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Excess Vacancies Induced by Disorder-Order Phase Transformation in Ni_3Fe

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The order-disorder transformation and lattice defects in Ni_3Fe have been studied by positron lifetime measurements. Anomalous vacancy-generation during ordering transformation, which was originally found on the ordering process of super-cooled disordered Cu_3Au, has been confirmed on the ordering transformation of Ni_3Fe. Disordered fcc solid solution of Ni_3Fe was brought to room temperature by quenching the specimen from temperatures above the order-disorder transformation point T_C. The ordering process into $L1_2$ structure was promoted by heating the sample isochronally or isothermally. It has been found that vacancies are generated in both heating processes, *i.e.*, during the ordering process of super-cooled disordered Ni_3Fe. Generated vacancies are not stable up to T_C and annealed out at temperatures below T_C.

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1. Introduction

The order-disorder transformation and vacancies in Cu_3Au were studied by positron lifetime measurement.[1)] It was found that vacancy-like defects are anomalously generated during the ordering process of super-cooled disordered Cu_3Au.[1)] The defects generated are most likely to be vacancies, although such formation of vacancies is hardly explained by the known mechanisms of vacancy formation. To confirm the anomalous vacancy-generation phenomenon and to investigate the origin and the mechanism, the ordering transformation and vacancies in Ni_3Fe have been studied by positron lifetime spectroscopy that is the most sensitive method to observe the vacancy-type lattice defects.

Below the transition temperature T_C, 780 K, Ni_3Fe has $L1_2$ ordered structure and above T_C it has A1 disordered fcc structure, which is the same with the case of Cu_3Au. In this paper, the results of positron annihilation study for the ordering processes of super-cooled disordered Ni_3Fe are reported and the origin and the mechanism of the vacancy-generation phenomenon are discussed.

2. Experimental Procedure

The Ni_3Fe sample ingot was prepared by melting 99.97% pure Ni and 99.96% pure Fe in pure argon atmosphere using an arc melting furnace. The sample ingot was given a homogenization treatment at 1323 K for 172.8 ks. The sample was cut into specimens suitable for positron lifetime measurements having a square surface of about $10 \times 10\,mm^2$ and 1.5 mm thickness. The specimens were quenched into water at 273 K after annealing at 1473 K for 1.8 ks in order to obtain super-cooled disordered Ni_3Fe. Then positron lifetime measurements were carried out during ordering processes promoted by isochronal annealing in steps of 1.8 ks/30 K or isothermal annealings at 623, 648, 673 and 698 K.

Positron lifetimes were measured at room temperature using a positron lifetime spectrometer with a time resolution (FWHM) of 175 ps. A 50 μCi positron source of ^{22}NaCl sealed with Kapton foils ($10\,g/m^2$) was sandwiched between two identical Ni_3Fe specimens. Each positron lifetime spectrum consists of more than 10^6 events and several spectra were accumulated for each measuring point in order to ensure the reproducibility of the data. Measured lifetime spectra were analyzed using the POSITRONFIT program, a part of the PATFIT-88[2)] software.

3. Results and Discussion

Before promoting the ordering process, the positron lifetime of as-quenched disordered Ni_3Fe specimens were measured. All of such specimens showed a lifetime value of 101 ± 1 ps, which is the same with the value for disordered Ni_3Fe standard specimen (quenched form 873 K). This clearly shows that there were practically no quenched-in vacancies in the as-quenched disordered specimens.

3.1 Ordering process on isochronal annealing

Figure 1 shows the change of mean positron lifetime τ_m in quenched Ni_3Fe on the isochronal ordering process. A considerable increase of τ_m is observed above 500 K. Above 600 K the τ_m begins to decrease with temperature and finally comes to the Ni_3Fe bulk value at temperatures below the order-disorder transformation point T_C, 780 K. Observed increase in τ_m strongly suggests the appearance of some defect during the ordering. Using the two-component analysis, the positron lifetime in the defect τ_d and the relative intensity I_d have been decomposed as shown in Fig. 2. The τ_d, about 155 ps at 530 K, increases with annealing temperature and reaches a maximum value of about 240 ps at 680 K, which is comparable with that for small vacancy clusters.

