CONF-860605--23 Received by OSTI

AUG 19 1986

MASTER

THE COLLAPSE OF DEFECT CASCADES TO DISLOCATION LOOPS DURING SELF-ION IRRADIATIONS OF Fe, Ni AND Cu AT 30, 300 AND 600 K*

M. A. Kirk, I. M. Robertson**, J. S. Vetrano**,
M. L. Jenkins*** and L. L. Funk
Materials Science and Technology Division
Argonne National Laboratory
Argonne, IL 60439

**University of Illinois, Urbana, IL 61801
***University of Oxford, Oxford OX1 3PH, England

CONF-860605--23

DE86 014530

February 1986

The submitted manuscript has been authored by a contractor of the U. S. Government under contract No. W-31-109-ENG-38. Accordingly, the U. S. Government retains a nonexclusive, royalty-free license to publish or reproduce the published form of this contribution, or allow others to do so, for U. S. Government purposes.

DISCLAIMER

This report was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government nor any agency thereof, nor any of their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof.

Submitted to the Thirteenth International Symposium on Effects of Radiation on Materials, June 23-25, 1986, Seattle, WA.

*Work supported by the U. S. Department of Energy, BES-Materials Sciences, under Contracts W-31-109-Eng-38 (ANL) and AC02-76ER01198 (Univ. of IL).

DISTRIBUTION OF THIS DOCUMENT IS UNLIMITED

THE COLLAPSE OF DEFECT CASCADES TO DISLOCATION LOOPS DURING SELF-ION IRRADIATIONS OF Fe, Ni AND Cu AT 30,300 AND 600 K*

M. A. Kirk, I. M. Robertson**, J. S. Vetrano**,
M. L. Jenkins*** and L. L. Funk
Materials Science and Technology Division
Argonne National Laboratory
Argonne, IL 60439

**University of Illinois, Urbana, IL 61801
***University of Oxford, Oxford OX1 3PH, England

February 1986

Submitted to the Thirteenth International Symposium on Effects of Radiation on Materials, June 23-25, 1986, Seattle, WA.
*Work supported by the U. S. Department of Energy, BES-Materials Sciences, under Contracts W-31-109-Eng-38 (ANL) and AC09-76ER01198 (Univ. of IL).

THE COLLAPSE OF DEFECT CASCADES TO DISLOCATION LOOPS DURING SELF-ION IRRADIATIONS OF Fe, Ni AND Cu AT 30,300 AND 600 K*

M. A. Kirk, I. M. Robertson**, J. S. Vetrano**,
M. L. Jenkins*** and L. L. Funk
Materials Science and Technology Division
Argonne National Laboratory
Argonne, IL 60439

**University of Illinois, Urbana, IL 61801
***University of Oxford, Oxford OX1 3PH, England

Abstract

The formation of dislocation loops by self-ion irradiations of Fe, Ni and Cu has been studied in situ in the ANL High-Voltage Electron Microscope as functions of ion dose and irradiation temperature. At low doses ($< 10^{12}$ ions/cm^2) at room temperature individual cascades were observed to collapse to vacancy dislocation loops in Cu with high probability, in Ni with lower probability, and in Fe with zero probability. Cascade collapse was observed at low doses at 30 K in Cu and Ni, but at rates less than their respective rates at room temperature. A loop formation rate for Ni at 600 K is also reported. At higher doses ($> 10^{13}$ ions/cm^2) where overlap of cascades becomes significant, loops were first observed in Fe and with a supra-linear build-up with dose. Also at higher doses a decrease in loop production rate in Cu and Ni was observed due to loop coalescence. The materials and temperature dependence of cascade collapse probabilities is suggested to be related to thermal spike mechanisms during the cascade formation lifetime.

Key Words: Defect Cascades, Dislocation Loops, Fe, Ni and Cu, Self-ion Irradiations, HVEM, 30, 300 and 600 K.

---

*Work supported by the U. S. Department of Energy, BES-Materials Sciences, under Contracts W-31-109-Eng-38 (ANL) and AC09-76ER01198 (Univ. of IL).

THE COLLAPSE OF DEFECT CASCADES TO DISLOCATION LOOPS DURING SELF-ION IRRADIATIONS OF Fe, Ni AND Cu AT 30,300 AND 600 K*

M. A. Kirk, I. M. Robertson**, J. S. Vetrano**,
M. L. Jenkins*** and L. L. Funk
Materials Science and Technology Division
Argonne National Laboratory
Argonne, IL 60439

**University of Illinois, Urbana, IL 61801
***University of Oxford, Oxford OX1 3PH, England

Introduction

In metals under fast neutron bombardment atomic defects, vacancies and interstitials, are initially produced in defect cascades. Typically hundreds of defect pairs are formed in such a cascade, within 10^{-12} second of the primary recoil event, and over a volume of 10 nm diameter. The fate of these defects and how they affect material properties depends critically on the metal and the irradiation temperature. Often many of these defects form dislocation loops which are visible in the Transmission Electron Microscope. These dislocation loops are important as point defect sinks (and sources at high temperatures), as potential nucleation sites for voids or for a second phase, and as precursors of heavily tangled dislocation structures resulting in embrittlement.

Most researchers have believed for some time that the vacancies are produced in a central region of the defect cascade and that these vacancies can collapse to a close packed plane initially producing a dislocation loop at the cascade site. This has been convincingly demonstrated at room temperature in the ordered alloy Cu_3Au irradiated both with ions and with neutrons [1]. We will show in this paper and in another paper in these proceedings [2] that

---

*Work supported by the U. S. Department of Energy, BES-Materials Sciences, under Contracts W-31-109-Eng-38 (ANL) and AC09-76ER01198 (Univ. of IL).

many dislocation loops form athermally from defect cascades in certain metals, probably within a cascade lifetime of 10^{-11} second.

We report in this paper some results from a study in progress on the probabilities for cascade collapse to loops in Fe, Ni and Cu as functions of irradiation temperature and ion dose (cascade overlap). Self ions of energies of 50 and 100 keV were employed to simulate cascades produced by fast neutrons. Iron, nickel and copper were chosen to represent a possible range of cascade collapse probabilities based on an interpretation of damage rate data in Fe at 5 K under fast neutron bombardment [3]. These and other neutron data suggest that loops form from individual cascades produced at 5 K with increasing probability from Fe to Ni to Cu and that the probability increases with dose in Fe, and possibly to a lesser extent in Ni. We have found this interpretation to be consistent with our own earlier results in Fe [4].

Experimental Procedures

Samples were produced from commercially pure Fe and high purity Ni and Cu foils and electropolished using standard techniques. All experiments were performed in the High-Voltage Electron Microscope - Accelerator Facility at Argonne National Laboratory [5]. Samples were irradiated in situ with self ions at dose rates near 3×10^{10} ions/(cm^2 sec) and at temperatures of 30, 300 and 600 K ± 2 K. The relative accuracy of ion dosimetry among various irradiations reported here is 10%. The absolute accuracy of ion dosimetry is believed to be about 20% and is currently under investigation using the disordered zone techniques in Cu_3Au [1]. Electron micrographs were taken at the irradiation temperatures as a function of ion dose and following annealing, often in the same sample area. The HVEM was operated at voltages below the threshold for electron damage in the foils: 200 keV for Fe and 300 keV for Ni and Cu.

