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# Effects of Hydrostatic Pressure on the Curie Temperature of Ni-Based Alloys (Ni-V, -Cu, -Pd, -Pt and -Rh)

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Hydrostatic pressure effects on the Curie temperature $\Delta T_c/\Delta p$, have been investigated for Ni-based alloys, Ni-V, -Cu, -Pd, -Pt and -Rh, over a wide composition range. The Curie temperature was determined as an inflection point of self-inductance vs temperature curve. For all the alloys, $\Delta T_c/\Delta p$'s decrease monotonically accompanying with a change in sign as solute concentration increases. $\Delta T_c/\Delta p$'s as a function of $T_c$ are classified into two types. The data on Ni-V, -Cu and -Pd alloys almost lie on a line and rather rapid decrease in a concave downward curve has been obtained for Ni-Pt and -Rh alloys. It is concluded from qualitative arguments that the $d$-band widening with pressure almost counterbalances with compression-induced $s-d$ transfer for the former type and the widening overcomes the transfer effect for the latter type.

## § 1. Introduction

Effects of pressure on the Curie temperature $\Delta T_c/\Delta p$, as well as on the spontaneous magnetization is an important project associated with the investigation of the electronic structure of $3d$ transition metals and alloys at normal pressure through the pressure effect. Previously our group had tackled with this project and the data on Ni-Cu[1] and -Pd[2] alloys have been briefly reported. Since then, the theoretical analysis based on the itinerant electron model has been made for Ni and Ni-Cu alloys by Lang and Ehrenreich[3] (referred to as L.E. hereafter) by considering the compression-induced conduction band effect ($s-d$ transfer) as well as the $d$-band widening effect with pressure, where both effects are related essentially to the state density at the Fermi level and the correlation energy. Similar but rather simple estimations have been successively made by Shiga[4] for Invars and by Edwards and Bartel[5] for $MnAs_xSb_{1-x}$ without taking $s-d$ transfer effect into account. And results obtained by them that $\Delta T_c/\Delta p$ consists of two terms which are proportional and inversely proportional to $T_c$ at first sight, have been currently employed. Moreover, the interest on $\Delta T_c/\Delta p$ near the critical concentration where ferromagnetism disappears has also been increasing, associated with the problem of the homogeneous alloy system.

Therefore, the investigations of $\Delta T_c/\Delta p$ for other Ni-based alloys over the entire ferromagnetic composition range seem to be valuable both experimentally and theoretically. A series of experiments has again started in such circumstances and the results of Ni-V[6] and -Pt[7] alloys have been briefly discussed in previous notes and it has been noted that Ni-V, -Cu and -Pd alloys, and Ni-Pt and -Rh alloys belong to different types in the functional form of $\Delta T_c/\Delta p$ against $T_c$.[8]

In the present paper, the data on $\Delta T_c/\Delta p$ for Ni-V, -Cu, -Pd, -Pt and -Rh alloys are summarized and qualitative discussions are made from the standpoints mainly of the competition between $d$-band widening and $s-d$ transfer effects. The pressure was applied hydrostatically up to 8 kb and $\Delta T_c/\Delta p$ has been determined from the self-inductance vs temperature curve.

## § 2. Experimental

Alloy ingots of Ni-V, -Pt and -Rh were prepared by melting in a plasma jet furnace in argon atmosphere. The purity of the starting material was 99.99% except vanadium. An ingot was turned over and remelted several times during the melting process and subjected to the subsequent annealing at 1000°C for 24 hours so as to homogenize the composition. The cylindrical specimens, 3 mm in diameter and

10 mm in length, were cut out from the ingots and annealed at 800°C for 24 hours to remove the internal strain induced during machining. Since the weight loss occuring in the melting process was less than 0.5%, the weight ratio of elements of mixture was adopted as the composition of the alloy. The specimen acted as a core of a coil and the self-inductance of the coil, which is proportional to the initial magnetic permeability of the specimen, was measured with a standard Maxwell bridge, the frequency of 1 kHz.

