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A Calculation on Pressure Dependence of Spin Wave Energy of Invar Alloys

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Abstract

Under the assumption that the change in volume of the system affects both the band width and molecular field coefficient but the shape of the density-of-states curve remains unaffected, a formula for the pressure dependence of exchange stiffness D is derived. For a system of two dimensional free electrons with step-type density-of-states curve, calculations on D and its pressure dependence dD/dp are performed with reasonable values of parameters. Calculated results well reproduce the observed composition dependence of the reduced values of D and dD/dp for fcc Fe-Ni alloy system, while the calculated absolute values themselves are smaller by about an order of magnitude than the observed ones.

§ 1. Introduction

In previous papers,[1-3] we proposed a simple theory on volume magnetostriction of itinerant electron ferromagnet and explained the large positive spontaneous volume magnetostriction of Invar alloys. There, we assumed that when the volume of the system varies, the band width and the molecular field coefficient change but the shape of the density-of-states curve remains unchanged. It was shown that the high peak existing at the top of the density-of-states curve plays an essential role for the large positive volume magnetostriction of Invar alloys. Now, is it able to explain the composition dependence of the change in exchange stiffness D (the coefficient in the dispersion relation of spin wave $\hbar\omega_q = Dq^2 + \cdots\cdots$) due to pressure p by the existence of the high peak at the top of the density-of-states curve?

In the present paper, we discuss pressure dependence of D based on the above mentioned model. In § 2, we give a formula for dD/dp and show that the negative pressure dependence of magnetization of Invar alloys is explainable

qualitatively if we regard the composition with $D=0$ as the critical composition for ferromagnetism of fcc Fe-Ni system.[4] In § 3 and § 4, we give formulae for D and for dD/dp, respectively, for step-type density-of-states curve, assuming for the electrons to be two dimensional free electrons. Numerical calculations for D and dD/dp of fcc Fe-Ni alloys are done in § 5. The observed composition dependence of reduced values of these quantities[5-7] is qualitatively reproduced by the calculated ones, but the calculated absolute values themselves are smaller by about an order of magnitude than the observed ones.

§ 2. Formulation

Within the random phase approximation, D is given by

$$D=\frac{I}{3N}\sum_{k}\left\{\frac{f^{+}_{k}+f^{-}_{k}}{2\Delta}\nabla^{2}_{k}\varepsilon_{k}-\frac{f^{+}_{k}-f^{-}_{k}}{\Delta^{2}}(\nabla_{k}\varepsilon_{k})^{2}\right\} \tag{1}$$

for cubic metal[8], where Δ, the exchange splitting, is given by $\Delta=nI\zeta$, ζ, the relative magnetization, is given by $\zeta=\sum_{k}(f^{+}_{k}-f^{-}_{k})/nN$, $f^{\pm}_{k}$ are the Fermi distribution function, n is the number of electrons per atom, N is the number of atoms of the system and I, the effective interaction, is twice J, which was used in our previous papers[1-3] as exchange parameter. For a strong ferromagnet at 0 K, we have

$$D=\frac{1}{6Nn}\sum_{\varepsilon_{k}\leqq\varepsilon_{F}}\left\{\nabla^{2}_{k}\varepsilon_{k}-\frac{1}{nJ}(\nabla_{k}\varepsilon_{k})^{2}\right\}, \tag{2}$$

where ε_{F} is Fermi level.

To take into account the effect of pressure or the effect of change in volume of the system, we assume that the main effects of the volume change are the shift of the energy levels as a whole, the change in the band width and the change in J and the density-of-states curve does not change its shape when the volume changes. If we take W as an appropriate measure of the band width and put $\varepsilon_{k}=x_{k}W$, the change in the volume of the system affects W and J, but x_{k} remains unaffected. In a strong ferromagnet, $x_{F}=\varepsilon_{F}/W$ does not change due to the volume change. (In a weak ferromagnet, the volume change is accompanied by a change in magnetization, which changes $x^{\pm}_{F}$ and the situation is more complex than that in the strong ferromagnet.) Expressing (2) as

