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Argonne National Laboratory

THE USE OF NUCLEAR NULL-MATRIX IN NEUTRON-DIFFRACTION STUDIES

by

S. S. Sidhu, LeRoy Heaton,
M. H. Mueller, F. P. Campos,
K. D. Anderson, and D. D. Zaubers

RELEASED FOR ANNOUNCEMENT
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ABSTRACT

Most nuclei scatter thermal neutrons with a 180° phase change, but nine are known to scatter neutrons with a zero-degree phase change. These properties were utilized in preparing a number of nuclear null-matrices that are substitutional solid solutions or stoichiometric alloys of different elements or isotopes of the same element with close-packed hexagonal or face-centered cubic structures. The atomic percentages and the coherent nuclear-scattering amplitudes of the randomly distributed atoms of the matrix are such that the resultant scattering amplitude is zero. Consequently, the structure factor is zero and no diffraction peaks appear in the neutron pattern of the null-matrix. The compounds of the null-matrix material with light atoms, crystallizing in CaF_2 -type, NaCl -type, and Mn_4N -type structures, were investigated to determine unambiguously the positions of light atoms among heavy atoms. The chemical and/or magnetic order-disorder phenomena were studied in both the null-matrix alloys and their compounds.

I. INTRODUCTION

The availability of thermal-neutron beams in sufficient intensity and the coherent nuclear-scattering amplitudes¹⁻¹⁶ for some 132 nuclei provide powerful tools for the study of solids as well as liquids. The scattering amplitudes possess certain unique properties that lend extremely well to crystal-structure determinations. As shown in the appendix, the amplitudes of the light and of the heavy nuclei have comparable magnitudes. The values of some of them, in fact, are practically the same. Nine elements and isotopes: H, Li, Li^7 , Ti, Ti^{48} , V^{51} , Mn^{55} , Ni^{62} , and Sm^{152} , have negative scattering amplitudes and scatter thermal neutrons with zero-degree phase change; the positive sign of a scattering amplitude is conventionally attributed to a nucleus that scatters thermal neutrons with 180° phase change. Hydrogen, lithium, titanium, nickel, and samarium have two or more isotopes,

at least one of which has a negative scattering amplitude. The short-range nuclear interaction of the neutron with the atomic nucleus, in contrast to the interaction of the magnetic moment of the neutron with the spin and orbital magnetic momenta of the atom, produces isotropic scattering. As a result, there is no decrease in the scattering amplitude with the scattering angle, as is the case in X-ray scattering.

The fact that coherent nuclear-scattering amplitudes for both the light and the heavy atoms for thermal neutrons are of the same magnitude makes it possible to determine structures of substances containing light atoms, such as hydrides, deuterides, carbides, nitrides, and oxides, by neutron-diffraction techniques. The use of a nuclear null-matrix may aid in the unique determination of the positions of light atoms among heavy atoms and in the study of the chemical and magnetic ordering in solids. A null-matrix is a random occupation of equivalent lattice sites by different elements or different isotopes of the same element whose scattering amplitudes are of different sign. The atomic percentages and the scattering amplitudes of the randomly distributed atoms in a null-matrix are such that the resultant scattering amplitude

$$b_n = p_A b_A + p_B b_B \quad (1)$$

is 0, where p_A and p_B are the fractions of all sites occupied by A- and B-type atoms, respectively, and b_A and b_B are their coherent scattering amplitudes, of which one is negative. Consequently, a neutron pattern of a null-matrix would show no coherent diffraction peaks. An X-ray diffraction pattern of the same null-matrix, on the other hand, would show regular characteristic diffraction peaks and would be similar to that of a disordered alloy.

The diffraction patterns of the compounds formed by the null-matrix alloys with light atoms such as hydrogen, deuterium, lithium, etc., would show some unique features. If the null-matrix atoms remained randomly distributed in the crystal lattice of the compound as they are in the lattice of the null-matrix alloy, the diffraction peaks in the neutron pattern would then result from the lattice of the light atoms only, whereas an X-ray diffraction pattern of the same compound would essentially be of the lattice of null-matrix atoms. Thus, the null-matrix technique offers unique advantages, by means of which it is possible to obtain diffraction patterns of the crystal lattice of light atoms and of heavy atoms in the same compound separately.

Several null-matrices and their compounds were prepared from nuclei with known scattering amplitudes, the null-matrices with close-packed hexagonal and face-centered cubic structures, and the compounds with CaF_2 -type, NaCl -type, and Mn_4N -type structures. The null-matrices were studied for chemical and/or magnetic ordering, and the compounds for both the unambiguous determination of the positions of light atoms

among heavy atoms, and chemical and/or magnetic ordering. Some of the results reported in detail previously have been modified or treated differently and are included here in abbreviated form in the interest of completeness.