Hydrostatic pressures were generated with a Bridgman press and transmitting mediums were petroleum ether and kerosene in a temperature range from 77 K to room temperature and above room temperature, respectively. The highest pressure applied was 8 kb. The details of the construction of the pressure generator and the connection with the pressure bomb are referred to the article of Tatsumoto *et al.*[9]

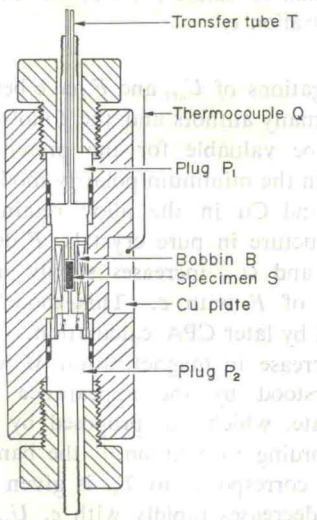


Fig. 1. Diagram of the pressure bomb. T: Stainless steel transfer tube to the pressure intensifier, $P_1$: Retaining plug, $P_2$: Plug for electrodes, S: Specimen, B: Bakelite bobbin, Q: Thermocouple.

Figure 1 shows the diagram of the pressure bomb. The bomb was made of steel, and the inner diameter, outer diameter and the length were 13 mm, 60 mm and 173 mm, respectively. In the figure, T is the stainless steel transfer tube connected with the pressure intensifier, $P_1$ the retaining plug and $P_2$ the plug for electrodes. The specimen S is inserted in the bakelite bobbin B and is slightly pressed with the cap of the bobbin, so that it may not be moved in the process of rise and drop of pressure. The bobbin can be screwed on the top of the plug $P_2$.

In the experiment for determining the pressure effect on $T_c$, $\Delta T_c/\Delta p$, in temperatures ranged from 77 K to 640 K, just above the $T_c$ of Ni. In the figure, Q is the chromel-alumel thermocouple and touched to the Cu plate wound on the pressure bomb. Prior to measurements, it was confirmed that the warming process at a rate of one degree per three minutes was enough to equalize the temperature of the specimen to the bomb. The cryostat used is referred to an article of Fujii.[10] In the measurement below 140 K, the freezing temperature of petroleum ether, the pressure bomb in which the pressure was applied in advance at a temperature where the ether is fluid, was slowly cooled in a temperature gradient in such a way that the solidification of the ether began from the bottom of the bomb. The same procedure has been taken also at liquid helium temperature range.[11]

The self-inductance vs temperature curves observed at normal pressure and a pressure are shown in Fig. 2 for $Ni_{90}Rh_{10}$ and $Ni_{80}Rh_{20}$, as examples. The curves drop as temperature rises, but the drop is steeper for the former specimen. The Curie temperatures $T_c$'s in the present work have been defined as the inflection points of the curves, indicated by arrows in the figure and the values of $\Delta T_c/\Delta p$ thus obtained are zero and $-0.51$ deg/kb for the former and the latter specimens, respectively.

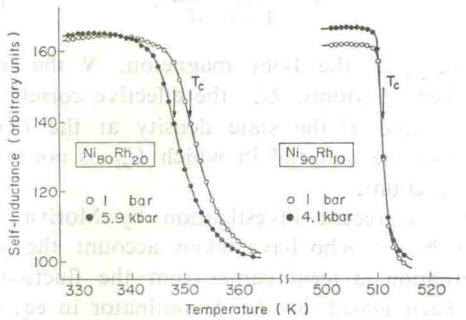


Fig. 2. Examples of the data on self-inductance as a function of temperature at normal pressure and a pressure for $Ni_{90}Rh_{10}$ and $Ni_{80}Rh_{20}$.

The curve of the initial permeability against temperature around $T_c$ generally depends on the internal strain in the specimen, the magnitude of the magnetic short range order above $T_c$ and on the local inhomogeneity of the composition

of the alloy. There is a limit, however, in the removal of latter two factors experimentally. When the local inhomogeneity exists, the transition temperature could not be represented uniquely, but distributes within a certain temperature range. Accordingly, the curve drops with a gentle slope around $T_c$. In such a case, Miyatani[12] has pointed out that $T_c$ may be defined as a temperature corresponding to a middle point of the total decrement of a permeability around the transition temperature, when the distribution is assumed to be Gaussian. However, the distribution will generally deviate from Gaussian and the calculation for such a case shows that temperature of the highest distribution corresponds to the inflection point of the curve. This inflection point coincides with the middle point when the distribution is Gaussian. The determination of $T_c$ in the present work will there-fore be reliable.

