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A Note on Pressure Effect on the Magnetic Moment

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where  $\alpha$  and  $\kappa$  are the linear thermal expansion coefficient and the volume compressibility, respectively.

The term  $(3\alpha T/\kappa) T_c^{-1}(\partial T_c/\partial p)$  in eq. (1) is practically small in comparison with 1, so that it may be neglected. Then eq. (3) reduces to

$$\sigma_s^{-1}(\partial \sigma_s/\partial p) = \sigma_{so}^{-1}(\partial \sigma_{so}/\partial p) - (T/\sigma_s)(\partial \sigma_s/\partial T)T_c^{-1}(\partial T_c/\partial p).$$
(2a)

Hereafter,  $\sigma_{so}^{-1}(\partial \sigma_{so}/\partial p)$ ,  $T_c^{-1}(\partial T_c/\partial p)$  and  $\sigma_s^{-1}(\partial \sigma_s/\partial T)$  in eq. (2a) will be denoted as  $C_1$ ,  $C_2$  and G(T), respectively, in order to simplify the notations. Then eq. (2a) is simply expressed as

$$F(T) = C_1 - C_2 T G(T), \qquad (2b)$$

where F(T) is  $\sigma_s^{-1}(\partial \sigma_s/\partial p)$ .

The discussions will be made on the basis of eq. (2b).

(I) As is pointed out in a previous section, the values of  $C_1$  and  $C_2$  should be required for investigating the pressure effect on the exchange interaction responsible for ferromagnetism, in either case where this investigation will be made on the basis of the localized electron model or of the collective electron model.

I<sub>a</sub>: The values of  $C_1$  and  $C_2$  can be evaluated from more than two observation equations like (2b) constructed by using the observed values of F(T) and G(T) at more than two temperatures. From this point of view, F(T), the temperature dependence of  $\partial \sigma_s / \partial p$ , is considered as worthwhile to investigate. I<sub>b</sub>: The measurement of  $\partial \sigma_s / \partial p$  at a single temperature can determine, from eq. (2b), either of  $C_1$  or  $C_2$ , but only when the other is known.

(II) As is found from eq. (2b), the function F(T), which is the basic observable quantity in the present discussion, varies with temperature as does G(T) which is determined by the functional form of  $\sigma_s(T)$ .

At low temperatures, approximately  $T < T_c/5$ , the spontaneous magnetization  $M_s$  observed has been satisfactorily represented from the spin wave theory by  $M_s = M_{so}(1 - AT^{3/2})$  with such a numerical constant A as of the order of  $10^{-6}$  deg.<sup>-3/2</sup> for Ni and Fe, for example. Here,  $M_{so}$  is the magnetization at 0°K. Then F(T) is given by

$$F(T) = C_1 + C_2 T \left( \frac{3}{2} \cdot \frac{A T^{1/2}}{1 - A T^{3/2}} - 3\alpha(T) \right), \tag{3}$$

where the relation  $M_s = \rho \sigma_s$  has been used. In the bracket in eq. (3), the 2nd term  $3\alpha(T)$  is practically small in comparison with the 1st term.

In the neighborhood of  $T_c$ ,  $M_s$  varies with T in accordance with  $(1-(T/T_c)^2)^{1/2}$  in the collective electron theory by Stoner<sup>6)</sup> or with  $((T_c-T)/T)^{1/2}$  in the molecular field theory. Then F(T) is given by

$$F(T) = C_1 + C_2 \frac{1}{2(1 - T/T_c)}$$
 in the collective electron theory. (4a)

$$=C_1+C_2\frac{(T/T_c)^2}{1-(T/T_c)^2}$$
 in the molecular field theory. (4b)

Although the term  $3\alpha$  should be introduced in eqs. (4a) and (4b) as in the similar form as in eq. (3), this term has been neglected due to its smallness. The second term on the right-hand side in eq. (4a) is larger than that in eq. (4b), but the difference between them is about 14% at  $T/T_c=0.9$  and it decreases as  $T/T_c$  increases.

In the intermediate temperature range,  $M_s$  decreases with temperature parabolically rather than  $T^{3/2}$  law.