II. CLOSE-PACKED HEXAGONAL NULL-MATRIX .

Titanium and zirconium metals crystallize in a close-packed hexagonal structure and form a continuous series of substitutional solid solutions. At a composition of approximately 67 a/o titanium and 33 a/o zirconium, these metals form a null-matrix for which the resultant scattering amplitude is

$$\begin{aligned}
 b_n &= p_{\text{Ti}} b_{\text{Ti}} + p_{\text{Zr}} b_{\text{Zr}} \\
 &= 0.67(-0.34 \times 10^{-12} \text{ cm}) + 0.33(0.70 \times 10^{-12} \text{ cm}) \\
 &\approx 0.
 \end{aligned} \tag{2}$$

Consequently, $|F_{hkl}|^2$ and I_{hkl} are zero for all hkl planes. As shown in Fig. 1, no diffraction peaks are observed in the neutron-diffraction pattern

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Fig. 1. Neutron-diffraction Patterns of Zirconium, 68a/oTi-32a/oZr Alloy, and Titanium; $\lambda = 1.07 \text{ \AA}$

of the null-matrix ($M_{\text{Ti-Zr}} = \text{Ti}_{0.68}\text{Zr}_{0.32}$), although the elements titanium and zirconium give usual neutron patterns.

A. Investigation of Chemical Order-Disorder in Titanium-Zirconium Hexagonal Null-matrix

There are two atoms per unit cell in the titanium-zirconium hexagonal null-matrix. If these atoms occupied ordered positions, such as one Ti at 0 0 0, and statistically 0.34 Ti and 0.66 Zr at $1/2 \ 2/3 \ 1/2$, the structure-factor expression for these positions would be

$$|F_{hkl}| = b_{\text{Ti}} + (0.34b_{\text{Ti}} + 0.66b_{\text{Zr}}) \exp \left[2\pi i \left(\frac{h+2k}{3} + \frac{\ell}{2} \right) \right]. \quad (3)$$

Since the numerical value of b_{Ti} is negative, the structure factors for Bragg reflections whose Miller indices make the second term in Equation (3) positive would be zero. This follows from the fact that, in general, in a hexagonal null-matrix of the titanium-zirconium type, the atomic fractions of titanium and zirconium atoms, p_{Ti} and p_{Zr} , respectively, in the unit cell satisfy the condition

$$2(p_{\text{Ti}} + p_{\text{Zr}}) = 2. \quad (4)$$

When $p_{\text{Ti}} > 0.5$ and one Ti is at 0 0 0, then statistically

$$2[(p_{\text{Ti}} - 0.5) + p_{\text{Zr}}]$$

is at $1/3 \ 2/3 \ 1/2$, and the structure factor becomes

$$|F_{hkl}| = b_{\text{Ti}} + 2 \left[(p_{\text{Ti}} - 0.5) b_{\text{Ti}} + p_{\text{Zr}} b_{\text{Zr}} \right] \exp \left[2\pi i \left(\frac{h+2k}{3} + \frac{\ell}{2} \right) \right], \quad (5)$$

and when

$$\exp 2\pi i \left(\frac{h+2k}{3} + \frac{\ell}{2} \right) = 1,$$

$$|F_{hkl}| = 2(p_{\text{Ti}} b_{\text{Ti}} + p_{\text{Zr}} b_{\text{Zr}})$$

$$= 0.$$

The Bragg reflections and the corresponding structure-factor squared values for this ordered arrangement are given in Table I. These may be summarized as follows:

	$ F_{hkl} ^2 \times 10^{24}, \text{cm}^2$
(1) $\ell = \text{even and } h + 2k = 3n$	0
(2) $\ell = \text{odd and } h + 2k = 3n$	0.471145
(3) $\ell = \text{even and } h + 2k = 3n \pm 1$	0.353269
(4) $\ell = \text{odd and } h + 2k = 3n \pm 1$	0.117817

where $n = 0, 1, 2, 3, \dots$, and $b_n = 0.3464 \times 10^{-12} \text{ cm}$.

TABLE I. Neutron Structure Factors Squared for Reflections for Ordered Titanium-Zirconium Null-matrix Alloy

hkl	$ F_{hkl} \times 10^{12}, \text{cm}$	$ F_{hkl} ^2 \times 10^{24}, \text{cm}^2$	hkl	$ F_{hkl} \times 10^{12}, \text{cm}$	$ F_{hkl} ^2 \times 10^{24}, \text{cm}^2$
001	$b_{\text{Ti}} + (b_n) e^{\pi i}$	0.471145	201	$b_{\text{Ti}} + (b_n) e^{\pi i/3}$	0.117817
100	$b_{\text{Ti}} + (b_n) e^{2\pi i/3}$	0.353269	004	$b_{\text{Ti}} + (b_n) e^{2\pi i}$	0
002	$b_{\text{Ti}} + (b_n) e^{2\pi i}$	0	202	$b_{\text{Ti}} + (b_n) e^{4\pi i/3}$	0.353269
101	$b_{\text{Ti}} + (b_n) e^{5\pi i/3}$	0.117817	104	$b_{\text{Ti}} + (b_n) e^{2\pi i/3}$	0.353269
102	$b_{\text{Ti}} + (b_n) e^{2\pi i/3}$	0.353269	113	$b_{\text{Ti}} + (b_n) e^{\pi i}$	0.471145
003	$b_{\text{Ti}} + (b_n) e^{\pi i}$	0.471145	203	$b_{\text{Ti}} + (b_n) e^{\pi i/3}$	0.117817
110	$b_{\text{Ti}} + (b_n) e^{2\pi i}$	0	005	$b_{\text{Ti}} + (b_n) e^{\pi i}$	0.471145
111	$b_{\text{Ti}} + (b_n) e^{\pi i}$	0.471145	210	$b_{\text{Ti}} + (b_n) e^{2\pi i/3}$	0.353269
103	$b_{\text{Ti}} + (b_n) e^{5\pi i/3}$	0.117817	211	$b_{\text{Ti}} + (b_n) e^{5\pi i/3}$	0.117817
200	$b_{\text{Ti}} + (b_n) e^{4\pi i/3}$	0.353269	114	$b_{\text{Ti}} + (b_n) e^{2\pi i}$	0
112	$b_{\text{Ti}} + (b_n) e^{2\pi i}$	0			