In previous measurements of $\Delta T_c/\Delta p$ on Ni-Cu[1] and -Pd alloys,[2] $T_c$ was determined from the measurements of transverse forced magnetoresistance.

## § 3. Results and Discussions

In the following discussions on $\Delta T_c/\Delta p$ in the present work, the Curie temperture $T_c$, defined as a temperature where enhanced susceptibility $\chi$ diverges, has been considered. Above $T_c$, $\chi$ is given by, on the basis of the itinerant electron model

$$\chi=\frac{2N\mu_B F}{1-U_{eff}F}\ , \qquad (1)$$

where $\mu_B$ is the Bohr magneton, $N$ the total number of atoms, $U_{eff}$ the effective correlation energy and $F$ the state density at the Fermi level of the material in which $U_{eff}$ is not taken into account.

In the recent investigation by Moriya and Kawabata[13] who have taken account the spin fluctuation, a term came from the fluctuation has been added in the denominator in eq. (1). In the present paper, however, discussions will be made after eq. (1).

### *3.1 Concentration dependence of $T_c$ at normal pressure*

Since $T_c$ determined from the condition of $U_{eff}(T_c)F(T_c)=1$ is proportional to $\sqrt{1-1/U_{eff}F}$,[4] the dependence of $T_c$ on solute (V, Cu, Pd, Pt and Rh) concentration $c$ of respective alloy may reflect the variation of $U_{eff}$ and $F$, especially in the form of product $U_{eff}F$ with $c$.

Figure 3 shows the Curie temperatures of Ni-V, -Cu, -Pd, -Pt and -Rh alloys observed as a function of $c$ and results are in agreement with literature data.[14,15] In order to discuss $\Delta T_c/\Delta p$, the variation of $U_{eff}$ and $F$ with $c$ at normal pressure may be required in advance for each alloy presently concerned.

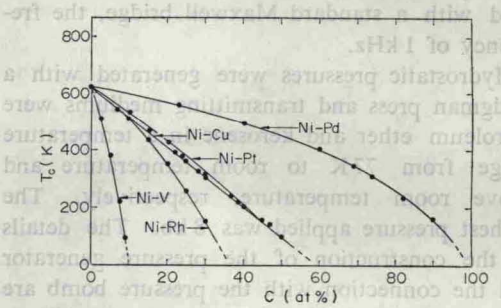


Fig. 3. Curie temperature at normal pressure as a function of solute (V, Cu, Pd, Pt and Rh) concentration $c$.

Investigations of $U_{eff}$ and $F$ have been roughly made by many authors and the following results seem to be valuable for the present purpose. Ni-Cu: In the minimum polarity model,[3] where both Ni and Cu in the alloy retain the electronic structure in pure crystals, $F$ decreases as $(1-c)F_{Ni}$ and $U_{eff}$ increases slowly due to the variation of $F$ with $c$. This model has been supported by later CPA calculation. Ni-V: The rapid decrease in magnetization $M$ with $c$ may be understood by the appearance of virtual bound state, which was proposed by Friedel.[16] Also according to Mathon,[17] the band splitting $\Delta$, which corresponds to $T_c$, is given by $U_{eff}M$. Since $T_c$ decreases rapidly with $c$, $U_{eff}$ seems to be almost constant and $F$ might change in accordance with the change in $M$. This constancy of $U_{eff}$ is also expected from that the critical concentration $c_F$ where ferromagnetism disappears is small for this alloy. The situation therefore will be similar to that of Ni-Cu. Ni-Pd and -Pt: Though Pd, Pt and Ni have been considered as isoelectronic, the variation of $T_c$ with $c$ for Ni-Pd and -Pt have been investigated in different ways. Ni-Pd alloys have been investigated on the basis of local enhancement model, in which Ni rather than Pd are attributable to the divergence of $\chi$ or to $T_c$. Estimations of Harris and Zuckermann[18] covered the

whole composition range by assuming almost constant $F$ and Levin *et al.*[19] discussed dilute Ni region. The latter authors have pointed out the appropriateness of decreasing $F$ with $c$. For Ni-Pt, on the other hand, Schindler[20] has investigated on the basis of uniform enhancement model, in which Pt as well as Ni is attributable to $T_c$ and $U_{eff}$ may be the weighted mean with $c$ of those of Ni and Pt. Also this alloy has been considered as homogeneous one[21] and above statements may be accepted. Ni-Rh: Levin *et al.*[19] have pointed that uniform enhancement model may be applied to dilute Ni region. With respect to $F$, $F(c)$ is not monotonic, but will take maximum judging from the state density of Ni and Rh,[19] since Rh has one less electron than Ni.