Dislocation loops were imaged in thin regions of the foil (30-40 nm) using strong 2-beam or very slightly s-positive (small positive deviation from Bragg condition) diffraction conditions. Images were recorded in dark field with operating diffraction vectors known to image most or all loops produced in these metals under these irradiation conditions: $\bar{g} = \langle 200 \rangle$ for all Frank loops in fcc Cu and Ni [6], $\bar{g} = \langle 110 \rangle$ in bcc Fe [4]. The smallest loops resolvable varied between 1 and 2 nm depending on metal, foil surface quality and sample stage temperature. Identification and densities of loops were measured independently by 2 or 3 researchers. Both characteristic black-white and sharp black spot images were counted. Sizes of loop images were also determined in some experiments. Average loop image diameters were measured with a digitizer on prints of 600,000 magnification along the black-white interface of a loop when available, or over the average extent of a black spot.

Results and Discussion

The effect of increasing dose (100 keV Cu^+) in copper at room temperature is illustrated qualitatively in the micrograph series of Fig. 1. Vacancy dislocation loops in the exact same area are imaged at each dose. (It has been determined in many earlier experiments [7] that vacancy loops are formed in many metals by low dose ion irradiation of thin foils at room temperature.) Subcascade structure, more than one dislocation loop formed per 100 keV defect cascade, is observed to probably occur in 10-20% of all visible events at the lowest dose. A probable example is arrowed in Fig. 1, at 0.2×10^{12} ions/cm^2.

A rather rapid saturation of the loop (defect) density, measured from the micrographs of Fig. 1, occurred in Cu at 300 K as shown in Fig. 2. (Representative error bars for density variation within one micrograph and among determinations by several researchers are shown in this and following figures. No correction was made for the presence of subcascades; all loops were counted.) This saturation is apparently accomplished by loop coalescence. An example is clearly observed at relatively low doses where two nearby loops, existing at a dose of 0.5 x 10^{12} ions/cm^2 (arrowed, Fig. 1), disappear at the next dose with the formation of a new loop (arrowed). This is probably triggered by a cascade event at the location of the new loop, and thus three loops become one. The mechanism by which this occurs is unknown. Configurations of existing loops were observed to change with increasing dose, and a decreasing loop production rate was measured. It is possible at the higher doses that local bending could become severe and consequently some small loops could go out of contrast and were therefore not observed. However the observations of a strong saturation behavior at doses > 1 x 10^{12} ions/cm^2 and the clear example of loop coalescence at lower dose certainly indicate a real effect of a decreasing production rate of loops with increasing loop density.

Under similar self-ion irradiation conditions, vacancy dislocation loops are formed in Ni at 300 K at a rate of about one third of the initial rate in Cu. This is shown in Fig. 2. A saturation behavior in the loop density was also observed in Ni, but at higher doses than in Cu. This, and an energy dependence of the loop production rate, is displayed in Fig. 3. Subcascades were observed to form less often in Ni than Cu at 100 keV, with a frequency < 10% of observed events. Preliminary size distributions (not shown) indicate that loops in Cu are larger than in Ni.

The lower loop production rate at 100 keV, as compared to 50 keV (Fig. 3), was reproduced in 2 experiments at each energy. This somewhat surprising effect is difficult to understand. The average projected ranges for self ions in Ni are calculated by the TRIM code [8] to be 12 nm and 24 nm for 50 and 100 keV respectively. Most vacancy loops will be formed within those distances from the foil surface. Only at 100 keV will some loops be formed in the boundary layer at a depth of about 13 nm (1/3 $\xi_{<200>}$ at 300 keV electron energy) and thus possibly be invisible in a single reflection at a single tilt value [6]. It seems very unlikely that fully one half of all loops would be invisible, even though the boundary layer falls at about the damage peak in the 100 keV case. Tilting experiments will be performed to further check this effect, but we believe this will prove incapable of explaining the entire difference between irradiation energies.

The increase in loop density with 50 keV self-ion dose at 30 K is shown qualitatively in Fig. 4. Due to changing local diffraction conditions it was not possible to follow the same sample area as a function of dose at 30 K or upon annealing to 300 K. Different areas representative of each dose are instead shown. Again a saturation behavior of the loop density was measured (not shown) and found to be similar to that observed in the 300 K irradiation data.

The dependence of the loop production rate (at relatively low self-ion doses) on irradiation temperature is shown in Fig. 5. The production rates at 30 and 300 K were reproduced in 2 runs at each temperature. Only one experiment at 600 K has been performed to date. No obvious loop annealing occurred at 600 K, so this data reflects the true loop production rate. The difference in loop production rates at 30 and 300 K is about a factor of 2. This difference easily exceeds the error in the loop counting method on any given micrograph (representative error bars in Fig. 5), but also well exceeds an observable difference in loop resolution at the two irradiation temperatures. The liquid He sample stage is more stable at room temperature than it is with flowing cold gas. The loop-size distributions at 30 K and upon annealing to 300 K are displayed in Fig. 6. There is a shift of 25% of the defects to smaller sizes (1.0-2.0 nm) at 300 K, some of which may be a real annealing effect, the bulk of which is probably a resolution effect. The difference in loop production rates at 30 and 600 K may not be outside the effect of a loss of resolution of small loops at 30 K. However, the differences between 300 K and 30 K and between 300 K and 600 K appear to be real.

In Table 1 a summary is given of the loop production rates for the three elements under the irradiation conditions we have investigated to date. The Ni values are taken from the linear fits to the low dose points displayed in Figures 3 and 5. The Cu value at 100 keV (300 K) is from the linear portion of the curve in Fig. 2 and the value at 50 keV (30 K) is from a single dose point. Uncertainties in the Ni and Cu values average ~0.03. The Fe values are previously published [4,9], where a measurable yield is found only at higher doses (~10^{13} ions/cm^2). The loop yield in Cu at 30 K is surprisingly low and must be confirmed by another experiment. The value is also less certain than others in the table due to a change in ion dosimetry methods. However there is no doubt that individual defect cascades collapsed to vacancy dislocation loops in Cu and Ni at 30 K, a temperature below which the free migration of interstitials or vacancies does not occur. Thus the formation of these loops is attributed to a process occurring during the cascade lifetime (~10^{-11} sec).

A comparison among the three elements of loop yield versus dose is shown in Fig. 7 for room temperature irradiations (100 keV self ions), and in Fig. 8 for irradiations at 30 K (50 keV self ions). In both figures the linear behavior (slope n=1) indicates an isolated loop production rate which is constant in dose. The slopes $n < 1$ at high defect densities in Cu and Ni are predominantly due to saturation behavior such as loop coalescence. The slope $n = 0.7$ in Fe at 300 K (Fig. 7) may also be due to a saturation type behavior. The slope $n = 1.5$ in Fe at 30 K (Fig. 8) is probably the net result of a low probability for isolated cascade collapse (linear loop production rate, slope $n=1$) combined with a quadratic loop production rate (slope $n=2$) due to simple overlap of two defect cascades resulting in one dislocation loop.

