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Thermal stability of energetic ion irradiation induced amorphization for Ni_3Nb and Ni_3Ta intermetallic compounds

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To investigate the thermal stability of the ion-irradiation induced amorphous state in bulk samples of Ni_3Nb and Ni_3Ta intermetallic compounds, they were irradiated with 16MeV Au ions at room temperature, and were subsequently annealed up to 773K and 973K, respectively. The lattice structures of the irradiated samples and the annealed samples were examined by means of the grazing incidence x-ray diffraction (GIXD). The hardness of the samples was also measured as a function of annealing temperature. The amorphous state induced by room temperature irradiation recovers to the intrinsic ordered structure for Ni_3Nb samples at 773K. On the other hand, for Ni_3Ta samples, the irradiation-induced amorphous state doesn't recover to the intrinsic ordered structure even at 973K, but it changed to another lattice structure by the subsequent annealing. The values of the hardness decreased by the subsequent annealing. For Ni_3Nb samples, the irradiation at elevated temperatures was also performed. The higher temperature irradiation tends to suppress the amorphization more strongly in Ni_3Nb samples. As a result, the increase in hardness becomes smaller for the higher temperature irradiation.

Key words: Ni_3Nb, Ni_3Ta, ion irradiation at elevated temperatures, thermal annealing, lattice structure transformation, Vickers hardness, amorphization

1. INTRODUCTION

Various lattice structure transformation phenomena induced by charged particle irradiations have been reported so far [1-4]. In the previous experiments, thin film samples have mainly been used to observe the lattice structures through transmission electron microscopes (TEM). In contrast, the present study has adopted bulk intermetallic compounds samples as irradiation targets. In addition to the structure analysis, it is easy to evaluate macroscopic properties such as mechanical properties by using bulk samples. We have recently reported that the amorphization and the surface hardness change are induced in Ni_3Nb and Ni_3Ta intermetallic compounds by the energetic ion irradiation at room temperature [5]. As the amorphous state of Ni_3Nb and Ni_3Ta at room temperature is far from the thermal equilibrium state, it is of great interest to investigate its thermal stability. In the present study, we have examined the thermal stability of the irradiation-induced amorphous state in Ni_3Nb and Ni_3Ta by the following two experimental methods; one is the thermal annealing after the room temperature irradiation and the other is the ion irradiation at elevated temperatures.

2. EXPERIMENTAL PROCEDURE

Buttons of Ni_3Nb and Ni_3Ta were prepared by arc melting of high purity nickel and niobium or tantalum in an argon atmosphere. They were homogenized at 1273 K for 96 and 144 hours, respectively. The buttons were sliced into sheets with the dimension of 10mm, 10mm and 1mm. Finally, their surfaces were mechanically polished. The Ni_3Nb and Ni_3Ta samples were irradiated with 16MeV Au^{5+} ions by using a 3MV tandem accelerator at Takasaki Advanced Radiation Research Institute, National Institutes for Quantum and Radiological Science and Technology. The sample temperature during the irradiation was room temperature, 423K, or 523K for Ni_3Nb and was room temperature for Ni_3Ta.

The samples which were irradiated with the fluence of $5\times10^{14}/cm^2$ at room temperature were used for the thermal annealing experiment. For the Ni_3Nb sample, the isochronal annealing experiments were performed in a vacuum of about 3×10^{-4} Torr at 373, 473, 573, 673 and 773K. For the Ni_3Ta sample, the annealing temperatures were 873 and 973 K in addition to 373, 473, 573, 673 and 773K. Each annealing time was 100 min.

As was discussed by using SRIM profiles in the previous report [5], the effect of the Au ion irradiation is localized only at the region from the sample surface to the depth of about 2 μm. Therefore, we have performed the grazing incidence X-ray diffraction (GIXD) to observe the surface lattice structure for the samples irradiated at elevated temperatures and for those irradiated at room temperature and subsequently annealed at elevated temperatures.

