

Magnetization of Gold-Iron and Gold-Nickel Solid Solutions

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THE magnetic properties of Cu-Ni¹ and Cu-Fe² alloys have been studied by two of the present authors and by many other investigators. The present investigation on Au-Fe and Au-Ni alloys was undertaken as an extension of that work with the idea that any similarities or differences between the copper and gold alloys should be of interest in connection with the theory of ferromagnetism.

Previous magnetic investigations of Au-Fe^{3,4} alloys have shown that the gold-rich alloys are paramagnetic with a large temperature dependence and become ferromagnetic at about 10 percent iron. The ferromagnetic Au-Ni alloys have been studied carefully,^{5,6} while the gold-rich paramagnetic alloys have received some attention.⁷ These investigations showed that the magnetic properties of the quenched Au-Ni alloys were very similar to those of the Cu-Ni alloys.

Neither of these alloy systems has extensive solid solubility at room temperature. However, at elevated temperatures about 25 percent of iron by weight will dissolve in gold, while the nickel-

gold alloys become soluble in all proportions.⁸ When these alloys are quenched, the solid solution which existed at the elevated temperature is retained in a supersaturated condition which to a first approximation, at least, consists of a uniform distribution of the atoms. This practically uniform solution is demonstrated first of all by x-ray diffraction patterns and secondly by the continuous variation of various physical properties with composition. However, there can be no doubt that fluctuations in composition do occur on an atomic scale and this situation may be of importance in connection with the magnetic behavior of the alloys. On the other hand, there is metallographic and magnetic evidence^{4,6} that the quenched solid solutions, when reheated, do not decompose with a continuous shift in composition but rather that the initial precipitation of the second phase (iron or nickel-rich solid solution) occurs only locally while the bulk of the solid solution retains the magnetic properties of the quenched material. This indicates that the quenched solid solution has definite properties and a certain stability which does not allow the iron or nickel atoms to agglomerate into clusters which can grow and progressively change the magnetic behavior.

TABLE I. Mass susceptibility, $\chi \times 10^6$, gold-iron alloys.

Temp. °K	Weight percent iron								
	0.18	0.38	0.79	1.23	1.90	3.2	4.3	5.2	7.9
14	2.16*	4.79*	25.7*	31.9*	39.5	41.4	38.7	28.5	
20.4	1.26	4.50*	22.1*	30.1*	27.8	40.1	32.5	54.1	
63						47.6			
77	0.321	1.01	2.71	7.0	17.9	49.1			
133	0.153	0.59	1.46	3.95	9.52	47.0	66.4		
192	0.075	0.386	1.00	2.44	6.00	18.8	47.3	86.5	
245	0.030	0.276	0.77	1.99	4.35	12.4	25.1	61.7	
297	0.0069	0.226	0.627	1.63	3.61	8.96	17.6	32.2	489*
573			0.288		1.57				
723			0.158					4.84	
823					0.97				
873								4.00	
1023			0.113		0.74			2.85	
1223			0.080		0.58			2.24	

* Initial susceptibility given.

¹ A. Kaufmann and C. Starr, *Phys. Rev.* **63**, 445 (1943).
² Bitter, Kaufmann, Starr, and Pan, *Phys. Rev.* **60**, 134 (1941).
³ J. W. Shih, *Phys. Rev.* **38**, 2051 (1931).
⁴ Pan, Kaufmann, and Bitter, *J. Chem. Phys.* **10**, 318 (1942).
⁵ V. Marian, *Ann. de Physique* **7**, 519 (1937).
⁶ Köster and Dannöhl, *Zeits. f. Metallkunde* **28**, 248 (1936).
⁷ E. Vogt and H. Krueger, *Ann. der Physik* **18**, 755 (1933).

PREPARATION OF ALLOYS

The alloys were prepared by melting about 5 grams of pure gold and iron or nickel in an

TABLE II. Intrinsic magnetic moment per gram, σ , gold-iron alloys.

