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Reprinted from the
JOURNAL OF SCIENCE OF THE HIROSHIMA UNIVERSITY
Series A-II, Vol. 34, No. 3, DECEMBER, 1970

J. Sci. Hiroshima Univ., Ser. A-II
Vol. 34, No. 3, pp. 259~269 Dec. 1970

A Method of Measurements of the Magnetic Moment Under Hydrostatic Pressures

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(Received September 21, 1970)

A method for measuring the magnetic moment under hydrostatic pressures is presented concerning the apparatus and procedures. The correction of the apparent demagnetizing field in measurements of the saturation flux under pressures is principally discussed and the experimental treatment of the correction is described. Finally, some examples of the measurements so far done are briefly reviewed.

Introduction

On the basis of the molecular field theory, the pressure dependence of the magnetic moment per unit mass σ_s and the Curie temperature T_c, $(\partial\sigma_s/\partial p)$ and $(\partial T_c/\partial p)$, which are both expectable to be gotten experimentally, would provide a knowledge of the exchange interaction responsible for ferromagnetism.[1]

Theoretically, it is required to have $(\partial\sigma_{so}/\partial p)$ at 0°K, not $(\partial\sigma_s/\partial p)$ at a temperature. Experimentally, however, $(\partial\sigma_{so}/\partial p)$ is practically impossible to measure, but it can be estimated from the temperature dependence of $(\partial\sigma_s/\partial p)$ which may be derived from thermodynamical consideration, if the measurement of $(\partial\sigma_s/\partial p)$ can be made at several temperatures.[2,3]

The measurement of $(\partial\sigma_s/\partial p)$ under hydrostatic pressures, done by Nagaoka and Honda for the first time,[4] has so far done by means of various techniques.[5~11] Greater part of the data, however, are unavailable for estimating $(\partial\sigma_{so}/\partial p)$, since the measurement has been made only at one or a couple of temperatures. Tatsumoto and collaborators[12~16] have made the measurement over a comparatively wide temperature range by means of a method developed by the present authors.

According to the measurements so far done, $(\partial\sigma_s/\partial p)$ has mostly been deduced from the measurement of saturation flux Φ_s under pressures, so that it is necessary to make an accurate measurement of Φ_s under pressures, since the variation of Φ_s with pressure is usually very small, and it is also necessary to make correction for the demagnetizing field, since the specimen is usually of finite size.

In the present paper, a method which has been applied to the measurement on Ni, Fe,[12~14] ferromagnetic Cu-Ni[15] and Pd-Ni[16] alloys is presented concerning the apparatus, procedures and examples. This method was developed by the present authors so as to make the measurement of the variation of magnetic flux in an electromagnet capable of generating a high field enough to magnetize the hard material to saturation and in a comparatively wide temperature range. In this measurement, the sample is usually required to be so small as to be placed between the pole pieces of an electromagnet. It is therefore necessary to make a correction for the demagnetizing field, because of a comparatively small dimension ratio of the sample.

For 24 at.% Cu–Ni alloy which is magnetically soft, one of the present authors Tange[17] has succeeded in the same measurement by using the samples in comparatively long size in a solenoidal coil. In his measurement, therefore, the correction for the demagnetizing field was negligibly small.

Experimental

The measurement of the pressure effect on σ_s in the present experiment is composed of the measurements of the pressure effect on Φ'_s and of the linear compressibility. Here, Φ'_s is the saturation flux actually picked up by a search coill.

For each material, three cylindrical rods have been prepared from an ingot after severe hot-forging. Two of the three rods, 14.5 mm in length and 5.5mm in diameter, are used for the measurement of the pressure effect on Φ'_s, and the remaining one, 38 mm in length and 1.7 mm in diameter, is used for that of the linear compressibility.

Prior to the measurements, all specimens are well annealed at a suitable temperature in order to remove stress and to get homogenous small grain size.

