many enlightening discussions concerning the crystal structure of ordered Au_3Mn, and to H. N. Treaftis for his able assistance in some of these measurements.

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Magnetic Properties of Au_3Mn Alloys

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The magnetic properties of Au_3Mn alloy, which was known to be antiferromagnetic were investigated. In the ordered state with an antiphase domain structure, the Néel temperature was found to be 140°K, the paramagnetic Curie temperature about 200°K and effective Bohr magneton 4.06 μ_B. Also the magnetic properties for the filed sample of the ordered alloy and the quenched alloy were investigated. The magnetization of ordered alloy was measured in static magnetic fields up to 80 KOe at 293°, 201° and 83°K. The observed magnetic behaviors were discussed based on a theory for the antiferromagnetic crystal with non-equivalent sublattices.

1. Introduction

It was known by many workers[1,2,3] that the ordered Au_3Mn alloy was an antiferromagnetic with the Néel temperature at 145°K. The behavior of magnetization at strong magnetic field was predicted by Meyer[4]. On the other hand, an antiphase domain structure in the ordered Au_3Mn was recently studied in detail by Watanabe[5]. In this paper the antiferromagnetic properties of Au_3Mn alloy in the ordered and disordered states, and the magnetizations in strong magnetic fields of this alloy are studied, and the origin of the magnetic properties is discussed in connection with the special atomic arrangement in this alloy.

2. Experimental results

Au_3Mn with a stoichiometric composition was prepared by the usual ceramic method. Specimens were subjected to following heat treatment: Specimen No. 1 was annealed below the order-disorder transformation point for twelve days. Specimen No. 2 was filed after this heat treatment. Specimen No. 3 was quenched into ice water from 700°C.

According to the X-ray analysis of the powdered specimen, Specimen No. 1 was confirmed to have an ordered structure with two-dimensional antiphase domain. The results of analysis are shown in Table I. Lattice parameters corresponding to the original

Table I. Observed and calculated lattice spacings and intensities.

HKL (hkl)*	d_{cal} d^*_{cal}	I_{cal}	d_{obs}	I_{obs}
110	7.283	11		
130	4.496	27		
210	3.960	40		
011	3.892	1		
140	3.621	38	3.67	vvw
101	3.599	38	3.59	vw
111	3.513	53		
230	3.253	5		
031	3.217	23		
150	3.005	10		
131	2.993	3	2.84	vvw
310	2.682	2	2.67	vvw
250	2.533	2		
051	2.516	13	2.50	vvw
330	2.428	6	2.46	vvw
151	2.405	3	2.41	w
241	2.334	1000	2.33	vs
(111)*	2.334*			
340	2.256	11	2.27	w
301	2.251	6		
311	2.230	25	2.18	w
170	2.221	1		
350	2.081	3	2.09	w
331	2.077	2		
400	2.040	160	2.02	w
080	2.020	156		
002	2.005	153		
(200)*	2.040*			
(020)*	2.020*			
(002)*	2.005*			
071	2.001	1		
171	1.943	6	1.91	vvw
112	1.933	1		
132	1.851	5	1.86	vvw
351	1.847	1		
411	1.807	2	1.80	vvw
212	1.788	2		
181	1.762	11	1.76	vvw
370	1.760	0.4		
142	1.754	13		
431	1.723	8	1.70	vvw
232	1.707	1.5		
152	1.668	3.5	1.65	vvw
290	1.644	3.6		
510	1.624	0.3		
371	1.612	3.4		
191	1.608	3.4		
312	1.606	0.2		
451	1.585	6.4	1.57	vw
530	1.562	1.7		
332	1.546	2.6	1.53	vvw
540	1.515	3.3		
501	1.512	3.3		
511	1.505	3.1	1.50	vw
381	1.504	6.5		
342	1.499	6.5		
172	1.488	0.6		
550	1.457	1.3		
531	1.456	0.4		
1,11,0	1.446	1.3		
352	1.444	2.5		
480	1.436	111.4	1.43	s
402	1.430	110.6		
082	1.423	109.9		
(220)*	1.436*			
(202)*	1.430*			
(022)*	1.423*			
272	1.419	5	1.39	vvw
0,11,1	1.380	2.3		
551	1.369	0.4		
013	1.332	0.1	1.34	vw
1,12,0	1.329	2.5		
372	1.323	1.4	1.32	vvw
103	1.319	2.5		
113	1.315	1.8		
033	1.297	2	1.29	vvw
3,11,0	1.292	2.6		
133	1.281	3.1		
292	1.271	4	1.26	vvw
512	1.262	0.5		
650	1.254	0.4		
053	1.235	2	1.23	vvw
532	1.232	1.9		
641	1.227	174.4	1.22	vs
2,12,1	1.218	177.0		
243	1.212	176.9		
(311)*	1.227*			
(131)*	1.218*			
(113)*	1.212*			
581	1.210	4.7	1.21	vvw
542	1.208	4.7		
3,12,0	1.206	2.3		
303	1.200	2.3	1.19	vw
313	1.196	1.9		
0,13,1	1.187	2	1.18	vvw
552	1.179	2	1.17	ms
333	1.171	0.3		
710	1.163	0.2		