$$\frac{D}{W}=\frac{1}{6Nn}\sum_{x_{k}\leqq x_{F}}\left\{\nabla^{2}_{k}x_{k}-\frac{W}{nJ}(\nabla_{k}x_{k})^{2}\right\}, \tag{3}$$

we have

$$\frac{d}{dv}\left(\frac{\widetilde{D}}{W}\right)\equiv\left(\frac{\widetilde{D}}{W}\right)'=\frac{1}{6Nn^2}\frac{W}{J}\left(\frac{J'}{J}-\frac{W'}{W}\right)\sum_{x_q\leqq x_F}(\nabla_q x_q)^2, \tag{4}$$

where $\widetilde{D}=D/a^2$, a is a measure of length of the unit cell of the system, $q=ak$, v is the volume per atom of the system, $J'\equiv dJ/dv$ and $W'\equiv dW/dv$. Since $(J'/J-W'/W)$ is evaluated to be positive,[2] we derive

$$(\widetilde{D}/W)'>0 \tag{5}$$

from (4). Because $\widetilde{D}'=W'(\widetilde{D}/W)+W(\widetilde{D}/W)'$, where $\widetilde{D}'\equiv d\widetilde{D}/dv$, we have

$$d\widetilde{D}/dp=-\kappa v\widetilde{D}'=\{-\kappa v(W'/W)\widetilde{D}\}+\{-\kappa vW(\widetilde{D}/W)'\}, \tag{6}$$

where κ is compressibility. The first term of (6) is positive when D is positive because W' is expected to be negative, and the second term is negative if (5) holds. It is generally expected from (6) for dD/dp to be positive for large D and negative for small D. This agrees qualitatively with experimental result[7] on $d\widetilde{D}/dp$ of fcc Fe-Ni system, that is, $d\widetilde{D}/dp>0$ in Ni-rich region with large D and $d\widetilde{D}/dp<0$ in Fe-rich region with small D. If we regard the composition with $D=0$ as the critical composition for ferromagnetism of fcc Fe-Ni system[4], then $d\widetilde{D}/dp$ is negative there as seen from (6) and the critical composition shifts towards Ni-rich side by application of pressure, resulting negative pressure dependence of spontaneous magnetization M. This is one of possible mechanism giving negative dM/dp of Invar alloys.

§ 3. D for an alloy system with step-type density-of-states curve

We take a step-type density-of-states curve shown in Fig. 1 as the simplest one which holds the essential feature of the density-of-states of 3d band in fcc transition metals. That is,

$$\nu(\varepsilon)=\begin{cases}\rho & \text{for } \varepsilon\leqq W_1\\ b\rho & \text{for } \varepsilon>W_1\end{cases}, \tag{7}$$

where $\nu(\varepsilon)$ is the density-of-states per atom per one spin direction and $0<b<1$. We take here $W\equiv 1/\rho$ as the measure of the band width. As a band structure to give the density-of-states shown in Fig. 1, we take that of two dimensional free electrons:

$$\varepsilon_k=\begin{cases}\alpha_0(k_1^2+k_2^2) & \text{for } \varepsilon\leqq W_1\\ \alpha_1(k_1^2+k_2^2) & \text{for } \varepsilon>W_1\end{cases}, \tag{8}$$

where

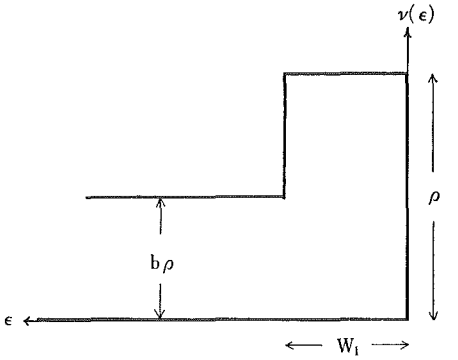

Fig. 1. Step-type density-of-states curve, the simplest one holding the essential feature of 3d band in fcc transition metals.

