Superconductivity of $\beta$-Gallium

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Elemental gallium can exist in several phases under ambient conditions. The stable $\alpha$ phase has a superconducting transition temperature, $T_c$, of 0.9 K. By contrast, the $T_c$ of the metastable $\beta$ phase is around 6 K. To understand the significant improvement in $T_c$ in the $\beta$ phase, we first calculate the electronic structure, phonon dispersion, and the electron-phonon coupling of gallium in the $\alpha$ and $\beta$ phases. Next, we solve the Eliashberg equations to obtain the superconducting gaps and the transition temperatures. Using these results, we relate the increased $T_c$ in the $\beta$ phase to structural differences between the phases that affect the electronic and phonon properties. The structure motif of the $\alpha$ phase is Ga$_2$ dimers, which form strong covalent bonds leading to bonding and antibonding states that suppress the density of states at the Fermi level. The $\beta$-Ga structure consists of arrays of Ga chains that favor strong coupling between the lattice vibrations and the electronic states near the Fermi level. The increased density of states and strong coupling to the phonons for the $\beta$-Ga chains compared to the $\alpha$ Ga$_2$ dimers enhance superconductivity in the $\beta$-Ga phase.

I. INTRODUCTION

The structural phase diagram of elemental gallium is complex. At ambient pressure, gallium crystallizes in the stable orthorhombic $\alpha$ phase. Several metastable phases, $\beta$, $\gamma$, $\delta$, and $\epsilon$, can also be synthesized under ambient conditions.1–4 Under pressure, Ga exhibits three additional phases, Ga-II, Ga-III, and Ga-V.5,6 Most gallium phases ($\alpha$, $\beta$, $\gamma$, $\delta$, Ga-II) undergo superconducting transitions. The superconducting transition temperature, $T_c$, of the stable $\alpha$-Ga phase is about 0.9 K.7 However, the metastable $\beta$, $\gamma$, and $\delta$ phases have much higher $T_c$'s. Metastable $\beta$-Ga is reported to have a $T_c$ of 5.9 to 6.2 K,8–10 while the transition temperatures of $\gamma$ and $\delta$ Ga are 6.9 to 7.6 K10,11 and 7.85 K,12 respectively.

The significant increase in the superconducting transition temperature from 0.9 K in the stable $\alpha$ phase to 5.8 K in the $\beta$, $\gamma$, and $\delta$ phases poses an interesting theoretical question. In the context of the recent discoveries of high-temperature superconductivity in hydrides at megabar pressure,13 there is great interest in understanding situations where long-lived metastable phases formed at ambient or high pressure might exhibit a higher $T_c$ than their stable counterparts. The gallium phases represent a unique opportunity to study this problem theoretically to understand the enhancement from the low-$T_c$ stable phase to the higher-$T_c$ metastable phases.

The phase stability of gallium was first studied by Gong et al. and Bernasconi et al. separately in the 1990s.14,15 Both studies found that the $\alpha$ structure is the most stable phase. Gong et al. pointed out that the structural motif of $\alpha$-Ga consists of Ga$_2$ dimers with strong covalent bonding and significant charge localization inside the dimers. A recent study based on the Wannier functions analysis of the chemical bonding in $\alpha$-Ga also supports covalent bonds within the Ga$_2$ dimers.16 Gong et al. characterize the metallic $\alpha$-Ga phase with its strong Ga-Ga bonds as “a metallic molecular crystal”.14

Recently, Campanini et al. reported an in situ characterization of the $\alpha$ to $\beta$ phase transition using a membrane-based nanocalorimeter9 and found that $\beta$-Ga is a strong coupling type-I superconductor with a $T_c$ of around 6 K.9 Khasanov et al. measured the thermodynamic critical field and specific heat of the high-pressure Ga-II phase as a function of temperature and demonstrated that it follows the same universal relations as in conventional superconductors.17 The significant improvement in $T_c$ as a result of the structure phase transition from the stable $\alpha$ structure to the metastable $\beta$ structure motivates us to examine various aspects of the two phases in search of factors that are important for improving $T_c$. In principle it would be interesting to study the even higher-$T_c$, $\gamma$ and $\delta$ phases as well, but the unit cell of these phases contain 20 and 66 atoms per primitive cell, respectively. By contrast, the primitive cells of the $\alpha$ and $\beta$ phases, contain 4 and 2 atoms, respectively.