The characteristic feature of the ordering in titanium-zirconium-type null-matrix material would be that the reflections for which $\ell = \text{even}$ and $h + 2k = 3$; that is, (002), (110), (112), (004), (114), etc., would not appear. Since no reflections appear in the null-matrix pattern, this would mean that the above reflections would not appear whether the material is ordered or disordered.

In contrast, if the same null-matrix were studied by X-ray-diffraction method, then, as shown in Table II, the reflections for which $\ell = \text{odd}$ and $h + 2k = 3n \pm 1$, that is, (001), (003), (111), (113), (005), etc., would not appear in the X-ray-diffraction pattern of the null-matrix, but

all reflections would appear in the pattern of the ordered alloy material. It is interesting to note that the X-ray structure-factor squared values for all reflections in the ordered material are higher than those for the null-matrix, except for those reflections that do not appear in the neutron patterns at all. These values remain the same in both the disordered and the ordered states.

TABLE II. X-ray Structure Factors Squared for Disordered and Ordered Titanium-Zirconium Null-matrix

hkl	$ F_{hkl} ^2$		hkl	$ F_{hkl} ^2$	
	Disordered	Ordered		Disordered	Ordered
001	0	134.17	112	975.31	975.31
100	478.08	564.43	201	731.48	733.15
002	1800.30	1800.30	004	894.61	894.61
101	1335.00	1359.89	202	188.65	271.31
102	346.63	417.85	104	196.93	253.41
003	0	113.17	113	0	74.18
110	1213.83	1213.83	203	543.31	561.24
111	0	113.78	005	0	66.44
103	785.85	807.81	211	513.74	530.77
200	249.42	311.45	114	654.59	654.85

To determine experimentally if chemical ordering could be produced in this null-matrix alloy, a sample composed of 67.6 a/o titanium and 32.4 a/o zirconium was annealed in the β region at 900°C for 2 hr, then cooled and held isothermally for 2 hr at 800, 700, 600, and 500°C in the β , $\beta + \alpha$, and α regions, and finally cooled to room temperature in approximately 4 hr. Both neutron- and X-ray-diffraction patterns were made at room temperature, but no ordering of atoms was observed in either pattern.

B. Nonstoichiometric Metal Hydrides of Titanium-Zirconium Null-matrix (CaF₂-type)

Titanium-zirconium null-matrix was used in the study of nonstoichiometric metal hydrides (metal deuterides) for the purpose of locating deuterium atoms uniquely in the crystal structures of these compounds. While the crystal structures of titanium, zirconium, and the null-matrix alloy are hexagonal, their dideuterides, TiD_{1.97} (Ref. 17) (below room temperature), ZrD_{1.98} (Ref. 18), and MD_{1.99} (Ref. 19) (at room temperature) crystallize in distorted CaF₂-type or tetragonal structures with axial ratios less than unity. There are four molecules per unit cell of the dideuteride with the following atom sites:

4 M at (0 0 0, 1/2 1/2 0, 1/2 0 1/2, 0 1/2 1/2)

8 D at $\pm(1/4 1/4 1/4, 3/4 3/4 1/4, 3/4 1/4 3/4, 1/4 3/4 3/4)$.

The structure-factor expression is

$$F_{hkl} = \left[1 + e^{\pi i(h+k)} + e^{\pi i(h+l)} + e^{\pi i(k+l)} \right] \left[b_M + C b_D \cos \frac{\pi}{2}(h+k+l) \right], \quad (6)$$

in which b_M is the resultant nuclear-scattering amplitude for metal atoms, b_D is for deuterium atoms, and C is the mole fraction of deuterium in a given deuteride. For reflections with all odd or all even indices, the expression reduces to

$$F_{hkl} = 4 \left[b_M + C b_D \cos \frac{\pi}{2}(h+k+l) \right]. \quad (7)$$

If the metal atoms in the dideuteride lattice are distributed at random as in the null-matrix, then b_M will be zero, and the structure factor for the deuteride becomes

$$F_{hkl} = 4 C b_D \cos \frac{\pi}{2}(h+k+l),$$

and

$$|F_{hkl}| = 0, \text{ when } (h+k+l) \text{ is odd}, \quad (8)$$

or

$$= 4 C b_D, \text{ when } (h+k+l) \text{ is even}.$$

The reflections observed in the neutron-diffraction pattern of $MD_{1.99}$, as shown in Fig. 2, are from the lattice of deuterium atoms only. Thus, these results confirm two facts unambiguously: (1) Deuterium atoms in the metal deuteride structures form a lattice of their own and occupy definite positions with respect to metal atoms. (2) The null-matrix atoms occupy random sites in the distorted CaF_2 -type structure.