Annealing results for the 30 K irradiations have been quite varied among the three elements. In Fe the loop density increased 20-30% at a dose of 8×10^{13} ions/cm^2. In Ni at low doses ($\sim 1 \times 10^{12}$ ions/cm^2) the increase was much greater, about 100%, but at the higher dose of 3×10^{13} ions/cm^2 the increase was only 10-20%. Such small increases upon annealing may be due primarily to an increase in loop resolution at room temperature as mentioned ealier. The larger increase of 100% is well outside this uncertainty, however. A single observation in Cu irradiated to low dose (3×10^{11} ions/cm^2) at 30 K and annealed to 300 K revealed a decrease in loop density of 25% (Fig. 9). From the distribution of loop sizes at both temperatures (not shown) it seems likely that many of the larger loops (4-7 nm) had slipped out of the Cu foil.

A video recording was made during three irradiations of Ni at 300 K with 50 keV self ions to investigate the possibility of loop loss to the sample surface occurring within one second to one minute of formation. Such loops would go unobserved in normal micrograph exposures made typically 10-20 minutes following irradiations. The video resolution was somewhat inferior to film micrographs such that only about half of the defects known to be formed could be observed. With this limitation it was still concluded that no substantial loss of large loops occurred within that time frame. No large loops were observed to suddenly disappear in the video recording. A very few large loops have been observed to disappear quite suddenly on the microscope screen, probably under the influence of the electron beam, 10 minutes or more following the ion irradiation. Their numbers were insignificant to the measured defect densities.

The differences in loop production rates among elements and between 30 and 300 K for any one element (Table 1) can be qualitatively explained within a thermal spike model for the evolution of the defect cascade. This model allows for a sufficient energy density and cascade lifetime to permit the defect motion required for the formation of vacancy dislocation loops. The difference among the three elements may be explained by differences in the average thermal spike lifetimes. The difference with irradiation temperatures, 30 and 300 K, may be explained by a more rapid quenching of the thermal spike at 30 K and thus a lower probability of loop formation. This latter idea is taken from a more complete cascade study in Cu_3Au reported briefly in this conference [2].

The room temperature yields, or loop production rates, in Fe, Ni and Cu can be compared with recent literature values. If our Cu yield could be corrected for subcascades, at the 100 keV incident self-ion energy, it would probably agree closely with the 30 keV result (0.4-0.5) of Stathopoulos [6]. This latter work is probably the most accurate yield determination in Cu. The Fe yield at 300 K agrees rather well with some recently reported results of Kitagawa, et al [10]. However, our Ni yield disagrees with those of Robinson [11] and Kitagawa [10]. The Robinson yield of 0.44 loops per incident 80 keV self ion was determined with a microscope of higher resolution, but reported size distributions are in approximate agreement with ours, leaving the bulk of the difference unexplained. The Kitagawa yield of 0.9 loops per incident 65 keV self ion, and size distribution peaking between 0.5 and 1.0 nm with no defect sizes > 1.6 nm, we feel are unreliable results due to their use of the weak beam technique. It is known that imaging and sizing small loops is most reliably performed and interpreted using strong 2-beam or slightly s-positive conditions (small positive deviation from Bragg conditions) [6,12].

Summary

1. Evidence for the saturation of dislocation loop densities with self-ion bombardment in Cu and Ni is presented. Loop coalescence is observed.
2. The production rate of loops at 300 K in Ni at a self-ion bombardment energy of 50 keV is twice that of 100 keV.
3. Vacancy dislocation loops are formed in Cu and Ni under self ion bombardment at 30 K, implying a cascade collapse process occurring during the cascade lifetime (10^{-11} sec).
4. In Ni the cascade collapse probability, or loop production rate, during 50 keV self ion irradiation increases by a factor of 2.5 from 30 K to 300 K, and decreases by a factor of 0.6 from 300 K to 600 K.
5. The loop yield differences in Fe, Ni and Cu at low doses of self ions is attributed to differences in cascade thermal spike lifetimes.
6. Comparing loop yields of Cu, Ni and Fe versus self ion dose, three types of behavior are distinguished: isolated cascade collapse, collapse due mainly to overlap of defect cascades, and a saturation behavior due to loop coalescence.

References

[1] Jenkins, M. L. and English, C. A., J. Nucl. Mater. 108 & 109 (1982) 46.

[2] Black, T. J., Jenkins, M. L., English, C. A. and Kirk, M. A., to be published in these proceedings.

[3] Dunlop, A., Pande, B. M., Böning, K., Rosner, P. and Schaefer, H. E., J. Nucl. Mater. 108 & 109 (1982) 83.

[4] Robertson, I. M., Kirk, M. A. and King, Wayne E., Scripta Met. 18 (1984) 317.

[5] Taylor, A., Wallace, J. R., Ryan, E. A., Philippides, A. and Wrobel, J. R., Nucl. Inst. Methods 189 (1981) 211.

[6] Stathopoulos, A. Y., Phil. Mag. A44 (1981) 285.

[7] Jäger, W., J. Microsc. Spectrosc. Electron. 6 (1981) 437.

[8] Biersack, J. P. and Haggmark, L. G., Nucl. Inst. and Meth. B7/8 (1980) 257.

[9] Kirk, M. A., Robertson, I. M., King, Wayne E., Ryan, E. A. and Philippides, A., Mat. Res. Soc. Symp. Proc. 41 (1985) 319.

[10] Kitagawa, K., Yamakawa, K., Fukushima, H., Yoshiie, T., Hayashi, Y., Yoshida, H., Shimomura, Y. and Kiritani, M., J. Nucl. Mat. 133 & 134 (1985) 395.

[11] Robinson, T. M. and Jenkins, M. L., Phil. Mag. A43 (1981) 999.

[12] Katerbau, K.-H., Dr. rer. nat. Thesis, Univ. of Stuttgart, Stuttgart 1978.

Table 1

	Fe		Ni		Cu	
Self Ion Energy (keV)	50	100	50	100	50	100
Irradiation Temperature (K)						
30	~.001	~.001	.08	(.13)*	(.12)*	--
300		~.001	.20	.11	--	.60
600	--	--	.12	--	--	--

*limited data

Figure Captions

Fig. 1. Dark field ($\bar{g}$ = <200>) micrograph series illustrating increase in dislocation loop density in the same area in Cu with dose of self ions at an energy of 100 keV at a temperature of 300 K. A possible subcascade cluster of loops is arrowed at a dose of 0.2 x 10^{12} ions/cm^2. Two loops are arrowed at a dose of 0.5 x 10^{12} ions/cm^2 which disappear at the next dose and one loop is arrowed at that dose (0.9 x 10^{12} ions/cm^2) which probably is related to disappearance of two nearby loops from the previous dose.

Fig. 2. Comparison of loop production in Cu and Ni during 100 keV self-ion irradiations at a temperature of 300 K.

Fig. 3. Comparison of loop production in Ni during self-ion irradiations of 50 and 100 keV energies at a temperature of 300 K.

Fig. 4. Dark field ($\bar{g}$ = <200>) micrograph series illustrating the increase in dislocation loop density in Ni during 50 keV self-ion irradiation at a temperature of 30 K, and upon annealing to 300 K.

Fig. 5. Comparison of loop production in Ni during 50 keV self-ion irradiations at temperatures of 30, 300 and 600 K.

Fig. 6. Comparison of size distributions of loops formed at 30 K with loops formed at 30 K and annealed to 300 K.

Fig. 7. Comparison of loop production in Cu, Ni and Fe during 100 keV self-ion irradiations at a temperature of 300 K.

Fig. 8. Comparison of loop production in Ni and Fe during 50 keV self-ion irradiations at a temperature of 30 K.

