



High-pressure Synthesis and Bulk Modulus of Non-centrosymmetric Superconductor $\text{Mo}_3\text{Al}_2\text{C}$

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High-pressure Synthesis and Bulk Modulus of Non-centrosymmetric Superconductor $\text{Mo}_3\text{Al}_2\text{C}$

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Abstract. $\text{Mo}_3\text{Al}_2\text{C}$ is a superconductor without inversion symmetry (the transition temperature $T_C \sim 9\text{K}$). We have succeeded in preparing high-quality samples of $\text{Mo}_3\text{Al}_2\text{C}$ using the high-pressure synthesis technique. The samples were characterized by powder x-ray diffraction (XRD) analysis. Furthermore, powder XRD patterns for the samples with synchrotron radiation have been studied under high pressures up to around 10 GPa and the volume versus pressure curve for the compound has been investigated. A bulk modulus was estimated to be 221 GPa.

1. Introduction

The Mo-based superconductor $\text{Mo}_3\text{Al}_2\text{C}$ crystallizes with the β -Mn type structure of space group $P4_132$ with a transition temperature $T_C \sim 9\text{K}$ [1, 2]. The structure of the compound does not have a center of inversion. Superconductors without inversion symmetry such as CePt_3Si recently received special attention due to their exotic properties [3]. A mixture of spin-singlet and spin-triplet pairs condensate is proposed to explain superconductivity in the non-centrosymmetric compounds. The absence of Hebbel-Slichter coherence peak, a power-law behavior of spin-lattice relaxation rate, specific heat deviating from BCS model and positive pressure dependence of T_C suggest that $\text{Mo}_3\text{Al}_2\text{C}$ is an unconventional superconductor [2]. However, the sample reported ref. 2 includes Mo_2C as an impurity phase. In order to study the superconductivity of $\text{Mo}_3\text{Al}_2\text{C}$ in detail, high-quality samples without Mo_2C , which is a superconductor with $T_C \sim 3\text{K}$ [4], are required.

We have succeeded in preparing high-quality samples of $\text{Mo}_3\text{Al}_2\text{C}$ without Mo_2C under high pressure using a multi-anvil type high-pressure press. In this study, we report the results of powder x-ray diffraction (XRD) measurements of $\text{Mo}_3\text{Al}_2\text{C}$ under high pressure to investigate the structural stability of the compound.

2. Experimental

Single-phase polycrystalline samples of $\text{Mo}_3\text{Al}_2\text{C}$ were synthesized at high temperatures and high pressures using a double-stage multi-anvil high-pressure apparatus. $\text{Mo}_3\text{Al}_2\text{C}$ was prepared by reaction of stoichiometric amounts of 3N (99.9% pure) Mo, 3N-Al and 5N-C powders at 4 GPa. The reaction temperature was 1200°C. The sample cell assembly is similar to that used for the high-pressure synthesis of $\text{CeRu}_4\text{As}_{12}$ [5]. The prepared samples were characterized by powder x-ray diffraction (XRD) using $\text{Co } K\alpha_1$ radiation and silicon as a standard.

Using synchrotron radiation, powder XRD patterns of $\text{Mo}_3\text{Al}_2\text{C}$ were measured with a diamond-anvil cell (DAC) and an imaging plate at room temperature and high pressures [6]. High-pressure



XRD experiments with $\lambda = 0.619 \text{ \AA}$ were performed at the beam line 18C of the High Energy Accelerator Research Organization (KEK) Photon Factory in Tsukuba. We employed the DAC with diamond culet diameters of $500 \mu\text{m}$. The sample was finely ground and loaded in the $100 \mu\text{m}$ diameter hole drilled in a stainless steel (T301) gasket. The pressure in the DAC was determined by a pressure shifts in the R-line fluorescence spectrum of ruby. A 4:1 methanol-ethanol solution was used as the pressure medium.

