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Phonons and superconductivity in the cubic perovskite Cr3RhN
Theoretical examination of superconductivity in the cubic antiperovskite Cr₃GaN under pressure

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We present results of a first-principles investigation of the lattice dynamics and electron-phonon coupling of Cr₃GaN under pressure within a linear response approach based on density functional perturbation theory. It is found that stable phonon modes are maintained throughout the Brillouin zone in the pressure range 0-100 GPa. Our results at zero pressure indicate that the material is a conventional electron-phonon superconductor with intermediate level of coupling strength. It is further found that the decrease in the density of states at the Fermi level and the increase of phonon frequencies under pressure are the main causes for a monotonic decrease of the electron-phonon coupling parameter and the superconductor transition temperature. © 2013 AIP Publishing LLC. [http://dx.doi.org/10.1063/1.4817072]

I. INTRODUCTION

In recent years, the cubic antiperovskite materials have received great attention since the discovery of 8 K superconductivity in MgCNi₃.¹,² This transition is unexpected due to the large Ni content in this material, which may favor a magnetic ground state rather than superconducting state. This raises a question on the effects of magnetism on the superconducting properties of this material. In the experimental work of Rosner et al.,³ it was suggested that magnetic interactions can support superconductivity in this cubic antiperovskite. Later studies found that this material shows an s-wave BCS superconductivity,⁴⁻⁷ and its physical, chemical, and superconducting properties can clearly be understood by considering its electronic structure. This led to several experimental and theoretical efforts to investigate the electronic properties of this cubic antiperovskite. Experimental investigations include photoemission and x-ray absorption measurements.⁸

On the theoretical side, it includes the self-consistent tight-binding linear muffin-tin orbital (TB LMTO),⁹ the linear muffin tin orbital (LMTO) method,¹⁰ the full-potential augmented plane wave (FLAPW) method,¹¹,¹² and density-functional theory within the local-density approximation.¹³⁻¹⁵ These experimental and theoretical studies showed that the significant feature of the electronic structure of MgCNi₃ is an extended van Hove singularity which creates a large density of states (DOS) just below the Fermi level. In order to achieve a better understanding of superconductivity, phonon properties of this material have been studied experimentally as well as theoretically. Time-dependent inelastic neutron-scattering measurements¹⁶ have been used to obtain the generalized phonon density of states. Phonon dispersion curves for this material have been measured by using inelastic x-ray scattering method.¹⁷

Dynamical properties of MgCNi₃ have also been studied by employing different levels of theoretical approaches.¹⁸⁻¹⁹ Among MgCNi₃’s isostructural cubic antiperovskites ACNi₃ (A = Cd, Zn, Al, and Ga), only CdCNi₃ demonstrates superconductivity.²⁰ This has led to studies of the electronic properties of CdCNi₃. Theoretical works indicate that this compound, like MgCNi₃, is almost ferro-magnetic due to the existence of a large peak in the density of states slightly below the Fermi level, which is favourable to superconductivity. Further theoretical work on phonon spectrum, phonon linewidths, and electron-phonon coupling parameter²¹ has confirmed the experimental work that CdCNi₃ is a BCS-type superconductor with electron-phonon interaction of medium strength.

Recently, the discovery of a new superconducting (Tₐ ≈ 3 K) cubic antiperovskite ZnNNi₃ has been reported in the experimental work of Uehara et al.²²,²³ This material is the third Ni-rich antiperovskite series and the first antiperovskite nitride superconducting material. The discovery of superconductivity in ZnNNi₃ has motivated several groups²⁴⁻²⁹ to study the structural and electronic properties of this material. Very recently, attention has been directed to Cr-rich intermetallic antiperovskite materials because many of Cr compounds show superconductivity.³⁰⁻³³ The band structures for Cr₃GaN and Cr₃RN intermetallic antiperovskites have been investigated using the Korringa-Kohn-Rostoker method.³⁴ This theoretical study pointed out the possibility of superconductivity onset in these compounds due to the calculated high values of McMillan-Hopfield parameter (electronic part of electron-phonon coupling constant). However, this work³⁴ did not consider the phonon part of electron-phonon coupling constant. Realising the importance of the phonon influence on the superconducting properties of this material, we have recently performed a first-principles theoretical investigation of the structural, electronic, vibrational, and superconducting properties of the cubic antiperovskite Cr₃RhN.³⁵ This study illustrated that Cr₃RhN is dynamically stable, as no instabilities in the phonon dispersion curves have been found. The calculated superconducting transition temperature Tₐ value of around 17 K for Cr₃RhN (Ref. ³⁵) is much higher than the corresponding value of 8 K for MgCNi₃.

