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Hydrostatic Compression Effects on Fifth-Group Element Superconductors Nb and Ta Subjected to High-Pressure Torsion

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Severe plastic deformation by high-pressure torsion (HPT) brings about strain at the unit-cell level as well as reduction in the grain size. We investigated the crystal structure of severely strained Nb and Ta materials under the hydrostatic compression, and discussed the origin of the change in superconducting transition temperature T_c. The large increase in T_c for HPT-processed Nb is triggered by the structural deformation in the unit-cell level, while the small increase in T_c for HPT-processed Ta originates from strengthening intergrain-contact.

1 Introduction

In the periodic table containing 118 elements, there are 29 superconductors at ambient pressure, and 11 elements of them exhibit the increase in superconducting transition temperature T_c under hydrostatic pressure (HP) [1, 2]. Three fifth-group element superconductors such as V, Nb, and Ta have nominal T_c of 5.3 K, 9.2 K, and 4.5 K, respectively. All of three are considered to belong to a group that exhibits the increase in T_c under the HP circumstances [2]. Indeed, smooth lineac-increase in T_c is observed only in V [3]. According to the study of Struzhkin *et al.*, nonstrained Nb shows the increase in T_c after its slight decrease, and has the optimal T_c of 9.9K at around P = 10 GPa [4]. Indeed, the initial dT_c/dP for Nb depends on experimental conditions such as the sample condition and kind of pressure transmitting medium (PTM) [3, 5]. Contrary to the above two, Ta exhibits the decrease in T_c over wide pressure range [3], and there is only one plot suggesting the increase in T_c at 43 GPa [4]. Thus, there are much variety in the pressure dependence of T_c among V, Nb, and Ta (see Fig. 1(a)). In the present study, we compare –the aforementioned HP effects on non-strained V, Nb, and Ta (see Fig. 1(a)) and the HP effects on severely strained V, Nb, and Ta (see Fig. 1(b)) considered as the so-called Josephson arrays of small grains.

Recently by using severe plastic deformation (SPD) [6, 7], the improvement of functionality in superconductors has been successfully conducted [8, 9]. The SPD process plays the role of accumulating dislocations, resulting in the grain refinement at the submicrometer or nanometer ranges. It is known to be effective in increasing T_c of superconductors such as Nb [8] and Re [9]. There we have to consider both the intergrain network via grain boundary and intragrain crystallinity as factors to influence T_c. The strain installed in the SPD process is relatively evaluated via semi-experimental manner: In the case of high-pressure torsion (HPT) [10, 11] which is one of the SPD techniques, the revolution number N is a parameter to pursue the grain refinement.

We are exploring the observation of new pressure response using the SPD materials. In the present study, we conduct the HP compression to strained materials subjected to the HPT process. Figure 2 shows a strategy the present study. There are two parameters such as N related with the initially installed strain [Fig. 2(a)] and pressure (P) related with additional strain [Fig. 2(b)]. Both HPT-Nb (N = 10) and HPT-Ta (N = 10) exhibit the maximum T_c at around P = 2 GPa.

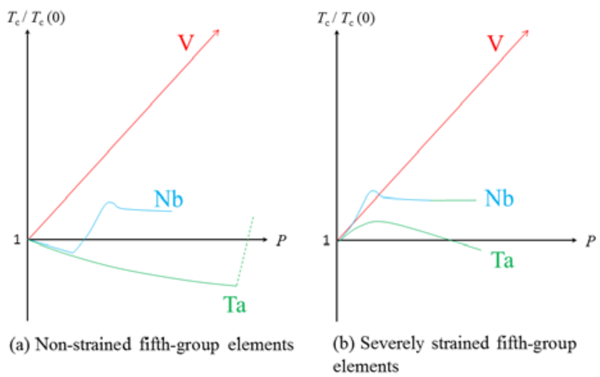

Fig. 1: Overview on pressure dependence of superconducting transition temperature T_c for (a) non-strained and (b) severely strained V, Nb, and Ta [3].

2 Experiment

As illustrated in Fig. 2(a), the disks of Nb, and Ta were subjected to HPT processing at room temperature under a selected pressure of P = 6 GPa and revolutions of N up to 20 with a rotation speed of 1 rpm. Specimens with dimensions of approximately 0.8 × 0.8 × 0.5mm^3 were cut from an HPT-processed disk using a wire-cutting electric discharge machine at the position of 2.5mm from the disk center. The grain size at steady state for Nb and Ta is 240 nm and 180 nm, respectively [12]

