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Atom transport efficiency in heavy ion irradiated metals

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Abstract

Multi-layer specimens were used to investigate diffusion near the irradiated surface in nickel single crystals under heavy ion irradiation at temperatures around the swelling rate maximum 800–950 K. Analysis of the data yields information on the effective fraction η of the displacement rate, with which transporting defects are produced (production rate of freely migrating defects), and on the radiation-induced sink strength. Upper limits of $\eta = 7\%$ are calculated for a self-ion irradiation at 950 K. In the investigated temperature range defect kinetics is dominated by the high sink strength of about 10^{15} m^{-2}. © 1999 Elsevier Science B.V. All rights reserved.

1. Introduction

The origin of microstructural changes observed in irradiated materials, e.g. void formation or phase transformations, is radiation-induced diffusion [1]. Two different mechanisms may be distinguished, which contribute to the radiation-induced diffusion coefficient: D_{mix}, resulting from the athermal relocation of atoms by the irradiating particles and dominating at lower temperatures, and D_{rad}, resulting from thermally activated migration of the radiation-induced point defects. The first mechanism is known as atomic mixing [2] and is rather efficient for cascade forming ion irradiation. The random displacements of atoms in the collision cascade suggest that this mixing effect is proportional to the displacement rate per atom, K (in units of displacements per atom per second, dpa/s). Typical values of the mixing efficiency D_{mix}/K of about 1 nm^2/dpa are experimentally observed for 300 keV Ni^+ ion irradiation of nickel [3].

At elevated temperatures the radiation-induced point defects contribute to transport according to their concentration and mobility. For example, the diffusion coefficient of the radiation-enhanced self-diffusion is given by $D_{\mathrm{rad}} = f_{\mathrm{v}} D_{\mathrm{v}} C_{\mathrm{v}} + f_{\mathrm{i}} D_{\mathrm{i}} C_{\mathrm{i}} +$ (additional terms) [1]. Here f is the correlation factor, D the diffusion coefficient, and C the concentration of transporting defects. The subscripts v and i indicate single vacancies and interstitial atoms, respectively, and the terms within the brackets allow for further (generally small) contributions arising from mobile defect clusters [1,4]. The magnitude of the radiation-enhanced diffusion under steady state irradiation conditions is determined by the dynamical equilibrium between the production and annihilation rates of the point defects [1].

For realistic damage modelling the knowledge of the production and annihilation rates as well as the mixing efficiency is required in order to allow valid predictions of transport effects for arbitrary conditions including the technically important case of neutron irradiation. While the determination of the mixing efficiency and its interpretation is straightforward [2], quantitative estimates of the defect production and annihilation rates are complicated. This proves to be true in particular for irradiation with heavy ions and neutrons, for which it is known that the effective sink strength is mostly radiation-induced and the effective production rate of mobile defects is only a small fraction of the displacement rate, and that no simple relation between these quantities and the corresponding irradiation parameters exists [4,5].

In this paper spatially resolved measurements of the self-diffusion coefficients at temperatures around the swelling rate maximum of ion irradiated nickel are presented and the radiation-enhanced diffusion near the irradiated surface is analyzed. The effect of the surface sink on the spatial variations of the mobile defect concentrations [1] and the limited range of the ion-induced damage together with the known defect properties [6]

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will be used to evaluate both, the effective production rate of transporting defects and the effective sink strength. Moreover, values of the mixing diffusion coefficient, D_{mix}, obtained at low temperatures will be reported.

2. Experimental details

Nickel multi-layer specimens were grown under UHV conditions at room temperature on well polished Ni single crystals with a diameter of 12 mm cut about 5° off the (1 1 2) zone axis. They contained four layers of about 1 nm thickness of the ^{63}Ni tracer about 100 nm apart starting with the first layer about 100 nm below the irradiated surface. For some experiments single crystal specimens with only one tracer layer in a depth of about 75 nm below the irradiated surface were produced. Details of the preparation and characterization of the specimens can be found elsewhere [7].

Only part of the specimens' surface was irradiated with 600 keV Kr^{++} or 300 keV Ne^{+}, Ar^{+} and Ni^{+} ions through an appropriate aperture of about 0.2 cm^2. By this procedure the irradiated and non-irradiated state can be analyzed in one and the same specimen such that the irradiation effect can be evaluated with high accuracy by direct comparison [3].