Motivated by our results for Cr₃RhN,³⁵ in this work, we present a study the structural and electronic properties of...
Cr-rich nitride cubic antiperovskite Cr$_3$GaN using the \textit{ab initio} pseudopotential method based on a generalized gradient approximation (GGA) of the density functional theory. Differences in the electronic structure and electronic density of states between Cr$_3$GaN and its isostructural compound Cr$_3$RhN (Ref. 35) are investigated and discussed. We have further carried out \textit{ab initio} linear response calculations of the lattice dynamics, electron-phonon interaction, and polarization characteristics of zone-centre phonon modes. The structural stability of Cr$_3$GaN has been studied under high pressure (from 0 GPa to 100 GPa). We have also presented the variations of the electron-phonon coupling parameter and $T_C$ with increasing pressure. Finally, differences in the Eliashberg function $\xi^2F(\omega)$ and superconducting parameters between Cr$_3$GaN and its isostructural compound Cr$_3$RhN are investigated and discussed.

II. THEORY

In this work, we have carried out first principle calculations based on the density functional theory, as implemented in the QUANTUM ESPRESSO code.\cite{36,37} The electronic exchange-correlation energy interaction has been treated within the GGA, using the Perdew-Burke-Ernzerhof (PBE) functional theory.\cite{38} The electron-ion interaction was described by using ultrasoft pseudopotentials.\cite{39,40} The wave function was expanded in plane waves with the energy cutoff of 60 Ry, and the electronic charge density was expanded with the energy cut off up to 240 Ry. The Kohn-Sham equations were solved using an iterative conjugate gradient scheme to obtain energy. Brillouin zone integration were performed using the $24 \times 24 \times 24$ Monkhorst-Pack $k$-point mesh.\cite{41}

Phonon spectrum and density of states for Cr$_3$GaN were obtained in the framework of the harmonic approximation to the force constants and using the linear response method\cite{36,37} which is realized in the QUANTUM ESPRESSO code.\cite{36,37} Within this scheme, second order derivatives of the total energy were calculated to obtain the dynamical matrix. A static linear response of the valence electrons was considered in terms of the variation of the external potential corresponding to periodic displacements of the atoms in the unit cell. The screening of the electronic system in response to the displacement of the atoms was taken into account in a self consistent manner. Integration up to the Fermi surface is done with the smearing technique with the smearing parameter $\sigma = 0.02$ Ry. For phonon calculations, we have used a $12 \times 12 \times 12\mathbf{k}$ mesh for the Brillouin zone integration. Dynamical matrices have been computed on a $4 \times 4 \times 4\mathbf{q}$ point mesh, and a Fourier interpolation has been used to calculate phonons for any chosen $\mathbf{q}$ point.

For calculating the superconducting properties, the Migdal-Eliashberg approach\cite{42,43} has been implemented using the linear response method.\cite{15,44,46} The matrix element for electron-phonon interaction and the Eliashberg function have been evaluated numerically, as described in our previous papers (see, e.g., Ref. 46). The superconductor transition temperature ($T_C$) was computed by using the McMillan modification of the Allen-Dynes formulation.\cite{47,49}

III. RESULTS

A. Structural and electronic properties

Cr$_3$GaN crystallizes in the cubic antiperovskite structure with the space group Pm3m. In this structure, as shown in Fig. 1, six Cr atoms occupy the face-centered positions of unit cell forming a three-dimensional network of Cr$_6$ octahedron, N atoms occupy the body-centered cubic position surrounded by the Cr$_6$ octahedron cage, and Ga atoms are located at the corners of the unit cell. Table I shows our calculated structural parameters, together with available experimental and other theoretical data.\cite{34} One can see that our calculated lattice constant is larger than the local-density approximation (LDA) value of 3.79 Å (Ref. 34) but only 1.4% smaller than the experimental value of 3.88 Å. Unfortunately, there are no experimental and theoretical data for bulk modulus ($B$) and its pressure derivative ($B'$) for us to compare with. We have also compared the calculated structural results for Cr$_3$GaN with those for its isostructural superconducting counterpart Cr$_3$RhN.\cite{35} The substitution of Rh by Ga mainly affects the bulk modulus value rather than the lattice constant value: the bulk modulus for Cr$_3$GaN differs from that for Cr$_3$RhN by 11%. The near equality of the lattice constants of these isostructural compounds is expected