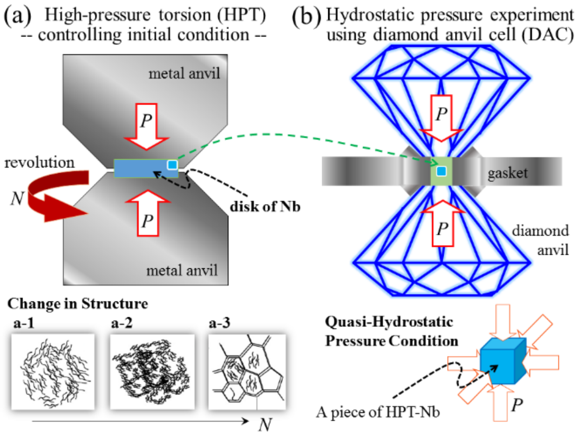

Fig.2: Strategy of the present study in the case of Nb [3, 5]. (a) In high-pressure torsion (HPT) for controlling the initial condition, pressurization and revolution are conducted by using two metal anvils. As the revolution number (*N*) increases, dislocations are accumulated (a-1), and the grain size largely changes. The small-angle grain boundaries gradually transform to large-angle grain boundaries with increasing *N* ((a-2)→(a-3)). Finally due to the balance between generation and annihilation of dislocations, the grain size remains unchanged despite the increase in *N*. (b) A few pieces of sample cut from the HPT-processed materials are placed in diamond anvil cell (DAC) to conduct the hydrostatic pressure experiment. There, the specimen is pressed in a quasi-hydrostatic pressure condition using the DAC.

We performed X-ray diffraction (XRD) analyses for Nb and Ta under high pressure up to P = 6.8 GPa at room temperature using a synchrotron radiation XRD system with a cylindrical imaging plate at BL-8B. The energy of the incident X-rays was 16 keV. Pressure was applied using a DAC that consisted of two diamond anvils with flat tips having a diameter of 0.8mm and a 0.3-mm-thick CuBe gasket. The small pieces of HPT-Nb or HPT-Ta were placed in a randomly oriented manner in a sample cavity with a diameter of 0.4mm along with a ruby of manometer and transparent PTM such as fluorinated oil (FC77, Sumitomo 3M Ltd.). Transparent FC77 undergoes solidification below 1 GPa at room temperature [13]. All the atomic positions in the body-centered cubic (bcc) structure are special positions, and therefore, the structural parameters can be evaluated just with the use of the diffraction peak angle. The lattice parameters were estimated using all of observed diffraction peaks: For Nb, we used five diffraction peaks of the plane indices (110), (200), (211), (220), and (310). For Ta, the diffraction peak of the plane index (310) was not used.

3 Results and Discussion

Figures 3 shows the pressure dependence of interplanar distance of (110), (200), (211), (220), and (310) for the HPT-Nb materials for N = 1 (c) and 10 (d). For reference, the data for both arc-melting (ARC) and AR materials are also shown in Figs. 3(a) and 3(b), respectively. At first, the AR material exhibits anisotropic transformation of the unit cell structure at around P = 2 GPa, suggesting that strain accumulated in the cubic lattice with high symmetry is released at a threshold pressure (P_c). This phenomenon does not occur in the ARC material without residual strain, suggesting the importance of residual strain. When the HPT procedure with N = 1 is subjected to the AR material, the grain refinement enhances the capacity of accumulating the additional strain, resulting in the increase in P_c to approximately 3.4 GPa. However, HPT with the level of N = 10 does not permit to accumulate any additional strain, so that anisotropic unit-cell deformation occurs even at quite small pressure.

Comparing with ARC, AR and HPT (N = 1) exhibits the one-direction shrinkage and two-direction expansion. On the other hand, the HPT (N = 10) material exhibits two-direction shrinkage for P < 2 GPa and two-direction expansion for P > 2 GPa. At around 2 GPa, the deformation manner changes, suggesting complex manner of strain release. In particular, the HPT (N = 10) material with heavy residual strain has a deformed bcc structure, which is close to the body-centered tetragonal (bct) structure for P > 3 GPa.

Figures 4 shows the pressure dependence of interplanar distance of (110), (200), (211), and (220) for the HPT-Ta samples for N = 0 (a), 2 (b), and 10 (c). In all of three, all of interplanar distances exhibit the shrinkage, and there are no anisotropic deformation of the bcc unit cell. For N = 0, at around P = 1~2 GPa, the shrinkage magnitude is suppressed similarly to that observed in AR-Nb. The above pressure region is consistent with that possessing the maximum of T_c. On the other hand, both N = 2 and 10 shows that the uniform contraction continues monotonously. Both HPT (N = 2) and HPT (N = 10) do not show anomalous change in a at around P = 1~2 GPa, suggesting that the increase in T_c cannot be explained only with the change in lattice constant. The normalized lattice constant exhibits no prominent N dependence, and it continues to decrease monotonously in the considered pressure region.

In fifth-group element superconductors V, Nb, and Ta, artificial material manipulation using the severe plastic deformation and additional hydrostatic compression was attempted to increase the superconducting transition temperature T_c. In HPT-Nb with the small N and HPT-Ta for N = 0, 2, and 10, the increase in T_c due to strengthening the intergrain contacts was observed. Further, the prominent increase in T_c for HPT-Nb for N = 10 originates in the anisotropic deformation of the unit cell. When the quasihydrostatic compression is yielded to the material with heavy residual strain in the unit cell, severe strain accumulated in the bcc unit cell with high symmetry is released at a small amount of additional pressure. In the case of Nb, this manner of lattice deformation is favorable for increasing T_c. The knowledge obtained there yields a new strategy to increase T_c from the viewpoints of material science. There would be further potentiality for increasing T_c in the border region between material science using severe plastic deformation and extreme condensed-matter physics using hydrostatic compression.

