Radiation Effects and Defects in Solids, 1993, vol. 126, pp. 111-114
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PHENOMENOLOGICAL MODEL OF DAMAGE CREATION BY HIGH ELECTRONIC EXCITATIONS IN PURE METALS

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INTRODUCTION

It is now well established that high electronic energy deposition (HEED) may induce damage creation in some pure crystalline metals.
Damage creation is usually evidenced :
(i) by transmission electron microscopy (TEM) observations : striations which are parallel to the incident ion beam direction are observed in Ti irradiated with 4.4 GeV lead ions at 15 K [1]. These striations consist in dislocation loops [2]. HEED may also induce latent tracks in some metallic alloys [3, 4].
(ii) by and/or in situ electrical resistivity measurements: the comparison of the measured cross section (σ_{exp}) to that awaited from nuclear elastic collisions (σ_{elas}) shows that the ratio ($\xi = \sigma_{exp}/\sigma_{elas}$) can be much higher than 1 in some metals [1,5].

The evolution of the defect population or of the damaging mechanism with increasing ion fluence can be followed by the differential defect production curve (DDPC) ($\dot{\Delta\rho} = d\Delta\rho/d\Phi$ versus $\Delta\rho$, where Φ is the irradiation fluence and $\Delta\rho$ the electrical resistivity increase). The linear behaviour of the DDPC observed in iron irradiated with uranium ions was interpreted as the result of HEED which induces a competition between two processes : defect creation in a cylinder around the ion path and recombination of the pre-existing defects in an outer cylinder [6]. But the recent results of the damage induced by GeV lead ions in Ti, Co and Zr show a positive curvature of each DDPC [7]. In order to confirm this latter result, two low temperature (T = 20K) irradiations have been performed with 4.5 GeV xenon ions and 4 GeV uranium ions.

EXPERIMENTAL and RESULTS

The usual experimental procedure as described in ref [1, 6] was applied. The thicknesses of the samples were selected in order to remain in the region in which energy losses in electronic excitations ($(dE/dx)_e$ ~ a few keV/Å) are much larger than those in nuclear elastic collisions ($(dE/dx)_n$ ~ a few eV/Å). The total fluences obtained 1.8 x 10^{13} (Xe) and 5x10^{12} (U) ions/cm^2 are sufficient to estimate the saturation electrical resistivity increase for some metals : Ti, Zr and Co.
The DDPCs were drawn for each sample in fig 1. We can easily notice that they show two distinct parts:

(i) the initial part of the curve (i.e at low fluences) is approximately linear and can be described by a simple equation

$$\dot{\Delta\rho} = \dot{\Delta\rho}_0 - p_0\ \Delta\rho$$

where $\dot{\Delta\rho}_0$ is the initial damage production rate and p_0 has the dimension of a surface.

(ii) the final part (i.e at high fluences) is also approximately linear and can be described by the following equation

$$\dot{\Delta\rho} = -\ p_\infty\ (\ \Delta\rho - \Delta\rho_\infty\ )$$

where $\Delta\rho_\infty$ is the saturation electrical resistivity increase and p_∞ has the same dimension as p_0 .
These four parameters are deduced from the experimental curves (fig 1) and quoted in table.I.

Target	Ion	Energy (GeV)	$(dE/dx)_e$ keV/Å	$\dot{\Delta\rho}_0$ $10^{-13}\mu\Omega cm^3$	p_0 10^{-13} cm^2	p_∞ 10^{-13} cm^2	$\Delta\rho_\infty$ $\mu\Omega$ cm	$\Delta\rho^e_\infty$ $\mu\Omega$ cm
Ti	Xe	5.03	1.09	0.41	4.54	0.38	0.42	24
	Xe	4.47	1.16	0.42	4.56	0.38	0.42	
	Pb	4.48	2.89	10.1	6.25	0.60	3.04	
Zr	Pb	4.09	3.74	2.05	5.58	0.27	2.22	25
	U	4.11	4.25	6.6	18.4	2.74	0.85	
Co	Pb	4.85	5.2	0.62	3.58	0.34	0.62	-
	U	3.52	6.69	1.6	20	3.06	0.18	
	U	3.11	6.88	1.5	24.2	3.6	0.16	

TABLE I. Average energy, electronic stopping power, parameters ($\dot{\Delta\rho}_0$, p_0, $\Delta\rho_\infty$ and p_∞) deduced from the experiments, and saturation resistivity obtained after 2.5 MeV electron irradiations [7].