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Fig. 2. Neutron-diffraction Pattern and Unit Cell of $MD_{1.99}$; $\lambda = 1.07 \text{ \AA}$

Another interesting feature of this study was that the distorted CaF_2 -type structures of $\text{TiD}_{1.97}$ and $\text{ZrD}_{1.98}$ could be changed to the cubic CaF_2 -type by raising the temperature of $\text{TiD}_{1.97}$ from below room temperature to room temperature and by changing the composition of $\text{ZrD}_{1.98}$ to $\text{ZrD}_{1.58}$. In contrast, however, as the concentration of deuterium in $\text{MD}_{1.99}$ was decreased to $\text{MD}_{1.2}$, the tetragonal structure of $\text{MD}_{1.99}$ did change but not to a CaF_2 -type cubic. Instead it formed a new phase with an entirely different structure in which the null-matrix atoms took ordered positions. When the concentration of deuterium atoms was again increased to dideuteride composition, the tetragonal structure was restored and the null-matrix atoms occupied random sites in the lattice. Thus, it was possible to change not only the structure of a deuteride phase by varying the concentration of deuterium atoms in the system, but also to produce order-disorder in the null-matrix atoms.

C. Carbide of Titanium-Zirconium Null-matrix (NaCl-type)

The use of the titanium-zirconium null-matrix was extended to a study of the carbide of the null-matrix for the purpose of locating carbon atoms among relatively heavy metal atoms uniquely. The null-matrix carbide, MC, like TiC and ZrC, crystallizes in a NaCl-type structure with the metal and the carbon atoms occupying the following positions:

$$\begin{aligned} 4 \text{ M at } & 0 \ 0 \ 0, \ 1/2 \ 1/2 \ 0, \ 1/2 \ 0 \ 1/2, \ 0 \ 1/2 \ 1/2, \\ 4 \text{ C at } & 1/2 \ 1/2 \ 1/2, \ 0 \ 0 \ 1/2, \ 0 \ 1/2 \ 0, \ 1/2 \ 0 \ 0; \end{aligned}$$

and the structure factor for the unit cell is

$$\begin{aligned} |F_{hkl}| = & b_M \left[1 + e^{\pi i(h+k)} + e^{\pi i(h+l)} + e^{\pi i(k+l)} \right] \\ & + b_C \left[e^{\pi i(h+k+l)} + e^{\pi i l} + e^{\pi i k} + e^{\pi i h} \right]. \end{aligned} \quad (9)$$

Since the metal atoms form a null-matrix, $b_M = 0$, and the structure factor is

$$\begin{aligned} |F_{hkl}| &= 4b_C; \text{ for } hkl \text{ all odd or all even} \\ &= 0, \text{ for mixed indices.} \end{aligned} \quad (10)$$

A neutron-diffraction pattern of MC and the unit cell derived from it are shown in Fig. 3. The calculated and the observed relative intensities of the diffraction peaks are given in Table III. It is apparent that these intensities result from coherent scattering by the carbon atoms only. The use of the nuclear null-matrix, therefore, makes it possible to determine positions of carbon atoms in a given NaCl-type structure uniquely.

Fig. 3. Neutron-diffraction Pattern and Unit Cell of MC;
 $\lambda = 1.07 \text{ \AA}$

TABLE III. Neutron-diffraction Relative Intensities for MC

hkl	I/I_0 (calc)	I/I_0 (obs)	hkl	I/I_0 (calc)	I/I_0 (obs)
111	100.0	100.0	400	15.7	16.2
200	56.7	56.4	331	54.4	52.0
220	58.6	56.8	420	52.2	46.6
311	87.3	81.4	422	45.4	35.7
222	26.9	26.7	333, 511	55.5	40.6

$R = \sum |I_0 - I_c| / \sum I_0 = 0.043$. Values of I/I_0 (obs) were not corrected for temperature factor.

D. Order-Disorder of Metal Atoms in a NaCl-type MC Structure

If the four null-matrix atoms in the MC structure occupied ordered positions, such as 2 Ti at $0\ 0\ 0$, $1/2\ 1/2\ 0$, 1 Zr at $1/2\ 0\ 1/2$, and statistically 0.69 Ti and 0.31 Zr at $0\ 1/2\ 1/2$, then the structure factor for the unit cell is

$$F_{hkl} = b_{Ti} \left[1 + e^{\pi i(h+k)} \right] + b_{Zr} e^{\pi i(h+l)} + (0.69b_{Ti} + 0.31b_{Zr}) e^{\pi i(k+l)} + b_C \left[e^{\pi i(h+k+l)} + e^{\pi il} + e^{\pi ik} + e^{\pi ih} \right] \quad (11)$$

and $F_{hkl} = 4b_C$ for hkl all odd or all even, since the expression

$$b_{Ti} \left[1 + e^{\pi i(h+k)} \right] + b_{Zr} e^{\pi i(h+l)} + (0.69b_{Ti} + 0.31b_{Zr}) e^{\pi i(k+l)} = 0. \quad (12)$$