$$\nu(\varepsilon)=\begin{cases}\rho\equiv 1/W & \text{for } \varepsilon\leqq W_1\\ b\rho & \text{for } \varepsilon>W_1\end{cases}$$

with $0<b<1$ and $n_0\equiv\rho W_1$.

$$\alpha_0=vW/4\pi a \text{ and } \alpha_1=vW/4\pi ab. \tag{9}$$

Here, a is length of the edge of the cube taken as the unit cell and $v=sa^3$ with $s=1$ for sc, 1/2 for bcc and 1/4 for fcc lattice. For the band of (8), we have

$$\nabla^2_{\boldsymbol{k}}\varepsilon_{\boldsymbol{k}}=4\alpha(\varepsilon) \text{ and } (\nabla_{\boldsymbol{k}}\varepsilon_{\boldsymbol{k}})^2=4\alpha(\varepsilon)\varepsilon \tag{10}$$

with

$$\alpha(\varepsilon)=\begin{cases}\alpha_0 & \text{for } \varepsilon\leqq W_1\\ \alpha_1 & \text{for } \varepsilon>W_1\end{cases}.$$

Because $\nabla^2_{\boldsymbol{k}}\varepsilon_{\boldsymbol{k}}$ and $(\nabla_{\boldsymbol{k}}\varepsilon_{\boldsymbol{k}})^2$ are independent of $\boldsymbol{k}$ depending on ε only, we can replace the summation over $\boldsymbol{k}$ in (2) by integration over ε. Defining P_F and Q_F as

$$P_F\equiv\sum_{\varepsilon_{\boldsymbol{k}}\leqq\varepsilon_F}(\nabla^2_{\boldsymbol{k}}\varepsilon_{\boldsymbol{k}})/N \quad\text{and}\quad Q_F\equiv\sum_{\varepsilon_{\boldsymbol{k}}\leqq\varepsilon_F}(\nabla_{\boldsymbol{k}}\varepsilon_{\boldsymbol{k}})^2/N, \tag{11}$$

we have

$$\left.\begin{aligned}P_F&=\int_0^{\varepsilon_F}\nu(\varepsilon)\,(\nabla^2_{\boldsymbol{k}}\varepsilon_{\boldsymbol{k}})d\varepsilon=\int_0^{\varepsilon_F}4\alpha(\varepsilon)\nu(\varepsilon)d\varepsilon,\\ Q_F&=\int_0^{\varepsilon_F}\nu(\varepsilon)\,(\nabla^2_{\boldsymbol{k}}\varepsilon_{\boldsymbol{k}})^2d\varepsilon=\int_0^{\varepsilon_F}4\alpha(\varepsilon)\varepsilon\nu(\varepsilon)d\varepsilon\end{aligned}\right\} \tag{12}$$

and

$$D=\frac{1}{6n}(P_F-\frac{1}{nJ}Q_F) \tag{13}$$

with

$$n=\int_0^{\varepsilon_F}\nu(\varepsilon)d\varepsilon. \tag{14}$$

(i) For the case of $\varepsilon_F\leqq W_1$, i.e., $n\leqq n_0\equiv\rho W_1$, we have

$$P_F=nsa^2W/\pi \text{ and } Q_F=n^2sa^2W^2/2\pi \tag{15}$$

and hence

$$D=\frac{sa^2W}{6\pi}\left(1-\frac{W}{2J}\right) \text{ for } n\leqq n_0. \tag{16}$$

The Stoner condition of ferromagnetism for this case is $2J>W$; the Stoner condition and the condition of $D>0$ hold simultaneously in this case.

