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AVERAGE MAGNETIZATION OF Fe-Al ALLOYS

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AVERAGE MAGNETIZATION OF Fe-Al ALLOYS

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ABSTRACT

It is shown that the average magnetization of Fe-Al alloys can be well described if the iron moment is assumed to change markedly with a given number of Al first neighbours of the Fe atom. For 4 Al first neighbours the Fe moment was determined from the reported magnetization data as $\mu_A = 1.82 \pm 0.02\ \mu_B$. To account for the magnetic behaviour observed above concentrations of 25 at% Al the presence of magnetic Fe_3Al type clusters is assumed.

РЕЗЮМЕ

Показывается, что средняя намагниченность сплавов Fe-Al может хорошо описываться, если предполагается, что магнитный момент атомов железа является характерной функцией числа первых соседних атомов алюминия. На основе сообщенных данных по намагниченности был определен момент атомов железа в случае четырех первых соседних атомов алюминия: $\mu_A = 1.82 \pm 0.02\ \mu_B$. Для описания магнитных свойств, наблюдаемых при концентрациях выше 25 ат.% алюминия предполагается существование групп типа Fe_3Al.

KIVONAT

Megmutatjuk, hogy a Fe-Al ötvözetek átlagmágnesezettsége jól leírható, ha feltesszük, hogy a vasatomok mágneses momentuma az első szomszéd Al atomok számának jellegzetes függvénye. A publikált mágnesezettség adatokból meghatároztuk a vasatomok momentumát 4 Al első szomszéd esetén, melyre $\mu_A = 1.82 \pm 0.02\ \mu_B$ adódott. A 25 at% Al feletti koncentrációknál megfigyelt mágneses viselkedés leirására mágneses Fe_3Al tipus clusterek létezését tettük fel.

INTRODUCTION

Fe-Al alloys have been observed to exhibit up to 54 at% Al concentrations three different structures of their b.c.c. lattice, namely, disorder from 0 to 18 at% Al, Fe_3Al /or DO_3/-type order from 18 to 38 at% Al and FeAl /or B2/ -type order above 38 at% Al. The b.c.c. lattice of the alloy comprises four f.c.c. sublattices, the sites of which are occupied by the different atoms as shown in Fig. 1 for Fe_3Al and FeAl order.

Fig. 2 shows the average magnetization data, as extrapolated to T = 0 ^{o}K [1,2,3,4]. For the explanation of the anomalous decrease in the average magnetization at about 30 at% Al observed by susceptibility measurement, the presence of an antiferromagnetic FeAl phase [1] or the presence of a finely dispersed ferromagnetic Fe_3Al phase with superparamagnetic behaviour in this antiferromagnetic FeAl phase [4,5] have been suggested. However, the diffuse X-ray scattering data /Fig. 3/ [6] and Mössbauer spectra [5,7,8] indicate a homogeneous transition. Neither the coexistence of two phases [5] nor the existence of a long range antiferromagnetic order in the FeAl phase [9] could be confirmed by experiment.

In contrast with the average magnetization [1,4] and neutron diffraction [9] measurements which do not show the existence of any long range magnetic order in the alloys with about 50 at% Al, it is apparent from Mössbauer spectroscopy [5,7] that only the Fe atoms at B- and D-type sites have a magnetic moment which can produce a short range order. This nearly ferromagnetic behaviour is confirmed also by the superparamagnetic specific heat anomaly observed at 48,8 at% Al [10]. If the concentration of iron atoms increases, their number at sites A with 4 iron neighbours at sites D statistically also increases and Fe_3Al type clusters can form which have sufficiently high moment to initiate a macroscopic magnetic ordering. However, at this stage the interaction between the clusters is still weak and that explains the observed anomalies of superparamagnetic nature [4].

The aim of the present work is to show that the changes in the average magnetization measured on Fe-Al alloys up to 50 at% Al can be accounted for by taking into consideration the changes in the occupation of the sublattice sites by using Beck's assumption [7, 11].