This follows from the fact that in the structure of MC the metal atom fraction p_{Ti} and p_{Zr} of titanium and zirconium atoms, respectively, per unit cell are

$$4(p_{Ti} + p_{Zr}) = 4,$$

and when $p_{Ti} > 0.5$ and $p_{Zr} > 0.25$, expression (12) becomes

$$b_{Ti} \left[1 + e^{\pi i(h+k)} \right] + b_{Zr} e^{\pi i(h+l)} \\ + 4 \left[\left(p_{Ti} - 0.5 \right) b_{Ti} + \left(p_{Zr} - 0.25 \right) b_{Zr} \right] e^{\pi i(k+l)}$$

and

$$= b_{Ti} \left[1 + e^{\pi i(h+k)} - 2e^{\pi i(k+l)} \right] + b_{Zr} \left[e^{\pi i(h+l)} - e^{\pi i(k+l)} \right] \\ + 4 \left(p_{Ti} b_{Ti} + p_{Zr} b_{Zr} \right) e^{\pi i(k+l)} \quad (13)$$

or

$$= 0, \text{ for } hkl \text{ all odd or all even.}$$

As given in Equations (10) and (11) the structure factor for the null-matrix atoms in MC in both cases is $4b_C$. This means that for crystal structures* in which the null-matrix atoms occupy face-centered cubic sites, that is, $0\ 0\ 0$, $1/2\ 1/2\ 0$, $1/2\ 0\ 1/2$, $0\ 1/2\ 1/2$, that part of the structure factor which results from the distribution of these atoms is zero for all-odd or all-even Miller indices reflections, whether these atoms occupy ordered sites or are randomly distributed.

III. FACE-CENTERED CUBIC NULL-MATRIX

The crystal structure of a stoichiometric $PtMn_3$ alloy is cubic with four atoms per chemical unit cell. The structure factor for the disordered state of the alloy is

$$F_{hkl} = \left(\frac{b_{Pt} + 3b_{Mn}}{4} \right) \left[1 + e^{\pi i(h+k)} + e^{\pi i(h+l)} + e^{\pi i(k+l)} \right], \quad (14)$$

where b_{Pt} and b_{Mn} are the coherent nuclear-scattering amplitudes of platinum and manganese, respectively. Since, $b_{Pt} = 0.95 \times 10^{-12}$ cm and $b_{Mn} = -0.36 \times 10^{-12}$ cm, the structure-factor squared values for all reflections

*NaCl, ZrS, CaF_2 , BiF_3 , Mn_4N , etc., type structures.

are practically zero and the structure becomes one of the typical face-centered cubic null-matrix.

A. Chemical Ordering in PtMn₃ Null-matrix

The PtMn₃ alloy orders with the platinum atom at the corner position and the manganese atoms at the face-centered positions. The structure-factor expression for the ordered state of the alloy is

$$F_{hkl} = b_{Pt} + b_{Mn} \left[e^{\pi i(h+k)} + e^{\pi i(h+l)} + e^{\pi i(k+l)} \right] \quad (15)$$

and

$$= b_{Pt} - b_{Mn} \text{ for } hkl \text{ mixed,}$$

or

$$= 0 \text{ for } hkl \text{ all odd or all even.}$$

Therefore, only the mixed indices would give reflections. Since these reflections result only when ordering takes place in the structure of the alloy and their intensities depend upon the degree of ordering, these properties were utilized to study the order-disorder phenomena in the PtMn₃ alloy.

X-ray and neutron-diffraction patterns were made of two samples, one ordered and the other that appeared to be disordered when examined with X rays. As shown in Fig. 4, the X-ray diffraction pattern of the ordered sample consists of reflections from all odd and all even, as well as mixed-indices planes. In contrast, the neutron pattern consists primarily of reflections from the mixed-indices planes. The presence of weak intensity peaks at 2θ values of approximately 18 and 29° is due to magnetic ordering, which is treated below. The neutron pattern of the sample that appears to be disordered when examined with X rays (Fig. 5) shows weak intensity reflections of mixed-indices planes, indicating that the sample is partly ordered.

The degree of disorder was determined by measuring the disorder parameter, γ , for two samples. If the lattice sites in the unit cell of PtMn₃ were occupied as given in Table IV, then the structure factor for the partly ordered alloy is

$$F_{hkl} = \left[(1 - \gamma) b_{Pt} + \gamma b_{Mn} \right] + \frac{1}{3} \left[\gamma b_{Pt} + (3 - \gamma) b_{Mn} \right] \left[e^{\pi i(h+k)} + e^{\pi i(h+l)} + e^{\pi i(k+l)} \right] \quad (16)$$

106-7795

Fig. 4. Diffraction Patterns of Ordered PtMn_3 Alloy. (a) Filtered Iron Radiation, (b) Neutron $\lambda = 1.08 \text{ \AA}$.

106-7794

Fig. 5. Diffraction Patterns of Disordered PtMn_3 Alloy. (a) Filtered Iron Radiation, (b) Neutron $\lambda = 1.08 \text{ \AA}$.

TABLE IV. Occupancy of Lattice Sites in Unit Cell of PtMn_3

	0 0 0	$1/2 \ 1/2 \ 0, 1/2 \ 0 \ 1/2,$ $0 \ 1/2 \ 1/2$	Total
Pt	$1 - \gamma$	γ	1
Mn	γ	$3 - \gamma$	3

where γ = fraction of platinum or manganese atoms that change sites.

