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Diffuse X-ray scattering studies of radiation defects in Ni and dilute Ni alloys

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Abstract

Pure Ni, dilute Ni–Cu and Ni–Fe alloys were irradiated with 2 MeV electrons at about 90 K. The resulting defect structures at room temperature were studied by X-ray scattering methods using synchrotron radiation. In Ni and dilute Ni alloys, major defects were vacancy loops (averaged size 5–7 Å). A larger number of vacancy loops was found to exist in the alloys than in pure Ni, and this is interpreted in terms of solute atoms causing an increase in the probability of loop nucleation.

1. Introduction

The X-ray scattering method with radiation of a wave length comparable to the atomic distances (~1 Å) is an appropriate technique to investigate microstructures of radiation defects in crystalline solids. Radiation defects induce diffuse X-ray scattering (DXS) around the Bragg peaks. The distribution of this diffuse scattering contains very detailed information on the radiation defects. The DXS method has the enough sensitivity to observe the intensity of typically 10–100 ppm of atomic size defects. The determination of the structural properties of such small agglomerates (defect types and size distribution of each defect) has, therefore, become a unique field of application for the DXS technique [1].

It is known that defect structure affecting material properties depends on irradiation conditions. The formation of relatively simple defect structures in pure metals and dilute alloys has been investigated using electron irradiation [2–4]. Although nickel is of great importance because of its use in alloys for nuclear reactors, its detailed defect structure has not been so well understood as those of other FCC metals. In an attempt to obtain a better understanding of the defect behavior in Ni-base alloys, a detailed investigation of the defect structure in pure Ni, dilute Ni–Cu and Ni–Fe alloys was performed in this study.

2. Theory

The intensity of DXS from dislocation loops is given by the following equation

$$I_{\text{diffuse}} = N_{\text{D}}(r_{\text{e}}f_{\text{a}})^2|A(K)|^2, \tag{1}$$

where N_{D} is the total number of dislocation loops, r_{e} the classical electron radius, f_{a} the atomic scattering factor, K the scattering vector and $A(K)$ the scattering amplitude associated with a dislocation loop. In this expression, $A(K)$ may be written as [5,6]

$$|A(K)|^2 = \left|\sum_m e^{iKr_m}(e^{iKs_m} - 1) \pm \sum_n e^{iK(r_n+s_n)}\right|^2. \tag{2}$$

The first term in Eq. (2) describes the sum of diffuse scattering amplitudes (total amplitude minus the Bragg reflection amplitude) from the m-th lattice atom displaced from its ideal position r_m by s_m due to the presence of the dislocation loops (distortion scattering). The second term represents the sum of the scattering amplitudes from the n-th additional or missing atom, which

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defines a lattice defect (Laue scattering). The scattering vector K is given by the sum of the reciprocal lattice vector h and the vector q relative to h ($K = h + q$).

3. Numerical calculations

For the calculation of the terms in Eq. (2) the displacement field of lattice atoms around a loop has to be evaluated numerically. For the dislocation loop on a {1 1 1} plane having Burgers vector a/3 ⟨1 1 1⟩ (Frank loop), which is predominant in irradiated FCC metals as observed by TEM, the calculation of displacement field was carried out using a formula reported by Ohr [7] and corrected later by Rao [8]. Using the displacement data, we calculated the scattering amplitudes corresponding to Eq. (2) for q parallel to the ⟨1 1 1⟩ reciprocal lattice vector. Thus the intensity calculated for q-dependence of scattering amplitude along the ⟨1 1 1⟩ reciprocal lattice vector is shown in Fig. 1, where the diffuse scattering intensities for both a single vacancy loop and a single interstitial loop with a radius $R = 10$ Å are employed. It was confirmed in the calculation for the different loop radius ($R = 5$–30 Å) that the peak position of scattering intensity depends on each dislocation radius. The difference of the peak position was used for the evaluation of the size distribution of defect clusters.

