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REPORT



Thermal conductivity and electrical transport properties of double-A-layer MAX phase $\text{Mo}_2\text{Ga}_2\text{C}$

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ABSTRACT

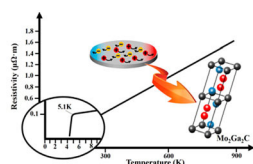
The present study reports the thermal conductivity and electrical transport properties of $\text{Mo}_2\text{Ga}_2\text{C}$, which is the first member of double-A-layer MAX phases. Dense bulk samples of $\text{Mo}_2\text{Ga}_2\text{C}$ were prepared by vacuum hot pressing. At room temperature, the thermal conductivity was measured to be $14.8 \pm 1.0 \text{ W/(m}\cdot\text{K)}$ and the electrical resistivity was $0.525 \pm 0.052 \mu\Omega\cdot\text{m}$. $\text{Mo}_2\text{Ga}_2\text{C}$ follows the Wiedemann-Franz law and the Lorenz number is $2.22 \times 10^{-8} \text{ W}\Omega\text{K}^{-2}$. $\text{Mo}_2\text{Ga}_2\text{C}$ is a superconductor with a transition temperature of ca. 5.1 K ($\pm 0.1\%$). The thermal conductivity and electrical resistivity are lower than those of most MAX phases probably due to the extra Ga layer.

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KEYWORDS

MAX phases; $\text{Mo}_2\text{Ga}_2\text{C}$; superconductivity; electrical transport; thermal conductivity



IMPACT STATEMENT

The present study prepared dense bulk samples of $\text{Mo}_2\text{Ga}_2\text{C}$, the first member of double-A-layer MAX phases, and characterized the thermal conductivity and electrical transportation. The results indicated $\text{Mo}_2\text{Ga}_2\text{C}$ to be a superconductor ($T_c = \sim 5.1 \text{ K}$).

1. Introduction

$\text{Mo}_2\text{Ga}_2\text{C}$ is a novel ternary carbide that was discovered in 2015 [1] and has similar structures to Mo_2GaC , a member of MAX phases. MAX phases are inherently nanolaminated materials, where M is an early transition metal, A is an A-group element (mostly IIIA or IVA), and X is C and/or N [2]. Although $\text{Mo}_2\text{Ga}_2\text{C}$ and Mo_2GaC are both laminated, Ga double-layers in $\text{Mo}_2\text{Ga}_2\text{C}$, instead of Ga mono-layers in Mo_2GaC , interleave the Mo_2C blocks in their crystal structure [3,4]. Thus $\text{Mo}_2\text{Ga}_2\text{C}$ is a double-A-layer MAX phase.

Recently, MAX phases were selectively etched to make two-dimensional MXenes [5,6], which show excellent performance in many fields [7–10]. $\text{Mo}_2\text{Ga}_2\text{C}$ is the only precursor to make Mo_2C MXene [11], which has better performance than other MXenes as electrocatalysts for hydrogen evolution [12], superconductor electrode

[13], or thermoelectric material [14]. Moreover, as the first member of double-A-layer MAX phases, exploration of the structure and properties of $\text{Mo}_2\text{Ga}_2\text{C}$ can help the discovering and preparing a series of double-A-layer MAX phases. For example, in 2016, another double-A-layer MAX phase, $\text{V}_2\text{Ga}_2\text{C}$, was predicted [15]. In 2017, double-A-layer $\text{Ti}_2\text{Au}_2\text{C}$ and $\text{Ti}_3\text{Au}_2\text{C}_2$ were prepared [16].

Numerous studies have reported on the theoretical prediction of the properties of $\text{Mo}_2\text{Ga}_2\text{C}$ following its initial discovery [3,17,18]. However, only a few experimental studies have reported these properties, [19,20] because synthesized $\text{Mo}_2\text{Ga}_2\text{C}$ has always been observed in the form of powders or films [1]. Only dense bulk samples can be used to measure many important properties, such as its mechanical, electrical, and thermal properties. The characterization of these properties are significant

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for the application and research of $\text{Mo}_2\text{Ga}_2\text{C}$ as well as other double-A-layer MAX phases.