(ii) For the case of $\varepsilon_F>W_1$, i.e., $n>n_0$, from the relations

$$\left.\begin{aligned} n-n_0&=\int_{W_1}^{\varepsilon_F} b\rho d\varepsilon=b(\varepsilon_F-W_1)/W,\\ n_0&=\int_0^{W_1}\rho d\varepsilon=W_1/W,\\ P_F-P^0{}_F&=\int_{W_1}^{\varepsilon_F} b\rho\cdot 4\alpha_1 d\varepsilon=v(\varepsilon_F-W_1)/\pi a,\\ P^0{}_F&=\int_0^{W_1}\rho\cdot 4\alpha_0 d\varepsilon=vW_1/\pi a,\\ Q_F-Q^0{}_F&=\int_{W_1}^{\varepsilon_F} b\rho\cdot 4\alpha_1\varepsilon d\varepsilon=v(\varepsilon_F{}^2-W_1{}^2)/2\pi a,\\ Q^0{}_F&=\int_0^{W_1} b\rho\cdot 4\alpha_0\varepsilon d\varepsilon=vW_1{}^2/2\pi a, \end{aligned}\right\} \tag{17}$$

we have

$$\left.\begin{aligned} P_F&=sa^2W\{n-n_0(1-b)\}/\pi b,\\ Q_F&=sa^2W^2\{n-n_0(1-b)\}^2/2\pi b^2 \end{aligned}\right\} \tag{18}$$

and hence

$$D=\frac{sa^2W}{6\pi b}\left[\left\{1-\frac{n_0(1-b)}{n}\right\}-\frac{W}{2bJ}\left\{1-\frac{n_0(1-b)}{n}\right\}^2\right] \text{ for } n>n_0. \tag{19}$$

As seen from (19), n_D, the value of n with $D=0$, is given by

$$n_D=n_0(1-b)/(1-2bJ/W). \tag{20}$$

It should be noticed that the present calculation of D is done for an open band, hence the first term of (2), i. e., the positive contribution to D is overestimated. We expect for the actual value of n_D to be smaller than that given by (20). The Stoner condition for ferromagnetism for this case is $2J>W$ for $n<2n_0$ and $2J>W/b$ for $n>2n_0$. In the case of $n>2n_0$, if J has such a value as $W/b>2J>(1+b)W/2b$, then the Stoner condition is unsatisfied but the strong ferromagnetism is favourable energetically than paramagnetism for such n as $n<n_c$, where n_c is given by

$$n_c=n_0\{2(1-b)/(1-2bJ/W)\}^{1/2}, \tag{21}$$

as seen from Shimizu's condition[9] for itinerant electron ferromagnetism.

§ 4. dD/dp for an alloy system with step-type density-of-states curve

(i) For the case of $\varepsilon_F\leqq W_1$, i. e., $n\leqq n_0$, putting

$$\tilde{D}\equiv D/a^2, \tag{22}$$

we have from (16)

$$\tilde{D}'\equiv\frac{d\tilde{D}}{dv}=\left(\frac{W'}{W}\right)\tilde{D}+\frac{sW^2}{12\pi J}\left(\frac{J'}{J}-\frac{W'}{W}\right), \tag{23}$$

and

$$\frac{d\ln\tilde{D}}{dp}=-\tilde{D}^{-1}\,\kappa v\tilde{D}'=-\kappa v\left(\frac{W'}{W}\right)\left[1+\frac{\left(\frac{J'}{W'}-\frac{J}{W}\right)}{\left(\frac{2J}{W}-1\right)\left(\frac{J}{W}\right)}\right]. \tag{24}$$

Here, since $-\kappa v(W'/W)>0$ ($\because W'<0$), the first term gives a positive contribution and the second term gives a negative or a positive contribution to $d\ln\tilde{D}/dp$ according to sign of $(J'/W'-J/W)$ because $2J>W$ when ferromagnetism appears. We have previously shown[2] that $(J'/W'-J/W)$ is negative if J is given by Kanamori's theory.[10] Therefore, it is expected that the second term of (24) gives a negative contribution to $d\tilde{D}/dp$.