4. Experimental

4.1. Samples and irradiation condition

High purity single-crystal samples of pure Ni and Ni alloys containing 0.04 at.% Cu and 0.05 at.% Fe were spark-cut from large single crystals grown by the Czochralski method. The sample surface parallel to a {1 1 1} plane was used for the DXS measurements. After removing the surface layer by electropolishing, the samples were annealed in vacuum at 900 K for 5 h. The thickness of all samples was about 1 mm. The irradiation with 2 MeV electrons was performed at JAERI Takasaki up to a dose of 1×10^{18} e^-/cm^2 at about 90 K.

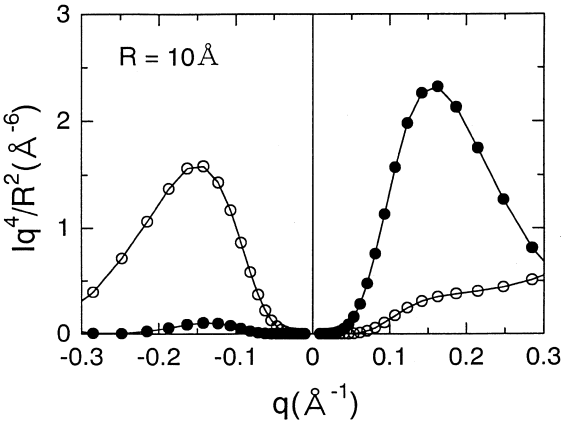

Fig. 1. Intensity of DXS scaled by q^4/R^2 calculated by Eq. (2) for vacancy (white symbols) and interstitial (black symbols) loop in Ni.

4.2. X-ray measurements

The DXS experiments on electron-irradiated samples were performed at room temperature with a four-circle diffractometer installed at BL-27B at the Photon Factory of Institute of Materials Structure Science, High Energy Accelerator Research Organization in Tsukuba. Diffuse scattering intensities were always measured near the (1 1 1) reflection in the [1 1 1] direction. The diffuse scattering intensity was obtained by subtracting the background scattering intensity obtained on the samples after complete annealing of the defects at 900 K for 1 h. The background scattering intensity after annealing was found to be almost identical to that before irradiation. The background scattering from the temperature diffuse scattering and the Compton scattering was also removed by taking a difference between as irradiated and annealed samples.

5. Results and discussion

The intensity of DXS close to the (1 1 1) Bragg reflection in the [1 1 1] direction measured for the electron irradiated Ni-0.04 at.% Cu alloy at room temperature is shown in Fig. 2 as a typical example where the intensity is the normalized value compared to that of the incident X-ray beam. Fig. 3 shows the q-dependence of diffuse scattering intensities for the pure Ni, the Ni–Cu and Ni–Fe alloys. Both q^{-2} and q^{-4} dependencies are seen indicating the presence of single vacancies and that of dislocation loops, respectively. From this figure one can also roughly estimate the average size of dislocation loop radius R_0 (5–7 Å for all alloys) according to the relation $q_0\ R_0 = 1$, where q_0 is the value at which the diffuse scattering intensity deviates from the q^{-4}-dependence [9]. Fig. 4 shows the diffuse scattering intensity multiplied by q^4 against q for the pure Ni, Ni–Cu and Ni–Fe dilute alloys. Detailed information on the defect structures is obtained by fitting the size distribution of different dislocation loops to these experimental scattering curves using the number of dislocation loops as a fitting parameter, since the contribution from single vacancies is thought to be negligibly small in the main fitting range of $q < -0.1$ Å^{-1}. For this data-fitting, loops of radii 5, 10, 20, 30 Å