Temp. °K	Weight percent iron					
	1.9	3.2	4.3	5.2	7.9	11.0
14	0.29	3.1	6.9	11.4		
20.4	0.36	3.1	7.2	10.8	22.7	31.1
63			1.8			
77	0.04	1.2	5.0	9.0	20.6	29.5
133	0.00	0.00	1.8			
192	0.01	0.02	0.08	1.04	15.5	
297	0.00	0.00	0.01	0.00	4.1	20.4

⁸ M. Hansen, *Der Aufbau der Zweistofflegierungen* (Verlagsbuchhandlung, Julius Springer, Berlin, 1936), pp. 223, 242.

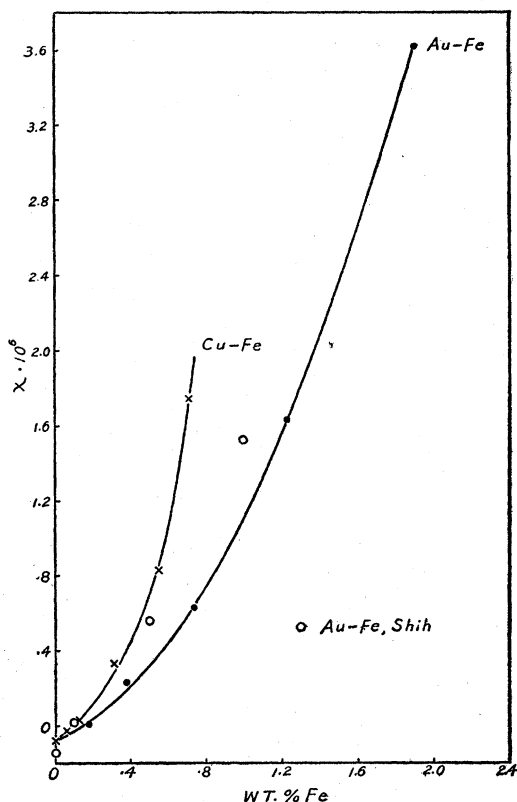


FIG. 1. Mass susceptibility, χ , of gold-iron and copper-iron alloys as functions of iron content.

alundum crucible with a hydrogen atmosphere at about 1500°C and then chill casting in a vacuum. The resulting material was annealed in hydrogen at 900°C for 10 hours and then quenched in water. This treatment was intended to give complete homogenization of the alloys and to retain the solid solution which existed at the heat treatment temperature.

METHOD OF MEASUREMENT

The apparatus used for making magnetic measurements at high and low temperatures and in fields up to 30,000 oersted has already been described.² The data obtained are plotted as magnetization curves which, in general, are straight lines at high fields. The slope of the linear portion of the magnetization curve determines the susceptibility. By extrapolating the linear portion to zero field and finding the intercept on the ordinate, the ferromagnetic moment can also be determined. This intercept

TABLE III. Mass susceptibility, $\chi \times 10^6$, gold-nickel alloys.

Temp. °K	Atomic percent nickel						
	12.5	25	37.5	50	62.5	75	87.5
14	0.271	1.02	3.52	13.4			
20.4	0.203	0.822	3.08	12			34.9
77	0.097	0.424	1.56	27.2	12.2		34.4
133	0.087	0.394	1.41				
192	0.088	0.388	1.42	6.08	54.6	20.6	
297	0.093	0.41	1.38	3.34	14.6	64.0	30.6
723	0.162						
823	0.174						
923	0.183						
1023	0.188						
1123	0.193						
1223	0.197						

should give a correct value for the intrinsic magnetization since any magnetization that is effected by the external field is removed. When the intercept is zero, the susceptibility is then the paramagnetic susceptibility in the usual sense. Values of the susceptibility, χ , and intrinsic magnetic moment, σ , per unit mass are given in Tables I and II for the gold-iron alloys and in Tables III and IV for the gold-nickel alloys. Occasionally, certain specimens which are on the border line between paramagnetism and ferromagnetism will exhibit a magnetization curve which does not become linear even at 30,000 oersted. In such cases the initial susceptibility, as determined by drawing a tangent to the magnetization curve at the origin, is given in Table I. In addition, the complete magnetization data for the gold-iron alloys which showed this behavior are given in Table V, since it is believed that the information may be of use in checking theory.