The depth dependent displacement cross section $\sigma_d(x)$ of the different irradiating ions in nickel was calculated by means of the binary collision code TRIM [8] with a threshold energy of 40 eV [9]. Values of $\sigma_{d,max}$, the displacement cross section in the damage maximum, are compiled in Table 1 together with the characteristic depth L of the damaged region, $L = \int dx \sigma_d(x)/\sigma_{d,max}$. The depth dependent displacement rate $K(x)$ in dpa/s is obtained from the ion flux density φ in the usual way as $K(x) = \varphi\sigma_d(x)$. Experiments were performed with ion flux densities φ between 1.4×10^{16} m^{-2}s^{-1} and 3×10^{17} m^{-2}s^{-1} and irradiation times between 250 and 2000 s.

Atom transport was measured from the broadening of the thin tracer layers by sectioning using sputter erosion in combination with secondary ion mass spectrometry (SIMS). Details of this measuring technique and of the determination of the diffusion coefficients are described elsewhere [3]. The error bar of the reported diffusion coefficients is about ±30%.

3. Results and discussion

Atom transport under ion irradiation at low temperatures is dominated by atomic mixing [2]. Hence it is expected that the spatial variations of the displacement rate, $K(x)$, which can be calculated, for instance, with a simulation program like TRIM [8], determine directly the local atomic mixing. The validity of this assumption has been verified by measuring the depth dependence of the diffusion coefficient under irradiation at 77 K [7,10]. The resulting mixing efficiencies, D_{mix}/K, for the ions used in this investigation are compiled in Table 1. They are independent of the depth below the irradiated surface in which D_{mix} was measured.

The radiation-enhanced diffusion is essentially caused by the radiation-enhanced concentration of mobile single interstitials and vacancies [4,5]. During homogeneous irradiation of an infinitely extended crystal these defect concentrations are determined by the dynamical equilibrium of defect production and annihilation. In the case of heavy ion irradiation they are also determined by diffusion due to defect gradients which arise because of the presence of the surface sink, where the defect concentrations are in thermal equilibrium, and because of depth dependent defect production rates and sink strengths. As the defects can migrate away from the location where they were produced it is in particular expected that considerable defect concentrations will build up in the crystal beyond the ion range, i.e. in parts which were not directly damaged by the irradiating ions.

It has been observed that for cascade forming irradiation at elevated temperatures usually different fractions of immobile vacancy and interstitial agglomerates are produced. This observation may imply different production rates of mobile vacancies and interstitials. Moreover, by this type of irradiation high sink strengths are produced. Consequently the sink dominated defect reaction regime can be presumed [4,7]. If effective values of the depth dependent defect production rate, $\eta K(x)$, and sink strength, $k^2(x)$, are used [10], then the steady

Table 1
Irradiation and transport parameters of nickel. The meaning of the different quantities is explained in the text

Ion	Irradiation		Transport (values in units of nm^2/dpa)		
	$\sigma_{d,max}$ (nm^2)	L (nm)	η/k^2_{max} ($T = 800$ K)	η/k^2_{max} ($T = 950$ K)	D_{mix}/K ($T = 77$ K)
300 keV Ne^{+}	0.041	240	3.2	14.1	0.42
300 keV Ar^{+}	0.11	142	3.4	15.6	0.56
600 keV Kr^{++}	0.26	139	4.5	25.0	0.7
300 keV Ni^{+}	0.20	95	26.1 [a]	285.0	0.49

[a] value obtained at 890 K.

state self-diffusion coefficient, $D_{rad}(x)$, is determined by the differential equation

$$\nabla^2 D_{rad}(x) + \eta K(x) - k^2(x) D_{rad}(x) = 0, \quad (1)$$

with the boundary condition of $D_{rad} = 0$ at the irradiated surface ($x = 0$) and at very large distance from it in the region of negligible defect production. The effective η comprises the contributions of the probably different fractional production rates for vacancies and interstitials.

A fit of a solution of Eq. (1) to the experimental data yields three parameters: the effective fraction of freely migrating defects, η, the maximum sink strength, k^2_{max}, near the irradiated surface and the sink strength k^2_0 at very large distance from the surface in the non-damaged region beyond the ion range [7,10].