(ii) For the case of $\varepsilon_F>W_1$, i. e., $n>n_0$, we have from (19)

$$\tilde{D}'=\frac{W'}{W}\tilde{D}+\frac{s\{1-n_0(1-b)/n\}^2}{12\pi b^2}\left(\frac{W^2}{J}\right)\left(\frac{J'}{J}-\frac{W'}{W}\right) \tag{25}$$

and

$$\frac{d\ln\widetilde{D}}{dp}=-\kappa v\left(\frac{W'}{W}\right)\left[1+\frac{\left\{1-n_0(1-b)/n\right\}\left(\frac{J'}{W'}-\frac{J}{W}\right)}{2b\left\{\frac{J}{W}-\frac{1-n_0(1-b)/n}{2b}\right\}\left(\frac{J}{W}\right)}\right]. \tag{26}$$

The first term of (26) gives a positive contribution and the second term gives a negative contribution, because $\{1-n_0\,(1-b)/n\}$ is positive, $(J'/W'-J/W)$ is negative and the denominator, which is propotional to D, is positive for positive D, i.e., for $n<n_D$. As n closes to n_D, the value of (26) closes to negative infinity.

§5. Numerical calculation of D and dD/dp for fcc Fe-Ni system

For fcc Fe-Ni system, the values of the parameters of the band are taken as[1)]

$$n_0=0.6,\ b=0.5 \text{ and } W=0.6 \text{ eV}. \tag{27}$$

If we take 3.5Å, the value of a of Ni metal[11)], as a and $s=1/4$, then the factor in (16) and (19) is evaluated as

$$sa^2W/6\pi \simeq 100 \text{ meV}\cdot\text{Å}^2 \tag{28}$$

and we have from (16), (19), (27) and (28)

$$D(\text{in meV}\cdot\text{Å}^2)\simeq\begin{cases}100\times(1-W/2J) \text{ for } n\leqq 0.6\\ 200\times\{(1-0.3/n)-\dfrac{W}{J}(1-0.3/n)^2\} \text{ for } n>0.6\end{cases} \tag{29}$$

Here, we expect that $0.91\leqq J/W<1.0$. The value of $J/W=0.91$ gives $n_c=2.0$, which corresponds to $Fe_{0.7}Ni_{0.3}$ provided the number of 4s electrons is 0.6 per atom, where n is regarded as the number of 3d holes per atom. For values of $J/W<0.91$, $Fe_{0.7}Ni_{0.3}$ is not ferromagnetic and for values of $J/W\geqq 1.0$, the Stoner condition is satisfied for any value of n. The values of D of each composition in fcc Fe-Ni system calculated by (29) with $J/W=0.91$ ($n_c=2.0$) and $J/W=0.94$ ($n_c=2.45$) are shown in Fig. 2. The calculated results reproduce qualitatively the observed composition of the value of D, which decreses with increasing Fe content, while the calculated value itself is smaller by about an order of magnitude than the observed one.

In this case, n_D and n_c are given by

$$n_D=0.3/(1-J/W) \text{ and } n_c=0.6/(1-J/W)^{1/2} \tag{30}$$

and always $n_D>n_c$. (Eq. (21) to give n_c holds only for $1>J/W>0.75$, i.e., for $n_c>1.2$.) This situation ($n_D>n_c$) conflicts with Katsuki's conjecture[4)] for the critical composition for ferromagnetism of fcc Fe-Ni system to be the composition with