This paper is structured as follows. Section II describes the details of the density-functional theory (DFT) calculations and the crystal structure of $\alpha$ and $\beta$ gallium. Section III compares the electronic structure, charge density distribution, and electron-phonon coupling for the $\alpha$ and $\beta$ phases of gallium. We show that the covalent Ga$_2$ dimers in $\alpha$-Ga suppress the electronic density of states (DOS) at the Fermi level, $E_f$, and that the chains in $\beta$-Ga lead to electronic states near $E_f$ that strongly couple to the phonons, thereby, increasing the superconducting transition temperature. Section IV, V summarize the results and discuss possible routes for improving the $T_c$ of elemental gallium.

II. COMPUTATIONAL METHOD AND CRYSTAL STRUCTURE

We perform the DFT calculations using the Quantum Espresso code18–20 with optimized norm-conserving
FIG. 1. The crystal structure (a,b) and electronic density of states (c,d) of α and β Ga. In the α phase (a), Ga atoms form dimers (or dumbbells), which lead to bonding and antibonding states that suppress the density of states at the Fermi level (c). In the β phase (b), the Ga atoms form arrays of one-dimensional chains along the c lattice vector direction that result in an increased density of states at the Fermi level (d).

Electronic structure. The electron configuration of the gallium atom is [Ar]3d104s²4p¹, with a filled 3d shell that does not participate in the chemical bonding. The 4s and 4p states hybridize and form bonds between the Ga atoms. Although the valence states of α and β-Ga consist of the same 4s and 4p orbitals, the bonding states and their orbital character near the Fermi level differ substantially in the two phases. Figure 1(c) shows that in α-Ga, states within 2 eV below the Fermi level are predominantly 4p states with negligible 4s character, while the 4s orbital contributes to one-third of the total DOS within 2 eV above the Fermi level. For β-Ga shown in Fig. 1 (d), the 4p orbitals dominate the DOS within 2 eV above and below the Fermi level, and the variation in DOS near the Fermi level is small. The most important feature in the DOS of α-Ga is the V-shaped pseudogap at the Fermi level that substantially reduces the number of states near $E_f$. This pseudogap has been attributed to covalent bonding, and the interplay between the electronic states and the Brillouin zone geometry.

Figure 2 shows how the band structures of α and β-Ga differ in several aspects that lead to the observed pseudogap in α-Ga and a larger DOS in the β phase. The electronic structure of the β-Ga consists of free-electron-like parabolic bands that span a few eV due to strong intra-chain and inter-chain hoppings. The band structure of the α-Ga, on the other hand, has a few bands that are dispersionless along certain $k$-paths in the Brillouin zone. The flat bands at 1.5 eV below the Fermi level along $X - S - R$ contributes to the DOS peak at $-1.5$ eV. The DOS peak at 1 eV originates from the flat bands along $\Gamma - X - S$.

III. RESULTS

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Fig. 3 illustrates the Fermi surfaces of the α and β-Ga. The Fermi surface of α-Ga is continuous across the entire Brillouin zone along the $\vec{a}^* + \vec{b}^*$ and $\vec{c}^*$ directions. Along the $\vec{a}^* - \vec{b}^*$ direction, the Fermi surfaces is confined to the region from $-0.2(\vec{a}^* - \vec{b}^*)$ to $+0.2(\vec{a}^* - \vec{b}^*)$, see Fig. S1 in the supplement for detail. The Fermi surfaces of β-Ga extend throughout the Brillouin zone and exhibit flat regions due to its one dimensional chain-like crystal structure, which in part explains the metastability of the β phase.

Charge density distributions. The pseudogap in the α-Ga DOS near the Fermi level strongly suggests the formation of filled bonding and empty antibonding states. Figures 4(a) and (b) show cuts through the charge density of α-Ga that include the Ga₂ dimer for the energy windows $[-1.5$ eV, $-0.5$ eV] and [0.5 eV, +1.5 eV], respec-
FIG. 2. The band structures of $\alpha$ and $\beta$-Ga projected on the $4s$ and $4p$ atomic orbitals show several nearly flat bands in the $\alpha$-Ga and nearly free-electron parabolic bands for $\beta$-Ga.

tively. The maximum in the charge density between the dimers for the states below the Fermi level and the valley for the states above demonstrate bonding and antibonding states below and above the Fermi level, respectively. Therefore, the formation of strong covalent bonds for the $\alpha$-Ge dimers results in localized states that are shifted away from the Fermi level and, hence, do not participate in the electron-phonon coupling process.