Fig. 9. Dark field ($\bar{g}$ = <200>) micrographs illustrating the decrease in loop density in Cu self-ion irradiated to 3.3 x 10^{11} ions/cm^2 at 30 K following an anneal to 300 K.

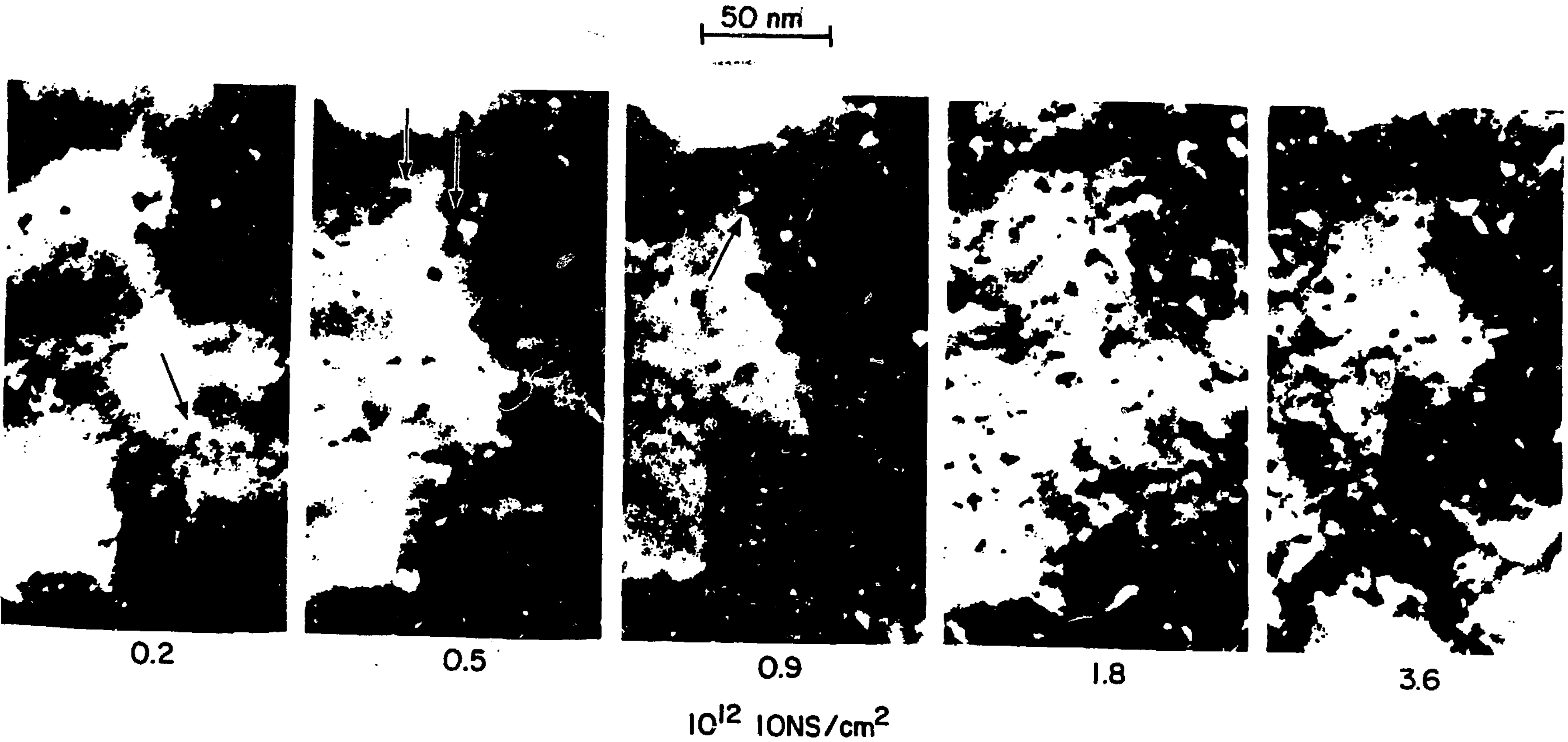


Fig.1

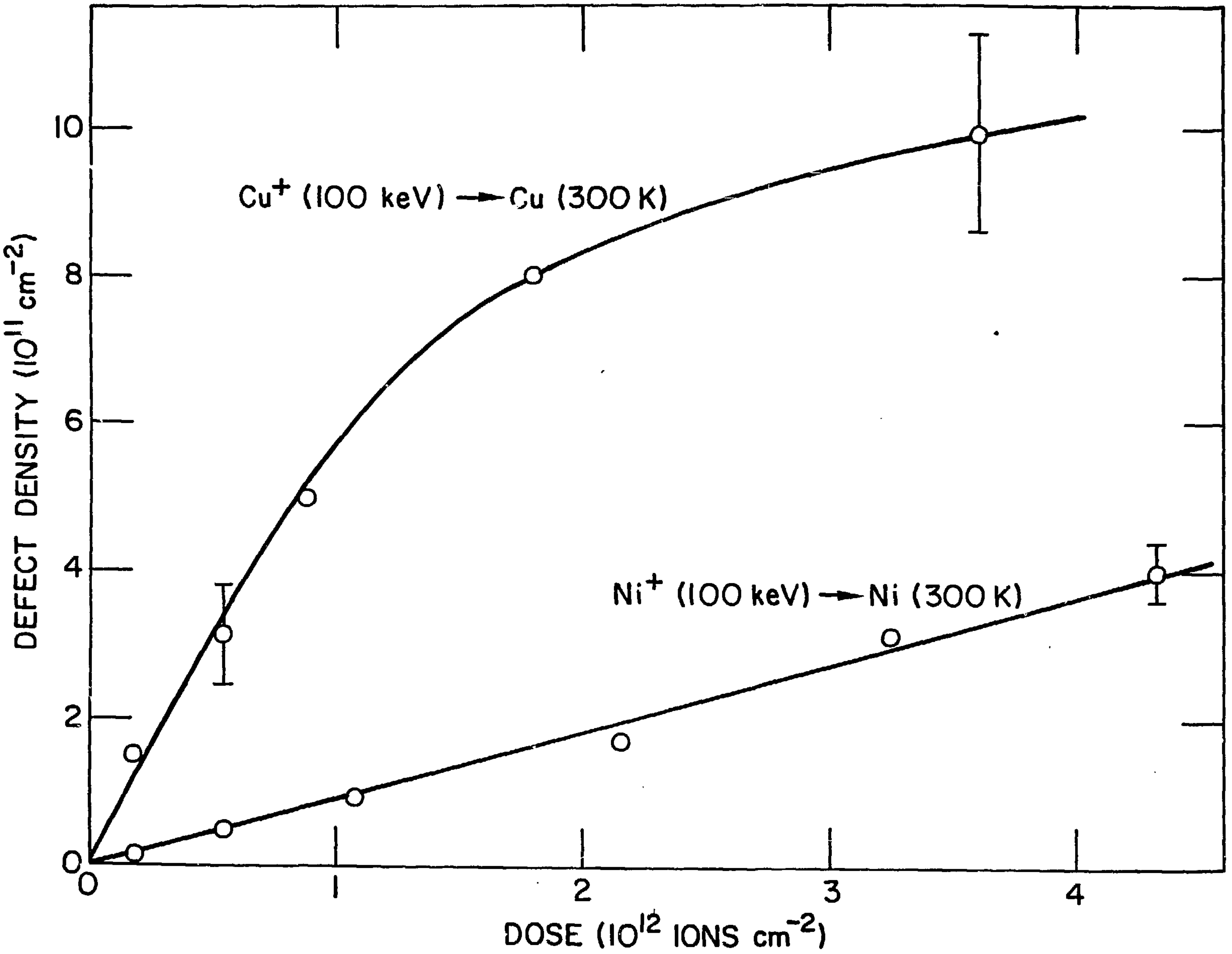

Cu+ (100 keV) → Cu (300 K)
Ni+ (100 keV) → Ni (300 K)
DEFECT DENSITY (10^11 cm^-2)
DOSE (10^12 IONS cm^-2)
0
2
4
6
8
10
0
1
2
3
4