Then it is found that F(T) is a monotonically increasing function with concave upward or decreasing with concave downward according as  $C_2 > 0$  or  $C_2 < 0$ . Moreover, |F(T)| becomes larger in the neighborhood of  $T_c$ . If  $C_2 = 0$ , then F(T) is independent of temperature.

On the basis of these discussions, the form of F(T) versus temperature curve may qualitatively be expected and six types of F(T) curves thus expected are schematically shown in Fig. 1 against the reduced temperature  $T/T_c$ . The curves  $A_1, A_2, \ldots$  in this figure correspond to the following cases:

$$C_1 > 0 ext{ and } C_2 > 0 : A_1,$$
  
= 0 :  $A_2,$   
< 0 :  $A_3,$   
 $C_1 < 0 ext{ and } C_2 > 0 : B_1,$   
= 0 :  $B_2,$   
< 0 :  $B_3.$ 

One may expect, therefore, that the temperature dependence of the pressure effect on  $\sigma_s$  observed in the form of the pressure coefficient  $\sigma_s^{-1}(\partial \sigma_s/\partial p)$  is given by any one of such curves as shown in Fig. 1 and also that the sign of  $\partial T_c/\partial p$  and  $\partial \sigma_s/\partial p$  is determined from the observed curve of  $\sigma_s^{-1}(\partial \sigma_s/\partial p)$ .

In order to ascertain the relations among the pressure effects on  $\sigma_s$ ,  $\sigma_{so}$  and  $T_c$  mentioned above, the data in previous measurements<sup>1-4)</sup> are useful.

The pressure effect on  $\sigma_s$  has been experimentally derived from the measurement of the pressure effect on the saturation flux and that of the compressibility. Then the pressure coefficient of  $\sigma_s$  is given by<sup>1)</sup>



 $\partial p$ ) as a function of reduced temperature  $T/T_c$ .

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$$rac{1}{\sigma_s}rac{\partial\sigma_s}{\partial p}=rac{1}{{oldsymbol{\varPhi}}_s}rac{\partial{oldsymbol{\varPhi}}_s}{\partial p}-rac{1}{3}\kappa,$$

(5)

where  $\Phi_s$  represents the flux picked up by a search coil wound directly on the specimen magnetized to saturation.

The pressure effect on  $\Phi_s$ , however, is hardly observed as already been pointed out<sup>2,7)</sup> and in previous papers<sup>2-4)</sup>, the pressure coefficient of  $\Phi_s$ ,  $\Phi_s^{-1}(\partial \Phi_s/\partial p)$ , in eq. (5) has been derived from the pressure coefficient of the observable flux  $\Phi'_s$ ,  $\Phi'_{s}^{-1}(\partial \Phi'_{s}^{-1}/\partial p)$ , for which a correction for flux leakage is required. This flux leakage results experimentally from the fact that the diameter of the search coil actually employed is larger than that of the specimen, and a detailed report of which will be made in the near future. The change in  $\Phi'_s$  with a pressure  $\Delta p$ ,  $\Delta \Phi'_s$ , has been measured on polycrystalline specimens of Ni and Fe,<sup>1,2)</sup> ferromagnetic Cu–Ni alloys up to 29 at. % Cu<sup>2,3)</sup> and Pd–Ni alloys up to 82 at. % Pd<sup>4)</sup> at 200°K, 273°K and various points between 273°K and 373°K under hydrostatic pressures up to 15 kbar.

The linear compressibility  $\kappa/3$  in eq. (5) has also been measured for each specimen at the respective temperatures, by utilizing an Advance wire which is usually used for the strain gauge wire. The technique has been developed by Tatsumoto et al.<sup>8)</sup> and has been briefly described.<sup>2)</sup>

Ni: A plot of  $\sigma_s^{-1}(\partial \sigma_s/\partial p)$  observed for Ni, as a function of reduced temperature  $T/T_c$ , is given by solid circles in Fig. 2. From these values of  $\sigma_s^{-1}(\partial \sigma_s/\partial p)$ , the values of  $C_1$  and  $C_2$  are estimated by the use of such observation equations as described in (I<sub>s</sub>) and the results are  $-2.7 \times 10^{-7}$  bar<sup>-1</sup> for  $C_1$ which is plotted with open circle and  $4.8 \times 10^{-7}$  bar<sup>-1</sup> for  $C_2$ .



