The Spatial Distribution of Irradiation Damage Spots

F. J. MINTER, A.J. E. FOREMAN
Metallurgy Division, United Kingdom Atomic Energy Authority Research Group, Atomic Energy Research Establishment, Harwell, Berks, UK

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The fine-scale damage spots observed in thin foils of neutron irradiated copper do not always appear to be entirely randomly distributed, showing in particular some apparent lining up of spots. A digital computer has been used to determine the number of linear triplets of spots in both a foil and a series of random arrays, the results being displayed pictorially by an electronic graph plotter. No significant difference between the results for the foil and the random arrays was found. The near randomness of the spots was confirmed by counts of the density distribution function for two foils, which both showed almost a Poisson distribution.

It is concluded that the apparent "structure" in the distribution of the irradiation damage spots is wholely compatible with that of random arrays. This result may have general implications in other branches of metallography.

1. Introduction

Electron microscope studies of thin foils of neutron-irradiated copper [1] show the existence of a small component of irradiation damage (< 50 Å diameter). Several workers have observed that these small spots do not appear to be entirely randomly distributed, and in particular often seem to fall in lines. The purpose of the present investigation is to determine the relative frequency of occurrence of such lines of spots in a foil as compared with a random distribution.

An example of the lining up of irradiation damage spots has been described by Rühle [2], who has observed in stereo 3 to 12 spots lying close together like "strings of beads" at intervals of 100 to 300 Å. He suggests that these form as a result of the reaction

$$C_u^{63} + n \rightarrow \alpha + C_0^{60} .$$

The α-particle has an energy of several MeV and could generate a sequence of 5 to 10 primary recoil atoms, resulting in the formation of a string of damage spots.

Whilst the formation of strings of spots is a very real possibility it is important to realise that an entirely random distribution of spots will also contain many such configurations. Fig. 1 shows a two-dimensional random array of 1600 spots generated by a computer, and which contains numerous examples of strings of spots of varying

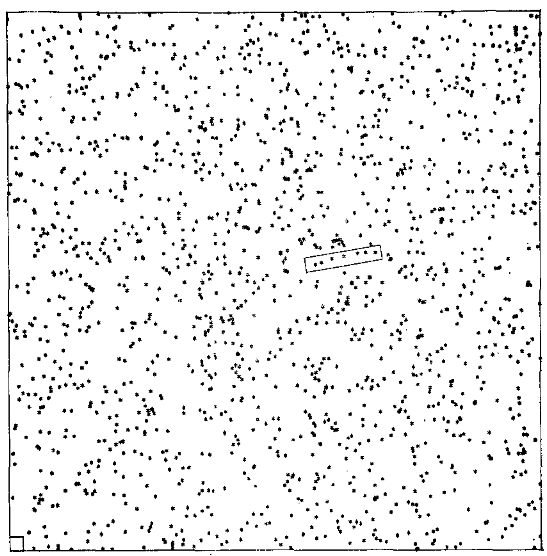


Figure 1 A random array of 1600 spots generated by a digital computer. Square box (bottom left) shows the average area per spot. A typical "string of beads" similar to that observed by Ruhle (1967) is shown.

size. It is clearly a property of a large random array that it will contain many apparently ordered groups of spots and it is essential to determine the relative frequency of occurrence of such groups. It was therefore decided to make a detailed comparison between a micrograph of a copper foil and a series of random arrays of spots of the same density.

2. Analysis of Spots into Linear Triplets

The electron micrograph shown in fig. 2 contains a typical distribution of irradiation damage and this was chosen for the analysis. It is from a copper foil 0.001 in. thick irradiated in the BEPO reactor to a total dose of 2×10^{18} neutrons/cm^2 (energy > 1 MeV) and annealed at 306° C for 251 min. The foil was electropolished in a solution of 56% orthophosphoric acid. The electron micrograph was taken on a JEOL JEM 7 high-resolution microscope and was of an area of foil approximately 1000 Å thick.

