PHYSICAL REVIEW VOLUME 184, NUMBER 2 10 AUGUST 1969

Orbital and Spin Magnetization in Fe-Co, Fe-Ni, and Ni-Co

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(Received 17 March 1969)

The effective electronic orbital contribution M_0 and the effective electronic spin contribution M_s to the spontaneous magnetization M_t for binary alloys of Fe, Co, and Ni are determined from Scott's measured magnetomechanical g'-factors using the relations $M_0/M_t=(2-g')/g'$ and $M_s/M_t=2(g'-1)/g'$, where g' is assumed to be independent of the strength of the magnetic field and the temperature. It is believed that M_0/M_t has several-fold greater accuracy when determined from the g' factors than when determined from ferromagnetic-resonance g factors. In terms of the Bohr magneton μ_B, the orbital magnetic moments for the pure elements at 300°K are: for iron, $(0.0918\pm0.0033)\mu_B$ (4.22% of M_t); for cobalt, $(0.1472\pm0.0034)\mu_B$ (8.81% of M_t); and for nickel, $(0.0508\pm0.0012)\mu_B$ (8.92% of M_t). The M_s values for the fcc phases of the alloys agree well with those reported by Meyer and Asch from their g-factor data. For these alloys, M_0 is much more sensitive to the crystalline structure than M_s and M_t are. The relationship $g'^{-1}+g^{-1}=1$ holds for Fe-Ni and Fe-Co alloys (with the exception of one point) within the accuracy of the g-factor measurements (~0.1–1.0%).

INTRODUCTION

IN this work we have determined the effective electronic orbital contribution M_0 and the effective electronic spin contribution M_s to the spontaneous magnetization M_t for binary alloys of Fe, Co, and Ni. To effect a separation of M_s and M_0 from M_t, we have used values of the magnetomechanical ratio g' determined experimentally by Scott,[1] using the Einstein–de Haas method. We believe Scott's experimental results lead to a high degree of precision in the determination of the relative proportion of spin and orbital contribution; in the determination of M_s, especially, the accuracy is limited by the error in M_t. Accuracy in g' measurements is essential in the determination of M_0, since this is, in most instances, a small fraction of M_t.

Kittel[2] has shown the g' values determined by the Einstein–de Haas method are related to M_0, M_s, the total electronic spin angular momentum J_s, and the total electronic orbital angular momentum J_0 by

$$g'(e/2m)=(M_s+M_0)/(J_s+J_0)\,, \tag{1}$$

from which it immediately follows that

$$M_0/M_t=(2-g')/g' \tag{2}$$

and

$$M_s/M_t=2(g'-1)/g'\,. \tag{3}$$

Thus, a determination of g' leads directly to the relative contributions of M_0 and M_s to M_t. The values of M_t available in the literature are obtained from saturation magnetization measurements, whereas the g' factors are measured at low magnetic fields. The g' factors were found to be independent of magnetic field strength in the range 2–20-Oe applied field.[1]

The dependence of M_s and M_0 upon small changes in g' is readily shown. For example, we may expand g' in powers of M_0/M_t and keep the first-order terms; then

$$M_0/M_t=\tfrac{1}{2}(2-g')\,, \tag{4}$$

which also gives for the M_s/M_t dependence,

$$M_s/M_t=\tfrac{1}{2}g'\,. \tag{5}$$

The orbital contribution is directly proportional to the departure of g' from 2, while the M_s dependence is proportional to the magnitude of g'. This fact clearly demonstrates the importance of accuracy in the values of g' used to calculate M_0/M_t.

Relationship of g' to the Spectroscopic Splitting Factor

In magnetic resonance experiments the orbital angular momentum changes are compensated by corresponding changes in the lattice angular momentum. In this case g is defined by[3]

$$g(e/2m)=(M_s+M_0)/J_s\,, \tag{6}$$

so we have for the ratio of g' to g,

$$\frac{g'}{g}=\frac{J_s}{J_s+J_0}=\frac{M_t-M_0}{M_t+M_0}=\frac{M_s}{M_s+2M_0}\,. \tag{7}$$

When the reciprocals of the definitions of g and g' are summed (after substitution of the expressions $M_0/J_0=e/2m$ and $M_s/J_s=e/m$) we obtain

$$g^{-1}+g'^{-1}=1\,. \tag{8}$$

In addition, an even simpler expression may be obtained provided g' differs only slightly from 2 (a relationship originally obtained by Kittel[3] and by Van Vleck[4]):

$$g-2=2-g'\,. \tag{9}$$

The departure of g and g' from the spin-only value of 2 is easily interpreted in terms of M_0 and M_t (provided

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[1] G. G. Scott, J. Phys. Soc. Japan **17**, 372 (1962); Phys. Rev. **148**, 525 (1966); G. G. Scott and H. W. Sturner, preceding paper, *ibid.* **184**, 490 (1969).