face centered orthorhombic cell are obtained as, a=4.08 Å, b=4.04 Å and c=4.01 Å. The intensities and the lattice spacings are calculated based on the superstructure with $A=2a$, $B=4b$ and $C=c$, shown in Fig. 1. Our results coincided with the structure observed by the electron diffraction[5]. For the Specimen No. 2 no superlattice reflection line was observed. Specimen No. 3 showed a partial ordered structure. In the crystal structure of ordered Au_3Mn shown in Fig. 1, the manganese atoms have two kinds of sublattice A, F and B, E, and a pair of nearest neighbor A-F-S located at the antiphase domain boundary. So, if we take the first and the second neighboring manganese atoms in an xy-plane, four atoms make a chain with a finite length, shown by E-F-A-B as well as C-D-G-H.

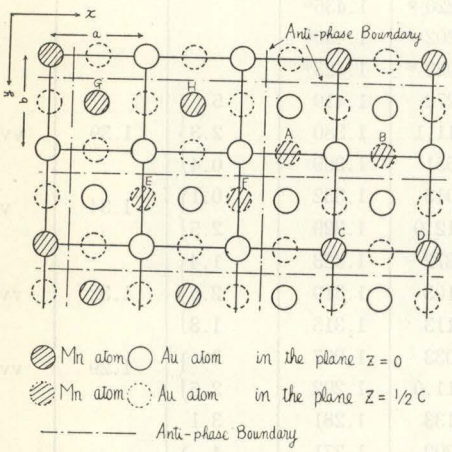

Fig. 1. Crystal structure of Au_3Mn.

The temperature dependence of magnetic susceptibilities for these specimens was observed; results are shown in Fig. 2. Both ordered and filed alloys show a typical antiferromagnetic behavior, while quenched alloy shows somewhat different behavior. In the ordered alloy, Néel temperature T_N was found to be 140°K. Based on the Curie-Weiss law, the effective magneton number μ_{eff} was estimated as 4.06μ_B, and the asymptotic Curie temperature θ_p is 200°K. These results agree with those obtained by Mayer. In the filed alloy, T_N was found to be 42°K, θ_p about 70°K, and μ_{eff} 5.24μ_B. The positive value of θ_p can be considered to support the existence of a strong ferromagnetic coupling between manganese atoms.

Further, the magnetization of the ordered alloy was measured in static fields up to 80,000 Oe at 293°K, 201°K and 83°K. Measurements were made by means of strain gauge type magnetometer, results being shown in Fig. 3. As can be seen in this figure, at a field strength of about 40,000 Oe the magnetization curve has an inflection point. This field is considered to be the critical field of spin flopping in this alloy. Above 80,000 Oe the magnetization seems to saturate. However, the magnetization at 80,000 Oe is about one half of the value of the completely aligned moment estimated from the data of susceptibility above T_N.