The preparation of dense bulk $\text{Mo}_2\text{Ga}_2\text{C}$ samples allows the measurements of their thermal/electrical conductivity, which are related to the mobility, concentration, and type of carriers (electrons and/or holes). In addition, measurement of the electrical conductivity at different temperatures can verify whether $\text{Mo}_2\text{Ga}_2\text{C}$ is a superconductor or not. Certain MAX phases exhibited the ability to be superconducting, such as Mo_2GaC [21], Nb_2SC [22], Ti_2InC [23]. These superconducting MAX phases are all strongly coupled nanolaminates [24]. Toth established Mo_2GaC as the first discovered MAX phase with superconducting character ($T_c = \text{c.a. } 4.0 \text{ K}$) in 1967 [21]. $\text{Mo}_2\text{Ga}_2\text{C}$ has a similar hexagonal structure to Mo_2GaC and both share the same elementary constituent. Therefore, it can be speculated that $\text{Mo}_2\text{Ga}_2\text{C}$ may also be superconducting. The theoretical calculations in Ref. [18] predicted that $\text{Mo}_2\text{Ga}_2\text{C}$ was less likely to be a superconductor than Mo_2GaC . Therefore, an experimental measurement should be conducted to verify the conjecture.

2. Materials and methods

Highly pure $\text{Mo}_2\text{Ga}_2\text{C}$ powders were synthesized from molybdenum carbide (Mo_2C) powders (Sigma Aldrich Co., USA, 99.5 wt.%, 325 mesh) and gallium (Ga) ingots (Beijing Xingrongyuan Co., China, 99.99 wt.%) at 650°C for 60 h [20]. Five g of the prepared $\text{Mo}_2\text{Ga}_2\text{C}$ powders (91.3 wt.% pure) were then placed in a graphite mold ($\phi 20 \text{ mm}$) and sintered at 750°C for 8 h in a vacuum hot-press furnace (ZT-40-21Y, Chenhua, Shanghai, China) [25].

The powders and sintered samples were tested by X-ray diffraction (XRD; Smart-lab, Rigaku Corporation, Japan) using $\text{Cu-K}\alpha$ ($\lambda = 1.5406 \text{ \AA}$) radiation. The fractured and polished surfaces of the sintered samples were observed by scanning electron microscopy (SEM; Merlin Compact, Zeiss, Germany) equipped with an energy disperse spectroscope (EDS; X-Max^N, Oxford, UK).

The Seebeck coefficient and electrical resistivity were simultaneously measured from room temperature (RT) to 900 K by a CTA-3 apparatus (Cryoall, Beijing, China) from a sintered sample, which was cut into cubes ($10 \text{ mm} \times 3 \text{ mm} \times 2 \text{ mm}$). The thermal diffusivity was measured by a LFA457 laser flash apparatus (Netzsch, Germany) from a cubic sintered sample ($10 \text{ mm} \times 10 \text{ mm} \times 2 \text{ mm}$). The accuracy of Seebeck coefficient, electrical resistivity or thermal diffusivity was 7%, 10% or 7%, respectively. The temperature-dependent resistance of a sintered sample from RT down to 2 K was measured by a four-point method in magnet-cryostat physical

property measurement system (PPMS-DynaCool, Quantum Design, Inc, USA). The temperature accuracy was $\pm 0.1\%$ for $T < 10 \text{ K}$ (typical) and $\pm 0.02\%$ for $T > 10 \text{ K}$ (typical).

3. Results and discussion

3.1. Microstructure characterization of the $\text{Mo}_2\text{Ga}_2\text{C}$ bulks

The sintered bulk samples had a disc shape with a diameter of 20 mm and a thickness of 2 mm. The density, which was measured by the Archimedes method, was 7.81 g/cm^3 . The relative density was calculated to be 98.8%. The XRD patterns of the initial powders and the sintered samples are shown in Figure 1(a). The secondary electron SEM images are shown in Figure 1(b-d). The inset in Figure 1(d) is a backscatter electron SEM image of the polished surface of a sintered sample.

