC}_2$ phase, the strontium atom is surrounded by 12 carbon atoms that form a heptagon and a pentagon above and below the Sr atom [see Fig. 6(a)]. In the $Immm \text{SrC}_2$ phase, the strontium atoms have 12 carbon neighbors arranged in a hexagonal prism with unequal bases [see Fig. 6(c)]. As mentioned above, these C atoms are

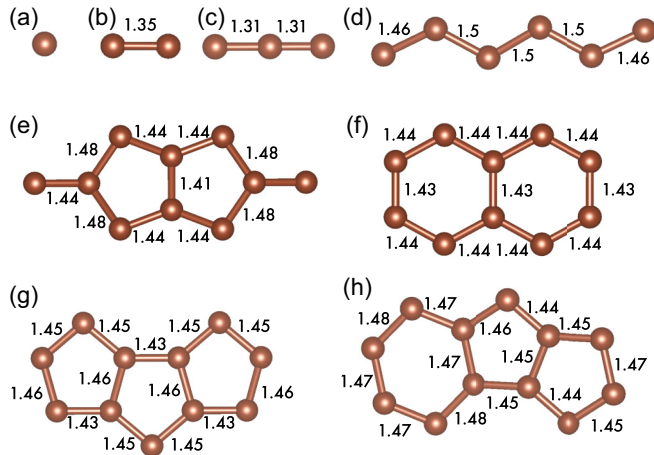


FIG. 4. Carbon patterns in the Sr–C system. Distances between atoms are measured in angstroms. (a) The isolated carbon anions in $Pnma \text{Sr}_2\text{C}$. (b) Carbon dimers (dumbbells) observed in the $C2/c \text{Sr}_3\text{C}_2$ and $C2/m$ structure of SrC (the structures were optimized at 50 and 25 GPa, respectively). (c) The carbon trimer presented in $P2_1/c \text{Sr}_2\text{C}_3$ at 50 GPa. (d) Zigzag C_6 groups observed in $P\bar{1} \text{SrC}$ at 50 GPa. (e) Carbon stripes in $P\bar{1} \text{SrC}_2$ at 50 GPa. (f) Carbon ribbons with hexagonal C rings in $Immm \text{SrC}_2$ at 90 GPa. (g) Carbon ribbons with pentagonal C rings in $Pnma \text{SrC}_3$ at 20 GPa. (h) Carbon ribbons with pentagonal and hexagonal C rings in $P\bar{1} \text{Sr}_2\text{C}_5$ at 50 GPa.

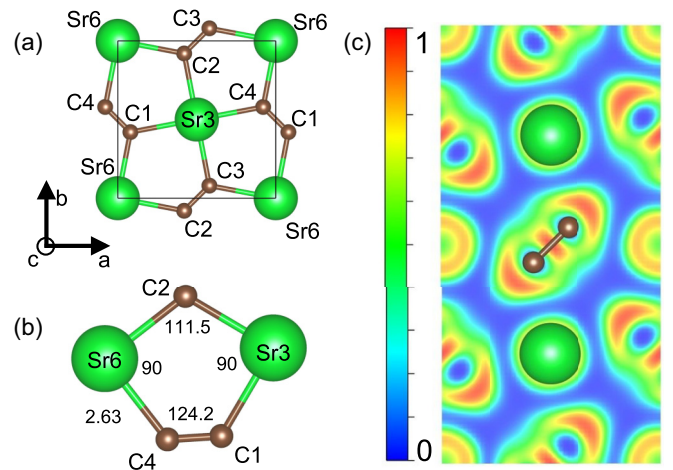


FIG. 5. (a) The Sr–C plane in the $P4/mbm \text{Sr}_3\text{C}_2$ structure contains the Cairo pentagonal tiling formed by Sr and C atoms with fourfold coordination of the Sr atom. (b) Angles (in degrees) and Sr6–C4 bond length (in angstroms) within the Sr–C plane of the $P4/mbm \text{Sr}_3\text{C}_2$ structure optimized at 30 GPa. (c) The 2D ELF of $P4/mbm \text{Sr}_3\text{C}_2$.