For mixed-Miller-indices planes, expression (16) becomes

$$F_{hkl} = \left(1 - \frac{4}{3}\gamma\right) (b_{\text{Pt}} - b_{\text{Mn}}). \quad (17)$$

For a completely ordered sample, $\gamma = 0$, and

$$F_{hkl} = b_{\text{Pt}} - b_{\text{Mn}}; \quad (18)$$

and for a completely disordered sample, $\gamma = 3/4$, and

$$F_{hkl} = 0. \quad (19)$$

The F_{hkl} values as a function of γ are given in Fig. 6.

The experimental values of F_{hkl} for the disordered and the ordered samples of PtMn_3 were obtained from the measured integrated intensities of a number of diffraction peaks in the neutron pattern of each sample and by calculating the value²⁰ of

$F_{hkl}(0)$ for $\sin \theta/\lambda = 0$. The numerical value of γ for the sample that was partly ordered was 0.667. This means the sample was 11% ordered. For the ordered sample, the value for γ was practically zero, indicating that ordering was nearly 100%.

106-8117

Fig. 6. Structure Factor $|F|$ as a Function of Disorder Parameter (γ)

B. Magnetic Ordering in PtMn_3

The susceptibility-temperature curve²¹ of PtMn_3 in the temperature range between 60 and 280°C shows a maximum, which is indicative of magnetic ordering. A series of neutron-diffraction patterns of

a fully ordered sample was made in the temperature range between liquid nitrogen and 375°C, which showed that PtMn_3 exhibits two types of spin arrangements. At liquid-nitrogen temperature, the magnetic unit cell of one is the same as the chemical cell. The Néel temperature of this form is 120°C. The second spin arrangement is associated with a tetragonal cell which is related to the chemical cell by

$$A_0 = \sqrt{2} a_0 \text{ and } C_0 = 2a_0; \quad H = h + k; \quad K = h - k \text{ and } L = 2\ell,$$

where the lattice parameters and the indices in capital letters are those of the tetragonal cell and the ones in small letters are those of the chemical cell. The observed interplanar spacings may be indexed in both systems as given in Table V.

TABLE V. Miller Indices (hkl) and Observed Interplanar Spacings (d) of PtMn_3 at 120°C

hkl	HKL	$d, \text{\AA}$	hkl	HKL	$d, \text{\AA}$
100	110	3.890	300, 221	330, 402	1.297
	111	3.479	310	420	1.230
110	200	2.751	320	510	1.079
	113	2.158	321	512	1.040
210	310	1.740	322, 410	514, 530	0.943
	131	1.698	330, 411	600, 532	0.917
211	312	1.588	421	622	0.849
	133	1.445	332	604	0.829

The magnetic-moment alignments in the two unit cells are related. The spin transformation from one to the other is such that the degree of alignment in the tetragonal cell is proportional to the disalignment in the chemical cell. The Néel temperature of the second form of arrangement is approximately 220°C.

Neutron-diffraction measurements made of an approximately 11% ordered sample of PtMn_3 showed that it also exhibits both types of spin arrangements. The intensities of magnetic peaks, however, are proportional to the degree of chemical ordering in the sample.

C. Chemical Ordering in Manganese-Cobalt Null-matrix

Manganese and cobalt form single-phase, face-centered cubic structures in the composition range 40 to 50 a/o manganese. At a composition of approximately 41 a/o manganese and 59 a/o cobalt, the structure becomes a nuclear null-matrix, since

$$p_{\text{Mn}} b_{\text{Mn}} + p_{\text{Co}} b_{\text{Co}} = 0.41(-0.36 \times 10^{-12} \text{ cm}) + 0.59(0.25 \times 10^{-12} \text{ cm}) = 0.$$

Two samples were filed from the as-cast alloy of composition $\text{Mn}_{1.75}\text{Co}_{2.25}$. One was unannealed, and the other was annealed at 750°C

for a week and cooled in the furnace at a rate of $1^\circ/\text{min}$. The X-ray-diffraction pattern of the unannealed sample consisted of reflections from all odd and all even indices planes and thus indicated that the alloy was disordered, although it would be difficult to observe with X rays if the sample is ordered since the difference in the atomic scattering factors for manganese and cobalt is small. However, as in the case of the face-centered cubic PtMn_3 null-matrix, the presence of mixed indices reflections such as (100) and (110) in the neutron pattern of the unannealed sample (Fig. 7) indicated that the sample was partly ordered. In the neutron pattern of the annealed sample, not only were the intensities of (100) and (110) reflections increased but there was an indication of the presence

106-8676

Fig. 7. Neutron-diffraction Patterns of $\text{Mn}_{1.75}\text{Co}_{2.25}$ Alloy; $\lambda = 1.069 \text{ \AA}$. (a) Unannealed Sample, (b) Annealed Sample.

of (201) and (211) reflections, showing that the atomic ordering in the alloy was enhanced by annealing it.