As shown in Figure 1(a), no new peaks were observed in the XRD pattern of the bulk sample after sintering. The peaks of impurity (Mo_2C and Ga_2O_3) were still very weak. From Rietveld analysis, the powder samples consisted of 91.3 wt.% $\text{Mo}_2\text{Ga}_2\text{C}$, 3.9 wt.% Mo_2C , and 4.7 wt.% Ga_2O_3 [20]. Due to strong preferred orientation of hot-pressed samples, Rietveld analysis cannot be used to obtain the compositions of bulk samples. However, it is convincing that the bulk sample, similar with powder sample, was still highly pure $\text{Mo}_2\text{Ga}_2\text{C}$ with a few impurities (Mo_2C and Ga_2O_3).

Figure 1(b) indicates that the $\text{Mo}_2\text{Ga}_2\text{C}$ powders had a grain size of $8 \pm 2 \mu\text{m}$. From Figure 1(c-d), the grain size of sintered samples was $1.8 \pm 0.2 \mu\text{m}$. Thus, the large grains of initial powders were broken into small grains in the hot-pressing process and the grain size did not significantly grow during sintering. From Figure 1(c), the sintered samples had a dense structure except for a few pores, which had diameters of $\sim 0.5 \mu\text{m}$ (red cycles in Figure 1(c-d)).

The back-scatter electron image (inset in Figure 1(d)) reflects the element composition of the sintered samples. The figure presents a dark gray color with some discrete dark areas and light-gray areas. The dark areas in the secondary electron image were determined to be pores. From EDS analysis, the main dark gray area has the molar ratio of $\text{Mo}:\text{Ga}:\text{C} = 1.4:1:2$, indicating that it is $\text{Mo}_2\text{Ga}_2\text{C}$. The discrete light-gray area has a molar ratio of $\text{Mo}:\text{Ga}:\text{C} = 8:0.1:8.4$, indicating that it is Mo_2C impurity. The presence of this impurities could decrease a little the electrical and thermal conductivity of the sample. However, these conductivities should be principally determined by the main phase, $\text{Mo}_2\text{Ga}_2\text{C}$.