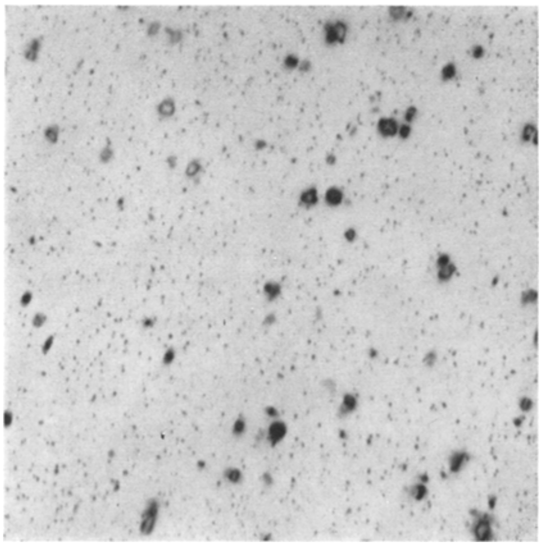


Figure 2 A copper foil irradiated to a dose of 2×10^{18} neutrons per cm^2 (energy > 1 MeV) and annealed at 306° C for 251 min.

For comparison purposes a set of random arrays of spots with the same density as the foil was prepared. The co-ordinates of the spots were determined by a pseudo-random number generator, using an IBM 7030 Stretch computer, and the arrays were plotted by a Stromberg-Carlson 4020 electronic graph plotter. A visual inspection showed a similar number of rows of spots in both the foil and random arrays. However, a high degree of personal choice was involved in counting the rows and it was considered preferable to use the computer to identify them.

The co-ordinates of 1353 fine-scale spots in the selected area of the micrograph (fig. 2) were measured and fed to the computer on punched cards. The graph plotter representation of these spots is shown in fig. 3, whilst fig. 4 shows a random array of the same density. The computer was programmed to search the distribution of spots from bottom to top for groups of three spots almost in a line, hereafter termed *linear triplets*. The criterion for linearity was that the perpendicular distance from the central spot on to the line joining the end spots should be less than 10% of the average spot spacing, i.e. similar to the size of the spots in the micrograph. In addition the angular deviation at the middle spot was required to be less than 10°, in order to minimise the number of triplets containing a pair of closely spaced spots.

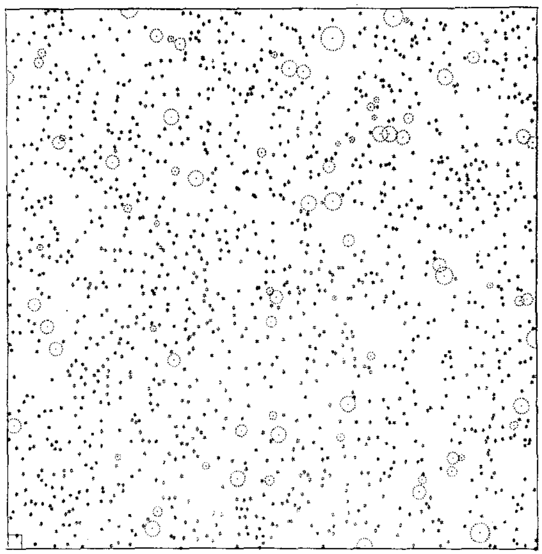


Figure 3 Graph plotter representation of the 1353 small defects observed in fig. 2. Dotted circles are an approximate representation of the areas of high strain contrast around large loops in the foil, where small spots cannot be observed.

Both the foil and a series of eight random arrays were searched for (i) a set of discrete linear triplets (no common points), and (ii) all possible linear triplets (common points allowed). The choice of discrete linear triplets in the foil is

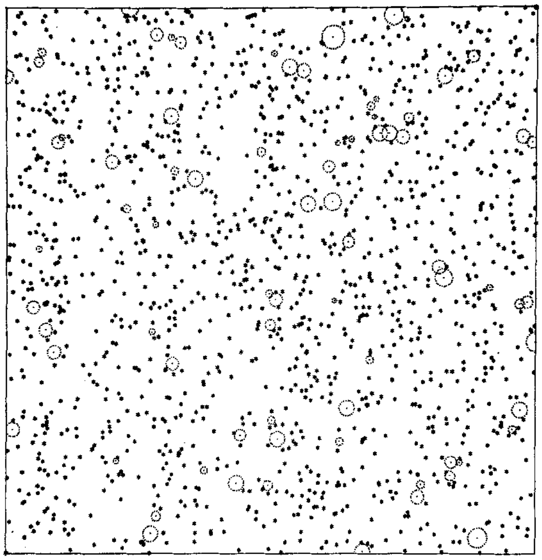


Figure 4 A random array of 1353 spots with the same density as the foil, Spots are excluded from the circular areas, which represent regions of dark contrast in the foil.