[2] C. Kittel, *Introduction to Solid State Physics* (John Wiley & Sons, Inc., New York, 1956), 2nd ed., p. 410.

[3] C. Kittel, Phys. Rev. **76**, 743 (1949).

[4] J. H. Van Vleck, Phys. Rev. **78**, 266 (1950).

$M_0/M_t \ll 1$). If we solve (4) for g' we obtain

$$g' = 2 - 2M_0/M_t, \tag{10}$$

while if we write a comparable expression for g we have

$$g = 2 + 2M_0/M_t. \tag{11}$$

Thus, g and g' both differ from 2 by twice the ratio of the orbital contribution to the magnetization, only with opposing signs.

RESULTS

A. M_s/M_t and M_0/M_t

Using Eqs. (2) and (3) we have calculated M_0/M_t for Fe-Co, Fe-Ni, and Co-Ni alloys for various compositions, from the g' values of Scott.[1] These values should be independent of temperature provided g' is. They are listed in Table I along with their probable errors. M_0/M_t curves are plotted in Fig. 1. The samples used in this work were slow-cooled from above 600°C, and therefore the structure in Table II is assumed.

B. M_o and M_s at 300°K

Based upon the experimental values of M_t (at 300°K) shown in Table II[5,6] we have calculated the spin and orbital magnetization for Fe-Co, Fe-Ni, and Co-Ni. The values of M_0 (at 300°K) are shown in Table III, together with the relative error. The relative error in our reported M_s values is, at most, slightly over 2%. A plot of M_s, as a function of effective atomic number, is shown in Fig. 2.

C. Kittel–Van Vleck g-g' Relationship

The value of $g'^{-1} + g^{-1}$ has been calculated for Fe-Co, and Fe-Ni alloys as a function of Z, as given in Table III and Fig. 3. The values of g are from the ferromagnetic resonance measurements of Meyer and Asch.[7] While g values are available for Co-Ni, they are not known at exactly the same concentrations as Scott's g' values (except $Z = 27.5$) and, because of the anomalous behavior of g' and g values as a function of Z for this alloy, interpolated values would be questionable. At $Z = 27.5$, the sum $g^{-1} + g'^{-1}$ for Co-Ni has the value 0.999. Fe-Ni alloys have a maximum deviation from unity of 0.1%, and for Fe-Co the maximum deviation is less than 1%.

The deviation from the expected value (dotted line) for $g^{-1} + g'^{-1}$ might possibly be correlated with the degree of crystalline anisotropy. We note the largest deviation from the Kittel–Van Vleck expression for Fe-Co appears at $Z = 26.5$, and it is at this value a superlattice is formed. The fit for Fe-Ni is better than expected.

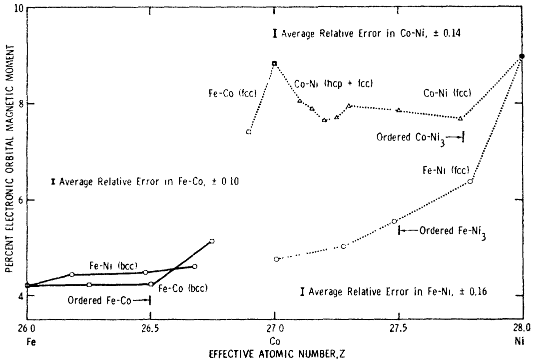

FIG. 1. Percent orbital magnetization ($M_0/M_t \times 10^2$), as a function of the effective atomic number. Note: $[(M_s/M_t) \times 10^2 = (1 - M_0/M_t) \times 10^2]$.