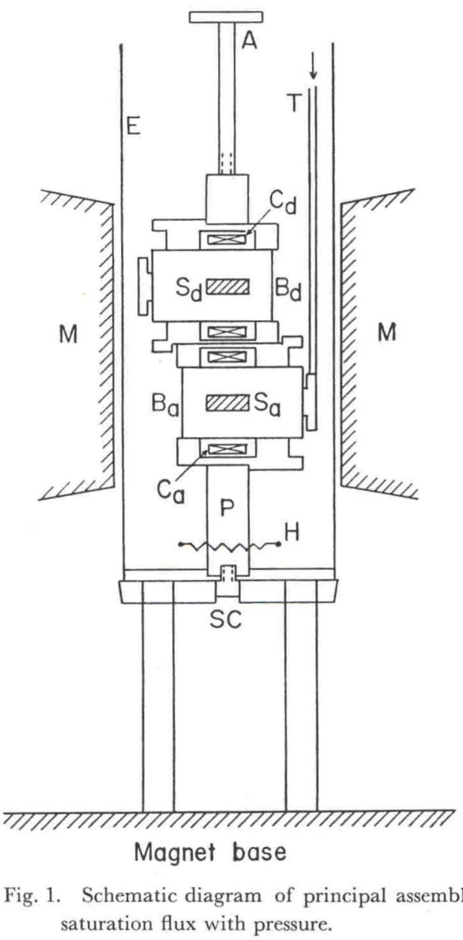
Hydrostatic pressures have been generated with a slightly modified Bridgman press which was made of nickel-chromium-molybdenum steel. The press bore the pressure test up to 18 kbar. Pressures up to 15 kbar have easily been generated by using petroleum ether as a transmitting medium. The imported stainless steel tube, 0.6 mm in inner diameter and 3mm in outer diameter, has been employed as a pressure transfer between the pressure generator and the pressure bomb, in which the specimen was inserted.

Details of the construction of the pressure generator and the technique of the connection between the generator and the pressure bomb are referred to the article by Tatsumoto et al.[18]

The pressure has been determined from the pressure dependence of the electrical resistance of a well annealed manganin wire, the standard pressure of which was calibrated with the freezing pressure of mercury at 0°C, 7.640 kg/cm^2.

In Fig. 1, the principle of the measurement of the variation of Φ'_s with pressure is schematically shown. In this figure, M is an electromagnet, the pole gap and the diameter of the pole surface being 80 mm and 100 mm, respectively. Two same pressure bombs B_a and B_d are made of precipitation hardened beryllium copper, being 42 mm in length, 5.5 mm in inner diameter and 25 mm in outer diameter. The bomb bore the strength test up to 18 kbar, but the highest working pressure which has usually been applied is 15 kbar. The same specimens are inserted in both the bombs. The lower bomb B_a is used as an active one, suffixed with a, in which the pressure is applied, and the upper bomb B_d as a dummy, suffixed with d, in which no pressure is applied.

The detailed construction and arrangement of the bomb assembly are schematically shown in Fig. 2, where the active bomb is represented. The bomb is fixed with six screw S_1–S_6, three at the one side of the bomb, to the coil holder CH made of copper. In the figure only S_1, S_2, S_4 and S_5 are



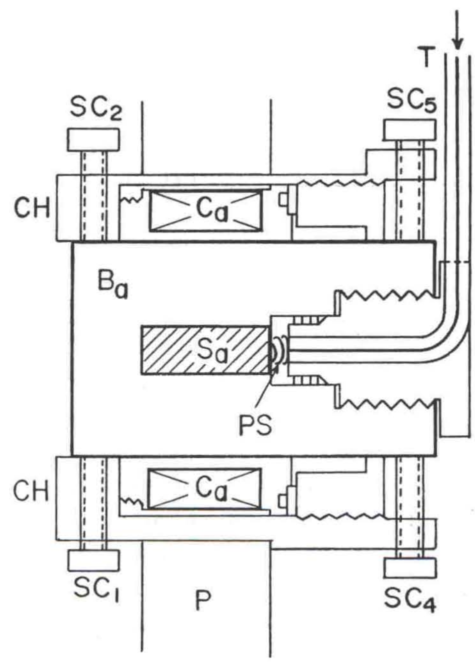


Fig. 2. Diagram of the active pressure bomb
B_a: Active bomb, S_a: Active specimen,
C_a: Active search coil, CH: Coilholder,
PS: Phosphor bronze spring,
SC_1, SC_2, SC_4 and SC_5: Screws,
P: Holding plate, T: Transfertube.