<table>
<thead>
<tr>
<th>Cubic antiperovskite</th>
<th>$a$ (Å)</th>
<th>$B$ (Mbar)</th>
<th>$B'$</th>
</tr>
</thead>
<tbody>
<tr>
<td>Cr$_3$GaN</td>
<td>3.82</td>
<td>2.30</td>
<td>4.46</td>
</tr>
<tr>
<td>LDA (Ref. 34)</td>
<td>3.79</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Experimental$^1$</td>
<td>3.88</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Cr$_3$RhN (Ref. 35)</td>
<td>3.81</td>
<td>2.59</td>
<td>4.32</td>
</tr>
<tr>
<td>LDA (Ref. 34)</td>
<td>3.71</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Experimental$^1$</td>
<td>3.86</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

![FIG. 1. The cubic antiperovskite structure of Cr$_3$GaN.](image-url)
because the atomic radius of Rh and Ga atoms are 1.35 pm and 1.34 pm, respectively.

Before discussing our electronic results, we have to mention that Cr$_3$GaN is not magnetic because the spin-down and spin-up electronic structures for this material are found to be exactly the same. Fig. 2 shows the electronic band dispersion curves of Cr$_3$GaN along several high symmetry directions of the simple cubic Brillouin zone, calculated at the equilibrium lattice constant using the GGA-PBE approximation. The overall band profile for Cr$_3$GaN is found to be in fairly good agreement with previous theoretical calculations. The total, and site decomposed, electronic DOS are displayed in Fig. 3(a). From the band structure, it is seen that the low-lying band has a parabolic shape along all the considered symmetry directions. This band is mainly dominated by N 2s electrons and lies between -18 and -15 eV. Above this band, there is a considerably flat band with an energy of -14.60 eV, originating from the 3d electrons of Ga atoms. For energy window from -9 to -5 eV, the Cr electronic states (of s, p, and d orbital nature) hybridize with N 2p and Ga 4s states. The bands between -5 and -2 eV are mainly build up from Cr 3d states with some contributions from the 4p orbitals of Ga atoms. The contributions from other electrons to these bands are negligibly small. Cr 3d states contribute significantly to the bands close to the Fermi level.

Because the DOS distribution near the Fermi level ($E_F$) plays an important role in determining superconducting properties, it is important to examine the DOS in the vicinity of the Fermi level. We have observed a sharp peak at $E_F \sim 0.7$ eV which is created by the 3d electrons of Cr atoms. We have to mention that a sharp peak in the DOS near $E_F$ has also been noted for MgCNi$_3$ and CdCNi$_3$ (Refs. 15 and 21) which are superconductors. In agreement with the superconductor transition metal carbides such as CrC, NbC, and VC, the Fermi level for Cr$_3$GaN lies on a sharply increasing peak at $E_F + 0.47$ eV. This peak is mainly dominated by the 3d orbitals of Cr atoms. A similar peak with an energy of $E_F + 0.34$ eV is found in the electronic density of states for the superconductor CrC. We note, however, that the numerical values of the DOS at the Fermi level ($N(E_F)$) is 5.00 states/eV for Cr$_3$GaN, which is lower than the corresponding value of 6.16 states/eV for Cr$_3$RhN obtained in our previous work. The contribution of Cr atoms to $N(E_F)$ accounts for 97% and Ga atoms for 2%, while the contribution of N atoms is rather small, only 1%. Thus, nitrogen and gallium atoms do not contribute significantly to the DOS at the Fermi level and therefore are not involved in the conduction properties of Cr$_3$GaN. In contrast, the corner Rh atom in Cr$_3$RhN makes a considerable contribution to $N(E_F)$. It is this difference that makes the value of $N(E_F)$ for Cr$_3$RhN higher than the corresponding value for Cr$_3$GaN. Obviously, it is the 3d states of Cr atoms that give rise to the electrical conductivity in Cr$_3$GaN, though d electrons are generally considered as less efficient conductors.