DISCUSSION OF RESULTS—GOLD-IRON ALLOYS

The mass susceptibility at room temperature is plotted as a function of composition in Fig. 1 along with the data of Shih³ and the data for some copper-iron alloys.² In both of these alloy

TABLE IV. Intrinsic magnetic moment per gram, σ , gold-nickel alloys.

Temp. °K	Atomic percent nickel					
	25	37.5	50	62.5	75	82.5
14	0.001	0.083	2.59			
20.4	0	0.082	2.45	10.5	20.8	34.9
77	0	0.073	0.085	8.9	20.1	34.4
133	0	0.061				
192	0	0.038	0.099	0.21	16.8	
297	0	0.002	0.10	0.23	8.5	30.6

TABLE V. Magnetization of gold-iron alloys, σ .

Wt. % Fe	Temp. °K	Field in oersted					
		5490	10,980	16,480	21,960	27,440	32,920
0.18	14	0.0114	0.0233	0.0353	0.0458	0.0541	0.0615
0.38	14	0.032	0.058	0.083	0.105	0.123	0.138
0.38	20.4	0.024	0.044	0.064	0.083	0.099	0.115
0.79	14	0.087	0.142	0.188	0.226	0.265	0.304
0.79	20.4	0.071	0.114	0.150	0.184	0.214	0.240
1.23	14	0.157	0.272	0.377	0.466	0.544	0.618
1.23	20.4	0.140	0.259	0.345	0.425	0.497	0.560
1.90	14	0.39	0.67	0.93	1.16	1.36	1.59
1.90	20.4	0.35	0.60	0.80	0.97	1.12	1.25
3.2	14	2.50	3.23	3.67	4.00	4.26	4.46
3.2	20.4	2.50	3.19	3.63	3.94	4.20	4.39
3.2	63	1.23	1.92	2.39	2.78	3.07	3.31
3.2	77	0.80	1.40	1.84	2.21	2.52	2.78
7.9	297	2.54	4.37	5.64	6.63	7.38	7.96

systems the susceptibility increases very rapidly with the iron content but not in a linear fashion. It is apparent that iron produces a larger effect in copper than in gold, particularly if the alloys are compared on the basis of atomic percentage. The discrepancy between the data of Shih and the present authors may be due to the fact that Shih's measurements were made in relatively low fields.

The susceptibility of these alloys has a large temperature dependence which is fairly well described by the Curie-Weiss law as shown by the plotted data in Fig. 2. In Fig. 2 the ordinate consists of the reciprocal of the observed susceptibility minus the susceptibility of the gold in the specimen and multiplied by the weight percentage of iron in the alloy. This is, then, the reciprocal of the susceptibility of a gram of dissolved iron. The mass susceptibility of gold is

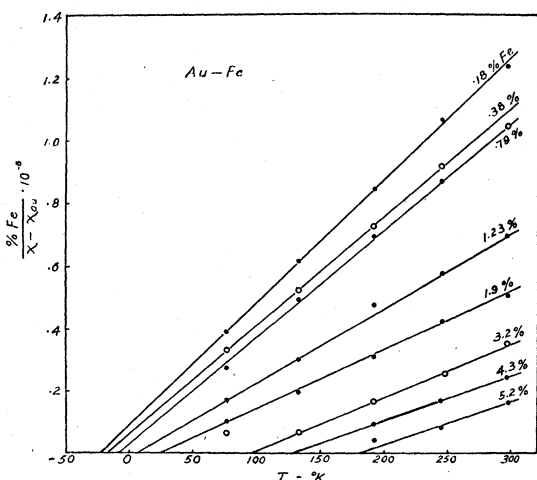


FIG. 2. Reciprocal of mass susceptibility per gram of dissolved iron in gold as a function of temperature.