### 3.2 *Pressure effects on $T_c$, $\Delta T_c/\Delta p$*

The pressure effects on $T_c$, $\Delta T_c/\Delta p$, obtained for the alloys are shown in Fig. 4 as a function of solute concentration $c$. Because $T_c$ changed linearly with applied pressure in the pressure range presently employed, $\Delta T_c/\Delta p$ could be determined uniquely from the slope of $T_c$ vs pressure curve.

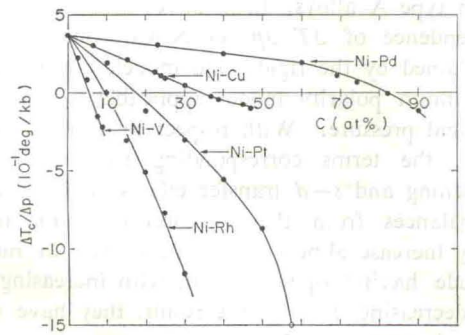


Fig. 4. A plot of the pressure effects on $T_c$, $\Delta T_c/\Delta p$, as a function of solute concentration $c$.

The experimental results shown in Fig. 4 are arranged as follows: (i) For all the alloys, $\Delta T_c/\Delta p$ decreases with increasing $c$. The initial rate of decrease is largest for Ni-V and smallest for Ni-Pd alloys, likewise the case of $T_c$ at normal pressure in Fig. 3. (ii) For each alloy, $\Delta T_c/\Delta p$ changes the sign from positive in Ni rich region to negative as $c$ increases. The reduced solute concentrations to $c_F$, $c/c_F$'s, where $\Delta T_c/\Delta p$ changes the sign are about 0.3 for Ni-Pt and -Rh, 0.6 for Ni-V and -Cu, and 0.85 for Ni-Pd alloys. The data on $\Delta T_c/\Delta p$ near $c_F$ have been obtained for Ni-Pd by Beille[22] and -Pt by Alberts *et al.*[21] and only the result for $Ni_{42.9}Pt_{57.1}$ is plotted in Fig. 3, since the behavior near $c_F$ is not the main object in the present work. (iii) For Ni-V and -Cu alloys, $\Delta T_c/\Delta p$'s change almost linear. For the others, curves are concave downward. Among them, the variation of the curve is rapid near $c_F$ for Ni-Pt and -Rh alloys. (iv) Unlike the case at normal pressure, no coincidence of the curves has been obtained for Ni-Cu and -Pt alloys. This result suggests[7] that the data on $\Delta T_c/\Delta p$ will play a part to the classification of characteristics of alloys at normal pressure.

In the current investigation of $\Delta T_c/\Delta p$ derived from $T_c$ in eq. (1), the following conditions have been assumed: (i) The band width $W$ of the $d$-band depends on the Heine's relation,[23] $W \propto R^{-5}$, where $R$ is the interatomic distance. (ii) The $d$-band is widened uniformly with pressure. (iii) The effective correlation energy $U_{eff}$ employed is the Kanamori type[24]

$$U_{eff} = \frac{U_b}{1+U_bK}\ , \qquad (2)$$

where $U_b$ is coulomb self-energy of an atomic orbital and $K$ is a quantity depending on the band shape.