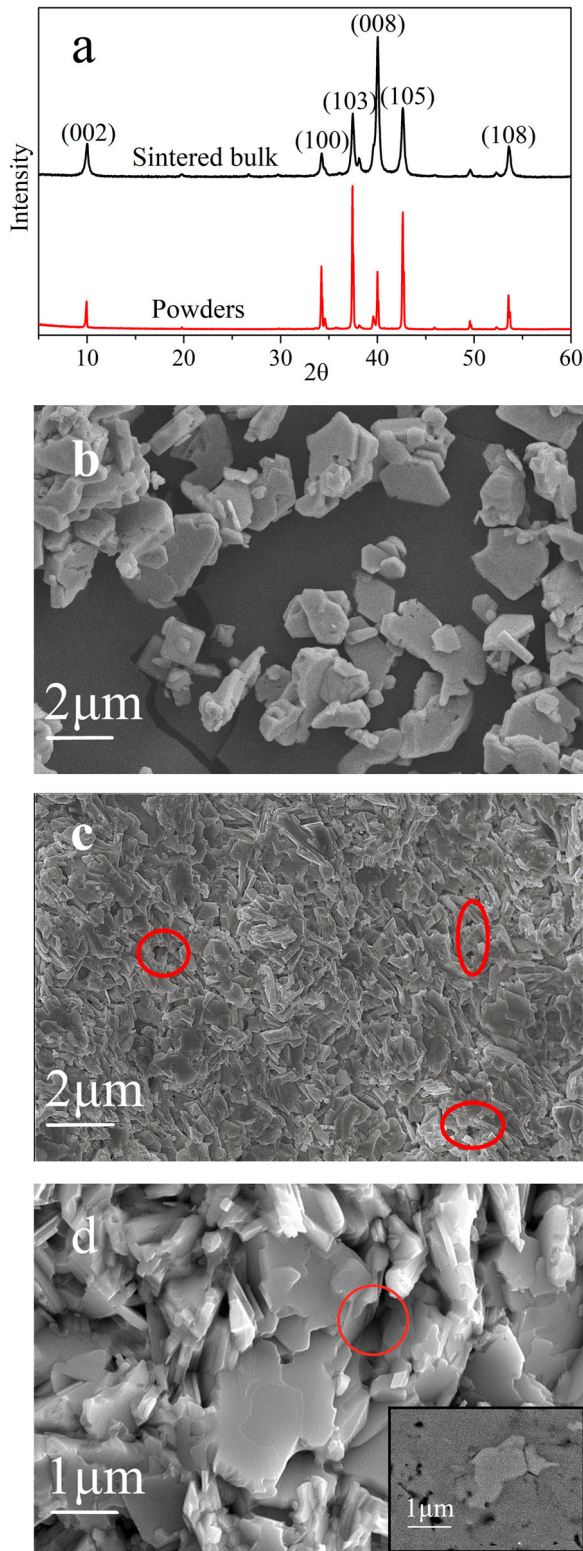


Figure 1. (a) XRD patterns of the powders and the sintered sample. (b) SEM image of the powders. (c) SEM image of the fracture surface of the sintered samples. (d) High magnification image of (c), the inset is back-scatter electron image of the polished surface.

3.2. Thermal conductivity and seebeck coefficient

The measured thermal diffusivity and Seebeck coefficient of the bulk samples are shown in Figure 2(a) and Figure 2(b), respectively. The thermal diffusivity (β) of the $\text{Mo}_2\text{Ga}_2\text{C}$ sample decreased smoothly with increasing temperature and gradually approached a fixed value.

The thermal conductivity (κ) of $\text{Mo}_2\text{Ga}_2\text{C}$ was calculated from the following equation:

$$\kappa = \beta \rho c \quad (1)$$

where ρ is density and c is the specific heat. Since the test was performed within the temperature range of 300 K to 900 K, *i.e.* in the high temperature region, the c was anticipated to follow the Dulong–Petit law [26]. Theoretical calculations showed the c of $\text{Mo}_2\text{Ga}_2\text{C}$ at a constant volume was approximate to a saturation value of 120 J/(mol·K) within the temperature range of 300 K to 1000 K [18]. Then, κ of $\text{Mo}_2\text{Ga}_2\text{C}$ in this temperature range was calculated from β by multiplying a constant (ρc). The κ values are shown in the right axis of Figure 2(a). The thermal conductivity of $\text{Mo}_2\text{Ga}_2\text{C}$

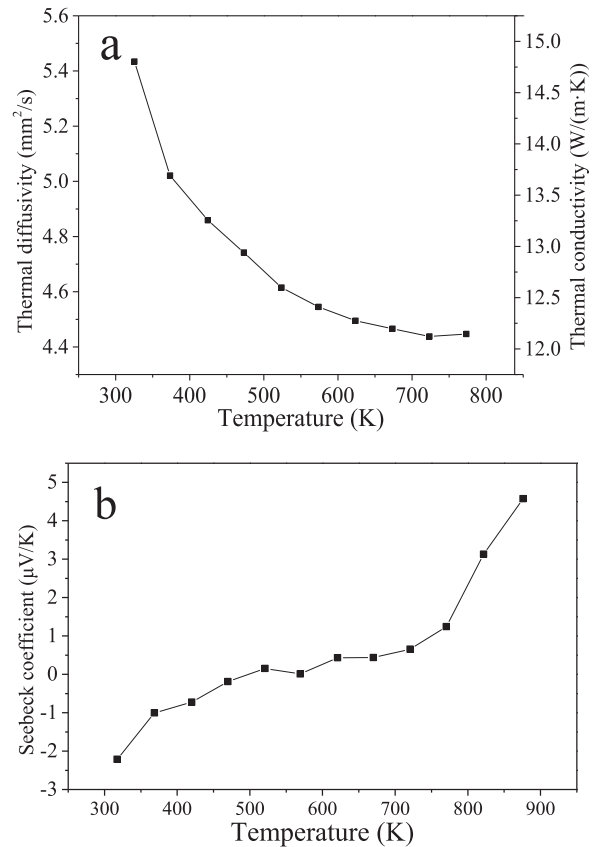


Figure 2. (a) Thermal diffusivity and thermal conductivity of $\text{Mo}_2\text{Ga}_2\text{C}$. (b) Seebeck coefficient of $\text{Mo}_2\text{Ga}_2\text{C}$.

at RT was calculated to be 14.8 ± 1.0 W/(m·K), which decreased with temperature and approached a fixed value of 12.1 W/(m·K).