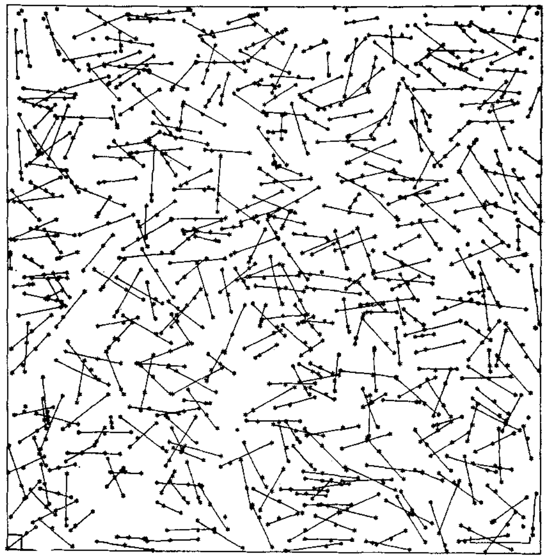
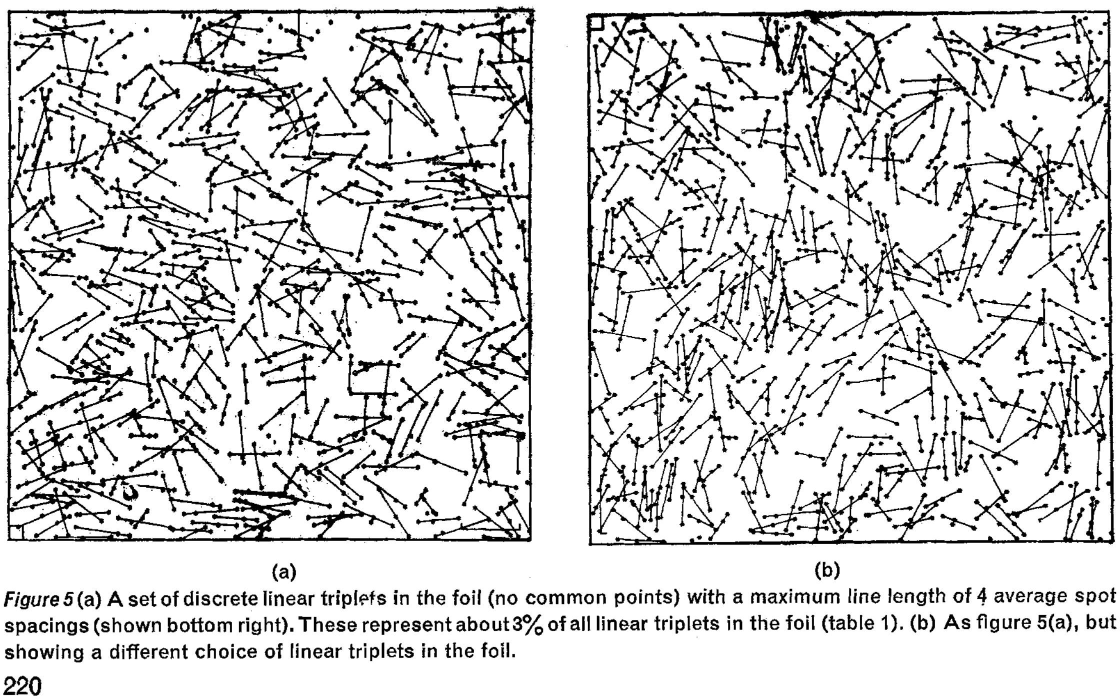


Figure 6 As fig. 5 (a), but with the random array of spots shown in fig. 4.

shown in fig. 5a; it is by no means unique and a second choice obtained by searching the foil traversely is shown in fig. 5b. Fig. 6 shows the discrete linear triplets in a typical random array (fig. 4). It will be noted that in order to remove a possible source of error the random spots have been excluded from circular areas that are approximately equivalent to areas of dark strain contrast in the foil.