Fig. 1 presents data of the depth dependent self-diffusion coefficient D_{rad} under irradiation with different ions. The solid lines are fits of a solution of Eq. (1) to these data. Details of the fitting procedure are given elsewhere [7]. As has been discussed in Ref. [7] the depth dependence of the sink strength for the self ion irradiations at 890 and 950 K and for the noble gas irradiations at 800 K was assumed to be proportional to the displacement cross section, while that of the noble gas irradiations at 950 K was assumed to be proportional to the implantation profile. In Fig. 1 exponential slopes of D_{rad} are visible beyond the damage length L (cf. Table. 1) in greater depth. The diffusion in this region arises from defects, which have migrated down their concentration gradient into the non-damaged part of the crystal. From these slopes the sink strengths k^2_0 are obtained, which are between 0.5×10^{14} m^{-2} and 3.5×10^{14} m^{-2} at the irradiation temperature of 800 K, and between 0.2×10^{14} m^{-2} and 1.5×10^{14} m^{-2} at the higher temperature of 950 K [7]. These sink strengths decrease slightly with increasing temperature but they are still higher than those expected for a well annealed single crystal. It may be speculated whether the long range, one-dimensional glide of small interstitial loops [11] out of the irradiated region and accumulation of these in the non-irradiated region is responsible for the observed magnitude of the sink strength k^2_0.

Fig. 2 presents the effective production rates of transporting defects, η, and the effective sink strengths, k^2_{max}, deduced from the fits shown in Fig. 1. Obviously,