METHOD OF CALCULATION

The relation between the magnetic moment of the Fe atoms and the number i of first neighbour Al atoms is approximated as

$$\mu_{Fe}(i) = \begin{cases} \mu_D & \\ \mu_A & \text{if} \quad i = 4 \\ 0 & \end{cases} \qquad \begin{matrix} i = 0, 1, 2, 3 \\ \\ i = 5, 6, 7, 8 \end{matrix} \qquad /1/$$

where $\mu_D = 2.21\ \mu_B$ is the moment of pure Fe, while μ_A was evaluated from the reported average magnetization data [1, 2, 3, 4] by the least-square method as $\mu_A = 1.82 \pm 0{,}02\ \mu_B$.

We take p_A, p_B, p_C and p_D to be the probabilities that the sites of type A, B, C and D, respectively are occupied by Fe atoms. It is known from experimental evidence that the sites A and C are equivalent, i.e. $p_A = p_C$ and obviously

$$2\,p_A + p_B + p_C = 4\,(1 - x) \qquad /2/$$

where x stands for the Al concentration.

We define now the order parameters α and β as

$$p_A = 1 - x + \alpha x \qquad /3a/$$

$$p_D = 1 - x + \beta x \qquad /3b/$$

and from /2/ we have

$$p_B = 1 - x - (2\alpha + \beta)x \qquad /3c/$$

Considering the occupation of the first neighbour sites we find that

sites B and D are surrounded by 8 A sites and the probability that n of these are occupied by Fe atoms is

$$p_8(n, p_A) = \binom{8}{n} p_A^n\,(1-p_A)^{8-n} \qquad /4/$$

sites A are surrounded by 4 D and 4 B type first neighbours. The probability that k of the D sites and l of the B sites are occupied by Fe can be expressed as

$$p_4(k, p_D) \cdot p_4(\ell, p_B) = \binom{4}{k} p_D^k (1 - p_D)^{4-k} \binom{4}{\ell} p_B^\ell (1-p_B)^{4-\ell} \quad /5/$$

The average magnetization per atom of the alloy is given by

$$\bar{\mu} = n_A \bar{\mu}_A + n_B \bar{\mu}_B + n_D \bar{\mu}_D \quad /6/$$

where $n_{A,B,D}$ stand for the relative numbers of iron atoms at the given type of sites and obviously:

$$n_A = 0.5 \cdot p_A, \qquad n_B = 0.25 \cdot p_B, \qquad n_D = 0.25 \cdot p_D$$

and $\bar{\mu}_{A,B,D}$ stand for the average magnetic moments at the given type of sites, thus

$$\bar{\mu}_A = \sum_{k=0}^{4} \sum_{\ell=0}^{4} p_4(k, p_D)\, p_4(\ell, p_B)\, \mu_{Fe}(k+\ell) \quad /7/$$

$$\bar{\mu}_B = \bar{\mu}_D = \sum_{n=0}^{8} p_8(n, p_A)\, \mu_{Fe}(n) = \sum_{n=5}^{8} p_8(n, p_A)\, \mu_D + p_8(4, p_A)\, \mu_A \quad /8/$$

On substitution into /6/ we get

$$\bar{\mu} = 0{,}5(1-x)\,(\bar{\mu}_A + \bar{\mu}_D) - 0.5\alpha x\,(\bar{\mu}_D - \bar{\mu}_A) \quad /9/$$

Let us look at some special cases.

1/ Complete disorder /α-phase/

$\alpha = \beta = 1$, thus $p_A = p_B = p_D = 1-x$ and therefore $\bar{\mu}_A = \bar{\mu}_D$

and

$$\bar{\mu} = (1 - x)\, \bar{\mu}_D.$$

2/ Perfect Fe_3Al-type order

The excess or deficient Al atoms due to the deviation from stoichiometry are responsible for the changes in the occupation of the D and B-type sites.