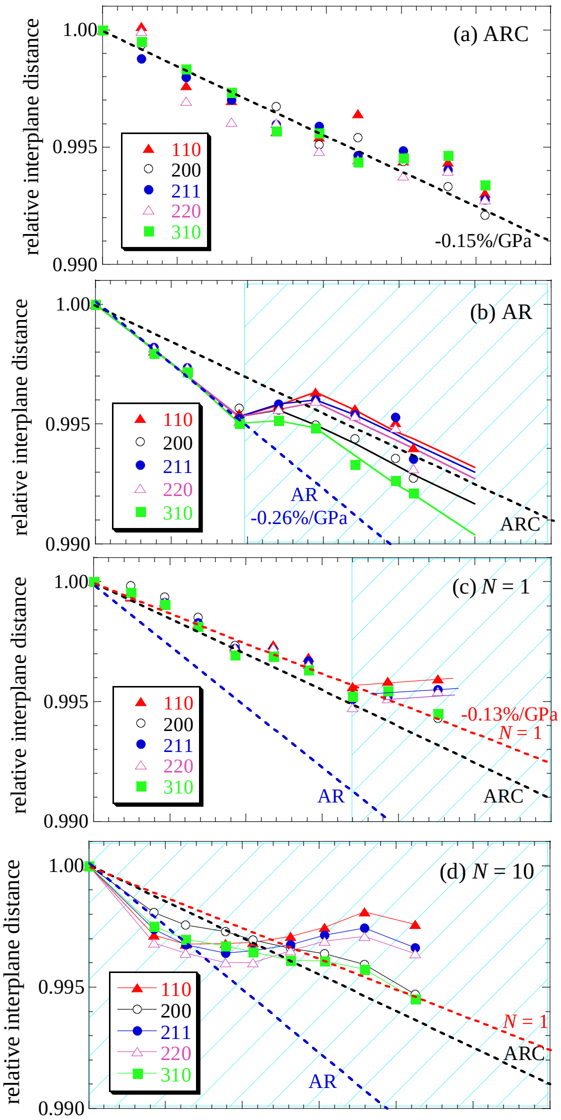

Fig.3: Pressure dependence of normalized interplanar distance of (110), (200), (211), (220), and (310) planes for ARC (a), AR (b), HPT-Nb (N = 1) (c), and HPT-Nb (N = 10) (d) [3]. The broken guide lines in all figures represent the interplanar distance change for ARC (black), AR (blue), and N = 1 (red). These initial slope values in the pressure regime where isotropic structure change occurs are also indicated. The pressure regime corresponding to anisotropic structure change is indicated in lightblue hatching.

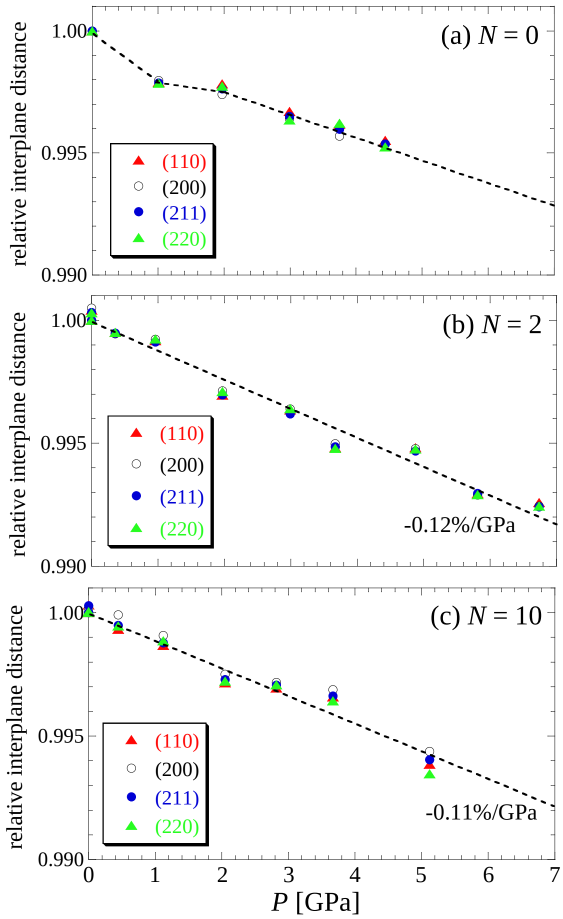

Fig. 4: Pressure dependence of normalized interplanar distance of (110), (200), (211), and (220) planes for HPT-Ta with N = 0 (a), 2 (b), and 10 (c) [3].

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