Fig. 1. Schematic diagram of principal assembly of apparatus for the measurement of the variation of saturation flux with pressure.
M: Electromagnet, B_a and B_d: Active and dummy bomb,
S_a and S_d: Active and dummy specimen,
C_a and C_d: Active and dummy search coil,
P: Holding plate, H: Heater, E: Stainless steel bath, T: Transfer tube, A: Adjusting screw,
SC: Screw.

shown. The coil holder, in which the active coil C_a is mounted, is fixed to the thick holding plate P, which is also made of copper. The active specimen S_a is slightly pressed against the left inside wall of the bomb with a phosphor bronze spring PS inserted between the specimen and the plug, so that it may be unmovable by the application of the magnetic fields. A stainless tube T for transferring the pressure from the generator is connected to the plug of the bomb. The assembly of the dummy bomb is just the same as that of the active bomb, except for the transfer tube.

In Fig. 1, the active bomb assembly is fixed to the holding plate P, while the dummy assembly is slightly movable up and down with an adjusting screw A, so as to vary the space between the active and dummy bombs. The holding plate is tightly fixed with a screw SC to the bottom of the stainless steel bath E which is mounted to the base of the magnet, as shown in Fig. 1.

The coils C_a and C_d were wound on a thin bobbin in the same way with the same total turn number. Their length, effective diameter and total turn number are 13 mm, 42 mm and 3,000, respectively. The bobbins were made of copper and bakelite. The copper bobbin was used for the measurement below room temperature and the bakelite bobbin for the measurement above it.

The active and dummy coils are set parallel to each other and they are connected in series and opposite sense, so that no resultant flux might be induced in the absence of pressures. In practice, however, the state of complete cancellation was not a stable one. A practically stable state could be obtained as the one with a small resultant flux resulting from the adjustment of the space between the active and dummy bomb assemblies.

The flux has been determined by means of a ballistic galvanometer, and lamp and scale, and given as a deflection occurring in the reversal of an applied magnetic field strong enough to magnetize the specimen to saturation. The reversal of the field could be done instantly with that of a mechanical switch. The change in Φ'_s, $\Delta\Phi'_s$, which is caused by an increase in pressure, Δp, is determined from the difference between the deflections of galvanometer in the absence and the presence of a pressure. One run of this measurement is composed of three steps: (i) under no pressure, (ii) applied pressure and (iii) again no pressure as is shown in Fig. 3. In this figure, the abscissa is the time and the ordinate is the deflection angle of galvanometer. At each step, several measurements were done at regular time intervals, as shown in the figure. The suitable interval was determined in advance, so as to get the stable base in the absence of pressure as shown in the figure. After making one run measurement, $\Delta\Phi'_s$ has been obtained from the deflection of the galvanometer, $2\Delta\theta$.

The cancellation of the fluxes in the coils C_a and C_d is scarcely changed by the fluctuation of temperature and field, since the active and dummy assemblies are just the same constitution and also at almost the same condition,