In order to investigate the effect of irradiation at elevated temperatures on the hardness change, we have performed the micro Vickers hardness measurements. We have already reported the effect of room temperature irradiation on the hardness for Ni_3Nb and Ni_3Ta samples [5]. Also, we have measured the hardness of samples after the thermal annealing. The applied load was 10 gf (98.07mN) to observe the irradiated region. The time interval of indentation was kept at 10 seconds.

3. RESULTS AND DISCUSSION

3.1 Results of the irradiation at elevated temperatures

First of all, the effect of irradiation temperature on the irradiation-induced lattice structure transformation will be discussed. Fig. 1a shows the GIXD spectra for the unirradiated Ni_3Nb sample and those irradiated with Au ions at room temperature [5]. For the spectrum of unirradiated sample, the peaks corresponding to the orthorhombic structure, which is the thermal equilibrium lattice structure of Ni_3Nb at room temperature [6], are observed. In the figure, the indices for the orthorhombic structure are shown. For the fluence of $1\times10^{13}/cm^2$, these peak positions hardly changes, meanings that the sample still keeps the orthorhombic structure. For the fluences of $5\times10^{13}/cm^2$ and $5\times10^{14}/cm^2$, the peaks for the orthorhombic structure completely disappear, and a broad peak clearly appears around 43°. The change in the GIXD spectra shows that the amorphous state is induced in Ni_3Nb by the irradiation. Fig. 1b shows the GIXD spectra for the Ni_3Nb samples irradiated at 423K. For the fluence of $5\times10^{13}/cm^2$, the spectrum shows that the sample is almost amorphized, but a small part of the sample remains the orthorhombic structure because tiny peaks corresponding to the crystalline structure can still be seen in the spectrum. For the irradiation with higher fluence ($5\times10^{14}/cm^2$), all the peaks for the orthorhombic structure disappear completely and only a broad peak is observed, i.e, the sample is completely amorphized by the ion irradiation. Figs. 1a and 1b imply that a larger fluence is necessary for the irradiation at higher temperature to induce the amorphous state in the Ni_3Nb samples. Such a tendency becomes more remarkable in the case of the Au irradiation at 523K (Fig. 1c). Even for the fluence of $5.0\times10^{14}/cm^2$, some peaks corresponding to the orthorhombic structure are still observed. Within the ion-fluence range of the present experiment, the complete amorphization was not realized under the Au irradiation at 523K. The irradiation temperature dependence of the amorphization can be explained as follows; the energetic ion irradiation deposits very large energy into narrow regions of the sample instantaneously. As a result, the atomic arrangement in the lattice is disordered and the long-range ordering is lost. Immediately after the disordering, atomic rearrangement occurs because the state induced by irradiation is a non-equilibrium state. In the case of irradiation at higher temperatures, the atomic rearrangement, which is assisted by the thermal diffusion of the constituent atoms, occurs more easily.

Fig. 2 shows that the hardness increases with increasing ion fluence for all the irradiation temperatures. Moreover the figure indicates that at the same fluence, the increase in hardness is smaller for the irradiation at higher temperature. These results suggest that the volume ratio of the amorphous state in the Ni_3Nb samples is an important factor for the hardness changes, because it has been reported conventionally that the amorphous state is harder than the crystal structure [7, 8]. The dependence of the irradiation temperature on the irradiation-induced amorphization has been reported by using TEM observation [9-11]. However, few researches have discussed the relationship between the lattice structure change and the mechanical properties because TEM samples are too thin to measure macroscopic properties.