Two cases have to be distinguished .

a/ $x \leq 0.25$

Then $\alpha = \beta = 1$, thus $p_A = p_D$ and $p_B = 1 - 4x$. By /7/ we have $\mu_D = \mu_D$ and by /8/ $\mu_A = \mu_D - (4x)^4 (\mu_D - \mu_A)$ thus from /9/

$$\mu = (1 - x)\ \mu_D - 0.5(4x)^4 (\mu_D - \mu_A).$$

b/ $x \geq 0.25$

Then $p_A = 1$, $p_B = 0$ and by /2/ $p_D = 2 - 4x$. By /7/ and by /8/ $\bar{\mu}_A = (2 - 4x)^4\ \mu_A$, thus

$$\bar{\mu} = (0.5 - x)\ \mu_D + 0.5\ (2 - 4x)^4\ \mu_A.$$

3/ Perfect FeAl-type order

Then $p_A = 1$, $p_B = p_D = 1 - 2x$, $\bar{\mu}_B = \bar{\mu}_D = \mu_D$ and

$$\bar{\mu}_A = \sum_{n=5}^{8} p_8(n, p_D)\ \mu_D + p_8(4, p_D)\ \mu_A\ ,\quad \text{thus}$$

$$\bar{\mu} = (0.5 - x)\mu_D + 0.5\ \bar{\mu}_A.$$

DISCUSSION

The solid line in Fig. 2 shows the average magnetization, as obtained from equation /9/ for the order parameters evaluated from the diffuse X-ray scattering data given in Fig. 3. The calculated values are not significantly sensitive to the order parameters, a 10 % change of the latter induces not more than about 2 % change in the former. The agreement with the measured values which is satisfactory up to 25 at%. Al becomes gradually worse as the Al concentration increases.

The difference between the predicted and measured values in the alloys with more than 25 at% Al can be explained by the increasing disorder of the Fe_3Al phase as ever more D sites are occupied at random by Al atoms and only the still remaining Fe_3Al type clusters can contribute to the average magnetization of the alloy in the measure that

$$\bar{\mu} = n_{Cl}\ \bar{\mu}_{Cl}\ , \qquad /10/$$

where n_{Cl} is the relative number of Fe_3Al type clusters, thus

$$n_{Cl} = 0.5 \cdot p,$$

where p is the probability that a Fe atom in an A-site has 4 Fe neighbours in D-sites, which is given as

$$p = (2 - 4x)^4.$$

The average moment of such a "magnetic cluster" is

$$\bar{\mu}_{Cl} = \mu_A + \bar{\mu}_D,$$

where μ_D is the average moment of Fe atoms at D-sites, i.e.

$$\bar{\mu}_D = (1 - 2x)\mu_D.$$

On substitution into eq./10/ the average magnetization produced by the randomly distributed Fe_3Al type clusters is given as

$$\bar{\mu} = 0.5\ (2 - 4x)^4 \left(\mu_A + (1 - 2x)\mu_D\right) \qquad /11/$$

The values calculated from eq./11/ are shown in Fig. 2 by the broken line which gives a good agreement with the experimental data even in the critical range of concentrations.

The Fe moment μ_A, for 4 Al first neighbours, as evaluated from the reported average magnetization data is in good agreement with $\mu_A = 1.8 \pm 0.1\ \mu_B$ obtained from Mössbauer data by using the expression

$$H_{Fe}(3) - H_{Fe}(4) = a\ (\mu_D - \mu_A) + \Delta H,$$

where $H_{Fe}(3) = 261$ kG [7] and $H_{Fe}(4) = 210$ kG [12] are the iron hyperfine fields measured at room temperature on alloys of about 25 at% Al for 3 and 4 Al first neighbours respectively; $a(\mu_D - \mu_A)$ is the change of the core polarization contribution, where $a = 65\ kG/\mu_B$ is the core polarization constant* and $\Delta H = 23$ kG [e.g. 7] is the change in the conduction electron contribution due to the replacement of a Fe atom at D site by an Al atom.

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*$a = 65\ kG/\mu_B$ was obtained from the hyperfine field of 145 kG corresponding to Fe atoms without Al first neighbours measured at about 50 at% Al at 4^oK [8, 13], where the presence of a paramagnetic line shows that the conduction electron contribution can be ignored.

The difference from $\mu_A = 1.5 \pm 0.1\ \mu_B$ that was determined from neutron diffraction on a Fe_3Al specimen [9] can be explained by either a slight deviation from stoichiometry or non-perfect order of the Fe_3Al specimen.

It is expected that the similar anomalous magnetic behaviour of the Fe-Si system can be explained in the same way.