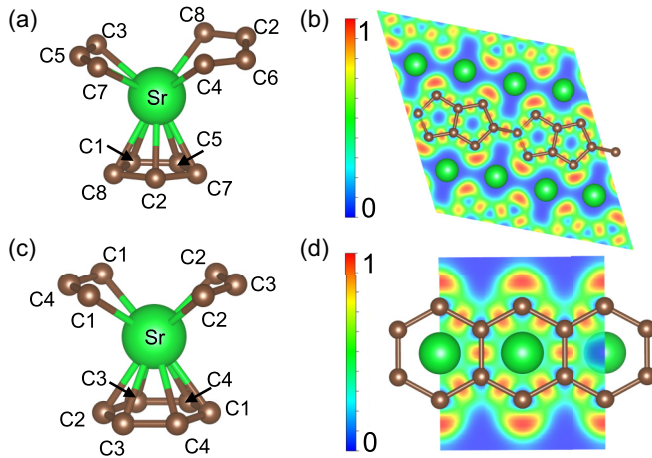


FIG. 6. The crystal structure of SrC_2 depicted along with the computed ELF. Green and brown spheres represent strontium and carbon atoms, respectively. (a) Coordination of Sr atoms by C atoms in $P\bar{1}$ SrC_2 at 30 GPa, involving a pentagonal base, (b) 2D ELF containing the pentagonal ribbons in the $P\bar{1}$ SrC_2 phase, (c) coordination environment of the Sr atom in the $Immm$ phase of SrC_2 at 90 GPa, and (d) ELF containing the carbon ribbons in the $Immm$ SrC_2 phase.

organized into polycyclic units of hexagonal rings, forming planar ribbons, and as a result, the carbon atoms create exotic one-dimensional extended polyanions. The hexagon features two angles of 122.5° and four angles of 118.8° . These angles, being close to 120° , along with the similar C–C distances, suggest that the carbon atoms are sp^2 hybridized. The shared edges of hexagons in $Immm$ SrC_2 are slightly shorter (by approximately $0.01 \text{ \AA}$) than the nonshared edges. This observation aligns with calculations for previously predicted isostructural high-pressure carbides (such as DyC_2 , CaC_2 , and YC_2) which showed shorter shared edges by about 0.02 – $0.03 \text{ \AA}$ [27]. Bader charge analysis suggests charge transfer from cationic Sr to C atoms (see SM Table 1). ELF maps of carbon ribbons shown in Figs. 6(b) and 6(d) suggest strong covalent bonds between carbon atoms and show localization of electrons on C–C bonds in pentagonal ribbons and hexagons.

Sr_2C_5 crystallizes in the low-symmetry space group $P\bar{1}$. C atoms in this structure feature extended ribbons consisting of C pentagons and C hexagons, as shown in Figs. 4(g) and 4(h). The C–C bond lengths range from 1.39 to $1.44 \text{ \AA}$, indicating an intermediate bond order between single and double C–C bonds. In the $P\bar{1}$ Sr_2C_5 structure, one Sr atom has a $+1.248$ Bader charge and is 13-fold coordinated, while another Sr atom has a $+1.202$ Bader charge and is 12-fold coordinated [see Fig. 7(a)]. In the fused C hexagon and C pentagon, carbon atoms are negatively charged. Therefore, it is evident that there is charge transfer from Sr to C atoms. The Sr atom with 13-fold coordination is surrounded by the C hexagon and a heptagon, while the Sr atom with 12-fold coordination is surrounded by the C pentagon and a heptagon. In the C hexagons, the four inner C atoms have three C–C covalent bonds each, while the two outer C atoms have two C–C covalent bonds and a localized nonbonding electron density each. The C–C bonds in the C hexagon form two different angles, α and β , where $\alpha = 112^\circ$ and $\beta = 124^\circ$. In C pentagons, the inner four C atoms have three C–C covalent

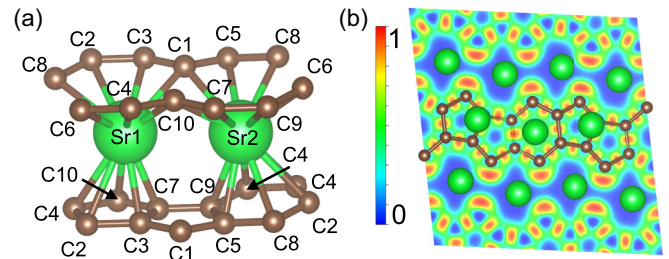


FIG. 7. (a) Coordination environment of the Sr atoms with the fused C hexagons and C pentagons in $P\bar{1}$ Sr_2C_5 at 50 GPa. (b) 2D ELF of the fused extended C ribbons, consisting of C hexagons and C pentagons.

bonds, while the outer C atom has two C–C covalent bonds and a localized nonbonding electron density. The nonbonded electron densities can be seen in the 2D electron localization function [see Fig. 7(b)].

Finally, SrC_3 crystallizes in the $Pnma$ space group at 20 GPa. In this structure, C atoms form infinite C ribbons consisting of pentagonal carbon rings [see SM Fig. 11(a)]. The Sr atom is surrounded by nine carbon atoms with a pentagonal base formed in a ring. The Bader analysis indicates a charge transfer from strontium to carbon atoms, with strontium atoms acquiring a positive Bader charge of $+1.343$. The carbon atoms, in contrast, have negative charges, with those not involved in ring fusion showing higher electron concentration. This is also evident in the 2D ELF plot [see SM Fig. 11(b)].