This result shows that during the ordering transformation of super-cooled disordered Ni_3Fe, anomalous generation of vacancy-like defects has occurred. Similar phenomenon was originally found on the ordering process of Cu_3Au.[1)]

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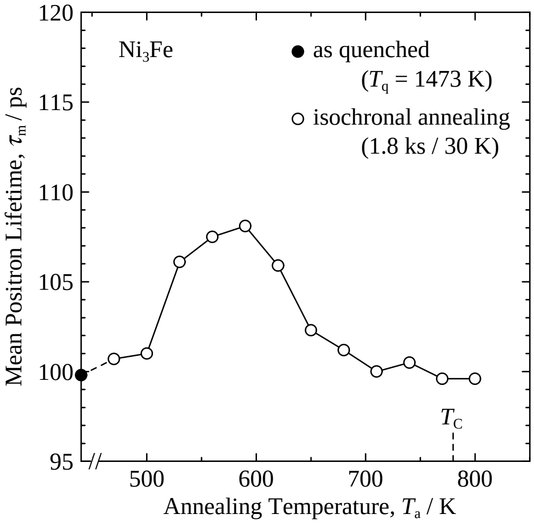

Fig. 1 Change of mean positron lifetime in quenched Ni_3Fe on isochronal annealing by 1.8 ks/30 K step.

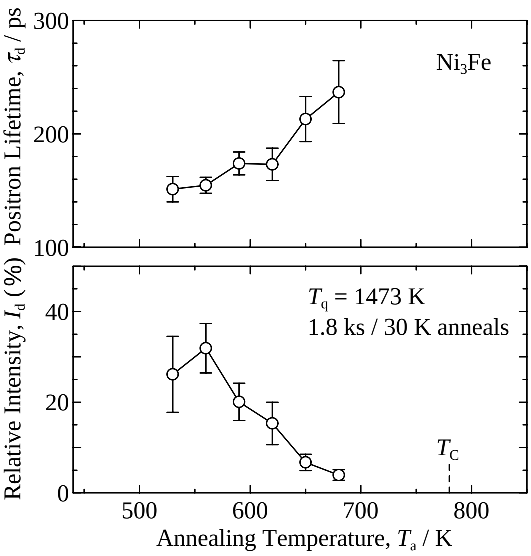

Fig. 2 Positron lifetime and relative intensity for the defect component appeared during isochronal annealing of quenched Ni_3Fe.

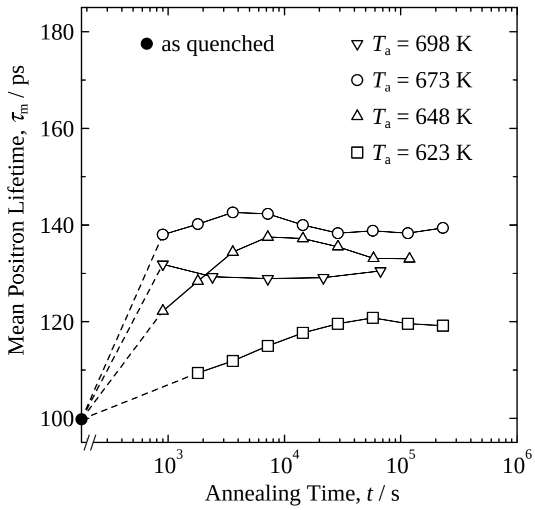

Fig. 3 Changes of mean positron lifetime in quenched Ni_3Fe on isothermal annealings at 623, 648, 673 and 698 K.

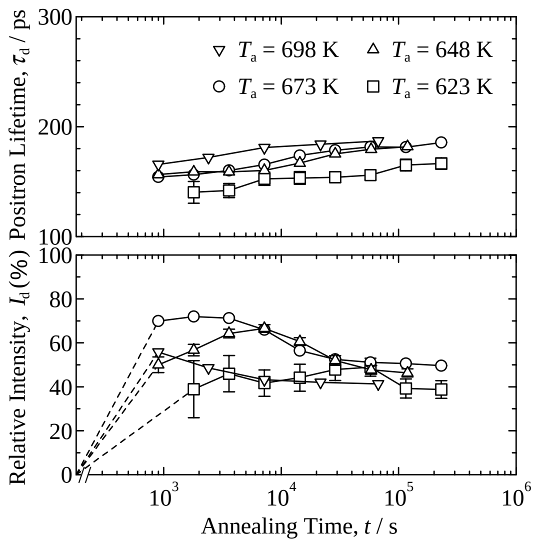

Fig. 4 Changes of positron lifetime and relative intensity for the defect component appeared during isothermal annealings of quenched Ni_3Fe.