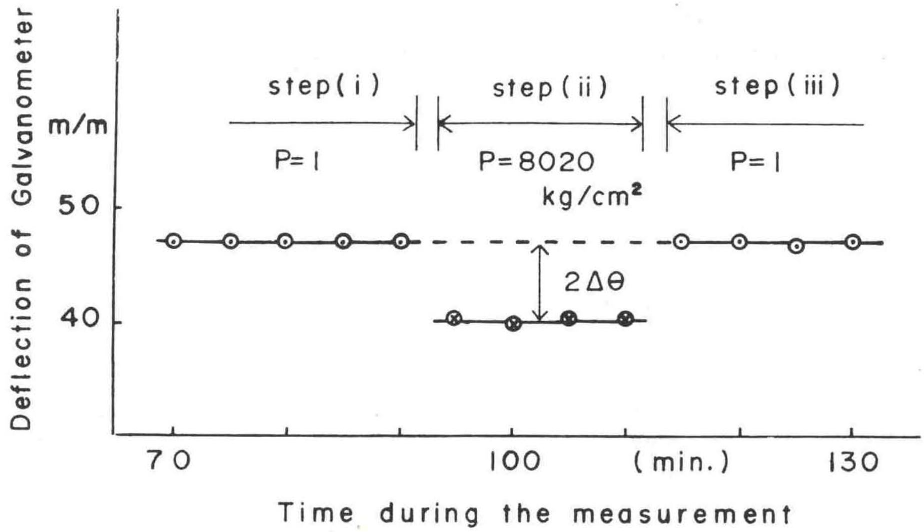


Fig. 3. A practical example of the results for obtaining $\Delta\Phi_s'$.
(Specimen Ni, Temperature $-73°C$ and magnetic field 5,100 Oe)
step (i): before the pressure is applied.
step (ii): under an applied pressure 8020 kg/cm^2.
step (iii): after the pressure is released.
Between the step (i) and (ii) the pressure is applied and between the step (ii) and (iii) the pressure is released.

so that a stabilized measurement was successfully accomplished in the whole course.

The measurement of Φ_s' itself, required to get the pressure coefficient of Φ_s', $\Phi_s'^{-1}(\Delta\Phi_s'/\Delta p)$, has been made in the same magnet using a search coil with 100 turns. The effective diameter and length of the coil are just the same as those of the coil C_a used in the measurement of $\Delta\Phi_s'$, in order to make the measurements of Φ_s' and $\Delta\Phi_s'$ in the same condition.

The measurements of $\Delta\Phi_s'/\Delta P$ and Φ_s' have been done at $-73°C$, $0°C$ and several points in a range from $0°C$ to $100°C$. The former two temperatures are obtained with stirring fans by the mixture of powdered dry ice and ethyl alcohol and that of erashed ice and water, respectively. The latter several constant temperatures were maintained in an oil bath with stirring fans and with a non-inductive heater (H in Fig. 1) made of nichrome wire, which was mounted at the bottom of the bath, as shown in Fig. 1.

The compressibility necessary for deriving the pressure effect on σ_s has also been measured at the same temperatures. This measurement was done with a compressimeter developed by Tatsumoto et al.[19]

Pressure effect on the saturation flux

The saturation flux Φ_s due to the saturation magnetization M_s in a specimen magnetized to saturation may ideally be picked up in a search coil wound directly on the cylindrical specimen of infinite length and is given by

$$\Phi_s = 4\pi M_s q n \tag{1}$$

with

$$M_s = D\sigma_s \tag{2}$$

Here, q, D and n are the cross sectional area, the density of the specimen and the total turn number of the coil, respectively. In this case, the cross sectional area q of the specimen may be assumed to be that of the search coil since the coil is wound directly, unless the thickness of the coil is large.

On differentiating Eq. (1) with respect to pressure p, the following familiar relation is obtained

$$\sigma_s{}^{-1}(\partial\sigma_s/\partial p) = \Phi_s{}^{-1}(\partial\Phi_s/\sigma p) - K/3, \tag{3}$$

where K is the volume compressibility of the specimen $-(1/V)\ (dV/dp)$, and $\sigma_s^{-1}(\partial\sigma_s/\partial p)$ and $\Phi_s^{-1}(\partial\Phi_s/\partial p)$ are pressure coefficient of σ_s and Φ_s, respectively. The second term $K/3$ in the right-hand side is derived from the pressure derivatives of both the cross sectional area q and the density D of the specimen, and this is evidently just equal to the linear compressibility of the specimen.