DISCUSSION

A. Orbital Magnetization

The orbital magnetic moment obtained for iron $(0.0918 \pm 0.0033)\ \mu_B$, differs from the value of $0.081\mu_B$ reported by Meyer and Asch.[7] This is readily understood, since their value for g differs by 0.017 $(2m/e)$ from that which we calculate using the Einstein–de Haas data and Eq. (8). The calculated orbital magnetic moment obtained for nickel, $(0.0507 \pm 0.0027)\mu_B$, and cobalt, $(0.1472 \pm 0.0034)\mu_B$, compares favorably with the values of 0.053 and $0.133\mu_B$ reported by Meyer and Asch. Meyer and Asch do not estimate their probable errors. Our results (Fig. 1) give more accurately the

TABLE I. Percent orbital magnetization for binary Fe, Co, and Ni alloys, as obtained from Scott's Einstein–de Haas gyromagnetic-ratio measurements.

Wt %			Effective		
Fe	Co	Ni	Z	g'[a]	$10^2 M_0/M_t$
100	0	0	26.00	1.919±0.002	4.22±0.12
90	0	10	26.18	1.915±0.004	4.44±0.22
75	25	0	26.24	1.918±0.002	4.28±0.09
75	0	25	26.48	1.914±0.004	4.49±0.22
50	50	0	26.49	1.916±0.002	4.38±0.09
65	0	35	26.68	1.912±0.002	4.60±0.09
25	75	0	26.74	1.902±0.002	5.15±0.10
10	90	0	26.90	1.862±0.002	7.41±0.15
50	0	50	26.98	1.908±0.004	4.80±0.24
0	100	0	27.00	1.838±0.002	8.81±0.09
0	90	10	27.10	1.851±0.003	8.05±0.16
0	85	15	27.15	1.854±0.002	7.88±0.08
0	80	20	27.20	1.858±0.003	7.64±0.15
0	75	25	27.25	1.857±0.002	7.70±0.15
35	0	65	27.28	1.904±0.004	5.04±0.20
0	70	30	27.30	1.853±0.002	7.93±0.08
25	0	75	27.48	1.895±0.004	5.54±0.22
0	50	50	27.51	1.846±0.003	8.34±0.16
0	25	75	27.75	1.849±0.002	8.17±0.08
10	0	90	27.79	1.880±0.006	6.38±0.32
0	0	100	28.00	1.835±0.002	8.92±0.09
Averages			Fe-Ni	±0.003	±0.16
			Fe-Co	±0.002	±0.10
			Co-Ni	±0.003	±0.14

[a] Values of the gyromagnetic ratio determined by Scott *et al.*, Ref. 1.

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[5] P. Weiss and R. Forrer, Ann. Phys. (Paris) **12**, 297 (1929).

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[7] A. J. P. Meyer and G. Asch, J. Appl. Phys. **32**, 3305 (1961).

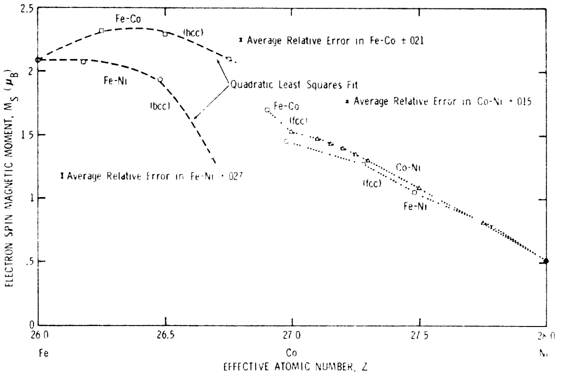

FIG. 2. Spin magnetization of binary Fe, Co, and Ni alloys at 300°K as a function of the effective atomic number. A least-squares fit of the bcc region for Fe-Co and Fe-Ni lead to the expressions $M_s(\text{Fe-Co}) = -1222.93 + 92.927Z - 1.7620Z^2$ and $M_s(\text{Fe-Ni}) = -2255.78 + 172.431Z - 3.2920Z^2$ with standard errors of ±1% for M_s(Fe-Co) and +5% for M_s(Fe-Ni). The ratio of the coefficients in these two expressions are remarkably alike:

Term	Ratio
0	1.845
1	1.851
2	1.860

amount of unquenched orbital contribution to the magnetization for Fe-Co and Fe-Ni alloy, the percent of orbital contribution increasing from 4.2 in Fe to 8.8 in Co to 8.9 in Ni.[2] The behavior of the orbital magnetic moment as a function of Z seems to be quite different for different crystal structures.[8] In the bcc structure of Fe-Co and Fe-Ni, the orbital magnetic moment changes less rapidly with concentration than it does in the fcc structure.