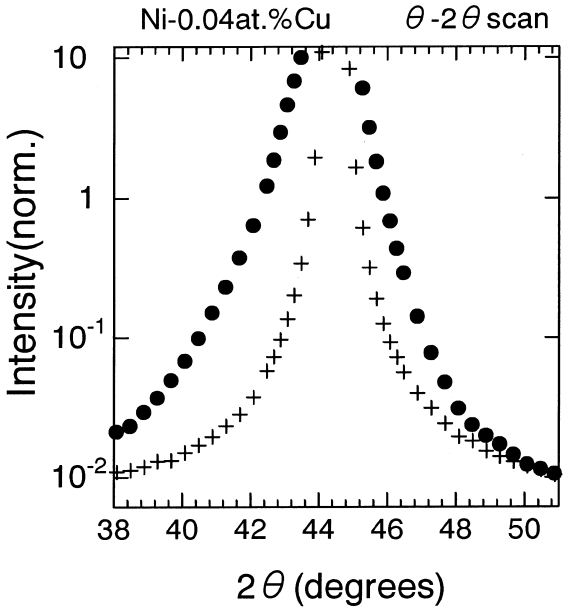

Fig. 2. Intensity of X-ray scattering from electron-irradiated Ni-0.04 at.% Cu alloy close to the (1 1 1) reflection in the [1 1 1] direction measured at room temperature. ●: After irradiation, +: after annealing.

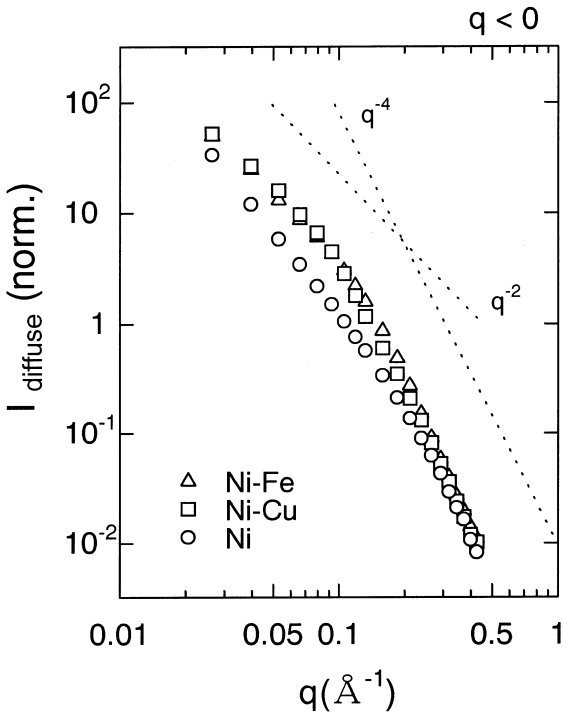

Fig. 3. The q-dependence of diffuse scattering intensity in pure Ni and dilute Ni alloys ($q < 0$). ○: Pure Ni, □: Ni–Cu, △: Ni–Fe.

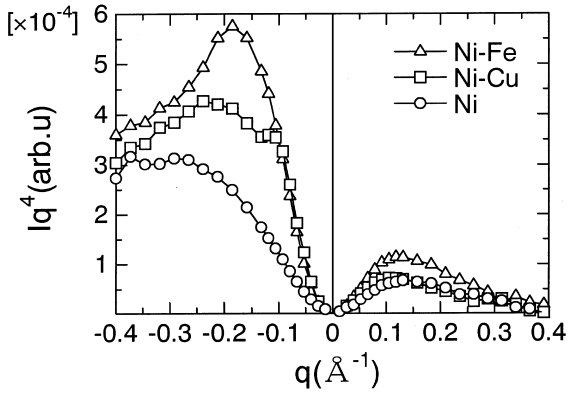

Fig. 4. Intensities of DXS scaled by q^4 obtained experimentally from electron-irradiated pure Ni and dilute Ni alloys. ○: Pure Ni, □: Ni–Cu, △: Ni–Fe.