Fig. 2. A plot of  $\sigma_s^{-1}(\partial \sigma^s/\partial p)$  vs.  $T/T_c$  for Ni. The points  $\bullet$  represent observed values. The points  $\bigcirc$  and  $\triangle$  are the estimated values from observation equations and from eq. (3), respectively.

As is found from this figure,  $\sigma_s^{-1}(\partial \sigma_s / \partial p)$  versus temperature curve belongs to type  $B_1$  in Fig. 1, which expects the negative and positive sign respectively for  $C_1$  and  $C_2$ , and this expectation is verified by the sign of  $C_1$  and  $C_2$  actually obtained. The negative sign of  $C_1$ can also be expected by extrapolating the curve observed back to 0°K.

The pressure effect on the saturation flux has been measured at  $4.2^{\circ}$ K by Kondorskii et al.<sup>9)</sup> and the value of  $C_1$  is  $-2.9 \times 10^{-7}$  bar<sup>-1</sup>. The direct measurements of  $\Delta T_c/\Delta p$ have been made by Patrick,<sup>10)</sup>

Bloch<sup>5)</sup> and Okamoto et al.,<sup>11)</sup> and the values of  $C_2$  observed are 5.6, 5.4 and 5.1 in unit of  $10^{-7}$  bar<sup>-1</sup>, respectively. For both  $C_1$  and  $C_2$ , the dis agreement

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between the estimated value and the observed one is not so remarkable.

The value of  $\sigma_s^{-1}(\partial \sigma_s/\partial p)$  at 100°K ( $T/T_c = 0.16$ ), where no measurement has been made, and also those at 200°K and 273°K may be obtained from eq. (3) in which  $C_1$  and  $C_2$  are the estimated values and A is  $8.6 \times 10^{-6}$  deg.<sup>-3/2,12)</sup> The values thus obtained are plotted with triangles in Fig. 2. At 100°K, the value appears to lie on a curve extrapolated from the observed curve back to 0°K, while at 200°K and 273°K, the values differ from the observed values in such a way as is shown. This result seems to be satisfactory, because eq. (3) derived from the spin wave theory is applicable only at low temperatures and the magnetization falls rather parabolically in the intermediate temperature range, as already mentioned.

Fe: The temperature dependence of  $\sigma_s^{-1}(\partial \sigma_s/\partial p)$  for Fe<sup>1,2)</sup> appears to be type  $B_3$ , although the temperature range actually employed was not wide enough to conclude definitely. The discussion on this point will be made in the next section where the data on the forced volume magnetostriction will be investigated.

**Cu–Ni alloys:** The observed values of  $\sigma_s^{-1}(\partial \sigma_s/\partial p)$  and the estimated value of  $C_1$  for 13.3 at. % and 23.8 at %. Cu–Ni alloys are plotted in Fig. 3. Two curves belong to type B<sub>1</sub>, but the curve for 23.8% Cu–Ni is more typical, because the Curie temperature of that specimen lies in a temperature range capable of the measurement of the pressure effect on  $\sigma_s$ . The curve for Ni shown in Fig. 2, therefore, will take the similar form to that of 23.8% Cu–Ni alloy in Fig. 3, in case where the measurement could be made up to  $T_c$  for Ni.

Since  $\Delta T_c/\Delta p$  has been measured on Cu-Ni alloys<sup>11</sup>,  $C_1$  may also be obtained directly from eq. (2b) in such



Fig. 3. A plot of temperature dependence of  $\sigma_s^{-1}$  $(\partial \sigma_s/\partial p)$  for 13.3 and 23.8 at. % Cu–Ni alloys. The points • aod  $\triangle$  represent observed values, and  $\bigcirc$  and  $\triangle$  are estimated from observation equations.

a way as  $(I_b)$ . For 24.7% Cu-Ni of which curve is not shown in Fig. 3,  $C_1$  thus obtained is  $-4.2 \times 10^{-7}$  bar<sup>-1</sup>, where the values of F(T) and G(T) observed at 200°K and the observed value of  $C_2$ , being  $2.5 \times 10^{-7}$  bar<sup>-1</sup>, have been used in eq. (2b). This value of  $C_1$  is fairly in good argreement with  $-4.0 \times 10^{-7}$  bar<sup>-1</sup> estimated from observation equations.