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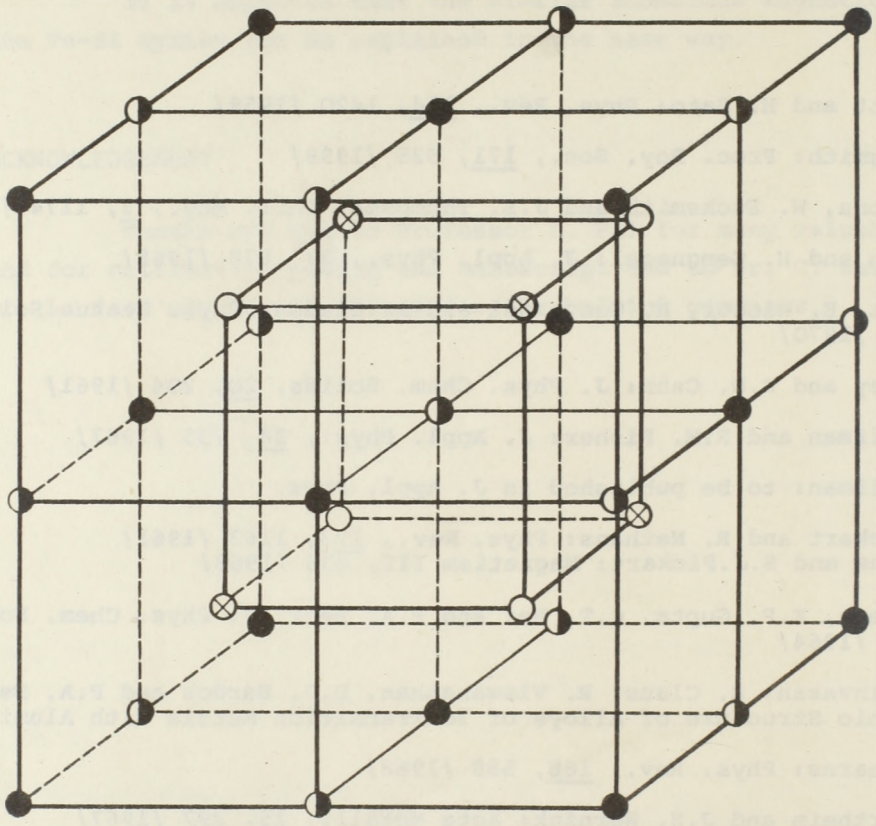