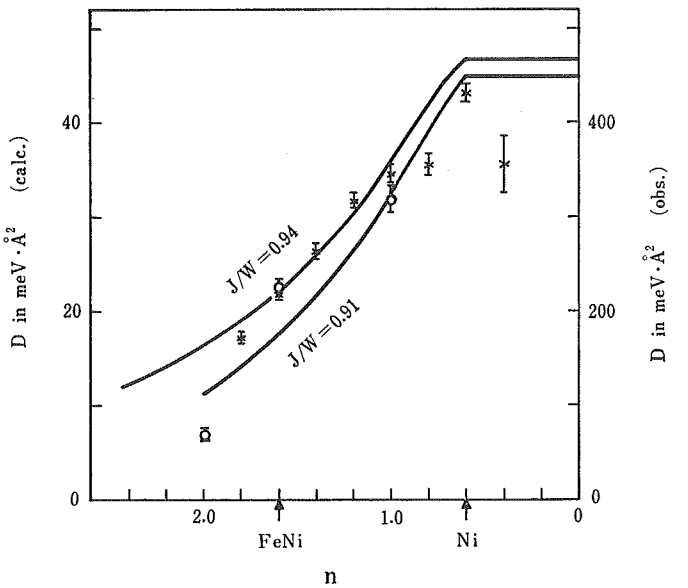

Fig. 2. D of fcc Fe-Ni and Ni-Cu alloys.
Solid lines : Calculated results by (29) with $J/W=0.91$ and 0.94.
Crosses and circles : Observed values.[5,6]
The scale of the ordinate on the left hand side is for the calculated values and the one on the right hand side is for the observed values.

$D=0$ (otherwise ferromagnetism holds), which includes implicitly the assumption of $n_D<n_c$. It should be noticed, however, that the value of n_D is overestimated by (20) as was commented before. Therefore, we must not say that Katsuki's conjecture has been contradicted decidedly by this situation.

The factor in (24) and (26), $-\kappa v W'/W$, is evaluated as

$$-\kappa v W'/W \simeq 1.0\times10^{-7}\ \mathrm{cm^2/kg}. \tag{31}$$

Here, we used the relation[2] between spontaneous volume magnetostriction ω_s and Δp, a dimensionless quantity related with energy difference between ferromagnetic and paramagnetic states[1], and observed value of $\omega_s=-4.2\times10^{-4}$ [12] and calculated value of $\Delta p=-4.5\times10^{-2}$ [1] for Ni metal. From (24), (26), (27) and (31) we have

$$\frac{d\ln\widetilde{D}}{dp}\ (\text{in } 10^{-7}\mathrm{cm^2/kg})\simeq\begin{cases}1+\dfrac{\left(\frac{J'}{W'}-\frac{J}{W}\right)}{\left(\frac{2J}{W}-1\right)\left(\frac{J}{W}\right)} & \text{for n} \leqq 0.6\\[2ex] 1+\dfrac{\left(\frac{J'}{W'}-\frac{J}{W}\right)}{\left\{\frac{J}{(1-0.3/n)W}-1\right\}\left(\frac{J}{W}\right)} & \text{for } n>0.6.\end{cases} \tag{32}$$

We have previously estimated as $J'/W'=0.75$ [1] from the composition dependence of spontaneous volume magnetostriction in fcc Fe-Ni system. Calculated results of $d\ln\tilde{D}/dp$ of each composition with this value of J'/W' and with $J/W=0.91$ and 0.94 are shown in Fig. 3. The calculated results fairly reproduce the feature of