In contrast to $\alpha$-Ga, Figures 4 (c) and (d) show that $\beta$-Ga displays a delocalized charge density along the Ga chains. Therefore, the breaking of the Ga dimers liberates the electronic states from the localized molecular bonds, leading to an increased number of states available near the Fermi level that can couple to the lattice vibrations and increase superconductivity.

**Phonon dispersion and electron-phonon coupling.** To identify the phonon modes that strongly couple to the electronic states, we plot the phonon dispersion of $\beta$-Ga in Fig. 5(a) along the high-symmetry path with the symbol sizes representing the electron-phonon coupling strength $\lambda^\nu_{\vec{q}}$, where $\nu$ denotes the phonon branch index and $\vec{q}$ the wavevector. The two lowest acoustic phonon branches display large electron-phonon coupling $\lambda^\nu_{\vec{q}}$ with noticeable variation along the high-symmetry path. We calculate the mean and standard deviation of $\lambda^\nu_{\vec{q}}$ for each phonon branch to quantify the strength and anisotropy of the electron-phonon coupling. Table I shows that the average coupling strength of the lowest two phonon branches are 0.36 and 0.25, which accounts for half of the total coupling strength of 1.16. The coupling constants of the third and fourth branches are 0.16 and 0.15, respectively, about 27% of the total coupling strength, and the highest two phonon branches contribute 19% of the total coupling.

Figure 5(b) compares the calculated electron-phonon coupling $\alpha^2 F(\omega)$ obtained with the EPW code for the fine $k$ and $q$-point meshes with the experimental $\alpha^2 F(\omega)$ from tunneling experiments$^{28}$ for $\beta$-Ga. The excellent agreement validates the computational methodology. Table II summarizes the electron-phonon coupling strength $\lambda$ and the frequency moments $\langle \omega^2 \rangle$ and $\langle \omega \log \omega \rangle$ of $\beta$-Ga. Using the Allen-Dynes equation with $\mu^* = 0.13$, we esti-
FIG. 4. Charge density of α and β-Ga projected on 1 eV energy windows surrounding the Fermi level. The charge densities for α-Ga demonstrate the presence of localized (a) bonding and (b) antibonding states between the Ga₂ dimers within 1 eV of the Fermi level. For the β phase, the charge densities (c) and (d) are delocalized along the Ga-chain, indicating one-dimensional extended states that couple strongly to the lattice.

TABLE I. The mean and standard deviation of λν|q integrated over the entire Brillouin zone for each phonon branch ν and in total. The electron-phonon coupling of the lowest branch is as large as the sum of the highest three branches.

| Phonon branch ν | 1    | 2    | 3    | 4    | 5    | 6    | Total |
|-----------------|------|------|------|------|------|------|-------|
| Mean (λν|q)       | 0.36 | 0.25 | 0.16 | 0.15 | 0.12 | 0.10 | 1.13  |
| Standard deviation | 0.34 | 0.11 | 0.07 | 0.08 | 0.04 | 0.04 | 0.43  |

determine a superconducting transition temperature of 6.5 K. Solving the isotropic Eliashberg equations, we obtain essentially the same superconducting transition temperature of 6.6 K. These values closely match the reported experimental Tc of 5.9 to 6.2 K.8–10 To characterize the anisotropy of the superconductivity, we also solve the anisotropic Eliashberg equations at several temperatures below Tc. Figure 6 compares the isotropic and anisotropic gap function as a function of temperature, which demonstrates that β-Ga is an isotropic superconductor.

Figures 7 and 8 illustrate the phonon spectrum and the eigenmodes at Γ for α-Ga, respectively. The primitive cell of the α-Ga structure contains four atoms that all lie on the $\mathbf{b} - \mathbf{c}$ plane, which represents a mirror plane. As a result, there are two types of phonon modes at the Γ point, in-plane and out-of-plane vibrations, as shown in Fig. 8. The first and the sixth optical phonon modes comprise of out-of-plane Ga-Ga bond bending, while for the third optical phonon mode, the two Ga-Ga dimers vibrate rigidly into and out of the $\mathbf{b} - \mathbf{c}$ plane. The second and the seventh optical phonon modes are the in-plane counterparts of the third optical mode. The eighth and
Fig. 7. (a) Phonon dispersion of α-gallium along high-symmetry path. (b) Eliashberg function of α-gallium.

Fig. 8. The nine optical phonon modes at the Γ point for the α-Ga. The plus and minus signs indicate the motion perpendicular to the plane. The two highest frequency modes (mode 11 and 12) are in-phase and out-of-phase Ga-Ga bond stretching modes, respectively.