For ferromagnetic Cu-Ni alloys with Cu content larger than 34 at.%, the sign of  $C_2$  has been found as negative,<sup>11)</sup> and the sign of  $C_1$  appears to be still negative judging from the curve of  $C_1$  versus Cu contents<sup>2,3)</sup> in which the available data have been plotted. Therefore, it can be expected that the

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curves F(T) for these alloys are type  $B_3$ , in case the measurement could be made.

**Pd-Ni alloys:** The observed values of  $\sigma_s^{-1}(\partial \sigma_s/\partial p)$  for 74 at. % Pd-Ni alloy



 $\sigma_s^{-1}(\partial \sigma_s/\partial p)$  for 74 at.% Pd-Ni alloy. The closed circles  $\bullet$  represent observed values. The points  $\bigcirc$ ,  $\triangle$  and  $\times$  are estimated from observation equations, eq. (4a) and eq. (2b) respectively. are given by solid circles in Fig. 4, together with  $C_1$  estimated from observation equations. The curve in this figure is type  $B_1$ , but it is comparatively flat in a wide temperature range. It is expected, therefore, that the curve will become type  $B_2$  and then  $B_3$  when Pd content increases to some extent, judging from the dependence of such curves on Pd contents,<sup>4)</sup> in other words, the Curie temperature would be expected to decrease with pressure.

Assuming that eq. (4a) is applicable to the case of Pd-Ni alloy, the value of  $\sigma_s^{-1}(\partial \sigma_s/\partial p)$  in the neighborhood of  $T_c$  may be obtained from eq. (4a) where  $C_1$  and  $C_2$  are the estimated and the observed value, respectively. For 74% Pd-Ni for example, the value of  $C_2$  is  $2.3 \times 10^{-7}$  bar<sup>-1</sup> resulting from the direct

measurement which will be reported in the near future and the values of  $\sigma_s^{-1}(\partial \sigma_s/\partial p)$  thus obtained are also plotted in Fig. 4 with triangles. Since the slope of the curve is very steep in the neighborhood of  $T_c$ , the disagreement between the estimated and observed value in this figure is not serious. The value of  $\sigma_s^{-1}(\partial \sigma_s/\partial p)$  plotted with cross in Fig. 4 is the one obtained from eq. (2b) where  $C_1$  is the estimated value and  $C_2$ , G(T) are the observed values.

## Comparison with the forced volume magnetostriction

From thermodynamics, the basic relation between the pressure effect on  $\sigma_s$  and the forced volume magnetostriction is given by<sup>13)</sup>

$$\left(\frac{\partial\omega}{\partial H}\right)_{P,T} = -\rho\left(\frac{\partial\sigma_s}{\partial p}\right)_{H,T},\tag{6}$$

where  $\partial \omega / \partial H$  is the forced volume magnetostriction, the volume strain per unit field strength in a strong magnetic fields, and this volume strain is associated with the field induced increase in spontaneous magnetization.

The data on the temperature dependence of  $\partial \sigma_s / \partial p$  in previous papers, therefore, may directly be compared, by the use of eq. (6), with the available

data on that of  $\partial \omega / \partial H$ . Here, it is to be noted that  $\partial \omega / \partial H$  required in the present discussion is the one obtained for the polycrystal composed of uniformaly distribused grains, in other words, the isotropic volume change. This isotropic volume change may also be obtained from the data for single crystal. In single crystal, however, the existence of the anisotropy in the forced volume magnetostriction has been pointed out<sup>14</sup> and actually been found in Fe for example.<sup>15</sup> The anisotropy in  $(\partial \omega / \partial H)_{single}$  means that  $(\partial \omega / \partial H)_{single}$  depends on the direction of spontaneous magnetization with respect to the crystallographic axes.<sup>15,16</sup> In the present paper, only the data have been quoted without making the detailed discussions on the derivation of  $\partial \omega / \partial H$ .