According to a previous report [27], the κ of MAX phases at RT was within the range of 12 W/(m·K) to 60 W/(m·K). The κ of $\text{Mo}_2\text{Ga}_2\text{C}$, 14.8 ± 1.0 W/(m·K), was also within this range, though it was close to the lower limit. Thus, the κ of $\text{Mo}_2\text{Ga}_2\text{C}$ should be lower than that of most MAX phases.

Generally, the thermal conduction of the materials attributed to the motion of phonons, electrons, and/or holes. Considering the conduction of $\text{Mo}_2\text{Ga}_2\text{C}$, the motion of the electrical carriers (electrons/holes) should be the main mechanism. The Seebeck coefficient was tested to further understand the effect of electrons/holes. The temperature dependence of the Seebeck coefficients is shown in Figure 2(b). Similar to other MAX phases [28], $\text{Mo}_2\text{Ga}_2\text{C}$ exhibited a weak Seebeck effect. A Seebeck coefficient of approximately -2.2 $\mu\text{V/K}$ was observed at 300 K and increased slowly with temperature. At ~ 520 K, the coefficient changed from negative to positive. The maximum coefficient reached 4.6 $\mu\text{V/K}$ at 880 K. That is to say, electrons and holes jointly affect the transport properties of $\text{Mo}_2\text{Ga}_2\text{C}$ within this temperature range. The motion effects of the electrons and holes are almost equal, thus eliminating the Seebeck effect. This agrees well with the previous report, which indicated that apparent electron and hole densities are roughly equal in most MAX phases [28]. In addition, this phenomenon was theoretically explained by the complicated function of the shape of their Fermi surface [29,30].

3.3. Electrical resistivity at room temperature and at high temperature

The resistivity of the measured $\text{Mo}_2\text{Ga}_2\text{C}$ samples is shown in Figure 3. Figure 3(a) presents a linear increase in the resistivity of $\text{Mo}_2\text{Ga}_2\text{C}$ ($R^2 = 0.99974$) as the temperature rose from RT to 900 K, which indicates metal behaviour. This result confirms a previous theoretical prediction [4]. The relation between resistivity (R) and temperature (T) can be described by the following equation [31]:

$$R = R_0[1 + \alpha(T - 300)] \quad (2)$$

where R_0 is the resistivity at 300 K, and α is the temperature coefficient of resistivity. The resistivity at RT was measured to be 0.525 ± 0.052 $\mu\Omega\cdot\text{m}$. The α value was calculated to be 0.0036 K^{-1} .