In light of the appreciable diversity of the observed carbon motifs demonstrated in Fig. 4, it is instructive to check whether at least some of these configurations can be explained using conventional chemical wisdom. For that purpose, we investigated the applicability of the Zintl-Klemm (ZK) concept, originally conceived to rationalize bonding patterns of polyanions, to the identified strontium carbides. To assess the validity of the ZK rules, the average charge of a C anion will be determined by assigning the formal $+2$ charge to the Sr atoms (Sr^{2+}) and imposing charge neutrality of the formal Sr_xC_y unit. In the case of $Pnma$ Sr_2C this would lead to a formal charge of the carbon anion of -4 . Formal charges are used for chemical interpretation within the Zintl-Klemm framework, whereas Bader charges describe the electron density distribution obtained from DFT calculations and therefore should not be expected to reproduce formal charges. By subsequently equating the structural motifs formed by that C anion to the isoelectronic neutral species, which in the case of C^{4-} would be a neon atom, the structure of the polyanion is inferred. Since neon is a noble gas, and Ne–Ne bonds cannot be formed, we expect no C–C bonds in Sr_2C , which we indeed observe. We performed this analysis for all stoichiometries discussed here, which we summarize in Table I. We find that for some of the strontium carbide compounds, namely, Sr_2C , Sr_3C_2 , and SrC , the C anion structure is consistent with the motifs predicted from the ZK rule. This finding is also in agreement with a previous investigation of high-pressure Ca–C phases [34].

However, in the other cases we find that for some stoichiometries, such as SrC_3 and Sr_2C_5 , the ZK concept cannot be directly applied. Double and triple bonds also lead to

TABLE I. Applicability of Zintl-Klemm (ZK) rules to the identified stoichiometries of strontium carbide. In cases where the formal charge was found to be fractional, the Zintl-Klemm rules cannot be applied (N/A) in a straightforward manner.

Compound	Formal C charge	Isoelectronic to	Expected anion structure	ZK rule fulfilled?
Sr_2C	-4	Ne	Monoatomic	Yes
Sr_3C_2	-3	F	Dimer	Yes
SrC	-2	O	Dimer	Yes
Sr_2C_3^a	-2	O	Linear	Yes
SrC_2	-1	N	Trigonal nonplanar	No
SrC_3	-2/3	N/A	N/A	N/A
Sr_2C_5	-2/5	N/A	N/A	N/A

^aThis formal charge assignment arises because the carbon atoms form trimers, where the central atom is charge neutral and the two edge atoms each carry a -2 charge.

exceptions from the ZK rule. A particularly notable example is the case of SrC_2 , in which the carbon species would be isoelectronic to a nitrogen atom. As a consequence, one would expect each carbon atom to form three covalent bonds. As illustrated in Fig. 4, we find that the carbon polyanion in *Immm* SrC_2 consists of planar, naphthalenelike arrangements. The planarity and similarity of all C-C bond lengths in that polyanion suggest that the configuration is aromatic. This implies that most of the carbon atoms form one single bond and one double bond, so that the bonding of these atoms in *Immm* SrC_2 is consistent with the ZK prediction. However, the two carbon atoms shared between the two rings must consequently have two single bonds and one double bond, which is not compatible with the isoelectronic N atom. The same argument applies to the planar structure of the anion in the *P1* phase of SrC_2 , so that both SrC_2 phases are in contradiction to the ZK rule.

IV. CONCLUSION

In summary, we produced a complete pressure-composition phase diagram for Sr-C compounds at pressures

up to 100 GPa. As a result we predicted 11 thermodynamically stable phases in the Sr-C system. Using electronic structure calculations and by performing Bader charge analysis, we unraveled very diverse chemistry in these compounds. Carbon atoms in these compounds form a diverse set of different environments: isolated C atoms in Sr_2C (methanide), dimers in Sr_3C_2 (ethanide) and SrC (ethylenide), linear trimers in Sr_2C_3 (allylenide), chains in SrC , and stripes with five-membered (C pentagons) rings in SrC_2 . There are also infinite ribbons consisting of five-membered (C pentagons) rings in SrC_3 , ribbons consisting of C pentagons in SrC_2 , and ribbons made of C pentagons and C hexagons in Sr_2C_5 . Bonding patterns in some of the obtained compounds can be described using the Zintl-Klemm rule.

We note that while powerful computational methods, such as the evolutionary crystal structure prediction used in this work, are capable of reliably predicting these exotic Sr-C compounds under pressure, experimental validation of the obtained results should be done.

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N.R. and E.M. initiated the study and performed crystal structure prediction calculations. P.G. performed ELF calculations and Bader analysis. A.R.O. and A.S. supervised the research. All authors participated in the writing, reviewing, and editing of the manuscript.

The authors have no conflicts of interest to disclose.

DATA AVAILABILITY

The data that support the findings of this article are openly available [66]. Requests for data or further information should be directed to the authors.

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