The measurement of the saturation flux is made practically by employing a specimen of finite length. Therefore, the search coil picks up not only Φ_s, but also a field inverse to the applied one, which is produced by the free magnetic poles appearing at both the ends of the specimen. Hereafter, this inverse field is called an apparent demagnetizing field, in a sense of the demagnetizing field generally used. The observed saturation flux Φ_s' picked up by a search coil with cross sectional area A is therefore given by

$$\Phi_s' = 4\pi M_s q n - N M_s A n, \tag{4}$$

where the intensity of the apparent demagnetizing field is assumed to be proportional to M_s everywhere in A, so that it may formally be represented with a proportional constant N, as NM_sAn.

In the case of $A=q$, the constant N introduced here is the so-called demagnetizing constant and is a function of only the dimension of the specimen. In a general case where $A>q$, N is a function of the dimensions of both the specimen and the coil; in other words, N has a constant value for the specimen and the coil given. The value of N, however, becomes smaller as the specimen gets longer in length.

By using Eq. (4), the pressure coefficient of σ_s in Eq. (3) is expressed as

$$\sigma_s^{-1}\left(\frac{\partial\sigma_s}{\partial p}\right)=\Phi_s'^{-1}\left(\frac{\partial\Phi_s'}{\partial p}\right)-\frac{1}{3}K+\frac{1}{4\pi q-NA}\left[\frac{2}{3}KNA+N\frac{\partial A}{\partial p}+A\frac{\partial N}{\partial p}\right]. \quad (5)$$

Comparing Eq. (5) with (3), not only the pressure coefficiet of Φ_s' is replaced for that of Φ_s, but also the 3rd term is added to the right-hand side.

In the present measurement, the coil is placed outside of the pressure bomb, so that it is unaffected by an applied pressure. Accordingly, as the 2nd term $\partial A/\sigma p$ in the brackets in Eq. (5) is zero, Eq. (5) reduces to

$$\sigma_s^{-1}\left(\frac{\partial\sigma_s}{\partial p}\right)=\Phi_s'^{-1}\left(\frac{\partial\Phi_s'}{\partial p}\right)-\frac{1}{3}K+\frac{1}{4\pi q-NA}\left[\frac{2}{3}KNA+A\frac{\partial N}{\partial q}\right]. \quad (6)$$

After rearranging the 3rd term in the bracket, the pressure coefficient of σ_s in the present experiment can be expressed as

$$\sigma_s^{-1}\left(\frac{\partial\sigma_s}{\sigma p}\right)=\Phi_s'^{-1}\left(\frac{\partial\Phi_s'}{\partial p}\right)-\frac{1}{3}K+\frac{1}{3}KC, \quad (7)$$

where C is a constant determined from the dimension of the specimen and the coil given.

Even if the dimensions of the specimen and the coil are changed in the present measurement, Eq. (7) will be generally accepted, but the value of C must be changed. The analytical estimation of C appears difficult except for an ellipsoidal specimen. The value of C, however, can be experimentally obtained in the following way.

In Eq. (7) the correction term $(1/3)KC$ can be neglected, if the specimen used has a large dimension ratio and the measurement can be done in a solenoidal coil. The pressure coefficient of σ_s is therefore obtained from

$$\sigma_s^{-1}\left(\frac{\partial\sigma_s}{\partial p}\right)=\Phi_s'^{-1}\left(\frac{\partial\Phi_s'}{\partial p}\right)_{sol.}-\frac{1}{3}K, \quad (8)$$

where the suffix sol. denotes that $\partial\Phi_s'/\sigma p$ is observed under the condition just mentioned. In the measurement the magnetically soft material is desirable for the specimen, because the solenoidal coil is used which hardly produces a high field.

Tange[17] has made the measurement in the solenoidal coil on magnetically soft material such as 24 at.% Cu-Ni alloy, and proved that Eq. (8) was available for a specimen of 75 mm in length and 5.5mm in diameter, and a search coil of 13 mm in length and 22mm in effective diameter.