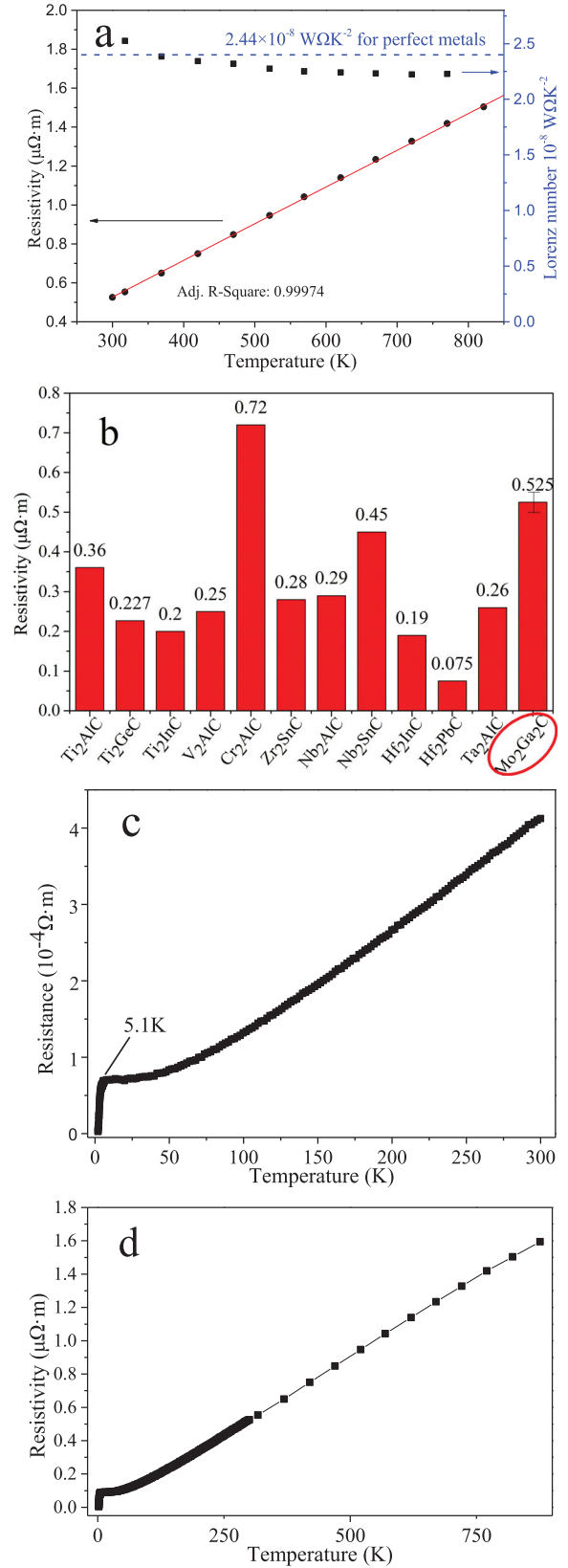


Figure 3. (a) Resistivity from RT to 900 K. The right vertical axis is the calculated Lorenz number, and the horizontal broken line is the number for perfect metals. (b) Comparison of $\text{Mo}_2\text{Ga}_2\text{C}$ resistivity with other MAX phases. (c) Resistance from 2 K to 300 K. (d) The resistivity-temperature relation from 2 K to 900 K.

The α of most MAX phases were within the range of 0.0021 K^{-1} – 0.0144 K^{-1} [2,27]. The α of the presented $\text{Mo}_2\text{Ga}_2\text{C}$ samples (0.0036 K^{-1}) is also within this range. The resistivity at RT was compared with those of the other MAX phases (Figure 3(b)). The resistivity of most MAX phases was within the range from $0.07 \mu\Omega\cdot\text{m}$ to $2.64 \mu\Omega\cdot\text{m}$ [27]. That of $\text{Mo}_2\text{Ga}_2\text{C}$ was also located within this range. However, $\text{Mo}_2\text{Ga}_2\text{C}$ exhibited a higher resistivity than most MAX phases with the exception of Cr_2AlC .

Based on the results and discussion in sections 3.2 and 3.3, $\text{Mo}_2\text{Ga}_2\text{C}$ exhibited a lower electrical conductivity and thermal conductivity, compared to most MAX phases. From the band structures and density of states, $\text{Mo}_2\text{Ga}_2\text{C}$ was predicted to be less conductive than its MAX phase counterpart Mo_2GaC , due to the additional Ga layer [4]. Thus, the additional A-layer theoretically decreased the conductivity of the double-A-layer MAX phase. It is possible that double-A-layer MAX phases have a lower conductivity of heat and/or electricity than regular MAX phase, though more experimental studies must be performed.

For the MAX phases, these conductivities were due to the joint effect of the motion of electrons and holes, as shown in the discussion of Figure 2(b). Thus, the lower conductivity must be due to the low mobility of the electrons and holes [32]. A previous study on Ti_3SiC_2 found that the electron mobility in TiC_x was significantly higher than that in Ti_3SiC_2 [33]. Thus, the A-layer in the MAX phases decreased the mobility of the carriers. In $\text{Mo}_2\text{Ga}_2\text{C}$ and other double-A-layer MAX phases, the extra A-layer can decrease more carrier mobility.