●A ◑C ○B ⊗D

Fe_3Al : ● ◑ ⊗ Fe ○ Al

FeAl : ● ◑ Fe ○ ⊗ Al

Fig. 1

Site designations for Fe_3Al and FeAl type superlattices

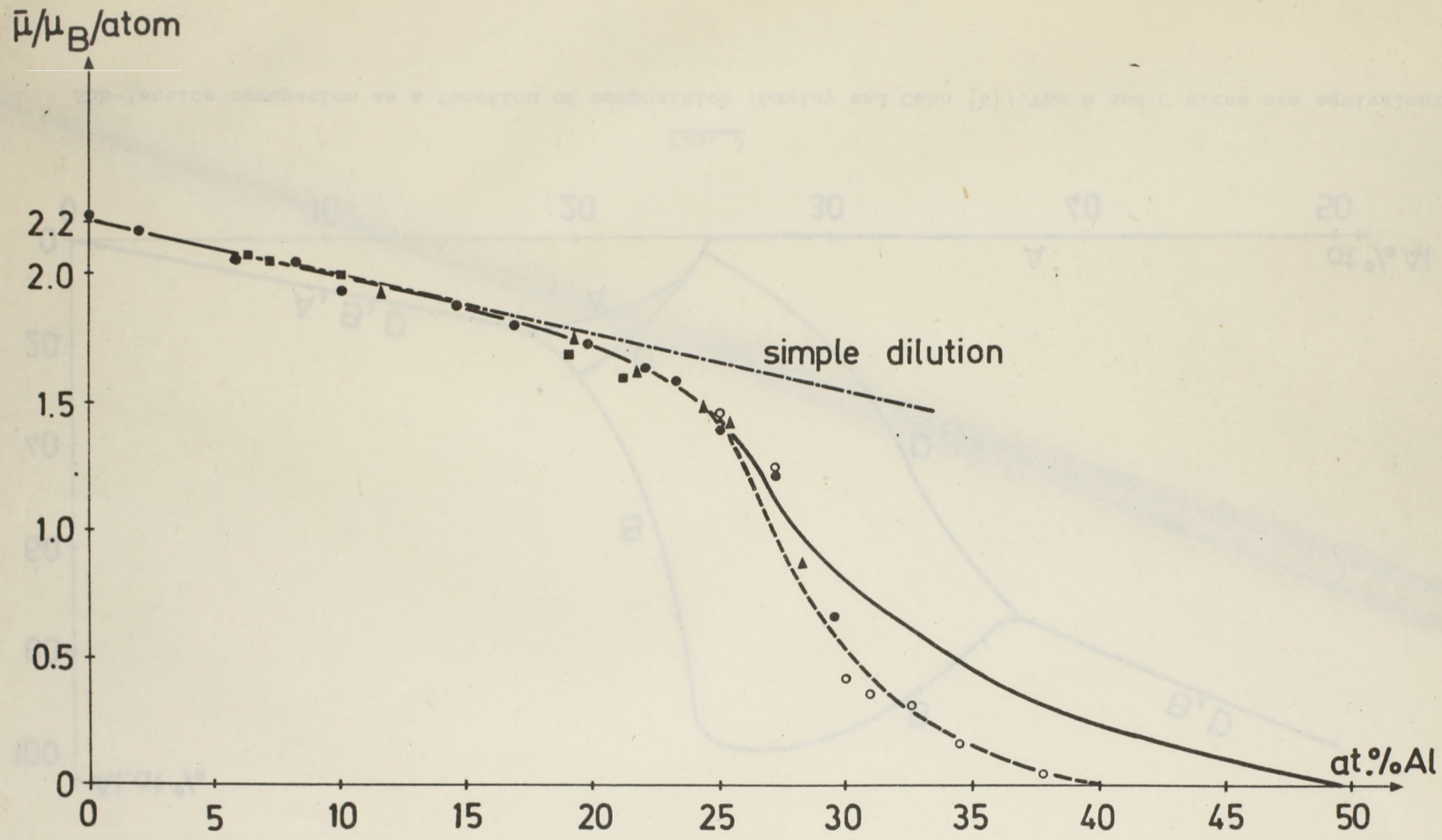


Fig. 2

Average magnetization of Fe-Al alloys as a function of concentration as extrapolated to $0^{\circ}K$.

▲ Arrott and Sato [1] ● Sucksmith [2] ■ Parsons et.al [3] o Danan and Gengnagel [4]