Table I shows the number of linear triplets found in both the foil and random arrays, taking the maximum length to be four average spot spacings. The numbers of both discrete and non-discrete triplets have been analysed into groups of various length and the results are shown in



(a) (b)

Figure 5 (a) A set of discrete linear triplets in the foil (no common points) with a maximum line length of 4 average spot spacings (shown bottom right). These represent about 3% of all linear triplets in the foil (table 1). (b) As figure 5(a), but showing a different choice of linear triplets in the foil.

TABLE I Numbers of discrete linear triplets (values in brackets show numbers of all possible lines, allowing common points).

	No. of lines	Spots on lines	Line length (units of average spot spacing) 0 – 0.5	0.5 – 1.0	1.0 – 1.5	1.5 – 2.0	2.0 – 2.5	2.5 – 3.0	3.0 – 3.5	3.5 – 4.0
Foil	439	1317	0	3	19	38	69	102	95	113
	(11 760)	(1353)	(0)	(21)	(206)	(620)	(1327)	(2149)	(3224)	(4213)
	438	1314	0	4	26	36	82	83	100	107
	(11 760)	(1353)	(0)	(21)	(206)	(620)	(1327)	(2149)	(3224)	(4213)
Random	437	1311	0	2	21	52	64	91	93	114
	(12 162)	(1353)	(0)	(34)	(217)	(667)	(1304)	(2275)	(3168)	(4487)
	432	1296	0	2	26	40	78	99	86	101
	(12 065)	(1353)	(0)	(38)	(215)	(601)	(1263	(2180)	(3250)	(4518)
	438	1314	0	6	28	41	65	90	108	100
	(11 681)	(1353)	(0)	(55)	(243)	(639)	(1282)	(2178)	(3003)	(4281)
	437	1311	0	3	16	48	72	96	89	113
	(12 234)	(1352)	(0)	(39)	(238)	(694)	(1341)	(2152)	(3310)	(4460)
	436	1308	0	4	27	36	70	84	110	105
	(11 933)	(1353)	(0)	(32)	(227)	(639)	(1334)	(2174)	(3218)	(4309)
	439	1317	0	6	30	59	68	77	86	113
	(12 045)	(1352)	(0)	(44)	(285)	(657)	(1315)	(2177)	(3187)	(4380)
	435	1305	0	4	20	48	71	89	103	102
	(12 118)	(1353)	(0)	(41)	(229)	(677)	(1368)	(2248)	(3135)	(4420)
	439	1317	0	5	22	44	81	81	102	125
	(11 584)	(1352)	(0)	(34)	(219)	(666)	(1206)	(2076)	(3154)	(4229)

histogram form in fig. 7 and 8, which give a direct comparison between the average counts of the foil and random arrays. In all cases the

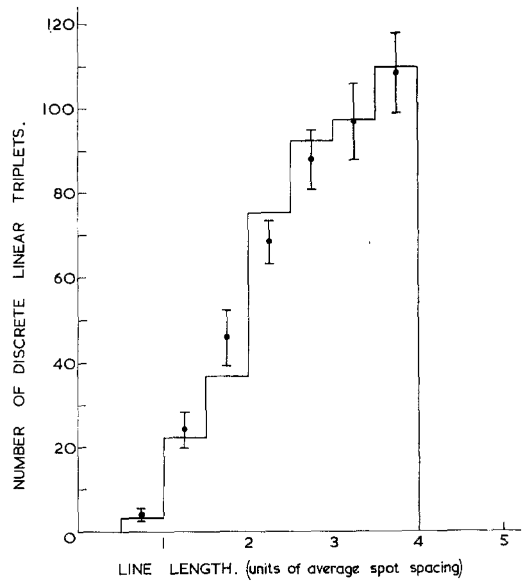


Figure 7 Histogram showing the distribution of lengths of the discrete linear triplets found in the foil (average of two choices). Solid circles show the average of the results from 8 random arrays (table 1) together with the rms deviation.