3.4. Wiedemann–Franz law

The thermal conductivity (κ) and the electrical conductivity (σ) of metals follow the Wiedemann–Franz Law [34], as described by the following equation:

$$\frac{\kappa}{\sigma} = LT \quad (3)$$

where L is the Lorenz number. According to the above results, $\text{Mo}_2\text{Ga}_2\text{C}$ has obvious metallic behaviours in the transport performance. Thus, $\text{Mo}_2\text{Ga}_2\text{C}$ is supposed to also follow the Wiedemann–Franz Law. From the κ in Figure 2(a) and σ calculated from R in Figure 3(a), L was calculated from Equation 3 and is shown in Figure 3(a). As expected, the calculated L between 300 and 800 K were close to the theoretical value for the perfect metals ($2.44 \times 10^{-8} \text{ W}\Omega\text{K}^{-2}$). Above 574 K, the L was stable at $2.22 \times 10^{-8} \text{ W}\Omega\text{K}^{-2}$. These results justify the validity of the measurements and that the $\text{Mo}_2\text{Ga}_2\text{C}$ is a metallic compound.

3.5. Resistance at low temperatures and superconductivity

The temperature-dependent resistance of a bulk sample was measured from RT down to 2 K (Figure 3(c)). The resistance clearly dropped precipitously at approximately 5.1 K ($\pm 0.1\%$) upon cooling, signaling the occurrence of superconductivity. This result indicates that, $\text{Mo}_2\text{Ga}_2\text{C}$, the first member of double-A-layer MAX phases, is a superconductor, which has never been previously validated experimentally. The transition temperature of $\text{Mo}_2\text{Ga}_2\text{C}$ was close to that of Mo_2GaC (c.a. 4.0 K) [21].

The resistivity of $\text{Mo}_2\text{Ga}_2\text{C}$ between 300 and 900 K is presented in Figure 3(a), and the resistance of a $\text{Mo}_2\text{Ga}_2\text{C}$ sample between 2 K to 300 K is presented in Figure 3(c). $\text{Mo}_2\text{Ga}_2\text{C}$ should have the same resistivity at RT in both Figure 3(a) and 3(c). Thus, the two figures were combined and the resistivity from 2 K to 900 K was then plotted (Figure 3(d)). The curve has the same slope (α value) at the low and high temperatures, which confirms the validity of the method to generate this figure. From this figure, it can be concluded that the resistivity of $\text{Mo}_2\text{Ga}_2\text{C}$ increased linearly with temperature within the range of 50 K to 900 K. In addition, Equation 2 is applicable in the large temperature range.

4. Conclusions

The present study measured and reported the thermal conductivity and electrical resistance of the $\text{Mo}_2\text{Ga}_2\text{C}$ bulk samples. The thermal conductivity was measured to be $14.8 \pm 1.0 \text{ W}/(\text{m}\cdot\text{K})$ at RT. From the Seebeck coefficient, the electrons and holes jointly affect the transport properties of $\text{Mo}_2\text{Ga}_2\text{C}$. The electrical resistivity of $\text{Mo}_2\text{Ga}_2\text{C}$ at RT was measured to be $0.525 \pm 0.52 \mu\Omega\cdot\text{m}$ and the resistivity increased linearly with temperature within the range of 50 K to 900 K. The thermal conductivity and electrical resistivity of $\text{Mo}_2\text{Ga}_2\text{C}$ were in the range of those of the MAX phases. However, they were lower than those of most MAX phases due to the extra Ga layer. $\text{Mo}_2\text{Ga}_2\text{C}$ follows the Wiedemann–Franz law, and the Lorenz number was calculated to be $2.22 \times 10^{-8} \text{ W}\Omega\text{K}^{-2}$. Moreover, $\text{Mo}_2\text{Ga}_2\text{C}$ is a superconductor with a transition temperature of ca. 5.1 K ($\pm 0.1\%$).

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Disclosure statement

No potential conflict of interest was reported by the author(s).

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