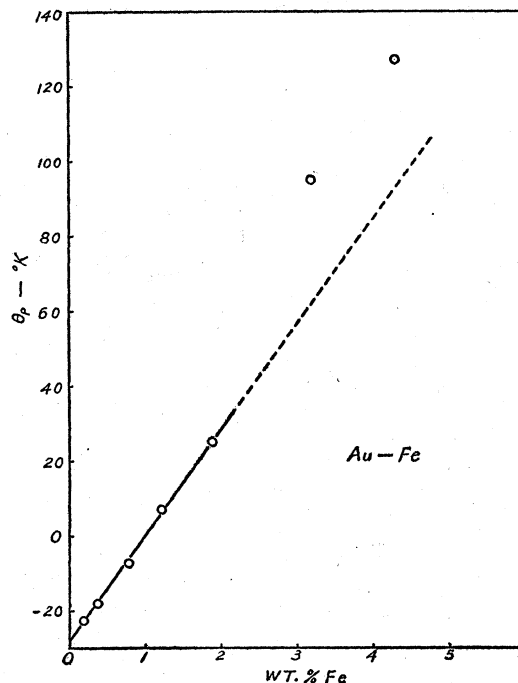


FIG. 3. Paramagnetic Curie temperature of dilute gold-iron alloys as a function of iron content.

taken as -0.142×10^{-6} for this purpose and this value was used at all temperatures since no measurements on its temperature dependence were made.

The experimental data for temperatures below 77°K do not lie on the straight lines shown and likewise the high temperature results obtained on three of the alloys do not lie on a continuation of these same straight lines. Such deviations from linearity indicate that the Curie-Weiss equation does not give a complete description of the observations. Nevertheless, it is interesting to note that the paramagnetic Curie temperature, θ_p , increases linearly with iron content from negative to positive values as shown in Fig. 3. It is perhaps significant that the dilute alloys which show this behavior do not become ferromagnetic even at 14°K while the specimens with enough iron to become ferromagnetic do not lie on the straight line.

From the slope, C , of the lines in Fig. 2 it is possible to calculate the Bohr magneton value per iron atom from the equation $\mu_B = 2.84(100M/C)^{1/3}$ where M is the molecular weight of iron. The values so obtained are tabulated in Table VI. It

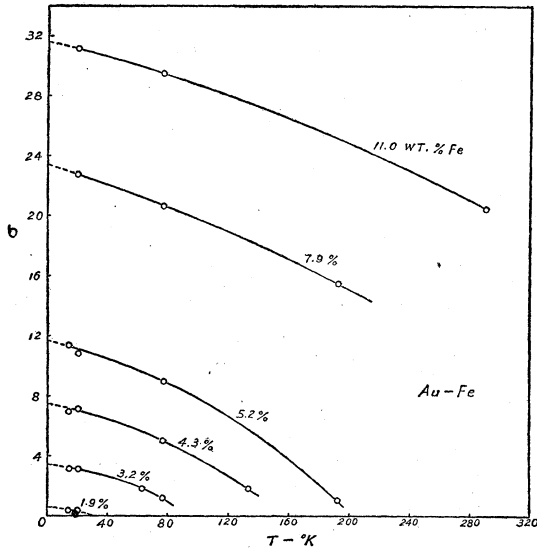


FIG. 4. Mass saturation value, σ , of gold-iron alloys as a function of temperature.

can be seen that the magneton value has the same order of magnitude as is found for iron ions⁹ in salts, but the fact that the value increases with iron content indicates that the iron atoms are not in the form of simple ions. It is possible that measurements above room temperature would indicate a more nearly constant magneton value as suggested by the observations on the alloy with 5.2 percent iron and as found for the copper-iron alloys.²

The saturation moment of the ferromagnetic alloys is plotted in Fig. 4 as a function of the absolute temperature. It is apparent that the ferromagnetic Curie temperature varies with iron content but is not well determined by these

TABLE VI. Bohr magneton number (p_B) for dilute gold-iron alloys.

Weight % Fe	p_B
0.18	3.4
0.38	3.6
0.79	3.7
1.23	4.4
1.9	4.9
3.2	5.2
4.3	5.7
5.2*	4.6

* Measured above room temperature.