DISCUSSION and PHENOMENOLOGICAL MODEL

These two linear behaviours observed in each DDPC seem to indicate that the damage creation is accomplished by two mechanisms, one of them will saturate much earlier than the other one. Using the model described in [6, 8] we can account for these observations. One incoming ion basically damages a cylinder of section $S_1 = \pi r_1^2$ around the ion path leading to a local resistivity increase ρ_1 inside the damaged cylinder. Some of the created defects pre-existing between two coaxial cylinders of radii r_1 and r_2 ($>r_1$) (i.e inside a cylindrical ring of section $S_2\text{-}S_1=\pi (r_2^2\text{-}r_1^2)$ around the trajectory of the ion) can be recombinated or/and agglomerated which leads to an overall resistivity increase ρ_2 in this partly annealed matter.
After an irradiation fluence Φ the fraction F_1 of the damaged but not annealed matter obeys :

$$dF_1 = (1\text{-}F_1)\, S_1\, d\Phi - (S_2\text{-}S_1)\, F_1\, d\Phi , \qquad (1)$$

the fraction F_2 of the damaged and subsequently annealed matter obeys :

$$dF_2 = (S_2\text{-}S_1)\, F_1\, d\Phi - S_1\, F_2\, d\Phi . \qquad (2)$$

We can deduce the total fraction $F = F_1 + F_2$ of perturbed matter as

$$dF = S_1\, (1\text{-}F)\, d\Phi \qquad (3)$$

and finally the average electrical resistivity increase is given by

$$\Delta\rho = \rho_1 F_1 + \rho_2 F_2 . \qquad (4)$$

Integrating equations (1) and (3) from 0 to a given ion fluence Φ and using (4) leads to :

$$\Delta\rho = \rho_2 [1\text{-} \exp(\text{-}S_1 \Phi)] + (\rho_1 - \rho_2)\, [1\text{-} \exp(\text{-}S_2 \Phi)]\, S_1 / S_2 \qquad (5)$$

Comparing the experimental measured parameters ($\dot{\Delta\rho}_0$, p_0, $\Delta\rho_\infty$ and p_∞) to their expressions given by the model, we deduce :

$$S_1 = p_\infty \qquad (6)$$

$$\rho_1 = \dot{\Delta\rho}_0 / p_\infty \qquad (7)$$

$$S_2 = \dot{\Delta\rho}_0\, (p_0 - p_\infty) / (\dot{\Delta\rho}_0 - p_\infty \Delta\rho_\infty) \qquad (8)$$

$$\rho_2 = \rho_1\, (S_2 - p_0) / (S_2 - S_1) \qquad (9)$$

The values of the latter parameters are listed in table II . The DDPC deduced from the model presents a good fit to the experimental data in fig.1.

The ratio ρ_1 / ρ_2 is almost equal to 3 except for the titanium irradiated with lead ions. This reduction of resistivity can correspond to an agglomeration of defects leading to vacancy or intertitial clusters for which the resistivity per defect is lower than that of an isolated defect. The size calculated for vacancy clusters to obtain such a factor of 3 is about 100 atoms [9]. The high reduction (ρ_1 / ρ_2 ~10) in the case of titanium irradiated by 4.4 GeV lead ions can be explained by either a creation of much larger defect agglomerates, indeed dislocation loops were observed by TEM [2], or by a very efficient recombination.

Target	Ion	Energy (GeV)	$(dE/dx)_e$ keV/Å	$S_1 = p_\infty$ 10^{-13} cm^2	S_2 10^{-13}cm^2	ρ_1 μΩ cm	ρ_1/ρ_2
Ti	Xe	5.03	1.09	0.38	6.8	1.08	2.8
	Xe	4.47	1.16	0.38	6.8	1.10	2.9
	Pb	4.48	2.89	0.60	6.9	16.8	9.8
Zr	Pb	4.09	3.74	0.27	7.5	7.59	3.8
	U	4.11	4.25	2.74	24.2	2.41	3.7
Co	Pb	4.85	5.2	0.34	4.9	1.82	3.4
	U	3.52	6.69	3.06	25.8	0.52	3.9
	U	3.11	6.88	3.6	33,4	0.42	3.2

TABLE II. Parameters deduced from table I using equations (6) to (9).

As expected, in a given metal, S_1 ($=p_\infty$) increases very rapidly with the amount of energy deposited in electronic excitation.

For Zr and Co, S_2 follows the same evolution as S_1 : the volume concerned by these annealing processes increases rapidly with HEED in such a way that the ratio of the damaged to partly annealed regions decreases as HEED increases, therefore $\Delta\rho_\infty$ decreases. As shown in tab.I $\Delta\rho_\infty$ is very low compared to that observed during electron irradiations.