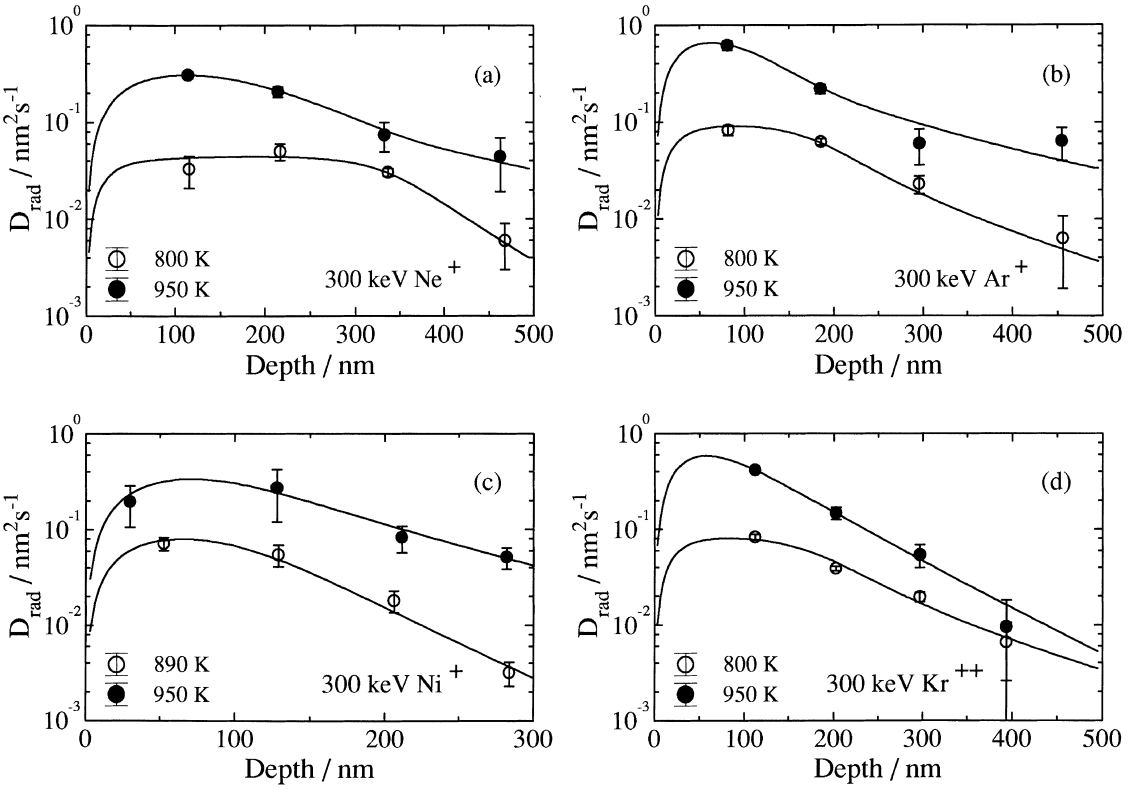

Fig. 1. Semilogarithmic plot of the depth dependence of the radiation-enhanced diffusion coefficient during irradiation with different ions at two temperatures. (a) 300 keV Ne^+, 3.0×10^{17} m^{-2}s^{-1}, 800 K: 435 s, 950 K: 250 s; (b) 300 keV Ar^+, 2.7×10^{17} m^{-2}s^{-1}, 800 K: 520 s, 950 K: 280 s; (c) 300 keV Ni^+, 890 K: 1.9×10^{16} m^{-2}s^{-1}, 2000 s; 950 K: 1.4×10^{16} m^{-2}s^{-1}, 500 s; (d) 300 keV Kr^{++}, 6.8×10^{16} m^{-2}s^{-1}, 800 K: 270 s, 950 K: 280 s. The solid lines are fits of a solution of Eq. (1) of the text.

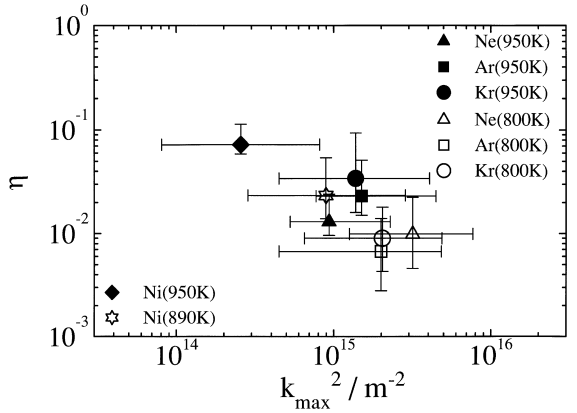

Fig. 2. Effective fraction of transporting defects, η, vs. the maximum sink strength, k^2_{max}, for the irradiation conditions indicated in Fig. 1.

the effective production rates cluster around about 1–2%. This low value is in general accordance with earlier results [3,12] and reflects not only the strong spontaneous recombination in the core of the displacement cascade but also the high degree of defect clustering for this type of irradiation. For the self-ion irradiation performed at 950 K a somewhat higher effective production rate of about 7% was observed. In a re-evaluation of earlier diffusion experiments performed at the same temperature [3] this high value is also obtained when the effect of the surface sink is taken properly into account.

The sink strengths k^2_{max} in the irradiated region are of comparable magnitude for the different irradiation conditions. They amount to about 2×10^{15} m^{-2}, except for the self-ion irradiation at 950 K, which yielded about 3×10^{14} m^{-2}. Fig. 2 moreover shows the tendency of a decreasing sink strength with increasing temperature. This is quite similar to the temperature dependence of the sink strength k_0^2 in the non-damaged region. For the self-ion irradiations this temperature dependence is particularly large, while the effect under noble gas irradiation is weaker, probably because noble gas bubbles are relatively stable. A decreasing sink strength with increasing temperature was observed already earlier and was interpreted as a thermal instability of the microstructure [13]. The correlation observed in Fig. 2 between η and k^2_{max} may be explained along the same lines by the thermal instability of small vacancy clusters at higher temperatures, which not only should lower the effective sink strength at these temperatures but also would yield an enhanced production rate of vacancies. This would result also in defect production rates effectively different for vacancies and interstitials, as was already mentioned above.

High sink strengths k^2 induced by heavy ion irradiation are well known. These high sink strengths result in a short diffusion length l for the defects of the order of $l = 1/k$ before they annihilate [7]. In the present investigation $k^2_{max} \approx 2 \times 10^{15}$ m^2 was observed and consequently the diffusion length is only about $l \approx 25$ nm. This length can be compared with L, the characteristic depth of the damaged region (cf. Table 1). The relation $L > l$ indicates that defects which are produced in the damage maximum near the depth L are unable to reach the surface, because they are annihilated at sinks before. On the other hand this means also that the effect of the surface sink on the defect concentrations is limited to a thin surface layer with a width of about l, in the present experiments of about 25 nm only. Therefore, for the present irradiation conditions the influence of the surface sink can be regarded as negligibly small near the location of the damage maximum and consequently the diffusion coefficient in this depth should be determined by the homogeneous defect kinetics [7].