3. Results and discussion

Figure 1 shows a powder XRD pattern of $\text{Mo}_3\text{Al}_2\text{C}$ prepared under high pressure. Most of the observed diffraction peaks were indexable using the β -Mn type structure. We were unable to detect other impurity phases including Mo_2C at the experimental resolution. The lattice constant determined by a least-squares fit to the data was 6.8623 \AA , in reasonable agreement with 6.8630 \AA as reported previously [2]. Specific heat and magnetic susceptibility measurements of $\text{Mo}_3\text{Al}_2\text{C}$ prepared under high pressure reveal superconductivity of the samples at around 9K, in agreement with the data reported earlier [2].

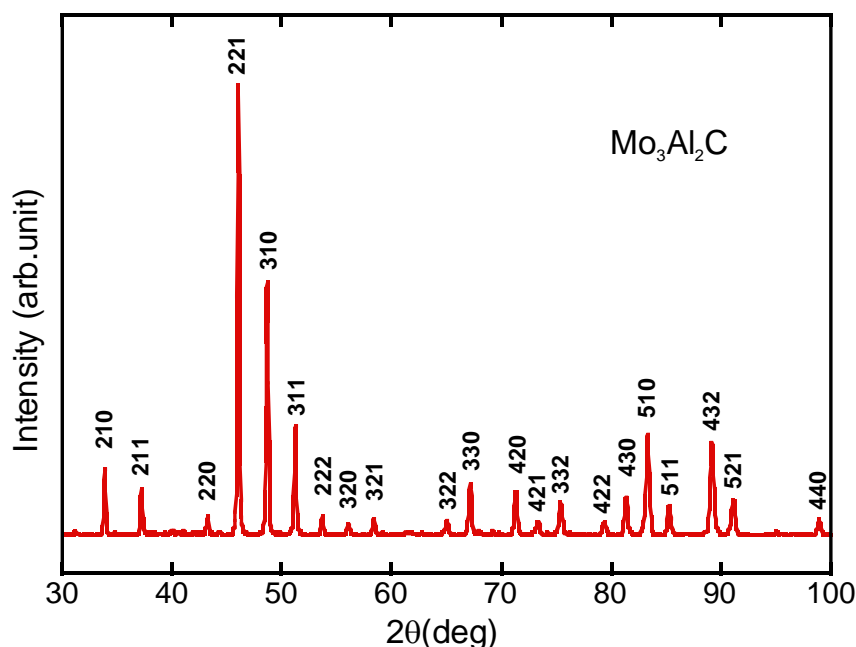


Figure 1. Powder x-ray diffraction pattern of $\text{Mo}_3\text{Al}_2\text{C}$ prepared under high pressure

Using a synchrotron radiation, the powder XRD of $\text{Mo}_3\text{Al}_2\text{C}$ has been studied under high pressures up to 9.74 GPa with the diamond anvil cell at room temperature. The d-values of the diffraction peaks decrease with increasing pressure. The lattice constant of $\text{Mo}_3\text{Al}_2\text{C}$ decreases to 6.7270 \AA at 9.74 GPa. No obvious change, such as the appearance of any new diffraction peaks, was observed. The result indicates that there is no change in crystal structure type within this pressure range. When pressure is reduced from 9.74 GPa to the ambient pressure, the d-values of the diffraction peaks return to those at ambient pressure. This behavior is completely reversible.

Figure 3 shows the relative cell volume (V/V_0) versus pressure curve for $\text{Mo}_3\text{Al}_2\text{C}$. Here, V_0 is the unit cell volume at ambient pressure. The cell volume of $\text{Mo}_3\text{Al}_2\text{C}$ monotonically decreases with increasing pressure up to around 10 GPa. This experimental data can be fitted by a Birch equation of state [7]:

$$P = \frac{3}{2}B_0 \left\{ \left(\frac{V}{V_0} \right)^{-\frac{7}{3}} - \left(\frac{V}{V_0} \right)^{-\frac{5}{3}} \right\} \cdot \left[1 - \frac{3}{4}(4 - B_0') \left\{ \left(\frac{V}{V_0} \right)^{-\frac{2}{3}} - 1 \right\} \right] \quad (1)$$

where B_0 is the bulk modulus, B_0' its first pressure derivative, V the volume, and P the pressure. A least-square fit to the data of $\text{Mo}_3\text{Al}_2\text{C}$ gives the following values: $B_0 = 221$ GPa and $B_0' = 2.3$.