D. Chemical Ordering in $(\text{Mn},\text{Co})_4\text{C}$

A carbide of composition $\text{Mn}_{1.75}\text{Co}_{2.25}\text{C}_{0.93}$ was prepared from manganese-cobalt null-matrix. Its crystal structure is of Mn_4N -type (C_2) with the metal atoms at $0\ 0\ 0$, $1/2\ 1/2\ 0$, $1/2\ 0\ 1/2$, $0\ 1/2\ 1/2$, and the carbon atom at $1/2\ 1/2\ 1/2$ position. The lattice parameter is $3.786 \text{ \AA}$, and the nearest distance of approach of metal atoms is $2.675 \text{ \AA}$. This structure has certain unique properties. While the metal atoms occupy mostly random positions in the structure of the alloy, they are ordered in the structure of the carbide. As seen in Fig. 8, the X-ray-diffraction pattern consists of reflections from hkl all odd and all even, and from mixed-indices planes for which $h + k + l$ is odd. The reflections from mixed-indices planes for which $h + k + l$ is even, that is, (110), (211), (310), (321), etc., are either absent or too weak to be observed. This would mean that the term $x(f_{\text{Mn}} - f_{\text{Co}})$ in the structure factor for these reflections, as given in Table VI, is of the order of $0.93f_{\text{C}}$, such that the $F_{hkl} = x(f_{\text{Mn}} - f_{\text{Co}}) + 0.93f_{\text{C}}$ is negligible.

Fig. 8

X-ray-diffraction Pattern of $\text{Mn}_{1.75}\text{Co}_{2.25}\text{C}_{0.93}$, Filtered Iron Radiation

106-7793

TABLE VI. X-ray Structure Factors for
 $\text{Mn}_{1.75} \text{Co}_{2.25} \text{C}_{0.93}$

F_{hkl}	
Miller Indices	
hkl all odd	$1.75f_{\text{Mn}} + 2.25f_{\text{Co}} - 0.93f_{\text{C}}$
hkl all even	$1.75f_{\text{Mn}} + 2.25f_{\text{Co}} + 0.93f_{\text{C}}$
Mixed Indices	
$h + k + l$ odd	$x(f_{\text{Mn}} - f_{\text{Co}}) - 0.93f_{\text{C}}$
$h + k + l$ even	$x(f_{\text{Mn}} - f_{\text{Co}}) + 0.93f_{\text{C}}$

Here f is the atomic scattering factor, and x is a constant of the ordering of the metal atoms ($x = 0.75$ in the present case).

Since $x(f_{\text{Mn}} - f_{\text{Co}})$ must be negative for F_{hkl} of mixed indices planes for which $h + k + l = \text{even}$, the lattice position, $0\ 0\ 0$, in the unit cell must be occupied by a manganese atom. Therefore, the intensities of reflections were calculated by using the following atomic ordering: 1 Mn at $0\ 0\ 0$; 0.75 Mn and 2.25 Co on the average at $1/2\ 1/2\ 0$, $1/2\ 0\ 1/2$, and $0\ 1/2\ 1/2$; and 0.93 C at $1/2\ 1/2\ 1/2$. The calculated and observed intensities, together with the interplanar spacings, are given in Table VII.

TABLE VII. Interplanar Spacings (d) and Calculated and Observed Relative Intensities (I/I_0) for $\text{Mn}_{1.75} \text{Co}_{2.25} \text{C}_{0.93}$

hkl	$d, \text{\AA}$	I/I_0 (calc)	I/I_0 (obs)	hkl	$d, \text{\AA}$	I/I_0 (calc)	I/I_0 (obs)
100	3.786	3.5	w	221, 300	1.262	0.6	vw
110	2.667	0.4	-	310	1.197	0	-
111	2.186	325.8	vs	311	1.142	199.6	s
200	1.893	173.8	s	222	1.093	82.5	m
210	1.693	1.1	vw	320	1.050	0.7	vw
211	1.546	0.1	-	321	1.012	0.2	-
220	1.339	123.1	m				

v = very; s = strong; m = medium; w = weak; - not observed.

The best comparison between the calculated and observed intensities was obtained from the atomic ordering given above and the atomic scattering factor of carbon atom as f_{C}^{+2} . Although no attempt was made to determine the exact valency state of the carbon atom, it was established that the

comparison was worse when (a) the metal atoms were assumed to occupy random positions, and (b) the valency of the carbon atom was zero or negative. These results imply that the carbide structure is ordered and the carbon atom in it is a positive ion. The latter seems to be confirmed by soft X-ray emission study of the carbon K band from the sample, in which the peak of the band is shifted towards higher energy as compared with the same peak from graphite. This is in agreement with the general conclusion drawn by Holliday from his soft X-ray emission studies²² of transition metals and their carbides, that is, "In the case of group VI transition metal carbides and beyond there is a definite transfer of charge from the carbon to the metal atom."

APPENDIX

Coherent Nuclear-scattering Amplitudes for Elements and Isotopes
(March 1966)