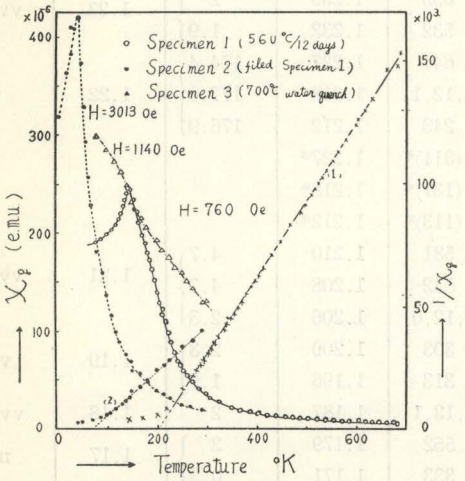

Fig. 2. Magnetic susceptibilities of Au_3Mn alloys.

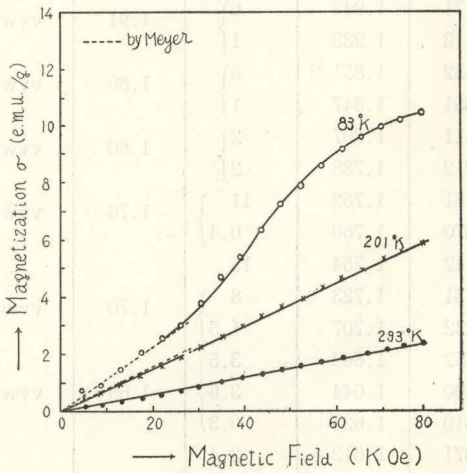

Fig. 3. Magnetization curves in ordered Au_3Mn.

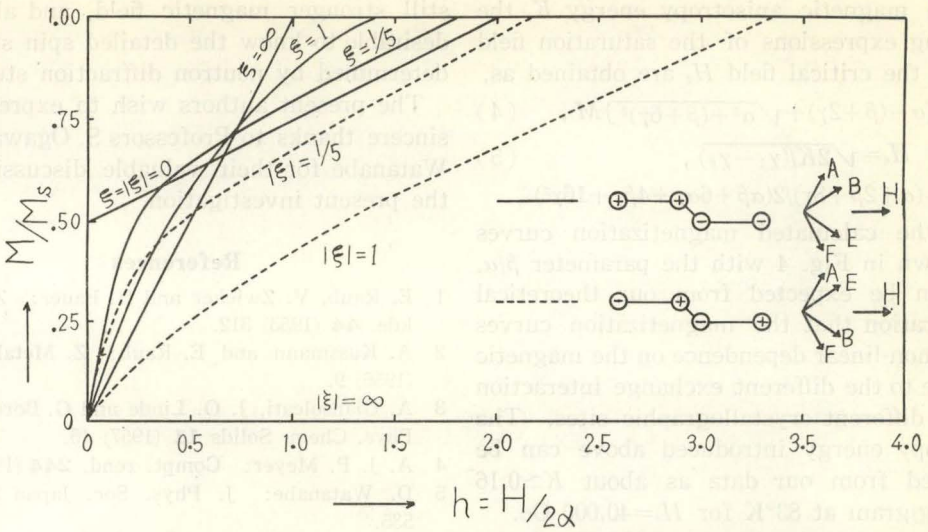

Fig. 4. Calculated magnetization curves with $\xi=\beta/\alpha$. ($M_A=M_B$, $\gamma=0$) $h_E=1-\xi+\sqrt{1+\xi^2}$ for $\beta>0$; $h_E=1+|\xi|+\sqrt{1+\xi^2}$ for $\beta<0$.