TABLE II. Related magnetic properties of binary Fe, Co, and Ni Alloys.

Wt %					
Fe	Co	Ni	g^a	M_t (μ_B)	Structure
100	0	0	2.091	2.175±0.01	bcc
90	0	10	2.096	2.170±0.02	bcc
75	25	0	2.080	2.423±0.01	bcc
75	0	25	2.100	2.025±0.02	bcc
50	50	0	2.090	2.393±0.01	bcc
65	0	35	2.102	1.386±0.02	bcc (?)
25	75	0	2.123	2.105±0.01	bcc
10	90	0	2.158	1.826±0.01	fcc
50	0	50	2.103	1.520±0.02	fcc
0	100	0	2.187	1.670±0.01	fcc (quenched)
0	90	10		1.604±0.01	hcp
0	85	15		1.559±0.01	hcp
0	80	20		1.513±0.01	hcp
0	75	25		1.463±0.01	fcc
35	0	65	2.100	1.341±0.02	fcc
0	70	30		1.413±0.01	fcc
25	0	75	2.097	1.115±0.02	fcc
0	50	50		1.182±0.01	fcc
0	25	75		0.887±0.01	fcc
10	0	90	2.135	0.844±0.02	fcc
0	0	100	2.183	0.569±0.01	fcc

[a] Interpolated values from Ref. 7.

TABLE III. Electronic spin and orbital magnetization.

Wt %					
Fe	Co	Ni	$g^{-1}+g'^{-1}$	M_0 300°K (μ_B)	M_s 300°K (μ_B)
100	0	0	1.009	0.0918±0.0033	2.083±0.023
90	0	10	0.9994	0.0962±0.0076	2.070±0.046
75	25	0	0.9986	0.1036±0.0037	2.319±0.026
75	0	25	0.9985	0.0910±0.0062	1.934±0.043
50	50	0	0.9920	0.1047±0.0037	2.288±0.025
65	0	35	0.9987	0.0638±0.0028	1.322±0.028
25	75	0	0.9930	0.1085±0.0034	2.096±0.021
10	90	0	0.9980	0.1353±0.0035	1.691±0.017
50	0	50	0.9987	0.0729±0.0048	1.447±0.032
0	100	0	1.002	0.1472±0.0034	1.523±0.015
0	90	10		0.1291±0.0041	1.475±0.018
0	85	15		0.1228±0.0031	1.436±0.018
0	80	20		0.1156±0.0038	1.397±0.026
0	75	25		0.1127±0.0028	1.350±0.015
35	0	65	1.001	0.0676±0.0043	1.273±0.028
0	70	30		0.1121±0.0028	1.301±0.015
25	0	75	1.005	0.0618±0.0037	1.053±0.024
0	50	50	0.998	0.0986±0.0031	1.083±0.013
0	25	75		0.0725±0.0018	0.815±0.009
10	0	90	0.999	0.0539±0.0027	0.790±0.019
0	0	100		0.0508±0.0012	0.518±0.006
Average			Fe-Ni	±0.0047	±0.027
			Fe-Co	±0.0036	±0.021
			Co-Ni	±0.0029	±0.015

Considerable complication arises in the properties of the Co-Ni alloys. In the region $27 < Z < 27.3$, the M_0/M_t curves (Fig. 1) show unexpected variations as a function of Z. It is tempting to assign this variation to the fact that, owing to the low phase-transition temperature, samples with this composition may be a mixture of hcp and fcc regions. Evidence against this is that pure cobalt in both hcp and fcc phases has the same g' value.[1] Nevertheless, it might be expected that the nickel atom affects the M_0/M_t value differently when it is present in the hcp structure than when it is in the fcc structure.