Fig.2

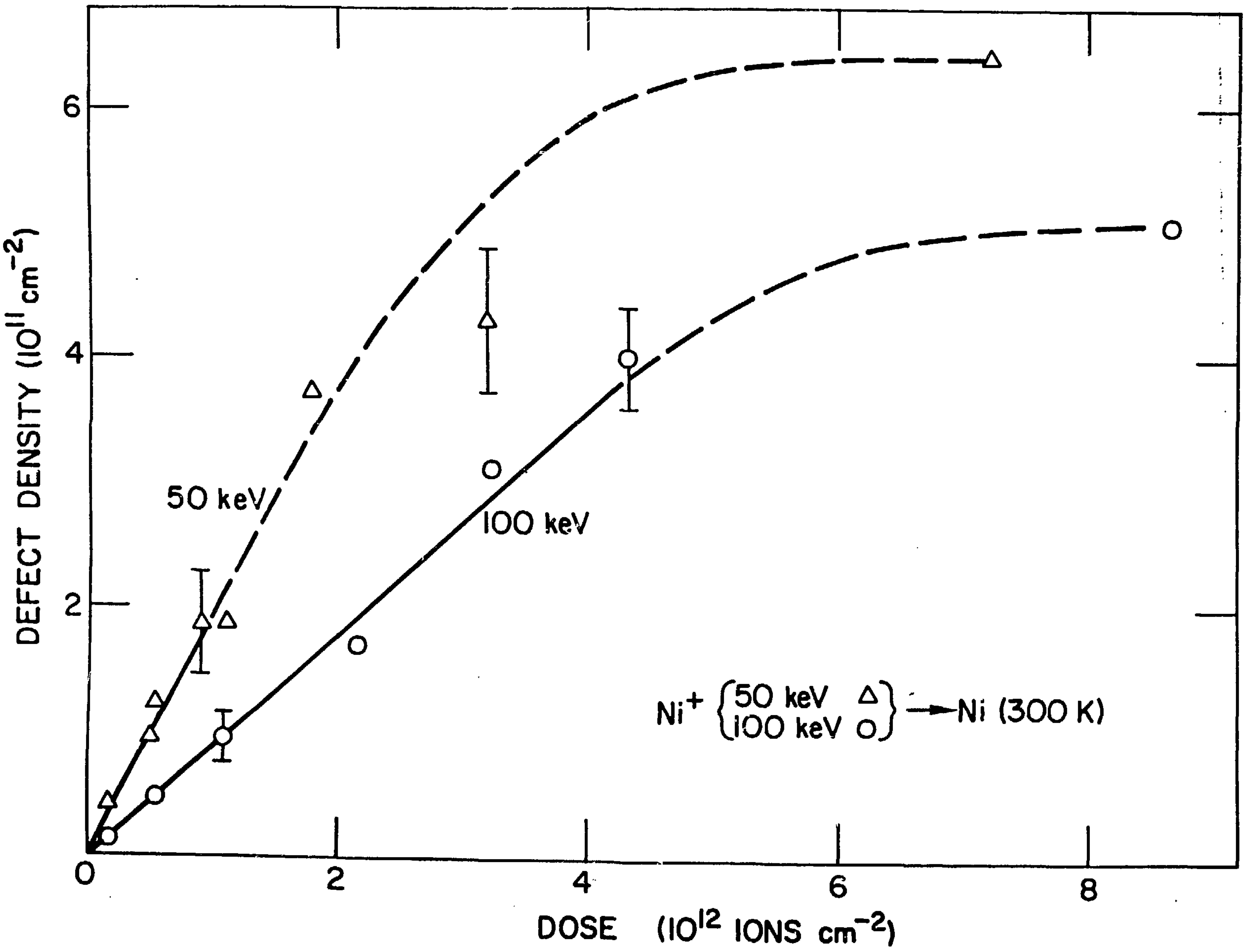

6
4
2
0
2
4
6
8
DEFECT DENSITY (10^{11} cm^{-2})
DOSE (10^{12} IONS cm^{-2})
50 keV
100 keV
Ni^{+} {50 keV △, 100 keV ○} → Ni (300 K)