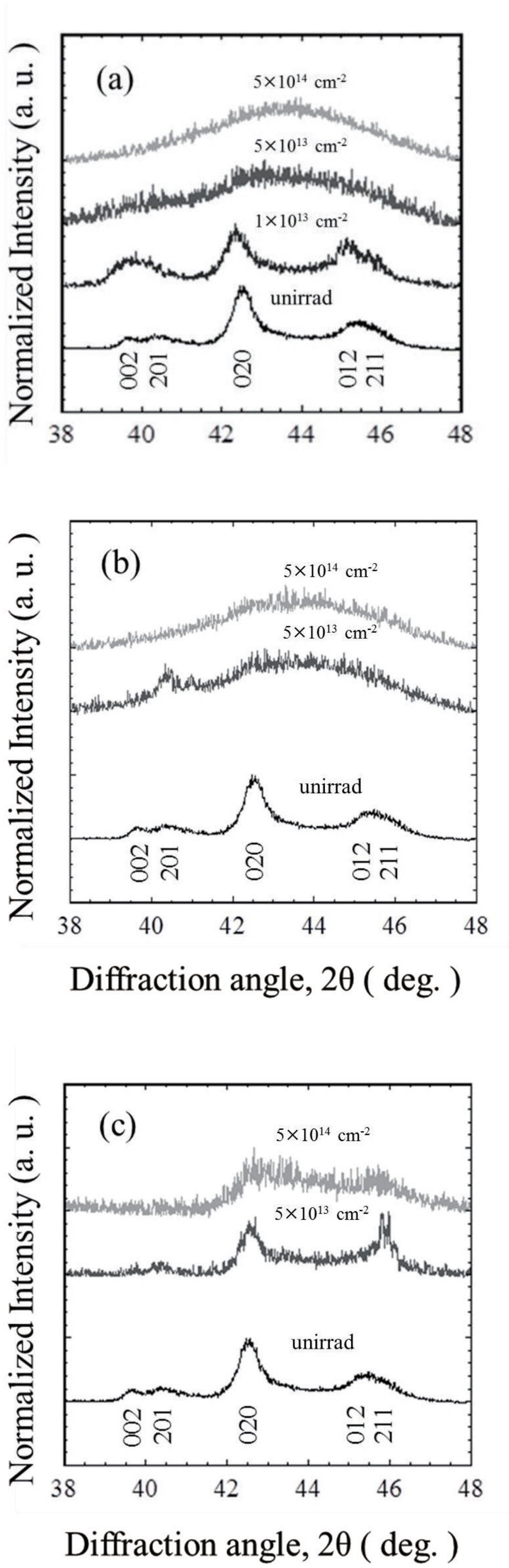

Fig. 1 GIXD spectra for the unirradiated Ni_3Nb sample and those irradiated with Au ions. Irradiation temperature is (a) room temperature, (b) 423K and (c) 523K.

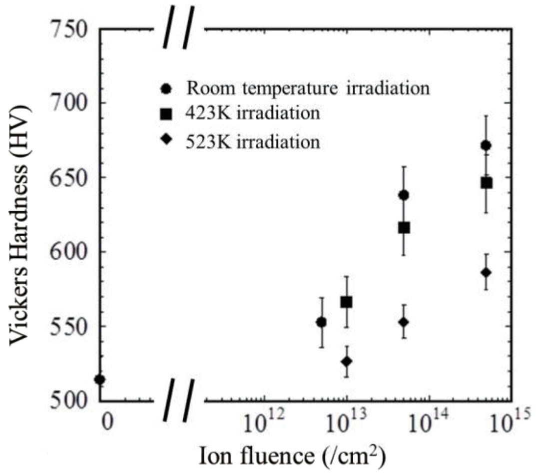

Fig. 2 Change in the Vickers hardness as a function of ion fluence for the Ni_3Nb sample for irradiation at room temperature, 423K and 523K.