⁹ E. C. Stoner, *Magnetism and Matter* (Methuen & Company, London, 1934), p. 312.

curves. The saturation value, σ_0 , at the absolute zero of temperature is obtained by a short extrapolation. These values of σ_0 are plotted in Fig. 5 and are listed in Table VII along with the moment per iron atom in Bohr magnetons. The alloys with more than 5 percent iron exhibit a larger moment per iron atom than is found in solid iron (2.2 Bohr magnetons). This fact, along with the dependence of Curie temperature on composition, indicates that the alloys have a definite ferromagnetism of their own which cannot be attributed simply to precipitated iron or to clusters of iron atoms in the solid solution. The variation of μ_B with composition is a consequence of the fact that σ_0 approaches zero at a finite iron content.

It is interesting to note that the iron content at which σ_0 is zero (2.5 percent) corresponds to the concentration below which any iron atom on the average will have less than one iron atom as a nearest neighbor. This is computed on the basis that a face-centered cubic lattice has twelve nearest neighbors for any lattice point. In view of the fact that the Heisenberg theory of ferromagnetism requires at least eight nearest neigh-

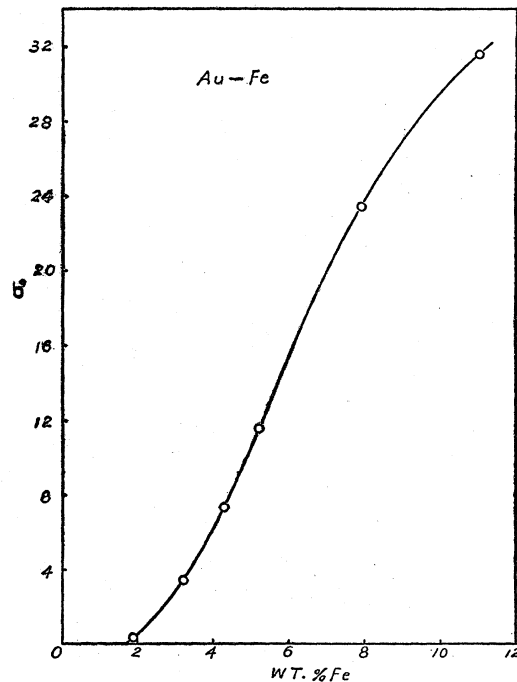


FIG. 5. Absolute mass saturation value, σ_0 , of gold-iron alloys as a function of iron content.

bors in order to have ferromagnetism,¹⁰ the present results are of considerable interest.

DISCUSSION OF RESULTS—GOLD-NICKEL ALLOYS

The susceptibility of the gold-rich alloys at room temperature and 14°K is plotted in Fig. 6 along with the corresponding data for the copper-nickel alloys. It is apparent that nickel produces a somewhat larger effect in copper than in gold but that in a qualitative sense its behavior is about the same in both metals. The temperature dependence of the susceptibility does not correspond at all to a Curie-Weiss behavior and is roughly described by the equation $\chi = aT + b + c/T$, which was applied to the dilute copper alloys.¹ The susceptibility of the alloy with 12.5 atomic percent nickel does increase at high temperatures but seems to approach a constant value rather than a straight line. The term in $1/T$ becomes predominate in those alloys with higher nickel content which approach the ferromagnetic compositions.

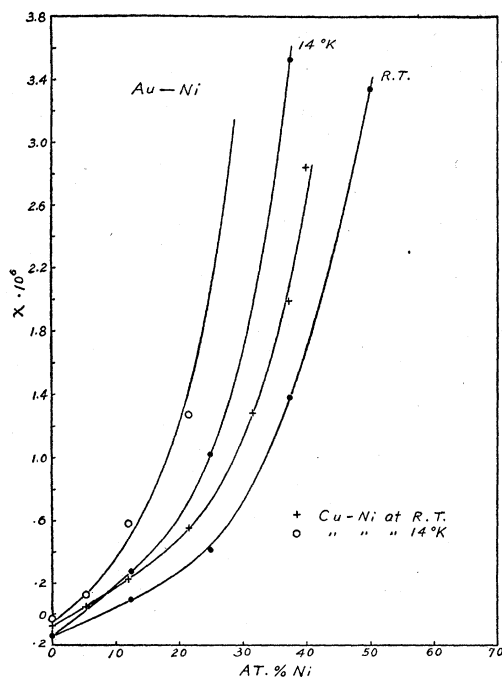


FIG. 6. Mass susceptibility, χ , of gold-nickel and copper-nickel alloys as functions of nickel content.