The pressure dependence of the DOS for Cr$_3$GaN is shown in Fig. 3(b). The effect of pressure is to redistribute the peaks close to the Fermi level: the peaks on either side of the Fermi level are decreased in their height. The weak shoulder at $E_F$ for $P = 0$ becomes sharper as the pressure increases. In order to a better understanding of the pressure effect on the electronic properties of the cubic antiperovskite Cr$_3$GaN, we have performed the calculation of total DOS at the Fermi level at different pressures. Fig. 4 shows the change of $N(E_F)$ with pressure. As can be seen from this figure, $N(E_F)$ decreases with increase of pressure. In particular, $N(E_F)$ diminishes from 5.00 States/eV to 3.20 States/eV as pressure increases from 0 GPa to 100 GPa. The effect of pressure on the electron-phonon coupling parameter $\lambda$ is directly related to the change in $N(E_F)$, following the McMillan-Hopfield expression $\lambda = \frac{N(E_F)}{M \langle \omega^2 \rangle}$, where $\langle \omega^2 \rangle$ is the averaged square of the electron-phonon matrix, $\langle \omega^2 \rangle$ is the averaged square of the phonon frequency, and $M$ is the mass involved.

B. Phonons and superconductivity

The primitive cell of this material contains five atoms, giving a total of 15 phonon branches including three acoustic branches. The calculated phonon dispersion curves along the high-symmetry lines in the Brillouin zone are shown in Fig. 5(a). The total phonon spectrum has an energy range of about 21 THz. In particular, the phonon dispersions show a set of 12 bands extending up to around 11 THz, separated by a gap of 8.4 THz from three high-frequency bands that lie between 19 and 21 THz. Also, all phonon modes exhibit positive frequencies, strongly suggesting that the optimized Cr$_3$GaN structure is dynamically stable. The highest three phonon branches are dominated by the motion of nitrogen atoms. These branches are dispersive but the highest of them is nearly flat along the M-X, X-R, and R-M symmetry directions. In the lowest region of the phonon spectrum, we have observed several bands which exhibit dispersive character. This indicates strong interaction between Cr and Ga atoms. The calculated total and partial phonon density of states are presented in Fig. 5(b). The partial DOS shows that the states from 4 THz to 6 THz arise from vibrations of Cr and Ga atoms and the features between 7 and 9 THz are created by the motions of Cr atoms with a very small admixture of N atoms. The sharp peak at 6.2 THz is due to the flatness of $T_{2u}$ mode along the $\Gamma - X$ symmetry directions. The sharp peak with the frequency of 10.7 THz also results from the displacements of Cr atoms. From this picture, it can be
expected that Cr vibrational modes would make a large contribution to electron-phonon interaction in Cr$_3$GaN. There is a large gap of 8.4 THz separating the upper sharp peaks at 20.2 and 21.6 THz. Both of these peaks are mainly localized on the N atoms but the lower peak includes a little contribution from Cr vibrations.

At the Γ point of Brillouin zone, the phonon modes in the cubic antiperovskite structure decompose according to Cr vibrational modes would make a large contribution to electron-phonon interaction in Cr$_3$GaN. There is a large gap of 8.4 THz separating the upper sharp peaks at 20.2 and 21.6 THz. Both of these peaks are mainly localized on the N atoms but the lower peak includes a little contribution from Cr vibrations.

At the Γ point of Brillouin zone, the phonon modes in the cubic antiperovskite structure decompose according to
the irreducible representation characteristic for the $O_h$ point group
\[ \Gamma_{ac} = T_{2u}^a \quad \text{and} \quad \Gamma_{opt} = T_{2u}^1 + T_{1u}^2 + T_{1u}^3, \]
for acoustic and optical modes, respectively. None of the triply degenerate optical modes is Raman active, as a perfect cubic structure, all lattice sites have inversion symmetry.

However, these phonon modes can be detected using infrared and neutron spectroscopic techniques. The frequencies of the optical phonon modes are calculated to be 2.86 THz ($T_{2u}$), 4.02 THz ($T_{1u}^1$), 6.24 THz ($T_{1u}^2$), and 19.16 THz ($T_{1u}^3$). The $T_{2u}$ mode is totally dominated by the vibrations of Cr atoms. The first $T_{1u}$ mode is created by the vibrations of all atoms in the unit cell with the maximum contribution coming from Cr atoms. The intermediate $T_{1u}$ mode results from opposing motions of Ga and Cr atoms. The highest optical phonon mode is due to the vibrations of N atoms with a small contribution from Cr atoms.