———— eq. /9/ -------- eq. /11/

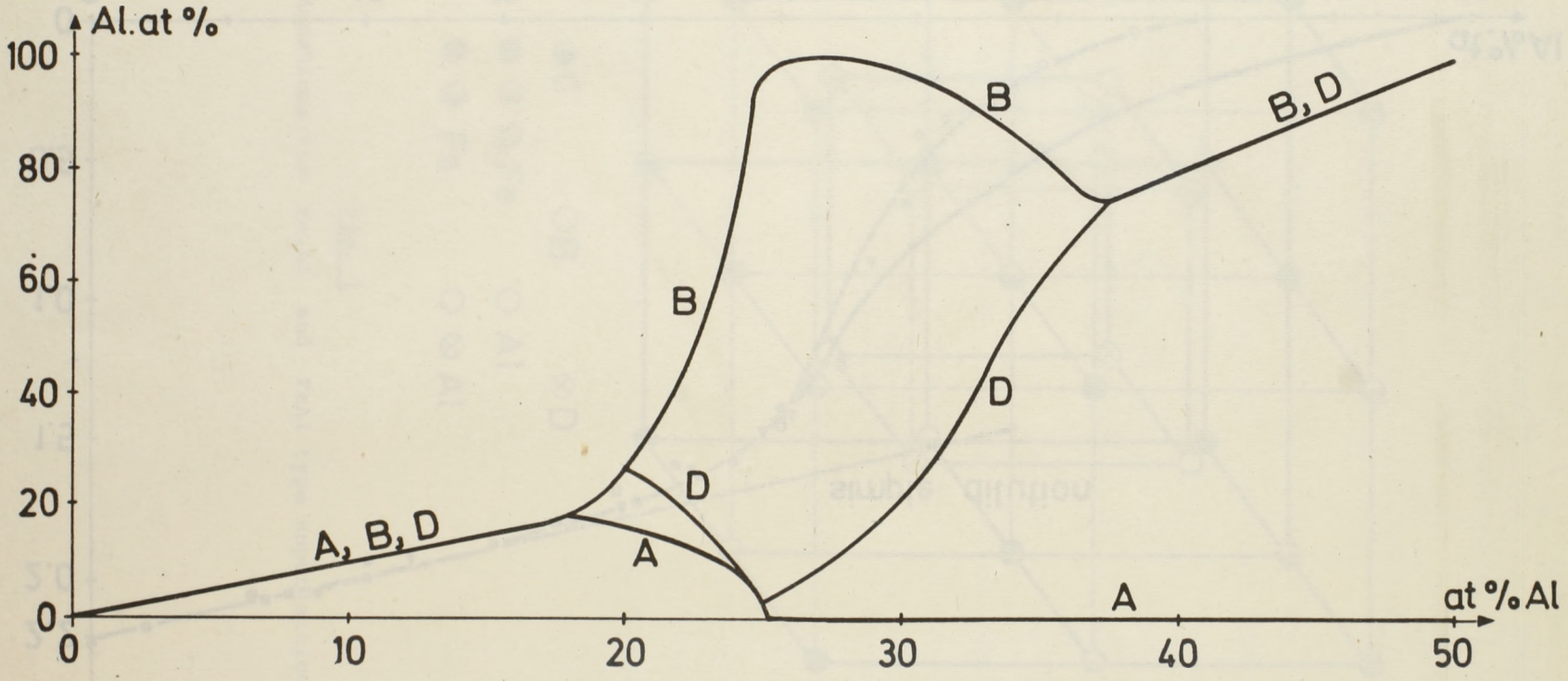
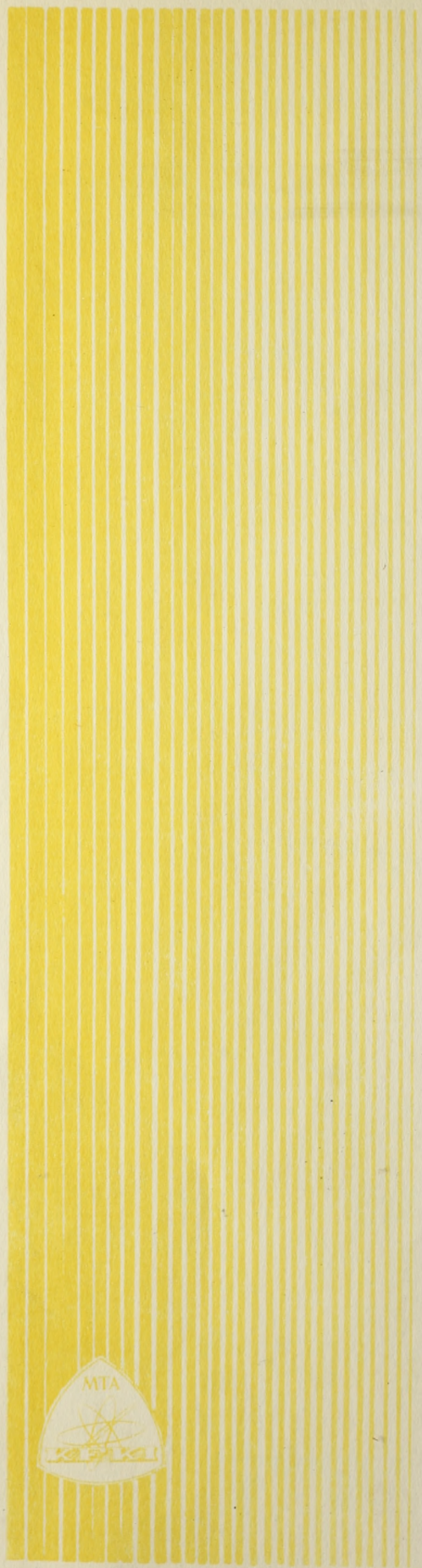


Fig. 3

Sub-lattice occupation as a function of composition /Lawley and Cahn [6]/.The A and C sites are equivalent.

61.304



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