Besides these assumptions proposed by L.E. for the analysis of $\Delta T_c/\Delta p$ of Ni and Ni-Cu alloys, L.E. have also introduced the compression-induced conduction band effect ($s-d$ transfer). The expression for $\Delta T_c/\Delta p$ derived by them is

$$\frac{dT_c}{dp} = \frac{5}{3}\kappa T_c + (\xi_1' + \xi_2 + \xi_3)\frac{5}{3}\kappa T_c\ , \qquad (3)$$

where $\kappa$ is the volume compressibility of the material. In eq. (3), the 1st term $(5/3)\kappa T_c$ corresponds to a simple case of $d$-band widening in which $U_{eff}$ is infinite and the effect of $s$-band is neglected. The term $\xi_1'$ is the contribution of the $d$-band in case of $U_{eff}$ being finite, referred to as $d$-band widening in the present paper, and $\xi_2+\xi_3$ come from the $s-d$ transfer with compression. The term $\xi_2$ in eq. (3), however, has not been considered here, since L.E. have pointed that this term is never of primary importance.

Comparing eq. (3) with the expression obtained by Shiga,[4] who has investigated $\Delta T_c/\Delta p$ of Invars without considering $s-d$ transfer, eq. (3) can be written as

$$\frac{dT_c}{dp} = \frac{5}{3}\kappa T_c + \frac{D}{T_c} \qquad (4a)$$

$$D=\frac{\kappa}{2k^2\alpha}\left[-\frac{5}{3}\frac{U_{eff}}{U_b}+\frac{N_c}{\tilde{F}+F_c}\left(\frac{F_s}{F}-K_s U_{eff}\right)0.36\right], \qquad (4b)$$

where $k$ is the Boltzmann's constant and notations $N_c$, $F_c$, $F_s$, $\tilde{F}$ and $K_s$ are referred to the article of L.E., and $\alpha$ to Shiga. It is understandable that (i) from eq. (4a), $\Delta T_c/\Delta p$ consists of two terms which are proportional and inversely proportional to $T_c$ at first sight, (ii) in eq. (4b), the 1st and 2nd terms in the brackets in $D$ are functions of $F$ and $U_b$ or $F$ and $U_{eff}$, where $K$ acts through $F$, and correspond to $\xi_1'$ ($d$-band widening) and $\xi_3$ ($s-d$ transfer) terms in eq. (3), respectively. Shiga's expression is the sum of $(5/3)\kappa T_c$ and the 1st term in $D$.

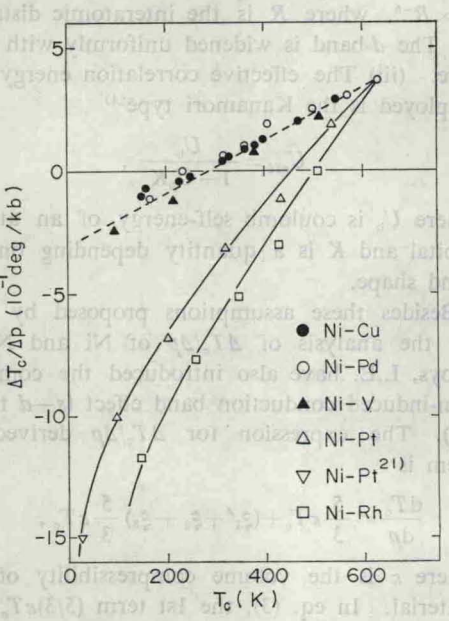


Fig. 5. A plot of $\Delta T_c/\Delta p$ as a function of $T_c$.

On the basis of considerations mentioned above, results for $\Delta T_c/\Delta p$ plotting as a function of $T_c$ will bear a rather profound meaning than plotting as a function of $c$ such as shown in Fig. 4. Figure 5 thus shows $\Delta T_c/\Delta p$ as a function of $T_c$. It is to be noted that the functional forms of $\Delta T_c/\Delta p$ in Fig. 5 are classified into two types, A and B. In type A, the linearity of $\Delta T_c/\Delta p$ with $T_c$ almost holds. Ni-V, -Cu and -Pd alloys belong to this type and the data almost lie on a line expressed as $\Delta T_c/\Delta p=(5/3)\kappa T_c-3\times10^{-1}$ in unit of deg/kb, using the compressibility $\kappa$ of Ni at room temperature.[25] The concentration and temperature dependence of $\kappa$ for the alloys may be neglected for the present purpose, judging from the experimental results.[25,26] For Ni-Pd, the deviation from the linearity near $c_F$ has been reported,[22] but the behavior near $c_F$ is not the main object in the present paper as is mentioned above. In type B alloys, on the other hand, $\Delta T_c/\Delta p$ vs $T_c$ curve is not linear, concave downward from Ni rich side. Ni-Pt and -Rh alloys belong to this type. In other words, type A or B is that in which the 2nd term $D/T_c$ in eq. (4a) is constant independent of $T_c$ or $c$, or a function of $T_c$ or of $c$.