3.2 Results of the subsequent thermal-annealing after room temperature irradiation

Fig. 3 shows the annealing temperature dependence of the GIXD spectra for Ni_3Nb irradiated with the fluence of $5\times10^{14}/cm^2$. Before the thermal annealing, the irradiated sample shows a broad peak which corresponds to the amorphous state. By the annealing below 673K, the broad peak remains unchanged. However, after the annealing at 773K, the broad peak disappears completely and the peaks for the orthorhombic structure appear. The figure clearly shows that the amorphous state induced by the irradiation recovers to the orthorhombic structure by the annealing at 773K. For Ni_3Ta, a more interesting result for the thermal annealing has been found. Fig. 4a shows the GIXD spectra of Ni_3Ta irradiated with the fluence of $5\times10^{14}/cm^2$ for various annealing temperatures. For comparison, the GIXD result for the unirradiated sample is also shown in Fig. 4a. The broad peak induced by irradiation hardly changes and the irradiation-induced amorphous state is stable by the thermal annealing from 373K to 573K. After the annealing at 773K, several peaks appear in the GIXD spectrum. These peaks do not correspond to the intrinsic monoclinic structure but they correspond to the orthorhombic structure which is not the thermal-equilibrium lattice structure of Ni_3Ta at room temperature. Fig. 4b shows the GIXD spectra for the rear surfaces of the irradiated Ni_3Ta samples. As the range of 16MeV Au ion is much smaller than the sample thickness, the rear surfaces of the irradiated samples were not affected by the irradiation, but, were exposed only to high temperatures during the thermal annealing treatments. Even after the annealing at 973K, the intrinsic monoclinic structure for the rear surface of the sample remains unchanged. The figure reveals that the structure change in Fig. 4a is not only due to thermal treatment, but is attributed to the combination of the room temperature irradiation and the subsequent thermal annealing. As the amorphous state induced by ion irradiation is non-equilibrium, it is difficult for the amorphous state to survive the thermal annealing at elevated temperatures. The thermal-equilibrium phase diagram of Ni_3Ta intermetallic compound which has been reported up to the present does not clearly show the existence of the orthorhombic structure. Therefore, the experimental result for the thermal annealing of the ion-irradiated Ni_3Ta can be explained as due to the thermally activated process from the amorphous state to the metastable orthorhombic state. The thermally activated process at the annealing temperatures of the present experiment is not sufficient to recover the lattice structure of Ni_3Ta to the intrinsic monoclinic structure.

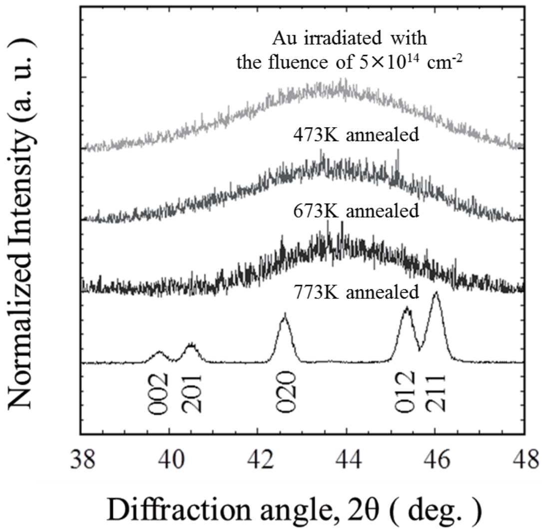

Fig. 3 GIXD spectra for Ni_3Nb irradiated with the fluence of $5\times10^{14}/cm^2$ and for Ni_3Nb thermally annealed at 473K, 673K and 773K.Indices below the spectrum of 773K annealed sample are for orthorhombic structure.

Fig. 5 shows the change in the Vickers hardness as a function of annealing temperature for the Ni_3Nb sample. After the 373K annealing, the hardness slightly decreases. After the annealing between 473K and 673K, the hardness nearly keeps a constant value. However, after the 773K annealing, the hardness rapidly decreases and recovers to the value before the irradiation. This rapid decrease in hardness at 773K is well correlated with the transformation from the amorphous state to the intrinsic orthorhombic structure at the same temperature (see Fig. 3). By contrast to the large decrease in hardness at 773K, the decrease in hardness at 373K is quite small, suggesting that this phenomenon is attributed to the recovery of irradiation-induced lattice defects with relatively simple configurations. Fig. 6 shows the change in the Vickers hardness as a function of annealing temperature for Ni_3Ta. After the annealing at 673K,