that are near the visibility limit of TEM were used. The result on the ratio of the number of vacancy or interstitial loops to that of the total loops expressed in percentage for both interstitial and vacancy loops with size R is summarized in Table 1. The size distribution of loops obtained theoretically by fitting is in good agreement with the rough estimation of average loop radii from the q-dependence ($R_0 \sim 6$ Å). The proportion of vacancy loops obtained from the theoretical fitting in this study is higher than that of interstitial loops. This result is different from that were reported by other researchers [2–4]. The theoretical calculation was carried out by Eq. (2) under the assumption that both vacancies and interstitials mainly form dislocation loops and there is little contribution from the stacking-fault tetrahedra (SFT) as reported previously [2–4]. The theoretical calculation by using Eq. (2) assuming that vacancies form SFT is difficult because there are no detailed calculations about the strain field around SFT. Moreover the strain field around SFT and thus the scattering intensity were reported to be not so different from those of vacancy loops in the q^{-4} region by Ehrhart and Averback [4]. The fact that major defects were vacancy loops in Table 1, therefore, seems to result from the experimental condition, especially irradiation temperature, i.e. the difference in mobility between interstitials and vacancies under the experimental condition. In this study the electron irradiation was carried out at 90 K followed by warming up to room temperature. Other researchers [2–4], on the other hand, investigated mainly the motion of interstitials in Ni alloys using the irradiation at low temperature (5 K), where the mobility of interstitials is very small, and subsequent isochronal annealing. The effect of the irradiation temperature seems mainly to have an effect on the resulting defect type. The estimation of point-defect type under similar irradiation conditions to ours has been

Table 1
The ratio of the number of loops to that of the total loops as a function of loop radius

Type	Loop radius (R) (Å)	Ni (%)	Ni–Cu (%)	Ni–Fe (%)
Vacancy	5	91.2	80.8	73.7
	10	8.1	18.3	25.5
	20	0.0	0.0	0.0
Interstitial	5	0.0	0.0	0.0
	10	0.0	0.0	0.0
	20	0.7	0.9	0.8

done by Kiritani and Tanaka [10]. They reported that major defect was vacancy at low temperature due to the small mobility of vacancies and that the interstitial atoms were lost at sinks such as surface or interstitial-loops. The higher diffuse scattering intensities from vacancies in dilute Ni alloys than that from pure Ni are probably due to the fact that the over-sized solute atoms, for example Fe and Cu in Ni, stabilize the nucleus of vacancy loops, as has been reported for interstitial loops [11]. In the case of interstitials, the higher intensities from dilute alloys than from pure Ni also seem to originate from the stronger binding between solute atoms and interstitials.

6. Conclusions

The defect structures in Ni alloys irradiated at about 90 K with 2 MeV electrons were investigated by DXS methods at room temperature. The defect structure was mainly composed of vacancy loops in all alloys. The effect of Fe and Cu additions to Ni on dislocation loop formation during irradiation was investigated and the large number of loops observed were explained on the basis of solute atoms stabilizing the nucleation sites for the formation of both vacancy- and interstitial-loops.

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References

[1] P. Erhart, J. Nucl. Mater. 216 (1994) 170.
[2] R.S. Averback, P. Erhart, J. Phys. F: Met. Phys. 14 (1984) 1374.
[3] P. Erhart, R.S. Averback, J. Phys. F: Met Phys. 14 (1984) 1365.
[4] P. Erhart, R.S. Averback, Philos. Mag. A 60 (1989) 283.
[5] B.C. Larson, Schmatz, Phys. Stat. Sol (b) 99 (1980) 267.
[6] P. Erhart, H. Trinkaus, B.C. Larson, Phys. Rev. B 25 (1982) 834.
[7] S.M. Ohr, Phys. Status Solidi B 64 (1974) 317.
[8] S.I. Rao, C.R. Houska, K. Grabowski, G. Ice, J. Sparks, J. Appl. Phys. 69 (1991) 8104.
[9] P. Erhart, U. Schlagheck, J. Phys. F: Met Phys. 4 (1974) 1589.
[10] M. Kiritani, H. Tanaka, J. Nucl. Mater. 69/70 (1978) 277.
[11] S. Kojima, T. Yoshiie, M. Kiritani, J. Nucl. Mater. 155–157 (1988) 1249.