¹⁰ E. C. Stoner, *Magnetism and Matter* (Methuen and Company, London, 1934), p. 360.

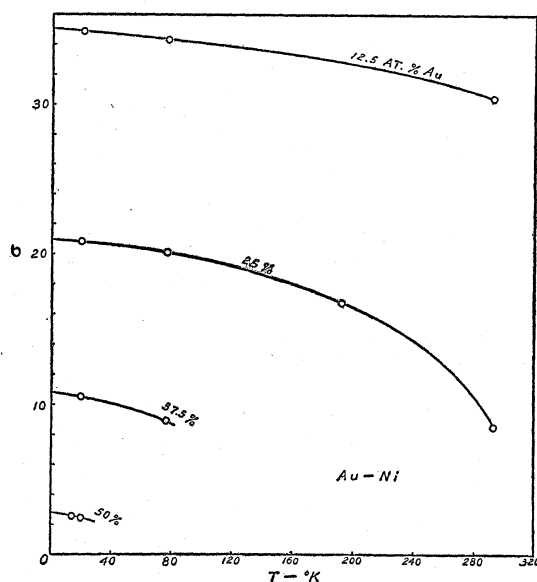


FIG. 7. Mass saturation value, σ , of gold-nickel alloys as a function of temperature.

The intrinsic magnetic moment of the ferromagnetic alloys is plotted as a function of temperature in Fig. 7. Again the Curie temperatures are not accurately fixed but the absolute saturation moment is definitely determined. This saturation moment as a function of gold content is plotted in Fig. 8 along with the results of V. Marian⁵ and the results on copper-nickel alloys.

The two investigations on gold alloys, which are in good agreement, show a non-linear variation of σ_0 with gold content and indicate that zero moment occurs at slightly less than 60 atomic percent gold. This is sufficiently different from the simple behavior of the copper-nickel alloys to indicate that other factors in addition to the electron concentration of the alloys will influence the saturation value. One such factor may be the change of lattice parameter with

TABLE VII. Mass absolute saturation, σ_0 , gold-iron alloys.

Wt. % Fe	Atom % Fe	σ_0	μ_B per Atom
11.0	30.8	31.6	2.89
7.9	23.2	23.4	2.97
5.2	16.2	11.7	2.26
4.3	13.7	7.4	1.73
3.2	10.5	3.4	1.07
1.9	6.4	0.4	

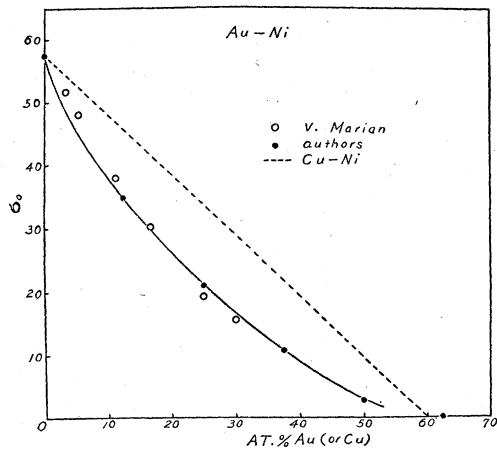


FIG. 8. Absolute mass saturation value, σ_0 , of gold-nickel and copper-nickel alloys as functions of gold content and copper content respectively.

composition. There is only a 2.5 percent difference in the parameters of pure nickel and copper

but a 16 percent difference in the case of nickel and gold.

CONCLUSIONS

There is a marked difference in the behavior of iron and nickel when dissolved in either copper or gold. The iron atoms in dilute alloys have a large magnetic moment and the susceptibility varies rapidly with temperature while the nickel atoms seem to lose their magnetic moment and the resultant susceptibility shows a smaller and more complex temperature dependence. The iron alloys with gold become ferromagnetic with as little as 8 atomic percent iron, while the nickel alloys with copper or gold require about 40 atomic percent nickel to accomplish this. To a first approximation copper and gold are equivalent when alloyed with iron or nickel, although in both cases the ferromagnetic elements produce a larger effect in copper than in gold.