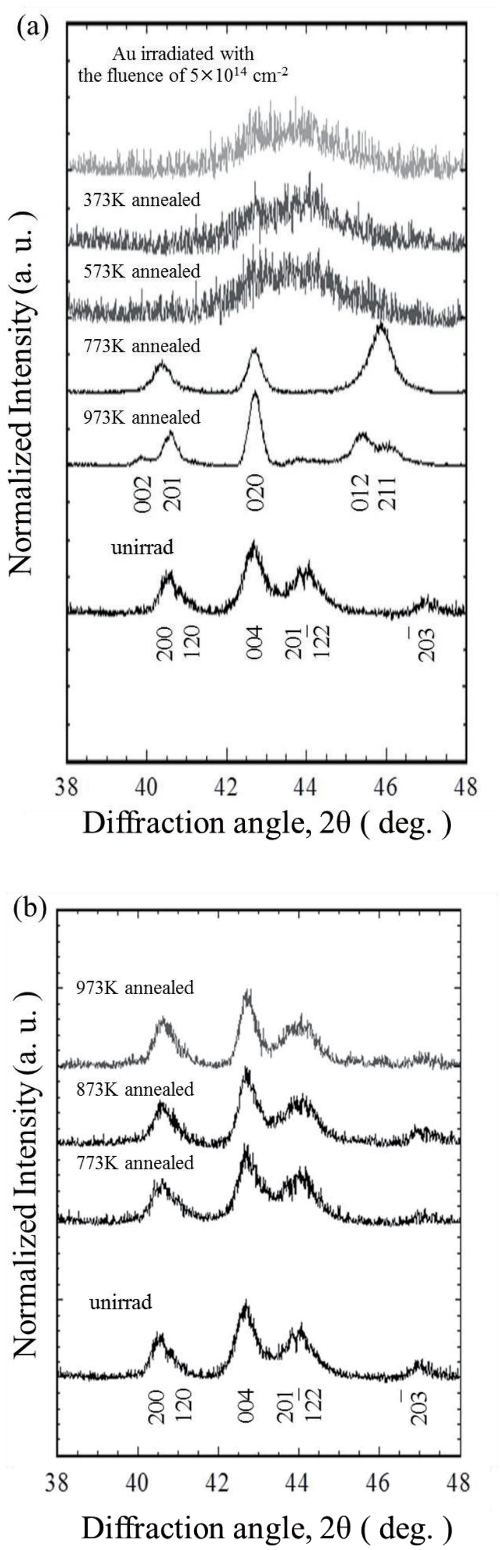

Fig. 4 GIXD spectra for irradiated surface (a) and rear surface (b) of Ni_3Ta which are thermally annealed at 373K, 573K, 773K, 873K and 973K after the irradiation. Irradiation fluence is $5\times10^{14}/cm^2$.

the hardness decreases largely. The temperature for the large decrease in hardness is coincident with the temperature for the appearance of crystal structure in the amorphous matrix. Similarly to the annealed Ni_3Nb sample, the hardness of the Ni_3Ta tends to recover to the value for the unirradiated sample during the thermal annealing. However, the complete recovery was not observed in the annealed Ni_3Ta within the annealing temperature range of the present experiment. When the annealing treatment was performed at even higher temperatures, the peaks corresponding to the intrinsic monoclinic structure would be observed, and the hardness would recover to the value before the irradiation.