In the present measurement, the dimension ratio of the specimen is small, so that Eq. (8) is unavailable. Substracting Eq. (8) from (7), the following relation is obtained

$$\Phi_s'^{-1}\left(\frac{\partial\Phi_s'}{\partial p}\right)_{sol.}-\Phi_s'^{-1}\left(\frac{\partial\Phi_s'}{\partial p}\right)_{mag.}=\frac{1}{3}KC. \quad (9)$$

Here the suffix mag. is used for $\partial\Phi'_s/\partial p$ in Eq. (7) and denotes the observation in an electromagnet with a specimen as in the present measurement. Equation (9) is generally accepted for obtaining the correction term $(1/3)KC$ experimentally, since C is theoretically independent of the saturation magnetization M_s of the specimen employed. Here it is noted that both $\Phi_s'^{-1}(\partial\Phi'_s/\partial p)_{\rm sol.}$ and $\Phi_s'^{-1}(\partial\Phi'_s/\partial p)_{\rm mag.}$ are dependent on the material of the specimen.

In the actual determination of the constant C in Eq. (9), Cu-Ni alloys have been employed, because some data of the relation between the apparent demagnetizing field and the dimension ratio of specimen and coil have already been obtained in the measurement by Tange. The determination of C has been made at $-73°C$ on the three specimens, 13, 18 and 24 at. % Cu-Ni alloys, so that a reliable value of C could be obtained as an average. The result showed that C was independent of the saturation magnetization M_s of the specimen employed as theoretically expected. The reason why the measurement was done at $-73°C$ is that the most stable measurement could be made at the temperature. The value of C thus obtained is 0.73 ± 0.03.

As referred to in section 1, many investigators have measured the pressure effect on Φ_s and some of them will be briefly reviewed from the view point of the apparent demagnetizing field caused by free poles appearing at both the ends of the specimen. Ebert et al.[5] have measured in the electromagnet and set the coil outside of the pressure bomb. The effective diameter of the coil was larger than that of the specimen and also the length of the specimenwas relatively short, but he was not careful of the apparent demagnetizing field. Knodorskii et al.[7] have measured in the solenoid and the specimen was 112 mm in length and 5.9 mm in diameter, respectively. Therefore, the influence of the apparent demagnetizing field seems to be negligible. Kouvel et al.[9] tried to reduce the demagnetizing field by making the specimen to be a part of the closed magnetic circuit.

Examples and discussions

The pressure coefficient of Φ'_s, $\Phi_s'^{-1}(\partial\Phi'_s/\partial p)$, is obtained from the measurements of $\Delta\Phi'_s$ and Φ'_s which were described in section 2.

The values of $\Delta\Phi'_s$ or $\Delta\Phi'_s/\Phi'_s$ observed at a temperature were almost linear with pressure over the pressures applied. The results for Fe and Ni in ref. (14) are again cited in Fig. 4 as an example. The values of $\Delta\Phi_s/\Phi_s$ in Fig. 4 are not $\Delta\Phi'_s/\Phi'_s$ which have been discussed in the present paper, but $\Delta\Phi'_s/\Phi_s$ in which the necessary correction to Φ_s is not made. The corrected value of $\sigma_s^{-1}(\partial\sigma_s/\sigma p)$ for Fe and Ni obtained from Eq. (7) with the value of C mentioned, is again plotted as a function of reduced temperature T/T_c in Fig. 5 which has been previously published.[2]

There is not any remarkable difference between the results in Fig. 5 and those of uncorrected ones[14] except for the absolute value.