These results will be discussed qualitatively on the basis of the following standpoints: (i) The descrimination between type A and type B might be made essentially from the dependence of $F$ and $U_b$ on $T_c$ or from the degree of contribution of the $d$-band widening and the $s-d$ transfer effects to $D$. (ii) The results obtained by Lang[27] and L.E. that $D$ shifts to the negative side regardless of its sign when $F$ and $U_b$ decreases, may be accepted as general tendency and applied to the alloys presently concerned, as far as discussions will be made from eq. (4a) and (4b).

In type A alloys, L.E. have pointed that $c$ dependence of $\Delta T_c/\Delta p$ for Ni-Cu could not be explained by the rigid band model, but by the minimum polarity model applicable to $T_c(c)$ at normal pressure. With respect to $D/T_c$ in eq. (4b), the terms corresponding to the $d$-band widening and $s-d$ transfer effects almost counterbalances from their numerical results that they increase almost in the same way in magnitude having opposite sign, with increasing $c$, or decreasing $T_c$. As the result, they have obtained constant $D/T_c$.

Above mentioned results for Ni-Cu obtained by L.E. could not be applied unconditionally to Ni-V and -Pd alloys belonging to type A, since the appropriate models and the detailed band shapes etc. are necessary to the final estimation. However, since the situation of $F(c)$ and $U_{eff}(c)$ for Ni-V would be similar to Ni-Cu as described in § 3.1, the competition between the widening and the transfer terms in Ni-V would vary as $c$ in a similar way to Ni-Cu. On the other hand, the simple argument unlikely explain the data on Ni-Pd, but the experimental results will support the similar situation.

For type B alloys, the competition between the widening and the transfer in $D/T_c$ should

be different from that of type A alloys. The dependence of $D/T_c$ on $T_c$ is shown in Fig. 6 for Ni-Rh as an example. The values of $D/T_c$ are obtained from the difference between $\Delta T_c/\Delta p$ observed and $(5/3)\kappa T_c$. For $\kappa$, the value of Ni at room temperature was again used. If $D$ is independent of Rh concentration, $D/T_c$ will vary like $-1/T_c$ and the curve normalized to Ni is represented by a dotted curve indicated as $D=$ constant. Therefore, the difference between the curve of $D/T_c$ and that of constant $D$ corresponds to the dependence of $D$ on $T_c$. In the corner of the figure, the values of $D$ estimated are given in arbitrary units as a function of $T_c$, where those for Ni-Cu and -Pt alloys are also given for comparison. For both Ni-Pt and -Rh alloys, $D$ shifts to negative side farther than that of Ni-Cu alloys. According to Lang's calculation, widening and transfer effects contribute negatively and positively to $D$, respectively. Therefore, the farther negative shift of $D$ would suggest that the widening effect overcomes the transfer effect in type B alloys comparing with type A alloys.

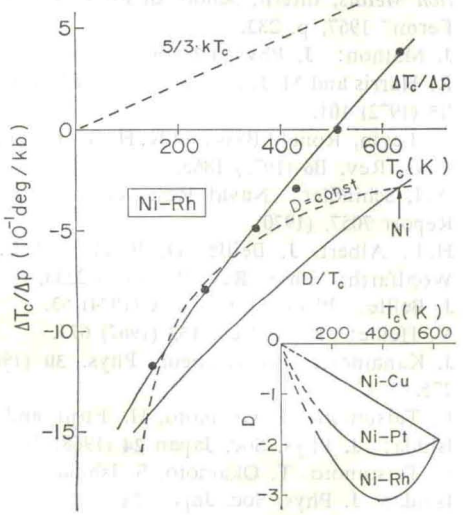


Fig. 6. Examples of the decomposition of $\Delta T_c/\Delta p$ into $(5/3)\kappa T_c$ and $D/T_c$, the 1st and 2nd terms in eq. (4a), for Ni-Rh alloys. In the corner of the figure, is given $D$ as a function of $T_c$ for Ni-Cu, -Pt and -Rh alloys.