We have previously reported the recovery of the irradiation-induced lattice structure change by the thermal annealing in Ni_3V and Ni_3Al intermetallic compounds [12, 13]. In the case of Ni_3V, the random fcc structure induced by the ion irradiation recovers to the intrinsic tetragonal structure ($D0_{22}$ structure) by the annealing at 773K. In the case of Ni_3Al, the disordered fcc structure recovers to the intrinsic ordered $L1_2$ structure by the annealing at 773K. In the present case of Ni_3Nb, although the thermal annealing induces the recovery of the lattice structure not from the disordered fcc structure, but from the amorphous state to the intrinsic lattice structure, the recovery of the lattice structure is also observed by 773K annealing. Then, we have found that irrespective of intrinsic lattice structure at room temperature or the irradiation-induced disordered state, the recovery of the irradiation-induced disordering around 700K is a common property for the three kinds of Ni-based intermetallic compounds. Ni_3Ta is, however, an exceptional case. By the annealing at 773K, The irradiation-induced amorphous structure changes to the orthorhombic structure, which is not the intrinsic monoclinic structure.

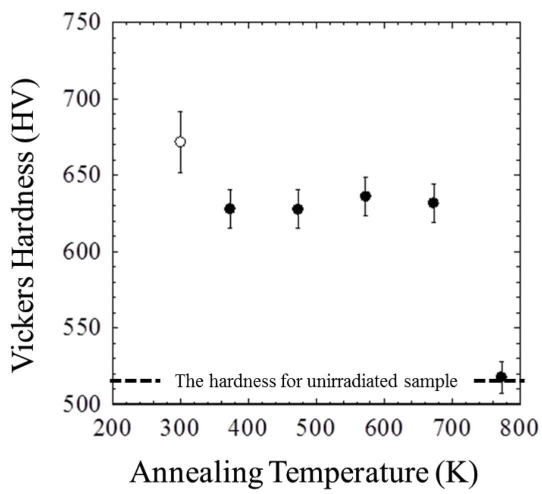

Fig. 5 Change in Vickers hardness as a function of annealing temperature for the Ni_3Nb sample. Open circle is the data for the sample before annealing.

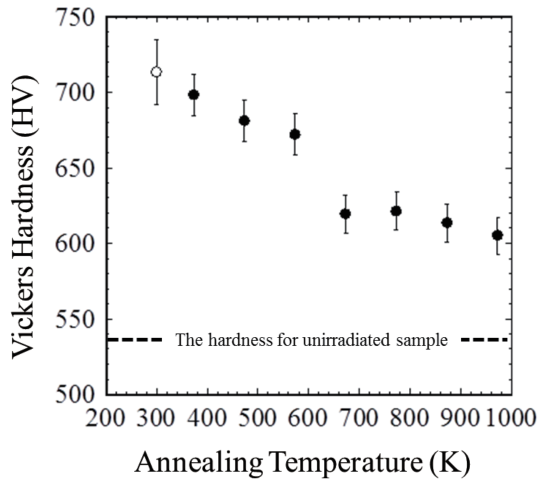

Fig. 6 Change in Vickers hardness as a function of annealing temperature for the Ni_3Ta sample. Open circle is the data for the sample before annealing.

5. SUMMARY

We have discussed the thermal stability of the irradiation-induced amorphous state and hardness changes of Ni_3Nb and Ni_3Ta through the thermal annealing experiment after the 16 MeV Au ion irradiation at room temperature and the irradiation at elevated temperatures. In the case of Ni_3Nb samples, they were almost amorphized under Au ion irradiation at room temperature and 423K, but by the irradiation at 523K, the crystal structure is still observed. The Vickers hardness increased with increasing ion fluence and with decreasing the irradiation temperature. After the subsequent annealing at elevated temperatures, the amorphous state induced by the irradiation recovers to the intrinsic orthorhombic structure in the Ni_3Nb sample. On the other hand, in the Ni_3Ta sample, the amorphous state induced by the irradiation changed to the orthorhombic structure, and did not recover to the intrinsic monoclinic structure. The present study indicates for the first time that by using energetic ion irradiation and subsequent annealing, a novel lattice structure, which does not exists in the thermal-equilibrium phase diagram, can be produced in Ni_3Ta intermetallic compound.

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