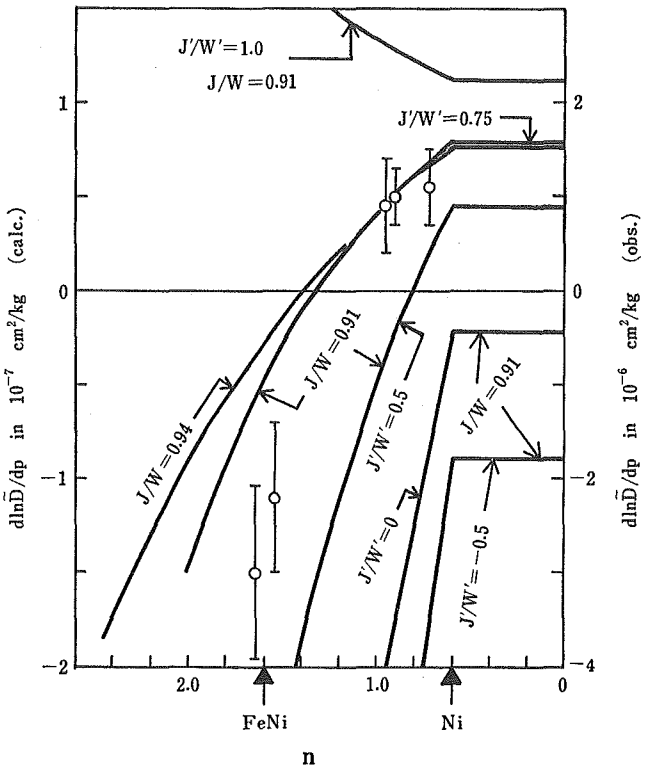

Fig. 3. $d\ln\tilde{D}/dp$ of fcc Fe-Ni alloys.
Solid lines : Calculated results by (32) with $J'/W'=0.75$ and $J/W=0.91$ and 0.94 ; and with $J'/W'=1.0$, 0.5, 0 and -0.5 and $J/W=0.91$.
Circles : Observed values.[7]
The scale of the ordinate on the left hand side is for the calculated values and the one on the right hand side is for the observed values.

the experimental result[7], that is, the change from positive value in Ni-rich region to negative value in Fe-rich region at $n\simeq 1.3$ and the gradual composition dependence in the former region and rather rapid one in the latter region, although the calculated absolute values are smaller by about an order of magnitude than the observed ones. In the present calculation, since the value of D is overestimated for large value of n, i.e., in Fe-rich region, we expect that the

composition dependence of $d\ln\widetilde{D}/dp$ is more rapid than the calculated ones in the region with negative $d\widetilde{D}/dp$.

For the sake of comparison, the calculated results with $J'/W'=1.0$, 0.5, 0 and -0.5 and with $J/W=0.91$ are shown in Fig. 3. (The calculated results with $J/W=0.94$ lie very closely to the corresponding curves with $J/W=0.91$.) For $J'/W'=0$ and -0.5, the absolute value of second term of (24) or (26), which gives negative contribution to $d\ln\widetilde{D}/dp$, is larger than the first term, hence $d\ln\widetilde{D}/dp$ is negative in whole range of composition. For $J'/W'=0.5$, the sign of $d\ln\widetilde{D}/dp$ changes from positive to negative at $n\simeq 0.80$ (0.83 for $J/W=0.94$) as n increases, while in the experiment[7] the sign seems to change at $n\simeq 1.3$. For $J'/W'=1.0$, both the first and second terms give positive contributions because $J'/W'-J/W>0$ in this case, hence $d\ln\widetilde{D}/dp$ is positive in whole range of composition and shows an opposite trend of composition dependence. It should be stressed that the value of the parameter $J'/W'=0.75$, which was evaluated previously from the composition dependence of spontaneous volume magnetostriction[1], is also essential to reproduce the observed composition dependence of $d\ln\widetilde{D}/dp$, as seen from the above discussion. Also, it should be noticed that $J'/W'>0$ means $dJ/dv<0$, which is in contrast to that usually expected from the Bethe-Slater curve for Invar alloys.

§ 6. Conclusion

Under the assumption that the change in volume of the system affects the band width and the molecular field coefficient but the shape of the density-of-states curve remains unaffected, a formula for the pressure dependence of D was derived. For the system of two dimensional free electrons with step-type density-of-states curve, calculations on D and $d\ln\widetilde{D}/dp$ were performed with reasonable values of the band parameters and of J/W and J'/W'. Calculated results well reproduced qualitatively the experimental results of D and $d\ln\widetilde{D}/dp$, although the calculated absolute values were smaller by about an order of magnitude than the observed ones. It was stressed that the value of $J'/W'=0.75$, which had been estimated previously from the composition dependence of spontaneous volume magnetostriction, was also essential to the composition dependence of $d\ln\widetilde{D}/dp$.

Mathon and Wohlfarth[13] discussed the pressure dependence of D of fcc Fe-Ni system on the basis of the itinerant electron model. They treated Ni and Ni-rich alloys as strong ferromagnet and Invar as very weak ferromagnet and fairly explained the observed close relation between the pressure dependence of the Curie temperature and that of the exchange stiffness[7]. Our calculation is also based on the itinerant electron model with somewhat different assumptions from their treatment.

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