B. Spin Magnetization

Crangle and Hallam[9] have discussed at some length various aspects of the magnetization of fcc and bcc

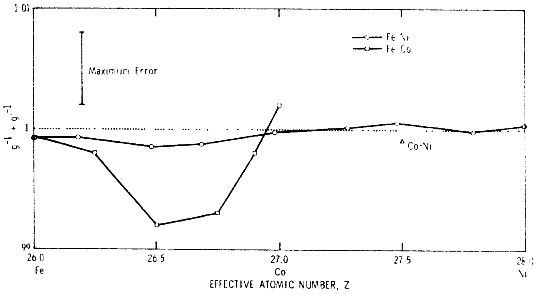

FIG. 3. Exactness of the g-g' relationship for binary Fe, Co, and Ni alloys.

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iron-nickel alloys. We have obtained results similar to theirs for the concentration dependence of the spin magnetic moment. For the bcc phase, M_s for both Fe-Co and Fe-Ni may be expressed as a quadratic function of Z, where the ratios of the coefficients for the two alloys is constant at approximately 1.85 (see caption of Fig. 2). The values of M_s for the fcc region are linear in Z with slope -1.01 per electron added, in reasonable agreement with the values reported by Meyer and Asch[7] (-1.05). The intercept $M_s=0$ occurs at $Z=28.55$.

The results for the spin magnetization in the bcc region previously reported by Crangle and Hallam[9] are difficult to interpret. Their value seems too high, since, if we assume the magnetic moments are distributed among the atoms in the same manner before and after alloying (and consider that each atom acts individually), their results would require 3.2 ± 0.1 magnetic electrons to be carried by the nickel atom to be consistent with the initial value of the slope. Our result reduces this number to $(1.1\pm0.1)\mu_B$ per atom.

RELATIONSHIP TO THEORETICAL CALCULATIONS

Using a band model, Mott[8] has calculated the total number of "holes" in iron to be 2.9 per atom, on the basis of a value of $2.1\mu_B$ for unbalanced spin with an orbital contribution of $0.12\mu_B$. Our value of M_0 of $0.092\mu_B$ changes Mott's results to 2.93 holes. This change will be significant when and if the uncertainty in the value of the polarization of the d bands is reduced.

Note added in proof. A. J. P. Meyer has pointed out in a private communication to the authors that the values of M_s and M_0 from Meyer and Asch[7] were obtained using M_t values for 0°K, whereas the M_0 and M_s reported here were obtained using values of M_t for 300°K.

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Theory of Ferro- and Antiferromagnetic Resonance in Solids Containing Ions of Complicated Level Structure*

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(Received 13 January 1969)

The time-dependent molecular-field (TDMF) approach to magnetic resonance is described and illustrated with six examples. For the simplest models, the random-phase approximation of TDMF is equivalent to Tyablikov spin waves. However, whereas Green's function and spin-wave techniques are forced into same-site decorrelation approximations when confronted with complicated single-ion dynamics, TDMF is general enough to include any single-ion terms without difficulty. Ions appear as coupled oscillators whose allowed transitions are pulled in forming collective modes. The procedure, being quite general, is readily automated and is thus convenient in actual calculations for physical problems.

I. INTRODUCTION

There is an enormous body of literature concerning excitations from ordered magnetic configurations.[1] Much of it aims at the difficult many-body aspects, restricting attention to simple Hamiltonians.[2] When the temperature is not zero, many methods are for static properties only.[3] For the dynamics of Hamiltonians containing single-ion terms, that is, complicated Hamiltonians, at finite temperatures there are few available techniques.[4,5] The best of these may be called the "time-dependent molecular-field" (TDMF) decoupling scheme. This paper is a description of that technique.

The TDMF has been used for treating rare-earth ions in the iron garnet,[6] but apparently its generality has not been widely appreciated.[7] The method can, within the same approximation, treat Hamiltonians consisting of exchange[8] plus one-ion terms of *arbitrary complexity.*

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[*] Part of this work was done at Osaka University, Toyonaka, Japan, under the auspices of the U. S.-Japan Cooperative Science Program, and part was done at Yale University with partial support from the National Science Foundation.

[1] See F. Keffer, in *Handbuch der Physik*, edited by H. P. J. Wijn (Springer-Verlag, New York, 1966), Vol. XVII/2, p. 28 ff.; S. Foner, in *Magnetism*, edited by G. Rado and H. Suhl (Academic Press Inc., New York, 1963), Vol. I, p. 383 ff, for many references.

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[8] The present work considers the common case of bilinear exchange. More complicated couplings may be handled by TDMF although at a considerable loss of efficiency.