3. Explanation of experimental results

In order to explain our experimental results illustrated above, we consider the exchange interactions of the first and the second neighbors of the intrachain *E-F-A-B* and one of the third neighbor of the interchain. For the Au_3Mn structure, the usual two-sublattice antiferromagnetic model[6] is not applicable. According to the empirical stand point for other manganese alloys and also according to an estimation* based on the theory of *s-d* exchange interactions[7], the following neighboring interactions can reasonably be assumed: the nearest neighbor interaction is antiferromagnetic (exchange field coefficient is denoted by $-\alpha$), and the second neighbor interactions along *y*- and *z*-directions are ferromagnetic (denoted by β and β_0 respectively). β_0 is regarded to be stronger than β, because β_0 is responsible to give the high Néel temperature in this substance. γ is the third neighbor interaction for the interchain spin arrangement, which is considered to be not so strong. Thus the stable arrangement of spins in the chain *E-F-A-B* is shown as $++--$. Based on the molecular field approximation, Néel temperature and susceptibility above T_N are expressed by the equations (1) and (2), respectively.

$$T_N=C[2\beta_0+(\alpha/2)\pm 2\gamma + \sqrt{\{(\alpha/2)\mp 2\gamma\}^2+(\beta\pm 2\gamma)^2}] \quad \text{for } \gamma\gtrless 0, \quad (1)$$

$$\frac{1}{\chi}=\frac{T-\theta_p}{C}-\frac{\tau}{T-T_h}, \quad (2)$$

where $\theta_p=C\{2\beta_0-(\alpha/2)+\beta+8\gamma\}$,

$T_h=C\{2\beta_0-(\alpha/2)-\beta-4\gamma\}$,

$\tau=C\{(\alpha/2)+2\gamma\}^2$ and $C=(N\mu_B^2/3k)g^2S(S+1)$.

In this expression T_N is found to be at a temperature higher than θ_p, this fact being contrary to the experimental evidence. The molecular field approximation is not so good in this case, because the atoms have only a few neighboring pairs. Therefore, it can not be expected to determine the correct value of interaction constants from the observed magnetic data such as $\chi(T_N)$, T_N and θ_p.

Next, if a magnetic field is applied to this substance, the free energy of this system is expressed by following equation (3):

$$E(\theta,\varphi)=-M_A^2\alpha\cos 2\theta-2M_AM_B\{(\beta+4\gamma)\cos(\theta-\varphi)+2\gamma\cos(\theta+\varphi)\} -H(M_A\sin\theta+M_B\sin\varphi). \quad (3)$$

In a given field, each spin on the *A*-site and *B*-site makes a different angle θ and φ from the antiparallel spin axis. Minimizing the free energy for θ and φ, and introducing an

* If the $3d^6\ 4s^1$ electron configuration of Mn atom in Au_3Mn alloy is assumed and the free electron approximation is applied and the cubic lattice parameter 4.03 Å and $k_f=1.21$ Å^{-1} are used in Yosida's expression, the ratio of interactions for the first, the second and the third neighbors is obtained as $-4:2:-1$.

uniaxial magnetic anisotropy energy K, the following expressions of the saturation field H_E and the critical field H_c are obtained as,

$$H_E=\{\alpha-(\beta+2\gamma)+\sqrt{\alpha^2+(\beta+6\gamma)^2}\}M\,, \quad (4)$$

and $$H_c=\sqrt{2K/(\chi_\perp-\chi_{//})}\,, \quad (5)$$

$$\chi_\perp(0)=(\alpha+2\beta+8\gamma)/2(\alpha\beta+6\alpha\gamma+4\beta\gamma+16\gamma^2)\,.$$

Then the calculated magnetization curves are shown in Fig. 4 with the parameter β/α.

It can be expected from our theoretical consideration that the magnetization curves have a non-linear dependence on the magnetic field due to the different exchange interaction for the different crystallographic sites. The anisotropy energy introduced above can be estimated from our data as about $K\simeq 0.16\times 10^6$ erg/gram at 83°K for $H_c=40{,}000$ Oe.

In order to obtain the knowledge for interactions between manganese atoms, it is neccessary to observe the magnetization in a still stronger magnetic field, and also it is desirable to know the detailed spin structure determined by neutron diffraction study.

The present authors wish to express their sincere thanks to Professors S. Ogawa and D. Watanabe for their valuable discussions for the present investigation.

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DISCUSSION

C. D. GRAHAM, JR.: It is remarkable to find that rather different results have led to quite similar conclusions, at least for the ordered Au_3Mn. The great differences between the quenched and filed alloys clearly merit further investigation.