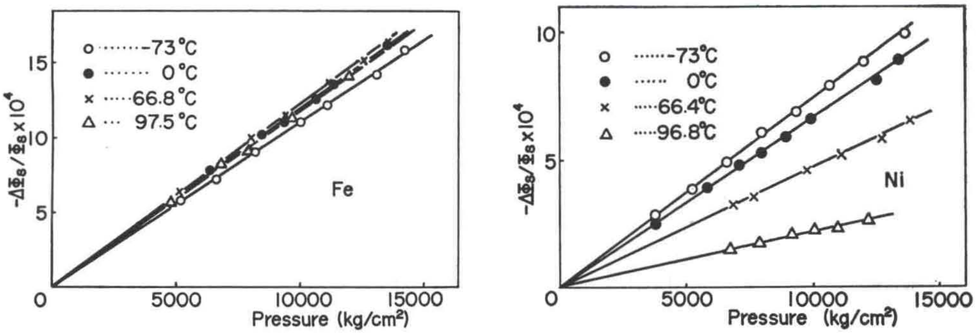


Fig. 4. A plot of $\Delta\Phi_s/\Phi_s$ for Fe and Ni as a function of pressure at four temperatures reproduced from reference (14).
The difference between $\Delta\Phi'_s/\Phi'_s$ in the present paper and $\Delta\Phi_s/\Phi_s$ in this figure is noted in the text.

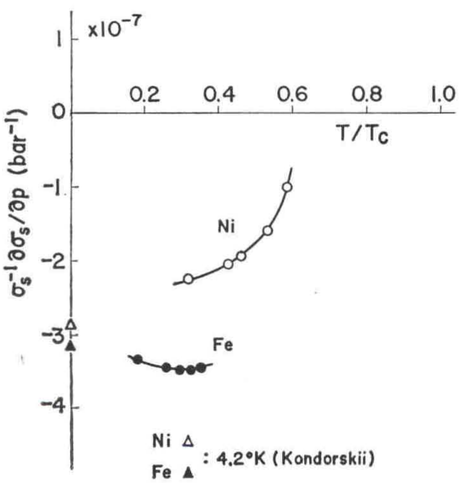


Fig. 5. A plot of $\sigma_s^{-1}(\partial\sigma_s/\partial p)$ as a function of reduced temperature TeT_c for Fe and Ni reproduced from reference (2).

In the results for Cu-Ni[15] and Pd-Ni[16] alloys, the correction described in section 2 has already been made.

From the value of $\partial\sigma_s/\partial p$ thus obtained over a wide temperature range, that of $\partial\sigma_{so}/\partial p$ has been estimated from thermodynamical consideration,[2] and the detailed procedures and discussion have been reported.[3] The pressure effect on σ_{so} would essentially provide a knowledge of the exchange interaction responsible for ferromagnet ism together with the pressure effect on T_c. Moreover, the pressure effect on σ_s at a temperature provides a direct contribution to the analysis of some magnetic properties of ferromagnetic metals and alloys. In the present paper, two examples will briefly be discussed. (i) Forced volume magnetostriction: The pressure effect on σ_s is combined with the forced volume magnetostriction $\partial\omega/\partial H$ in the following relation

$$\partial\omega/\partial H = -D\partial\sigma_s/\partial p. \tag{10}$$

Apart from the detailed analysis and discussion on the forced volume magnetostriction, the relation (10) has been used as a quantitative check of the pressure effect on σ_s, and vice versa. Precise measurements of the forced volume magnetostriction over a wide temperature range have recently been made by Tange et al.[20] for Ni, and their results are in fair agreement with those obtained from the pressure effect on σ_s obtained from Eq. (7) by the present authors. (ii) Linear compressibility: The linear compressibility, which appeared in Eq. (3) or Eq. (7), is one of the valuable quantities which reflects the exchange interaction, as clearly pointed out by Ishida.[21] The value of $\partial\sigma_s/\partial p$ is necessitated for the theoretical estimation of the linear compressibility based on a molecular field theory made by Ishida. The agreement between the calculated and observed values of the linear compressibility was fairly good for Cu-Ni[21] and Ni-Pd alloys.[22]

Acknowledgements

The present authors wish to express their hearty thanks to Mr. K. Kusumoto, an executive director of Toyo Kogyo Co., Ltd., for preparing the several kinds of pressure cylinders. The appreciations are also due to Prof. T. Kamigaichi, Dr. Y. Kato and other members who have engaged in the present work. The present work has been supported by the scientific research grant from the Ministry of Education of Japan.

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