Fig.3

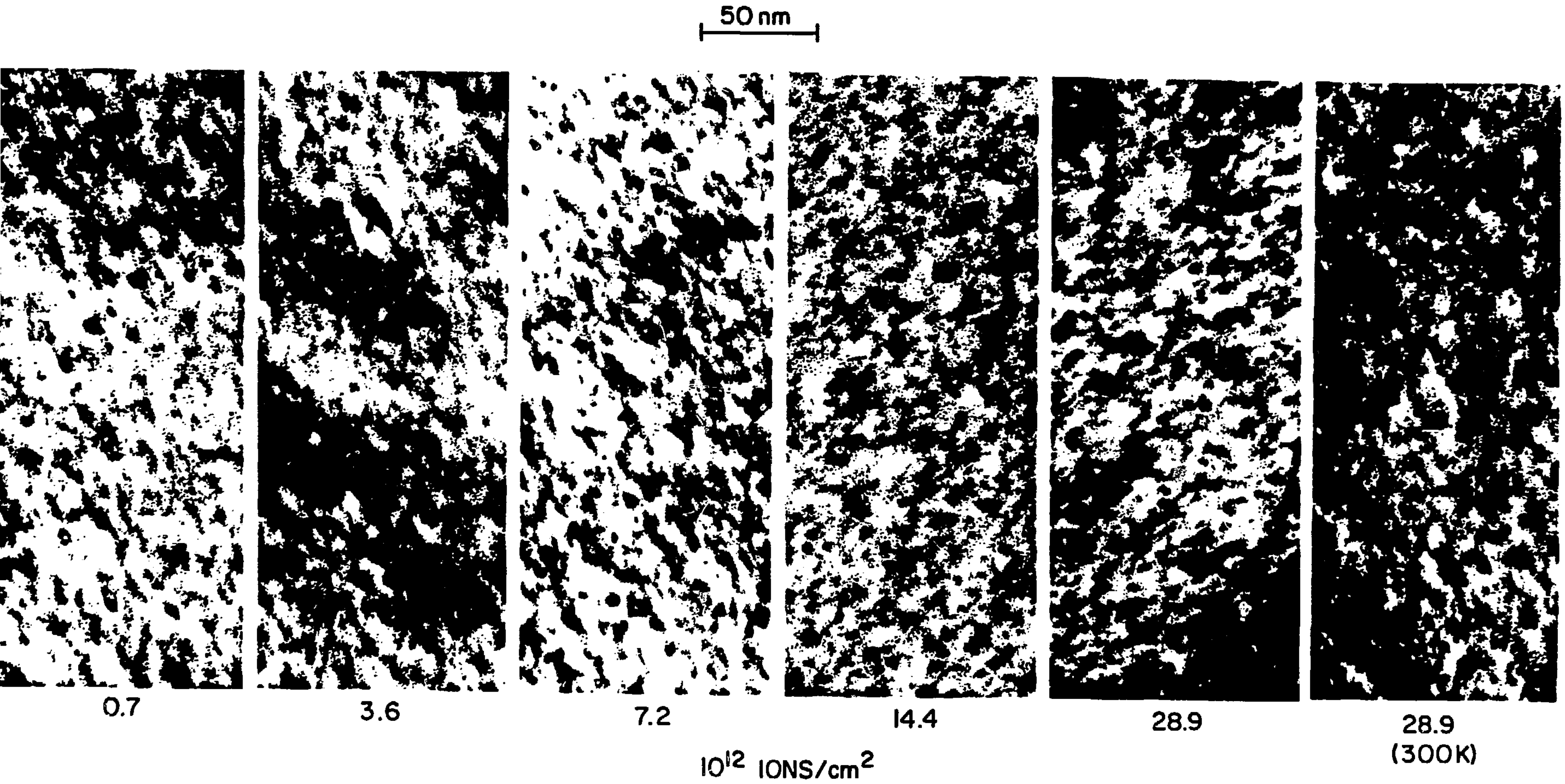
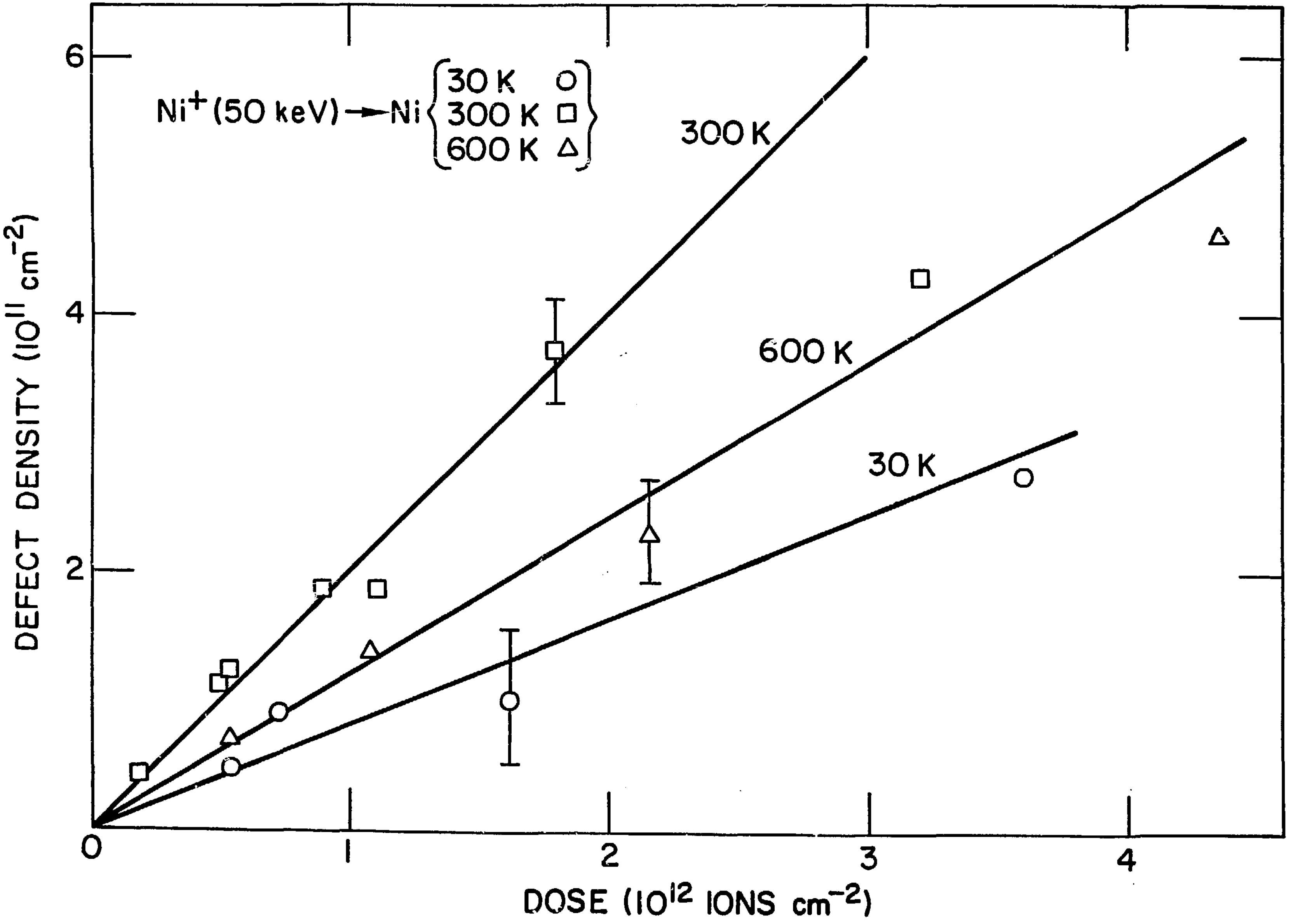
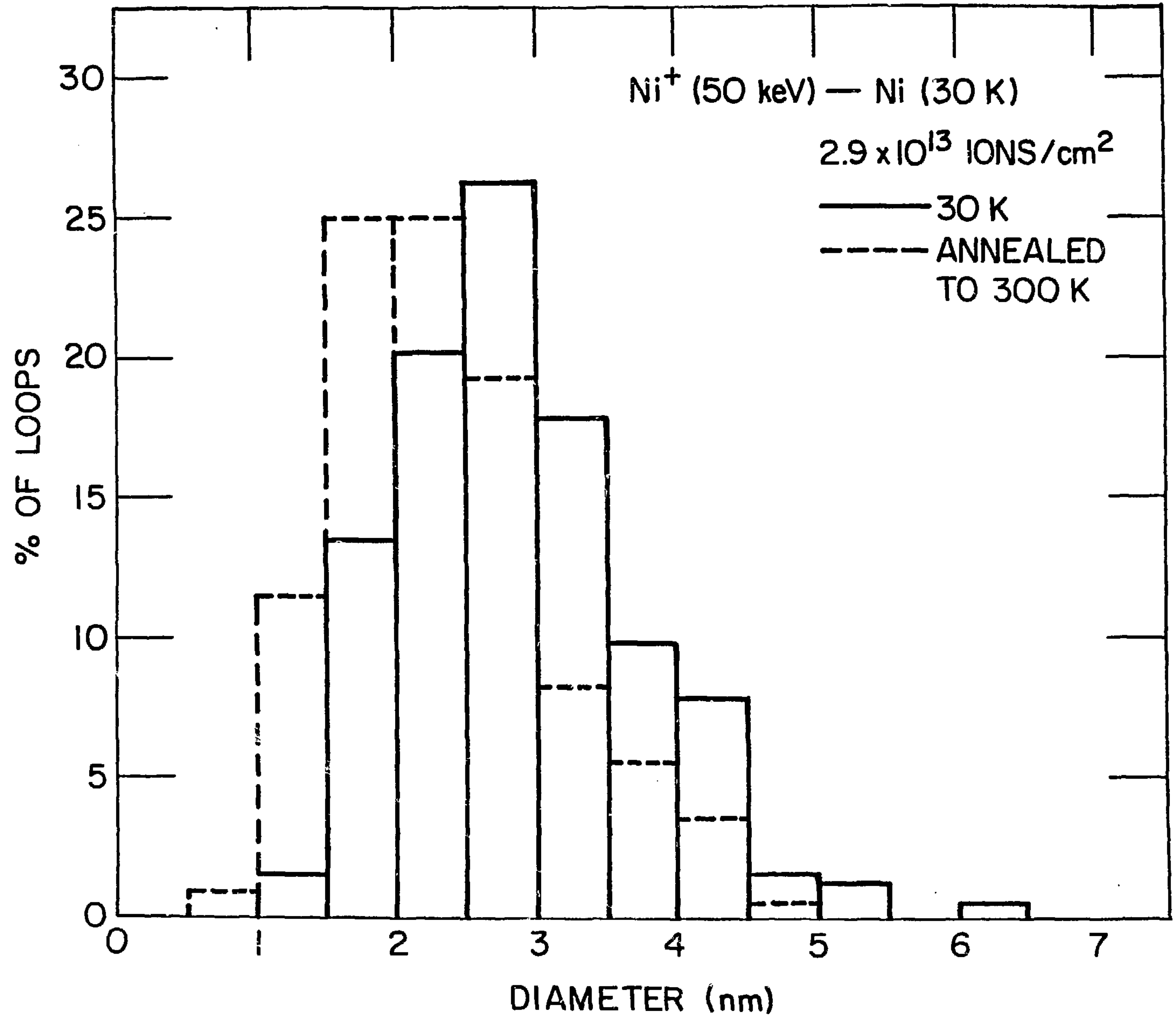