Fig. 3 presents the displacement rate dependence of the radiation-enhanced diffusion coefficients as measured under noble gas irradiation at 800 K for displacement rates between 7×10^{-4} dpa/s and 6×10^{-2} dpa/s in a depth of about 75 nm below the irradiated surface. The fluences used in these experiments were between about 3 and 6 dpa. According to the above considerations, the effect of the surface sink can be neglected and the data are representative of a homogeneous irradiation. The linear relation of $D_{rad}/K = \eta/k^2 \approx (5.5 \pm 2)$ nm^2/dpa clearly indicates the sink dominated defect reaction regime [1] with a sink strength which is independent of the displacement rate. Individual values of η/k^2 derived for the different irradiation conditions from the fit procedure shown in Fig. 1 are summarized in Table 1. A comparison with the mixing efficiencies shows that ion mixing effects are of minor importance at the higher irradiation temperatures.

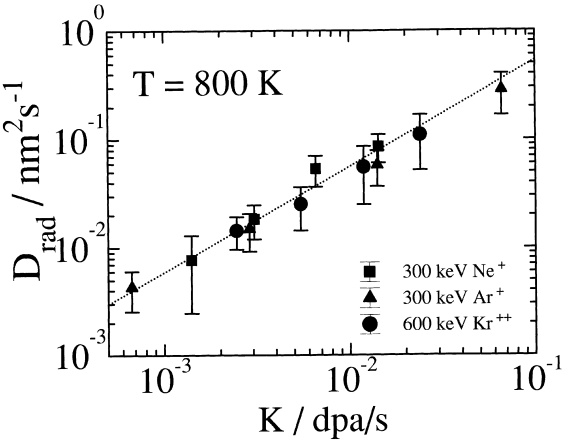

Fig. 3. Dependence of the radiation-enhanced diffusion coefficient on the displacement rate at a depth of about 75 nm for irradiation with noble gases.

The radiation-enhanced diffusion under noble gas irradiation at 950 K was about a factor of five more effective than at 800 K (cf. Fig. 1 and Table 1). Furthermore, it was also found to be determined by a sink dominated defect reaction regime. At this higher temperature, however, a weak displacement rate dependence of the sink strength yielded a slightly less than linear displacement rate dependence of the diffusion coefficient in accordance with recent observations on ion irradiated stainless steels [4].

The importance of the radiation-induced sink strength for the atom transport under ion irradiation at elevated temperatures has been demonstrated also by recent experiments of surface segregation performed on NiSi [14] and CuAu alloys [15]. In these experiments it was shown that the sink strength introduced near the irradiated surface by heavy ions like 400 keV Ne^+ is considerably higher than that produced by 1.5 MeV He^+. Moreover, the results indicated that the sink strength is higher *during* the irradiation than *after* an equivalent pre-irradiation. This supports earlier conclusions drawn from an order-of-magnitude discrepancy between sink strengths deduced from electron microscopic observations of the radiation-induced microstructure and from diffusion measurements [4]. It should be noted that the quantitative conclusions drawn with respect to the segregation kinetics [12] were obtained by assuming a recombination dominated defect reaction regime which is in disagreement to the present results. Further investigations of alloys are necessary in order to shed more light on the influence of the radiation-induced sinks on the segregation kinetics.

4. Summary

Irradiation of nickel with heavy ions at temperatures around the swelling rate maximum is characterized by a high radiation-induced sink strength and a low efficiency for the production of mobile point defects. The quantitative results obtained in the present investigation indicate that the radiation-enhanced diffusion is determined by a sink dominated defect kinetics. The reported effective defect production rates and sink strengths are slightly temperature dependent because of the thermal instability of the radiation-induced microstructure: while the sink strengths decrease with increasing temperature, the effective production rates increase. Both are not sensitive to the individual contributions of vacancies or interstitials, since the transport is determined by the sum of vacancy and interstitial fluxes which in the sink dominated defect regime do not interact.

The effect of the spatially inhomogeneous displacement rate on the radiation-enhanced diffusion coefficients was quantified. It was shown that considerable transport can be expected beyond the penetration depth of the irradiating ions. The influence of the surface sink in heavy ion irradiated specimens on the defect kinetics in the bulk is limited to a surface layer with a thickness of $1/\sqrt{k^2}$, i.e. the radiation-induced sink strength screens the bulk from the surface sink.

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