With respect to $F$ and $U_b$, the decrease in $F$ or $U_b$ results in the negative shift of $D$, as is already mentioned. Ni-Pt: The decrement of $F$ with $c$ may be smaller than that of Ni-Cu, since Pt has holes, while Cu has no hole. Therefore, $U_{eff}$ will decrease considerably with $c$ from the condition for determining $T_c$, since $c_F$'s are almost the same for Ni-Pt and -Cu. This decrease in $U_{eff}$ results in the decrease in $U_b$, as is found from eq. (2), assuming that $K$ will not change much due to the small change in $F$. If the uniform enhancement model could be applied to the concentrated alloys,[20] $U_b$ may be given as a weighted mean value of those of Ni and Pt. In this case, the decrease in $U_b$ with $c$ means that $U_b$ for Pt is considerably smaller than that of Ni, although no information has been available for $U_b$ of Pt. In conclusion, $U_b$ may contribute more effectively to the farther negative shift of $D$ shown in Fig. 6 than $F$, while the latter will be effective for type A alloys. Ni-Rh: The arguments for Ni-Rh may be similar to Ni-Pt alloys. Furthermore, it may be expected as already mentioned that the Fermi level passes the peak of the state density curve of Ni as Rh concentration increases,[19] and the sign of $F_s$ in eq. (4b), representing the gradient of the state density at the Fermi level, changes. Therefore, the transfer effect will be greatly reduced as $c$ increases, which results in the dominancy of widening effect and farther negative shift of $D$ than Ni-Pt alloys.

### *3.3 Relation between d-band widening and s—d transfer*

The functional forms of $\Delta T_c/\Delta p$ for Fe-Ni and Fe-Pt[28,29] and $MnAs_xSb_{1-x}$,[5] for example, are like $-1/T_c$ and shows the dominancy of the 2nd term $D/T_c$ in eq. (4b). Edwards and Bartel[5] have pointed that $D/T_c$ term is dominant in case of weak ferromagnetics, by taking only widening term into account and their calculated values fairly explain the experimental ones, like Shiga's estimation. Ni-Pt and -Rh alloys in the present experiment also have $-1/T_c$ character, although the characteristic is less remarkable than materials mentioned above. In type B alloys, therefore, the widening effect is not necessarily dominant but an important factor to $\Delta T_c/\Delta p$.

The importance of the transfer effect for $\Delta T_c/\Delta p$ is clear for type A alloys as is described in § 3.2. In the pressure effect on the spontaneous magnetization of Ni,[17] the transfer effect is dominant at 0 K and widening effect is negligible. As temperature rises, the former effect decreases and the latter increases. Also the transfer term in the pressure effect on the residual resistivity has been discussed by Beyerlein and Lazarus[30] on dilute Ni-Pd alloys.

Based on the considerations mentioned above and in § 3.2 from the standpoints of *d*-band widening and the $s-d$ transfer effect on $\Delta T_c/\Delta p$, it may be concluded that the former effect is important and becomes dominant near $c_F$ in type B, and both terms are important equivalently in type A alloys, in Ni-based alloys presently concerned. And these circumstances will also be accepted generally.

At concentration where the widening effect in a wide sense including $(5/3)\kappa T_c$ term and the $s-d$ transfer effect counterbalances, $\Delta T_c/\Delta p$ becomes zero and situation may occur regardless of the magnitude of both effects. However, the reduced Curie temperature $T_c$(alloy)/$T_c$(Ni) where $\Delta T_c/\Delta p$ becomes zero and changes the sign will be large for the alloy in which the *d*-band widening tending to decrease the magnitude of $\Delta T_c/\Delta p$ from positive side in Ni rich region overcomes rapidly the transfer effect and the experimental results that reduced Curie temperatures of type B alloys are larger than those of type A alloys support this argument.

The detailed theoretical investigations of the concentration dependence of $F$ and $U_{eff}$ (or $U_b$) for the alloys presently concerned would be desired for the quantitative analysis of $\Delta T_c/\Delta p$ and further investigation should take the spin fluctuation into account.

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