The values of  $\partial \omega / \partial H$  for Ni Ni: obtained from the measurements of  $\partial \sigma_s / \partial p$  given in Fig. 2 and from the direct measurements, are plotted in Fig. 5 as a function of  $T/T_c$ . The measurements of  $\partial \omega / \partial H$  quoted in this figure have been made by Stoelinga et al.<sup>17)</sup> and Lourens et al.<sup>18)</sup> on single crystals. At room temperature,  $\partial \omega / \partial H$  obtained by Snoek<sup>19)</sup> using polycrstal lies on the curve in this figure and the value,  $3 \times 10^{-10}$  oe<sup>-1</sup>, obtained by Hall<sup>20)</sup> using single crystal is omitted from the figure. As is found in Fig. 5, the



the pressure effect on  $\sigma_s$ , by the use of eq. (6). The points  $\bigcirc$ ,  $\square$  and  $\times$  represent measurements made by Stoelinga et al., Lourens et al. and Snoek, respectively.

verification of eq. (6) at various temperatures appears to be qualitatively given.

In polycrystals, the measurements, at room temperature, using dilatometric method such as made by Snoek give the positive sign to  $\partial \omega / \partial H$ . While previous measurements using strain gauge gave the negative sign opposite to that obtained from  $\partial \sigma_s / \partial p$ , a recent accurate measurement made by Tange et al.<sup>21)</sup> using strain gauge gives the positive sign and also the quantitative verification of eq. (6) given by them appears to be satisfactory. At higher temperatures, it is expected from the data on  $\partial \sigma_s / \partial p$  that  $\partial \omega / \partial H$  changes the sign from positive to negative, and the adiabatic measurements made by Döring<sup>22)</sup> using dilatometric method actually gave the negative sign to the isothermal  $(\partial \omega / \partial H)$ .<sup>23)</sup>

Fe: The data for comparing  $\partial \omega / \partial H$  and  $\partial \sigma_s / \partial p$  are given in Fig. 6. The pressure effect on  $\sigma_s$  at 4.2°K has been made by Kondorskii et al.<sup>9)</sup>. As is shown in this figure, the values of  $\partial \omega / \partial H$  observed<sup>15,17,19,24,25)</sup> are fairly scattered even at room temperature, independently of using single or polycrystal. Therefore, further examination should be required to the measurements of  $\partial \omega / \partial H$ , including the derivation of the isotropic volume change from the data

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The points 
and 
are obtained from the pressure effect on  $\sigma_s$ . The points  $\bigcirc, \square, \times,$  $\triangle$  and  $\bigtriangledown$  represent measurements made by Stoelinga et al.,17) Hasuo,15) Sneok,19) Kornetzki<sup>24)</sup> and Calhoun<sup>25)</sup>, respectively.

temperatures would also be desired.

Fe<sup>1,2)</sup> appears to be type B<sub>3</sub>, although the temperature range actually employed was not wide enough. If the curve of  $\sigma_s^{-1}(\partial \sigma_s/\partial p)$  versus temperature is type  $B_1$  or  $B_2$ ,  $\partial \omega / \partial H$ , at lower temperature range, may be expected to decrease when temperature is increased. However, the results obtained by Stoelinga et al.<sup>17)</sup> are temperature independent, as is plotted in Fig. 6, although the investigations of the anisotropy which is associated with the isotropic change in volume have not been made so thoroughly as has been made by Hasuo.<sup>15)</sup> The measurements of  $\partial \omega / \partial H$  or  $\partial \sigma_s / \partial p$  at higher

**Cu–Ni alloys:** For Cu–Ni alloys, the direct comparison of  $\partial \sigma_s / \partial p$  with  $\partial \omega / \partial H$ can not be made for the specimens with same Cu content. The sign of  $\partial \omega / \partial H$ obtained by Kornetzki<sup>24(a))</sup> for 33% Cu is consistent with that expected from the data on  $\partial \sigma_s / \partial p$ , while the signs obtained by Tsuji<sup>26)</sup> for 20 and 30% Cu-Ni alloys are unlikely to be consistent with those expected, except in the neighborhood of  $T_c$ . This disagreement in sign may be explained by the remark made be Tange et al.<sup>21)</sup>.

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