Since Cr$_3$GaN is isostructural to Cr$_3$RhN and the ionic radii and local bonding environment of these compounds are similar, the major difference in their phonon spectra is expected to arise from the mass difference between Ga and Rh. The zone-centre phonon frequencies of these two compounds are compared in Table II. As one can expect, the highest optical phonon mode lies at nearly the same frequency for both isostructural materials due to the light mass of N atom. The $T_{2u}$ and $T_{1u}$ phonon modes in Cr$_3$GaN have higher frequencies than their counter-parts in Cr$_3$RhN due to the mass difference between Ga and Rh atoms. In contrast, the $T_{1u}$ mode of Cr$_3$RhN lies at a slightly higher frequency than that of Cr$_3$GaN although the mass of Rh atom is heavier than that of Ga atom. This can be explained by using the separation of the phonon frequency into a short-range "normal" part $\omega_n^2$ and a long-range "anomalous" part $\omega^2 = \omega_n^2 + \omega_a^2$. The first part depends only upon short-range interaction, while the second part is always negative and depends on the electronic screening effects. As the corner atom is changed from Rh to Ga, although $\omega_n$ hardens due to decreasing mass, the negative contribution of $\omega_a^2$ becomes higher. This competition leads to a decrease in $\omega^2$ for the $T_{1u}$ mode of Cr$_3$GaN. As a result, due to stronger electronic screening effects in Cr$_3$GaN, the $T_{1u}$ mode for this material has a slightly smaller frequency than the corresponding phonon mode of its isostructural compound Cr$_3$RhN. In order to achieve a clear understanding of the mass effect on the vibrational modes at $\Gamma$, we have performed calculations of phonon modes in Cr$_3$GaN using the force constants for its isostructural compound Cr$_3$RhN (Ref. 35) but replacing the mass of Rh by that of Ga. We have observed that the frequencies of those $T_{1u}$ and $T_{1u}$ modes that contain considerable contributions from the corner atom (Ga or Rh) are affected by the mass replacement, whereas the frequencies of other species do not change.

Now we discuss the interaction between electrons and phonons. The electron-phonon coupling constant $\lambda$ is usually extracted from the Eliashberg function as described in our

### TABLE II. Calculated vibration modes (in THz) at $\Gamma$ for Cr$_3$GaN as compared with its isostructural compound Cr$_3$RhN.

<table>
<thead>
<tr>
<th>Material</th>
<th>$T_{2u}$</th>
<th>$T_{1u}^1$</th>
<th>$T_{1u}^2$</th>
<th>$T_{1u}^3$</th>
</tr>
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<tbody>
<tr>
<td>Cr$_3$GaN</td>
<td>2.86</td>
<td>4.02</td>
<td>6.24</td>
<td>19.16</td>
</tr>
<tr>
<td>Cr$_3$RhN (Ref. 35)</td>
<td>1.26</td>
<td>4.76</td>
<td>5.29</td>
<td>19.80</td>
</tr>
<tr>
<td>Cr$_3$GaN$^a$</td>
<td>1.26</td>
<td>4.86</td>
<td>5.90</td>
<td>19.80</td>
</tr>
</tbody>
</table>

$^a$Vibration modes at $\Gamma$ of Cr$_3$GaN obtained using the force constant matrix of Cr$_3$RhN.
previous papers (see, e.g., Ref. 46). It may be expected that the phonon modes which interact strongly with high 3d density of states at the Fermi level in Cr3GaN involve low-frequency vibrations of Cr atoms. These are the acoustic phonon modes as well as the low-frequency T2 mode which directly modifies the Cr-Cr distance. The electron-phonon coupling parameter for different zone-centre phonon modes in Cr3GaN is compared with those for Cr3RhN in Table III. Clearly, the largest electron-phonon coupling parameter for Cr3GaN is compared with those for Cr3RhN in Table III. The structural stability of Cr3GaN under high pressure has been examined in Fig. 6. The structural stability is confirmed by the lack of any imaginary phonon frequencies in the whole Brillouin zone in the pressure range 0–100 GPa. It is clear from this figure that all the phonon modes harden as pressure increases. In particular, the frequency of the T2u mode at the Γ point, dominating the electron-phonon coupling, is greatly increased from 2.86 THz at 0 GPa to 4.99 THz at 100 GPa. The enhanced frequency of this mode contributes to the increase of \((\langle \omega^2 \rangle)\) in the McMillan-Hopfield equation and thus results in the reduction of the electron-phonon coupling parameter. We have observed that the electron-phonon coupling parameter for T2u is considerable decreased from 0.72 at 0 GPa to 0.48 at 100 GPa. The origin of considerable decrease of \(\lambda\) for T2u mode with increasing pressure is mainly attributed to the enhanced frequency of this phonon mode.