Atomic Number	Element or Isotope	b, 10^{-12} cm	Atomic Number	Element or Isotope	b, 10^{-12} cm
1	H ¹	-0.378	24	Cr	0.352
	H ²	0.65		Cr ⁵²	0.490
2	He ⁴	0.30	25	Mn ⁵⁵	-0.36
3	Li	-0.18	26	Fe	0.96
	Li ⁶	0.18 ⁽³⁾		Fe ⁵⁴	0.42
	Li ⁷	-0.21 ⁽³⁾		Fe ⁵⁶	1.01
4	Be ⁹	0.774		Fe ⁵⁷	0.23
5	B	0.540 ⁽¹³⁾	27	Co ⁵⁹	0.25 ⁽³⁾
6	C	0.66 ⁽²⁾	28	Ni	1.03
	C ¹²	0.661		Ni ⁵⁸	1.44
	C ¹³	0.60		Ni ⁶⁰	0.30
7	N ¹⁴	0.940		Ni ⁶¹	0.75 ⁽⁶⁾
8	O ¹⁶	0.577		Ni ⁶²	-0.87
9	F ¹⁹	0.55		Ni ⁶⁴	- (6)
10	Ne		29	Cu	0.79
11	Na ²³	0.351		Cu ⁶³	0.67
12	Mg	0.516 ⁽¹⁰⁾		Cu ⁶⁵	1.11
13	Al ²⁷	0.35	30	Zn	0.59
14	Si	0.42	31	Ga	0.72 ⁽⁴⁾
15	P ³¹	0.53	32	Ge	0.84
	P ³¹	0.505 ⁽⁶⁾	33	As ⁷⁵	0.64 ⁽⁴⁾
16	S	0.31 ⁽²⁾	34	Se	0.89
	S ³²	0.31	35	Br	0.67
17	Cl	0.99	36	Kr	0.76 ⁽²⁾
18	Ar ⁴⁰	0.20	37	Rb	0.85 ⁽⁶⁾
19	K	0.35		Rb ⁸⁵	0.83 ⁽⁶⁾
	K ³⁹	0.37 ⁽⁶⁾	38	Sr	0.57
20	Ca	0.49	39	Y ⁸⁹	0.80 ⁽⁹⁾
	Ca ⁴⁰	0.49		Y ⁸⁹	0.78 ⁽¹⁴⁾
	Ca ⁴⁴	0.18	40	Zr	0.70 ^(2,6,7)
21	Sc ⁴⁵	1.18	41	Nb ⁹³	0.691
22	Ti	-0.34	42	Mo	0.661
	Ti ⁴⁶	0.48	43	Tc ⁹⁹	0.68 ⁽⁶⁾
	Ti ⁴⁷	0.33	44	Ru	0.73
	Ti ⁴⁸	-0.58	45	Rh ¹⁰³	0.591 ⁽⁶⁾
	Ti ⁴⁹	0.08		Rh	0.585 ⁽¹²⁾
	Ti ⁵⁰	0.55	46	Pd	0.59 ⁽³⁾
23	V ⁵¹	-0.051			

Element			Element		
Atomic Number	or Isotope	$b, 10^{-12} \text{ cm}$	Atomic Number	or Isotope	$b, 10^{-12} \text{ cm}$
47	Ag	0.61	67	Ho ¹⁶⁵	0.85
	Ag ¹⁰⁷	0.83	68	Er	0.79
	Ag ¹⁰⁹	0.43	69	Tm ¹⁶⁹	0.55 ⁽²⁾
48	Cd	0.38 + 0.12i	70	Yb	1.26 ⁽³⁾
	Cd ¹¹³	0.80 + 0.75i ⁽²⁾	71	Lu	0.73
49	In	0.39 ⁽⁴⁾	72	Hf	0.88
50	Sn	0.61	73	Ta ¹⁸¹	0.70
51	Sb	0.54 ^(1,4)	74	W	0.466
52	Te	0.56	75	Re	0.92
	Te ¹²⁰	0.52	76	Os	1.07 ⁽⁶⁾
	Te ¹²³	0.57		Os ¹⁸⁸	0.78 ⁽⁶⁾
	Te ¹²⁴	0.55		Os ¹⁸⁹	1.10 ⁽⁶⁾
	Te ¹²⁵	0.56		Os ¹⁹⁰	1.14 ⁽⁶⁾
53	I ¹²⁷	0.52		Os ¹⁹²	1.19 ⁽⁶⁾
54	Xe	0.476 ⁽⁸⁾	77	Ir	1.06 ^(5,6)
55	Cs ¹³³	0.49	78	Pt	0.95
	Cs	0.75 ⁽¹⁶⁾	79	Au ¹⁹⁷	0.76
56	Ba	0.52	80	Hg	1.3
57	La ¹³⁹	0.83	81	Tl	0.89
58	Ce	0.46	82	Pb	0.96
	Ce ¹⁴⁰	0.47	83	Bi ²⁰⁹	0.864
	Ce ¹⁴²	0.45	84	Po	
59	Pr ¹⁴¹	0.44	85	At	
60	Nd	0.72	86	Rn	
	Nd ¹⁴²	0.77	87	Fr	
	Nd ¹⁴⁴	0.28	88	Ra	
	Nd ¹⁴⁶	0.87	89	Ac	
61	Pm		90	Th ²³²	1.01 ^(9,11)
62	Sm ¹⁵²	-0.5	91	Pa	
	Sm ¹⁵⁴	0.8	92	U ²³⁵	0.98 ⁽¹¹⁾
63	Eu	0.63 ⁽⁴⁾		U ²³⁸	0.85 ^(9,11)
64	Gd	1.5 ⁽¹⁵⁾	93	Np	
65	Tb ¹⁵⁹	0.76 ⁽³⁾	94	Pu	0.75 ⁽⁹⁾
66	Dy	1.69 ⁽²⁾	95	Am	

The coherent scattering cross sections may be calculated for a bound atom from $\sigma_{\text{coh}}^b = 4\pi b^2$ and for a free atom from $\sigma_{\text{coh}}^f = \mu^2 \sigma_{\text{coh}}^b$, where μ is the reduced mass.

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