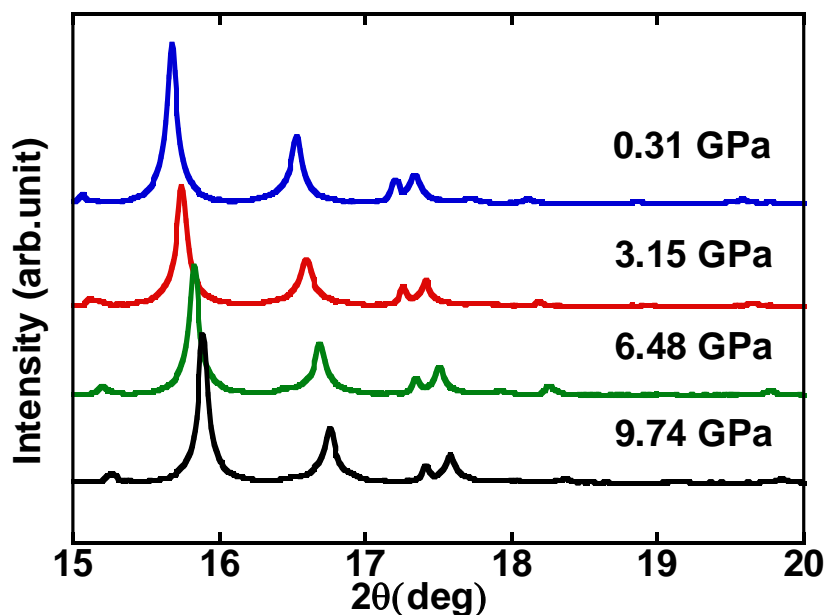


Figure 2. Powder x-ray diffraction patterns of $\text{Mo}_3\text{Al}_2\text{C}$ at each high pressure

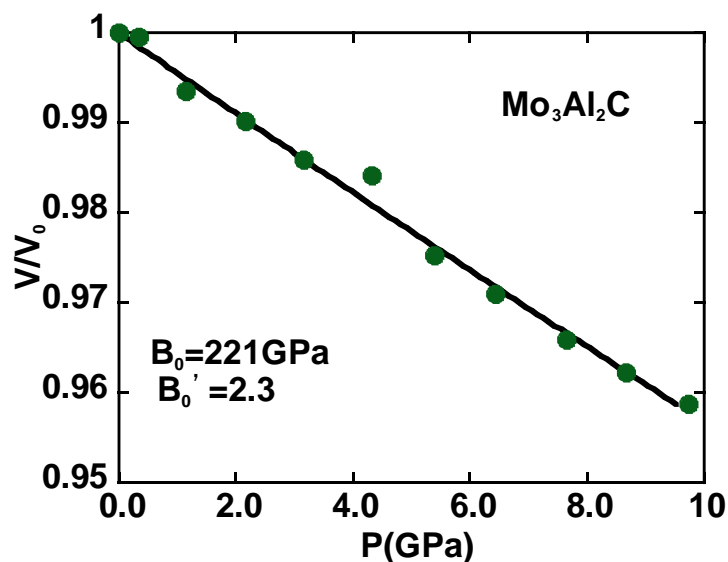


Figure 3. Pressure dependence of the relative unit cell volume V/V_0 of $\text{Mo}_3\text{Al}_2\text{C}$ in the pressure range up to around 10 GPa. The line shows the compression curve of the bulk modulus $B_0 = 221$ GPa, which was obtained by fitting of a Birch equation of state to data.

4. Summary

We have synthesized high-quality samples of $\text{Mo}_3\text{Al}_2\text{C}$ without Mo_2C using the high-pressure synthesis technique. The crystal structure of $\text{Mo}_3\text{Al}_2\text{C}$ is found to be stable at high pressures up to 9.74 GPa. The bulk modulus of $\text{Mo}_3\text{Al}_2\text{C}$ has been estimated from the XRD data under high pressure. The value of B_0 for the compound is 221 GPa. In order to investigate the superconducting properties of $\text{Mo}_3\text{Al}_2\text{C}$ in detail, photoelectron spectroscopy and NMR measurements are currently in progress.

Acknowledgments

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