Fig.4



Ni$^+$(50 keV) → Ni {30 K ○, 300 K □, 600 K △}
300 K
600 K
30 K
6
4
2
0
1
2
3
4
DEFECT DENSITY (10^{11} cm^{-2})
DOSE (10^{12} IONS cm^{-2})

Fig.5



Ni$^+$ (50 keV) — Ni (30 K)
2.9 x 10^{13} IONS/cm^2
30 K
ANNEALED TO 300 K
% OF LOOPS
0
5
10
15
20
25
30
DIAMETER (nm)
0
1
2
3
4
5
6
7

Fig.6

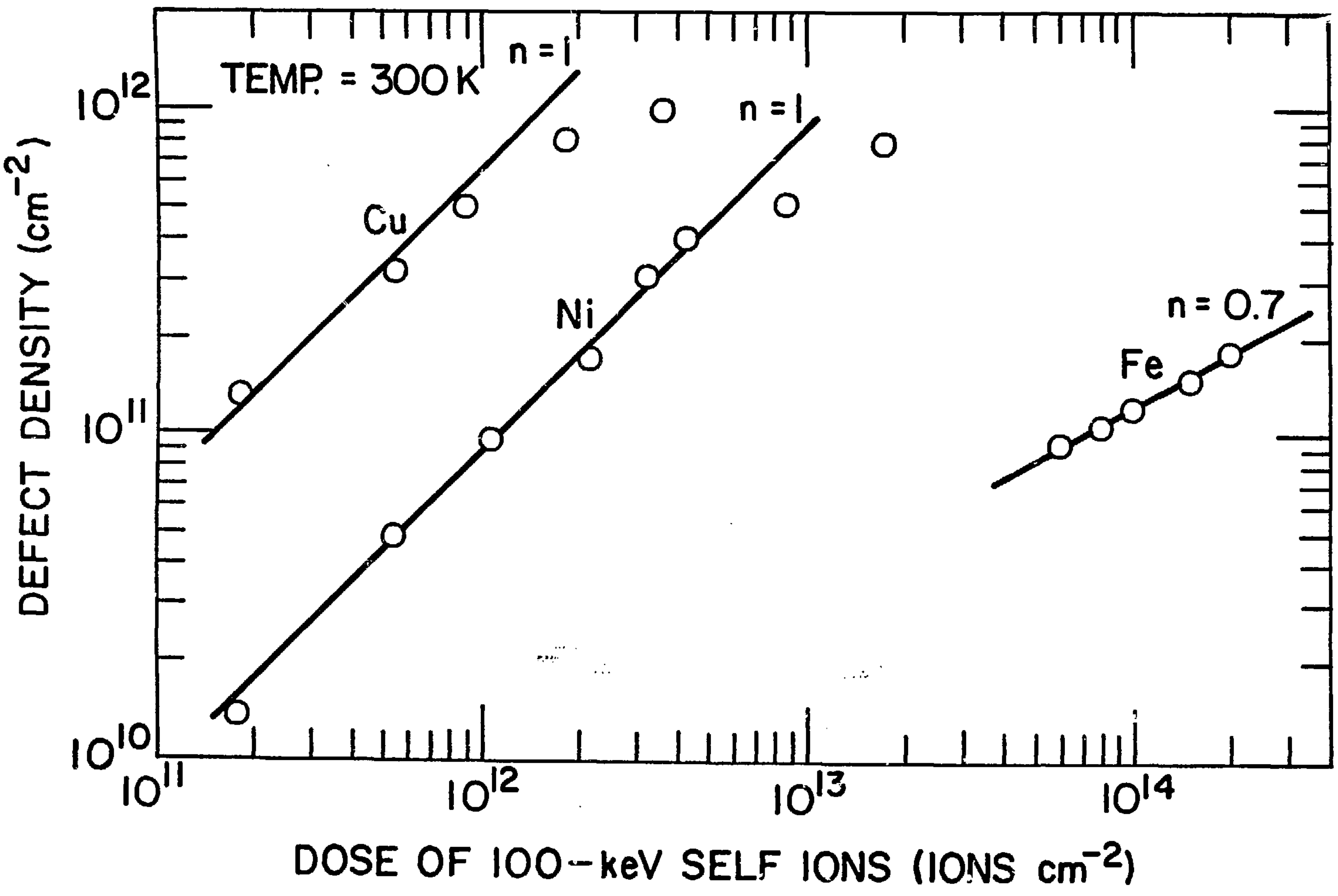

TEMP. = 300 K
n = 1
Cu
n = 1
Ni
n = 0.7
Fe
10^{12}
10^{11}
10^{10}
DEFECT DENSITY (cm^{-2})
10^{11}
10^{12}
10^{13}
10^{14}
DOSE OF 100-keV SELF IONS (IONS cm^{-2})