The Eliashberg function \(\xi^2F(\omega)\) of Cr3GaN is compared with that of Cr3RhN in Fig. 7(a). From this figure, we can say that the electron-phonon interaction in Cr3GaN is relatively weaker than the corresponding interaction in its isostructural compound Cr3RhN. Now, we can compare the average electron-phonon coupling parameter \(\lambda\) for Cr3GaN with Cr3RhN. Our computed value of \(\lambda\) for Cr3GaN is found to be 0.53 which is nearly half of the average electron-phonon coupling value of 1.03 for Cr3RhN.34 It should be noted that \(\lambda\) for Cr3GaN was estimated to be 1.3 in the theoretical work of Wiendlocha et al.34 This result is higher that our result of 0.53. We should, however, note that Wiendlocha et al.34 estimated this parameter without taking phonon properties into account. For Cr3GaN, we obtain for the logarithmically averaged frequency \(\omega_{ln}\) = 306 K. Now, in order to obtain the superconductor transition temperature for Cr3GaN, we need to know the value of \(\mu^*\), the effective screened Coulomb repulsion constant. Previous studies47–49 on transition metals showed that \(\mu^*\) takes values between 0.1 and 0.16. As Cr is a transition metal, we consider values of \(\mu^*\) between 0.1 and 0.13 in our study. Inserting the calculated values of \(\lambda\) and \(\omega_{ln}\) into the Allen-Dynes formula47–49 with \(\mu^* = 0.10, 0.11, 0.12,\) and 0.13, we have obtained the superconductor transition temperature \(T_C = 4.78, 4.17, 3.60,\) and 3.08 K. Our results clearly show that the cubic antiperovskite Cr3GaN is a superconductor with the value of \(T_C\) around 4 K.

All phonon frequencies harden with increasing pressure. This will affect the values of the averaged square of the phonon frequency \(\langle \omega^2 \rangle\) and the logarithmic-averaged phonon frequency \(\omega_{ln}\). These quantities are very important for the electron-phonon coupling parameter because it is well known that the hardening of phonon frequencies make a negative contribution to this parameter. The values of \(\langle \omega^2 \rangle\) and \(\omega_{ln}\) increase with increasing pressure. As can be seen from the McMillan-Hopfield expression, the frequency hardening is considered to contribute to the decrease of \(\lambda\) with increasing pressure. Finally, we have illustrated the pressure dependence of the electron-phonon coupling parameter (\(\lambda\)) and the superconductor transition temperature (\(T_C\)) in Figs. 7(b) and 7(c). We have observed a monotonically decreased \(\lambda\) from 0.53 at zero pressure 0.43 at 100 GPa. Due to the decrease of \(\lambda\), \(T_C\) also decreases with increasing pressure. \(T_C\) has been found to monotonically decrease from 0 to 100 GPa and reaches the value of 2.88 K at 100 GPa for \(\mu^* = 0.11\).

<table>
<thead>
<tr>
<th>Material</th>
<th>(T_{2u})</th>
<th>(T^1_{1u})</th>
<th>(T^2_{1u})</th>
<th>(T^1_{1u})</th>
</tr>
</thead>
<tbody>
<tr>
<td>Cr3GaN</td>
<td>0.72</td>
<td>0.14</td>
<td>0.03</td>
<td>0.02</td>
</tr>
<tr>
<td>Cr3RhN (Ref. 35)</td>
<td>1.98</td>
<td>0.14</td>
<td>0.17</td>
<td>0.02</td>
</tr>
</tbody>
</table>

**TABLE III.** The zone-centre electron-phonon coupling parameters for Cr3GaN as compared with those of its isostructural compound Cr3RhN.