But for Ti, S_2 is constant and $\Delta\rho_\infty$ increases as the electrical stopping power increases. As above-mentioned the damage creation mechanism seems to be different in titanium than in Zr and Co. Furthermore the electrical resistivity increase of Ti after an irradiation with 2.2 GeV U ions up to a fluence of 4x10^{12} ions/cm^2, is about 50 $\mu\Omega$ *cm* although the saturation resistivity increase $\Delta\rho_\infty$ is still not reached. This latter value is greater than the saturation resistivity observed during electron irradiations ($\Delta\rho^e_\infty$~24 $\mu\Omega$ *cm*). Moreover, a detailed study of this overdamaged titanium by TEM and X-ray diffractions has confirmed that the very strong HEED of uranium ions has induced a phase transformation (α-->ω) in this target [10]. The strong electrical resistivity increase may be due to the apparition of the ω phase.

This type of DDPC showing two distinct regimes has not been observed during irradiation of iron with 1 GeV uranium ions at 80 K [6]. At this temperature the defects might be totally recombined (i.e ρ_2 ~0) so that S_2 could only be determined (S_2 ~5x10^{-13} cm^2). The value estimated for S_1 (~0.3x10^{-13} cm^2) is coherent with the present results.

CONCLUSION

The above results show that the damaging mechanism resulting from HEED can be explained, as described in [6], as the competition between two processes : (i) damage creation in a cylinder of section S_1 around the trajectory of the ion and (ii) annealing of some defects pre-existing between two coaxial cylinders of section S_2 -S_1 . The annealing process is more important as the irradiation temperature increases.

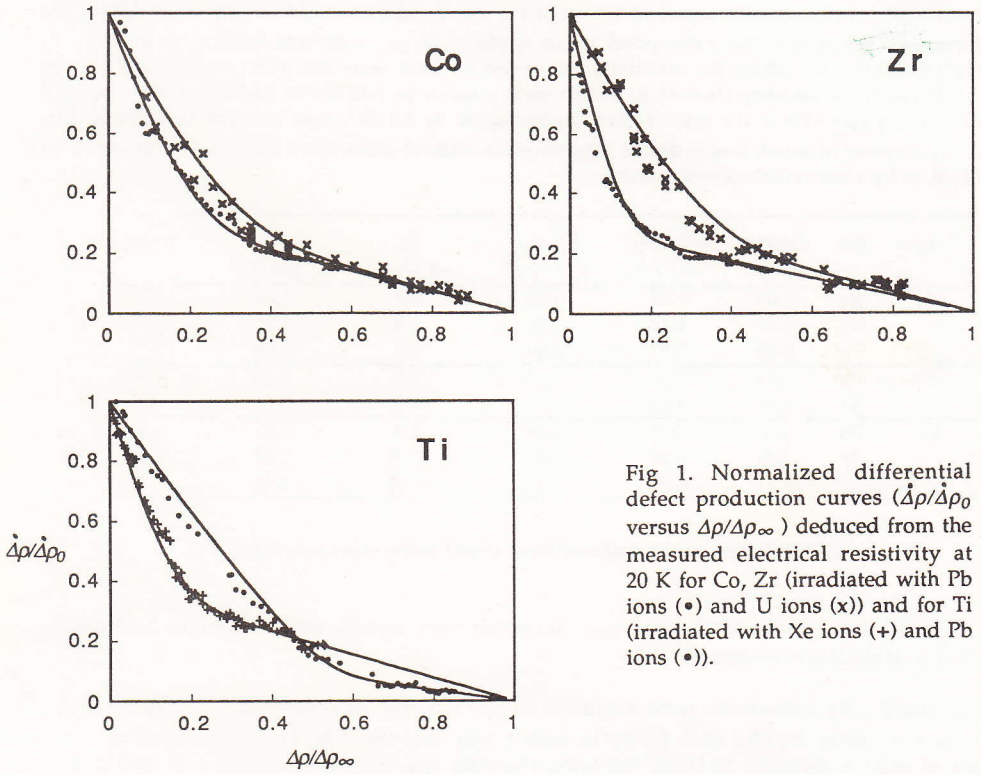

Fig 1. Normalized differential defect production curves ($\dot{\Delta\rho}/\dot{\Delta\rho}_0$ versus $\Delta\rho/\Delta\rho_\infty$) deduced from the measured electrical resistivity at 20 K for Co, Zr (irradiated with Pb ions (•) and U ions (x)) and for Ti (irradiated with Xe ions (+) and Pb ions (•)).

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