Fig.7

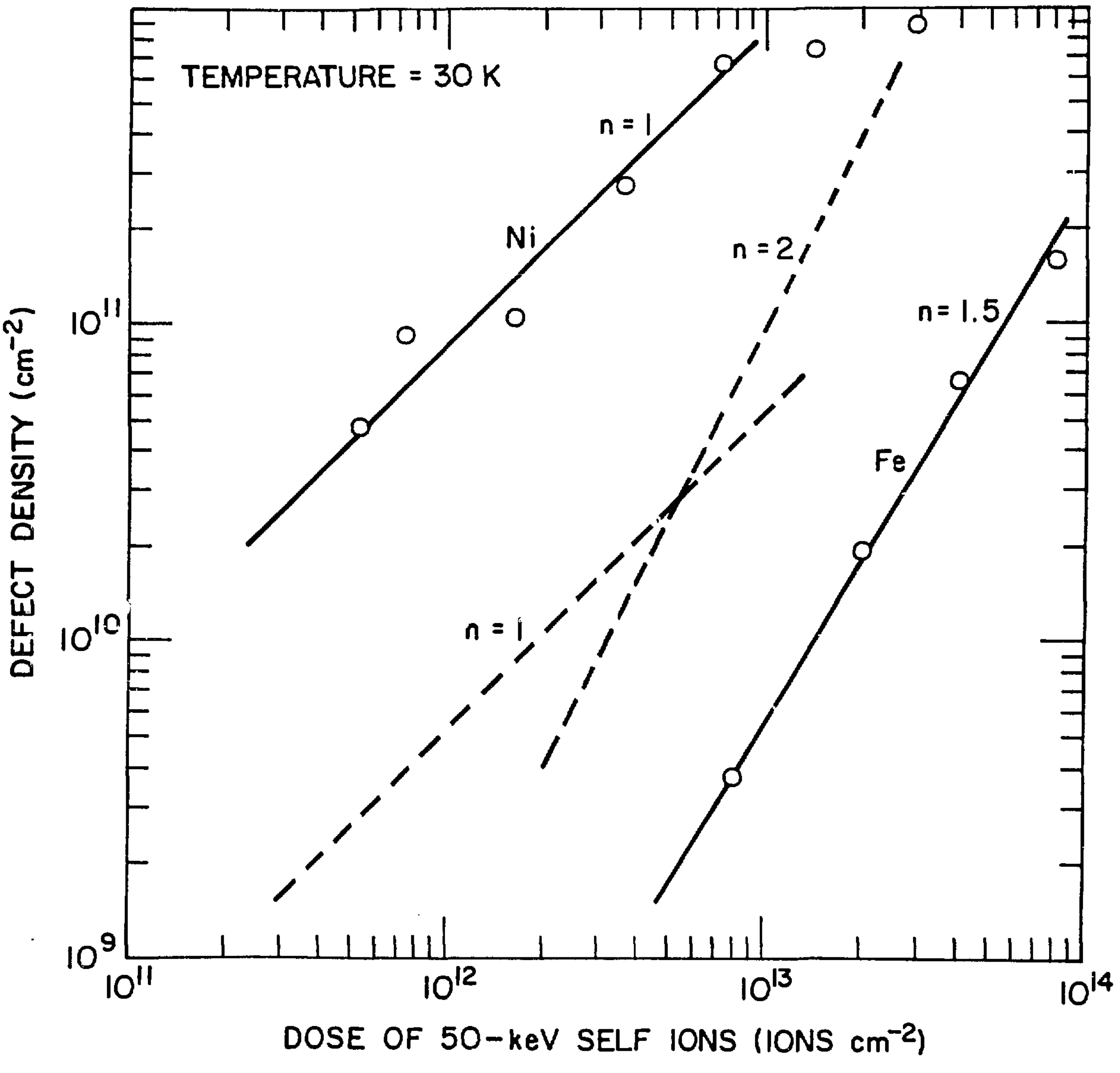

TEMPERATURE = 30 K
n = 1
Ni
n = 2
n = 1.5
Fe
n = 1
DEFECT DENSITY (cm^{-2})
10^{11}
10^{10}
10^{9}
10^{11}
10^{12}
10^{13}
10^{14}
DOSE OF 50-keV SELF IONS (IONS cm^{-2})

Fig.8

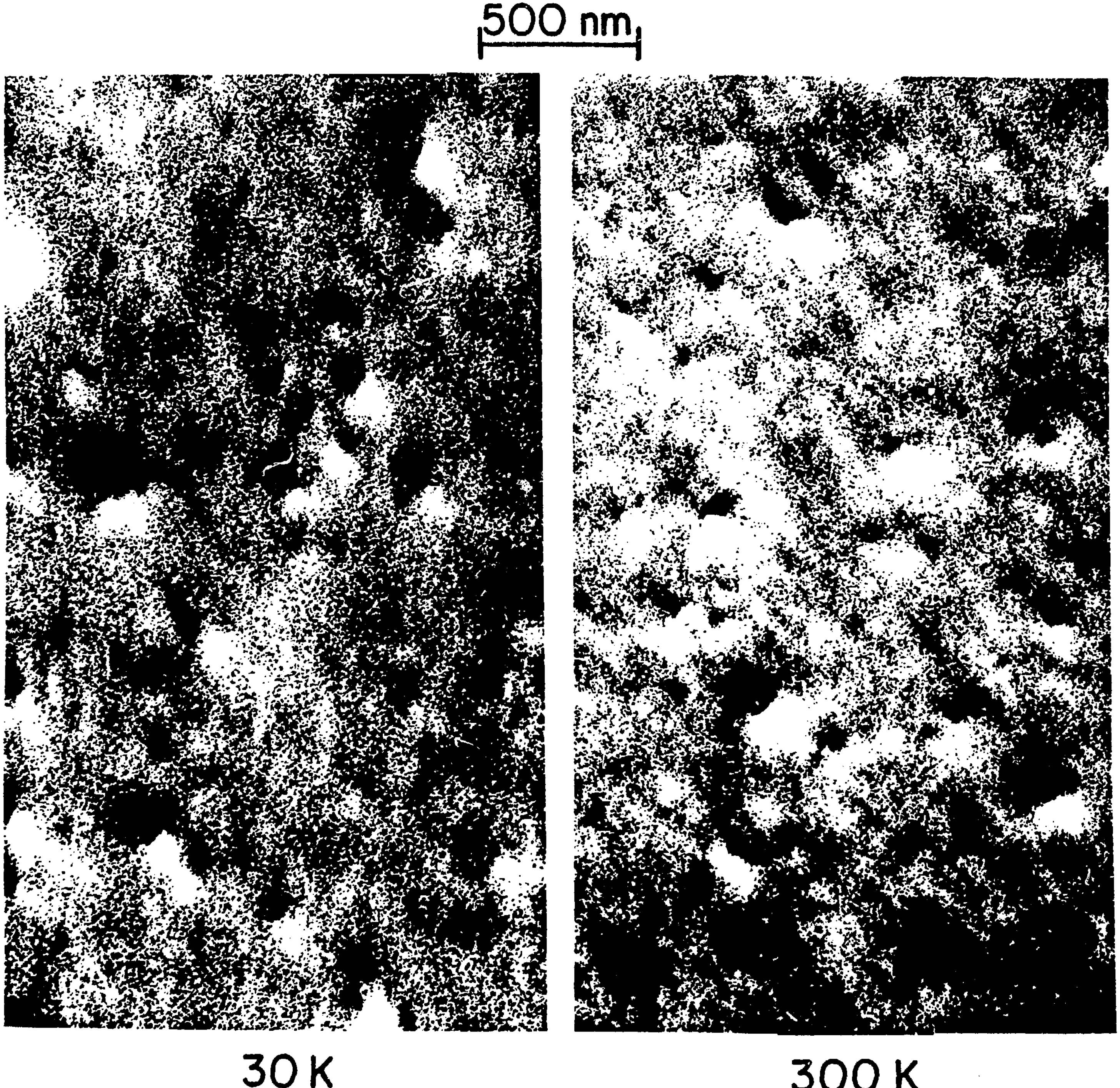


Fig.9