![Fig. 6. Calculated phonon spectra of Cr3GaN for (a) normal pressure (0 GPa) and (b) 100 GPa, respectively.](image-url)
We now make a comparison as regards superconductivity between Cr$_3$GaN and its isostructural compound Cr$_3$RhN by analysing their electronic and phonon structures. We have presented a comparison of the superconducting parameters for these compounds in Table IV. As noted before, three main factors effect $T_c$: the electronic DOS at the Fermi level $N(E_F)$, the logarithmic average phonon frequency $\omega_L$ (or the averaged square of the phonon frequency $\langle \omega^2 \rangle$), and the strength of electron-phonon coupling parameter $\lambda$. As regards the electronic structure, the calculated $N(E_F)$ value of 5.00 states/eV for Cr$_3$GaN is smaller than the corresponding value of 6.16 states/eV for Cr$_3$RhN, making superconductivity comparatively less favourable for Cr$_3$GaN. The lower energy phonon modes in Cr$_3$GaN are harder than the corresponding phonon modes in Cr$_3$RhN due to the small mass of Ga atom as compared to the mass of Rh atom. Although harder phonon frequencies lead to larger $\omega_L$ and $\langle \omega^2 \rangle$ values, they make a negative contribution to the strength of electron-phonon coupling parameter. Consequently, the $T_c$ value of around 4.2 K for Cr$_3$GaN is considerably lower than the corresponding value of 17.7 K for Cr$_3$RhN.

### IV. SUMMARY

In this work, we have presented a theoretical analysis of the structural and electronic properties of the cubic antiperovskite Cr$_3$GaN by using the generalised gradient approximation of the density functional theory and the plane wave pseudopotential method. Our results show metallic d-like bands crossing the Fermi level, indicating that the transition metal Cr may play more active role in the electronic and superconducting properties of this antiperovskite. The effect of pressure is found to redistribute the peaks in the electronic density of states close to the Fermi level: the peaks on either side of the Fermi level are decreased in their height. With pressure increase, $N(E_F)$ decreases monotonically, leading to a reduction of electron-phonon coupling parameter $\lambda$. We have shown that the phonon modes related to the vibrations of Cr atoms make a large contribution to the average electron-phonon coupling parameter $\lambda$. The computed value of $\lambda$ is 0.53, and the superconducting critical temperature is estimated to be around 4.2 K.

This compound is predicted to be structurally stable, as there are no imaginary phonon frequencies in the whole Brillouin zone in the pressure range 0.0–100 GPa. All phonon modes shift to higher frequencies when pressure increases. With increasing pressure, the values of logarithmic-averaged phonon frequency $\omega_L$ and averaged square of the phonon frequency $\langle \omega^2 \rangle$ increase rapidly. On the other hand, the values of $N(E_F)$, $\lambda$ and $T_c$ decrease with increasing pressure. We strongly believe that reduced $N(E_F)$ and the enhanced frequencies of phonon modes are the main causes for the monotonic decrease of $\lambda$, and consequently of $T_c$ with increasing pressure.

A comparison of the superconducting parameters for Cr$_3$GaN and Cr$_3$RhN shows that the value of $T_c$ for Cr$_3$GaN is lower than the corresponding value for Cr$_3$RhN. This is related to the relatively lower values of the electronic density of states at the Fermi level, the Eliashberg function, as well as the lower average electron-phonon coupling parameter for Cr$_3$GaN.

### TABLE IV. Comparison of superconducting state parameters for Cr$_3$GaN and Cr$_3$RhN. The parameter $\mu^*$ is taken to be 0.11.

<table>
<thead>
<tr>
<th>Perovskite</th>
<th>$N(E_F)$ (States/eV)</th>
<th>$\omega_L$ (K)</th>
<th>$\langle \omega^2 \rangle^{1/2}$ (K)</th>
<th>$\lambda$</th>
<th>$T_c$ (K)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Cr$_3$GaN</td>
<td>5.00</td>
<td>306</td>
<td>3.88</td>
<td>0.53</td>
<td>4.17</td>
</tr>
<tr>
<td>Cr$_3$RhN (Ref. 35)</td>
<td>6.16</td>
<td>255</td>
<td>339</td>
<td>1.03</td>
<td>17.74</td>
</tr>
</tbody>
</table>

FIG. 7. (a) The calculated electron-phonon spectral function $\pi^2 F(\omega)$ for Cr$_3$GaN as compared with that of Cr$_3$RhN. (b) The electron-phonon coupling parameter ($\lambda$) as a function of pressure for Cr$_3$GaN. (c) The superconductor transition temperature ($T_c$) as a function of pressure for Cr$_3$GaN. Two choices for $\mu^*$ have been made: 0.11 and 0.13.
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