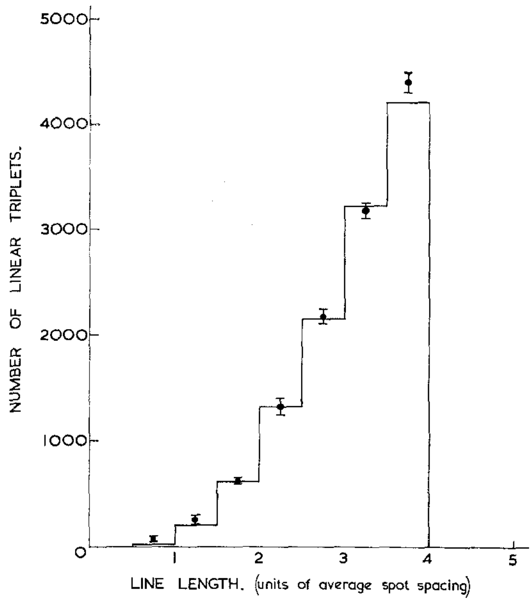


Figure 8 As fig. 7, but for all linear triplets (common points allowed).

results for the foil lie either within or close to the root mean square deviation from the mean of the random array values. Thus the number of linear triplets in the foil shows no significant deviation from that in a random array*.

*As a further test for randomness the degree of correlation between the x and y co-ordinates of the foil spots was determined and was found to be only 3%. with comparable values for the random arrays.

3. Density Distribution Function

A more general test to determine the degree of randomness of the fine-scale irradiation damage spots has been made, following the method of Ashby and Ebeling [3]. The procedure is to record the fluctuations in the local density of spots and compare with those for a random distribution. This test should readily detect any tendency of the spots to form clusters.

Two foil plots were each divided up into $28 \times 28 = 784$ identical squares and the number of spots in each square counted, omitting those squares containing large areas of strain contrast. The number of squares N containing n spots is shown in fig. 9. For a random array the probability $P(n)$ of finding n spots in a square may be shown to have a Poisson distribution.

$$P(n) = \frac{\mu^n}{n!} e^{-\mu},$$

where μ is the average number of spots per square. Fig. 9 shows the counts for the two foils and a typical random array, together with the Poisson distribution for each case.

It will be seen from fig. 9 that the deviation from a Poisson distribution for the two foils is slightly larger than, but comparable with, that for the random array. Thus the density fluctuations in the foils do not show any really significant deviation from randomness.

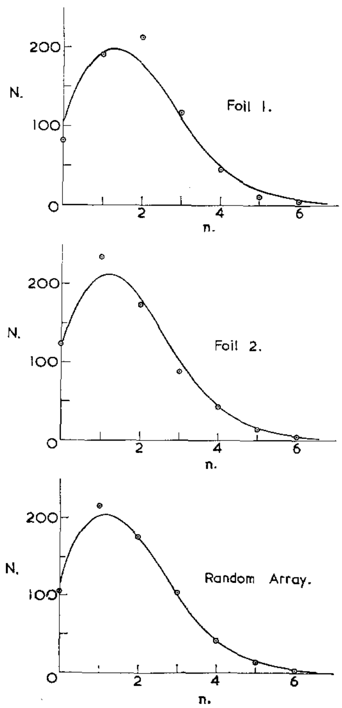


Figure 9 The density distribution function for two foils and a random array. Solid lines show the Poisson distribution curve for each case.

4. Conclusions

The computer calculation of the density of linear triplets in a foil shows no significant deviation from the results of a series of eight random arrays. The count of the density distribution function for two foils gives results close to a Poisson distribution. Also the correlation between the x and y co-ordinates of the foil spots is found to be negligible.

Thus the present results do not show any significant deviation from randomness. However, it has not been proved conclusively that the distribution of the spots is entirely random, since longer rows of spots or other groupings may have gone undetected in the present tests. In addition, the micrographs are only a two-dimensional projection of a three-dimensional array of spots. Fewer rows of spots would be found if a three-dimensional analysis were made, using stereo techniques, and it is possible that some significant effect might then be found.

It is concluded that considerable caution is needed in attaching any significance to patterns observed in micrographs of irradiation damage in metals. This is equally true for other types of defects found in various branches of metallography, e.g. precipitate and impurity particles, and any apparent "structure" should always be compared with the wide range of structural features occurring in random arrays.

Acknowledgements

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