The ninth phonon modes are bond stretching modes with much higher frequencies than the in-plane bond bending modes (modes 4 and 5). In addition, the phonon dispersions of the highest two phonon branches are gapped from the rest of the phonon spectrum, see Fig. 7. The integration of $g(\omega) = \frac{2\omega^2 F(\omega)}{\omega}$ over the frequency range from 25 to 30 meV turns out to be 0.03, about 13% of the total electron-phonon coupling strength.

IV. DISCUSSION

Several members in the boron group have played an important role in the quest for high-temperature superconductivity. Boron in MgB$_2$, for example, has $p$ orbitals that form delocalized metallic $\sigma$ and $\pi$ bonds. Although $\alpha$-Ga has strong bonds, they are localized and either below or above the Fermi level. Electron or hole doping might shift the Fermi level towards the DOS peaks at +1 or −1.5 eV, respectively, which could improve the superconducting transition temperature of the stable $\alpha$ phase.

Applying external pressures could be another route for improving the superconducting transition temperatures of $\alpha$ and $\beta$-Ga. Elemental boron per se is not a superconductor under ambient conditions, although it can transform into a superconductor with a $T_c$ of 4 K at 160 GPa. Further increasing the external pressure to 250 GPa can improve the $T_c$ to 11 K.

Gallium is already a superconductor at ambient pressure. Its $T_c$ ranges from 1 K in the stable $\alpha$ phase to 5–7 K in the metastable $\beta$, $\gamma$, $\delta$ phases. Under external pressure, several new phases are predicted to form. The superconductivity of Ga-II, one of several phases of gallium that emerge under pressure, is measured to be 6.46 K at 3.5 GPa. But the superconductivity of gallium under megabar pressures has not been explored systematically. Given that boron, the lightest member of the boron group, undergoes several structure phase transitions before it arrives at its superconducting phase under 160 GPa, new Ga phases with higher $T_c$ might also emerge under extreme pressure.

V. SUMMARY

We demonstrated that the disparate superconducting properties of gallium’s $\alpha$ and $\beta$ phases originate from their difference in bonding using DFT and presented first-principles calculations of the electronic structure, phonon dispersion, electron-phonon coupling, and superconducting properties. For the $\alpha$ phase, the Ga$_2$ dimers form bonding and antibonding states that result in a V-shaped suppression of the DOS at the Fermi level, which significantly reduces the number of states available for coupling to the phonons. The DOS of metastable $\beta$-Ga, on the other hand, is nearly free-electron gas-like due to strong intra- as well as inter-chain hoppings. The charge density confirms the strongly localized bonding and antibonding states below and above the Fermi level in $\alpha$-Ga, while it shows delocalized states near the Fermi level in $\beta$-Ga that are more likely to respond to lattice vibrations. The $T_c$ of $\beta$-Ga is estimated to be 6.5 K, which agrees with the experimental values of 5.9 to 6.2 K$^{8-10}$ and is much higher than the is much higher than the $T_c$ of $\alpha$ Ga. Also, the calculated electron-phonon coupling $\alpha^2 F(\omega)$ for $\beta$-Ga closely matches experiment, validating the computational approach. Therefore, the formation of Ga$_2$ dimers in the $\alpha$ phase suppresses its superconductivity.

We hope that this understanding of the electronic and phononic structure of the higher-$T_c$ metastable $\beta$ Ga phase can pave the way to understanding and designing ambient pressure metastable phases of other superconductors with higher $T_c$s.

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Supplementary material

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I. FERMI SURFACES

The Fermi surfaces of the α and the β gallium are presented in Fig. S1 and Fig. S2 respectively. The Fermi surfaces of the α gallium are mostly confined inside a region between $-0.2(\vec{a}^* - \vec{b}^*)$ to $0.2(\vec{a}^* - \vec{b}^*)$, see Fig. S1.

![Fig. S1](image1)

**FIG. S1:** The Fermi surfaces of the α gallium are shown in the figure above. FS1 to FS4 correspond to the four bands that cross the Fermi level.

![Fig. S2](image2)

**FIG. S2:** The Fermi surfaces of the β gallium corresponding to the three bands that cross the Fermi level are shown in the figure above.

II. PHONON MODES

We have visualized the lowest two phonon modes of β-Ga at Y and F points, see F1.mp4, F2.mp4, Y1.mp4 and Y2.mp4, which help understand the collective Ga vibrations that can induce strong electronic response. The phonon modes are visualized using Phononweb.