3.2 Ordering process on isothermal annealing

In order to confirm the anomalous phenomenon, the ordering was promoted by isothermal annealings. Figure 3 shows changes of mean positron lifetime τ_m in quenched Ni_3Fe on isothermal ordering processes at 623, 648, 673 and 698 K. Remarkable increases of τ_m are observed in all ordering processes. During the ordering at 673 K, τ_m increases dramatically to the peak value of 143 ps after annealing for 3.6 ks. On the ordering at 648, 623 K, τ_m increases to the peak of 138, 121 ps after 7.2, 57.6 ks annealing, respectively. During the ordering at 698 K, the maximum of τ_m seems to appear with a shorter annealing time than 0.9 ks. The increase of τ_m found on the isochronal ordering manifests more obviously on the isothermal ordering processes.

The measured lifetime spectra were analyzed into two components to obtain the positron lifetime in the defect τ_d and the relative intensity I_d. The results are shown in Fig. 4. On the ordering at 648 K, about 160 ps of τ_d appears at the beginning and its intensity I_d increases with annealing time to the peak at 7.2 ks annealing. For longer annealing, τ_d increases gradually. Correspondingly, I_d decreases with annealing time but remains around 50% even after 115 ks annealing. On the ordering at other temperatures, the changes of τ_d are almost similar to that at 648 K. At higher annealing temperatures (at 673 and 698 K) the peak positions of I_d seems to be shifted to shorter annealing times and at lower annealing temperature (at 623 K) vice versa.

3.3 Vacancy generation during ordering

In both ordering processes, isochronal and isothermal, the anomalous increase of positron lifetime is observed. The positron lifetime in the defect τ_d is initially about 155–160 ps, which is similar to that for single vacancy in Ni (160 ps)[3] and Fe (175 ps).[4] During the following ordering process, the increase of τ_d are accompanied with the decrease of I_d, which is the characteristic positron lifetime change for the clustering of excess vacancies. The defects generated anomalously

during the ordering process are most likely to be vacancies.

The origin of the anomalous generation of vacancies during ordering transformation is considered to be due to the difference in lattice constant between ordered and disordered phases. In both Ni_3Fe and Cu_3Au, the transformation from the disordered state to the ordered phase involves the volume contraction, by 2.2% in Ni_3Fe and 9.9% in Cu_3Au calculated with the lattice constant.[5,6] It is well known that the ordering transformation in the alloys proceeds along the homogeneous nucleation-growth type process of the diffusional transformation. During the ordering, the ordered phase precipitates homogeneously in the disordered matrix by the nucleation-growth mechanism and this process brings about the volume contraction locally around the precipitated ordered phase. The volume contraction produces high strain energy around the interface of ordered and disordered phases. This strain energy may cause vacancy generation to accommodate the volume contraction.

It is interesting to notice that the result of isochronal annealing at 680 K for 1.8 ks suggests the existence of vacancy clusters, *i.e.*, the positron lifetime is about 240 ps (Fig. 2), but such a long lifetime is not observed on the isothermal annealing at 698 K even after a longer annealing time than 1.8 ks. This is probably caused by the difference in the microstructure formed on the isochronal and isothermal heat treatments. In the ordering process during the isochronal annealing starting from relatively low temperatures, a lot of small ordered domains are formed with vacancies. Such small ordered domains become unstable at higher temperatures and subjected to the domain coarsening. Excess vacancies should migrate during the domain growth and form vacancy clusters eventually. In contrast to that, relatively bigger domains are formed during the isothermal annealing from the very beginning. In this case, generated vacancies are trapped in the strain around ordered domains without forming vacancy clusters.

In the study[7] of the mechanical property and ordering of Ni_3Fe, a low activation energy of ordering transformation was deduced by Takasugi *et al.*,[7] and they suspected the existence of excess vacancies. In this study, it has been confirmed by positron lifetime method that anomalous excess vacancies are generated during the ordering transformation.

4. Conclusion

The order-disorder transformation and lattice defects in Ni_3Fe have been studied by positron lifetime measurements. Anomalous vacancy-generation during the ordering transformation of super-cooled disordered Ni_3Fe has been found. The vacancies are possibly generated to reduce the strain energy at the interface of ordered and disordered phases by accommodating the lattice contraction due to the difference in lattice constant between ordered and disordered Ni_3Fe.

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