# The Magnetic and Electric Properties of (Co_<1-x> Mn_x)_2B Crystalline compounds and Amorphous Alloys

# The Magnetic and Electric Properties of $(Co_{1-x}Mn_x)_2B$ Crystalline Compounds and Amorphous Alloys*

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## Synopsis

The manganese concentration x dependence of the average magnetic moment $\mu o$, the Curie temperature Tc, the pressure effect on Tc and the electric resistance for the ferromagnetic amorphous alloys$(Co_{1-x}Mn_x)B$ were investigated. The Curie temperature decreases linearly with increasing x and $\mu o$ had a maximum around x=0.15. The magnetic susceptibility vs. temperature curves for all the prepared amorphous alloys obey the Curie-Weiss law above Tc. The pressure effect on Tc is that Tc has a value of 1.06 K/kbar at x=0 and is decreasing with increasing x and becomes zero at x=0.4. These results are analyzed on the basis of the pair interaction model and the local enviroment effect. There arise two kinds of minimum in the resistance vs. temperature curves. That the resistance minimum at high temperature has a strong correlation with Tc is assured by measuring the transverse effect of the resistance with amorphous alloys x=0.4. The resistance minimum for the amorphous alloys and crystalline compounds are found at low temperature under ferromagnetic state.

## I. Introduction

There are three kinds of compounds $Co_3B$, $Co_2B$ and CoB in the crystal structure of which is DO11 for $Co_3B$, C16 for $Co_2B$ and B27 for CoB in the Co-B system. The compounds $Co_3B$ and $Co_2B$ are ferromagnetic and magnetic moment of Co atom was determined as 747 K, 1.12 $\mu_B$ for $Co_3B$ and 422 K, 0.896$\mu_B$ for $Co_2B$[1]. The variation of the average magnetic moment per

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magnetic atom in (Me, Me´)-B compounds (Me, Me´ =Cr, Mn, Fe, Co and Ni) was discussed by Lundquist et al.[2] with the band model. According to their discussion the electronic state of the cobalt atom in the $Co_2B$ compound is very similar to that in the pure nickel. On the other hand, the magnetic moment per cobalt atom ($\mu o$) in $Co_{1-x}Mn_x$ binary amorphous alloys decreases with increasing boron concentration and its concentration dependence of $\mu o$ shows that the values of $\mu o$ at x=0.25 and 0.33 are almost the same as that in the crystalline compounds $Co_3B$ and $Co_2B$ respectively and the magnetic moment of the cobalt will become zero near x=0.48 from $\mu o$=1.3 at x=0.12. It is known that most of the ferromagnetic Co-metalloid amorphous alloys show the similar metalloid concentration dependence of $\mu o$. The number of nearest neighbor atoms around a cobalt on $Co_{1-x}Mn_x$ amorphous alloys is in the range of 10 to 13 for $x \leq 0.4$ and then the cobalt atom may be surrounded with those as in packing of fcc.

When the manganese atoms replace cobalt atoms partially in $Co_2B$ crystalline compound the average magnetic moment per magnetic atom vs. manganese concentration curve has a maximum around 25 at% Mn and the Curie temperature curve as a function of manganese concentration also has a maximum at 10 at% Mn. It was reported by Kadomatsu et al.[3] that the compounds $(Co_{1-x}Mn_x)_2$ are ferromagnetic for $x \leq 0.6$ and for $x > 0.6$ may be paramagnetic. They have interpreted the x dependence of $\mu o$ and Tc on the basis of the pair interaction model which was proposed by Kouvel[4] to explain the magnetic properties of Ni-Mn alloys. It was concluded that the exchange interaction of Co-Co and Co-Mn should be ferromagnetic and that of Mn-Mn should be antiferromagnetic. Obi et al.[5] performed the same analysis for $(Co_{1-x}Mn_x)_{100-y}B_y$ ternary amorphous alloys and obtained the average manganese magnetic moment per magnetic atom curve as a function of B concentration. The value of the magnetic moment of MnB crystalline compound which is ferromagnetic appears well on the above curve. $Mn_2B$, however, may be paramagnetic[1].

Recently the pressure effect in the Curie point of $(Co_{1-x}Mn_x)_2B$ compounds has been reported by Kodomatsu et al.[3]. When the hydrostatic pressure is applied the Curie temperature decreases and the absolute value of its pressure deviation (dTc/dP) increases with increasing Mn concentration. The thermal expansion of Fe-B amorphous alloys showed the anomaly below the Curie point and its behavior is invar-like[6]. The thermal expansion of Co-B amorphous alloys, however, did not show the anomalous behavior near the Curie point even if the pressure effect on Tc for Fe-B and Co-B amorphous alloys yields in the same order. The resistivity vs. temperature curves for the ferromagnetic amorphous alloys Fe-B and Co-B have a min-

imum at low temperature. While the ferromagnetic amorphous alloys Re-nickel[7] (Re:rear earth metals) show a minimum in electric resistivity vs. temperature curve near the Curie temperature. It was reported that there appear 2 kinds of minimum in the resistivity vs. temperature curves for Fe-(Ni,Co)-Zr alloys[8]. One of the two minima appears near Curie temperature and another at low temperature. It is interesting that there appears a minimum in the electric resistivity vs. temperature curve under the ferromagnetic state . It is useful for understanding the electronic state of Co atoms to study the resistance minimum in both crystalline and amorphous Co-B alloys having the same constitution.

In this paper, the Curie temperatre, pressure effect on the Curie temperature and electric resistance on $(Co_{1-x}Mn_x)_2B$ amorphous alloys are reported and discussed in the basis of the local enviroment effect and the pair interaction model comparing with those of the crystalline compounds with the same composition.

## II. Experimental Procedure

The $(Co_{1-x}Mn_x)_2B$ ( $0 \leqq x \leqq 0.4$ ) amorphous alloys were prepared a single roller quenching technique in the form 30 $\mu$m thick ribbon. The structure of all the prepared specimens was examined by X-ray diffraction and confirmed to be amorphous. The temperature dependence of magnetization and susceptibility were measured at temperatures between 4.2-600 K in a magnetic field of 6 kOe by a magneto balance. Hydrostatic pressure was applied to a specimen in a Teflon pressure cell filled with kerosine or silicine oil by using a piston cylinder type device[9]. The pressure was calibrated by using the Hg-solid-liquid transition temperature. The Curie temperature was determined by an ac transformer method. Density was measured by a normal Arckimedean method The error of density is smaller than about 0.1 % . The crystalline $(Co_{1-x}Mn_x)_2B$ (all elements 3N pure) in the desired proportion under a purified argon gas atomosphere the products were checked to be in a single phase by X-ray diffraction. The specimens were cut carefully into a patallelepoped (1x2x4 in mm ). Electric resistance was measured by a four probes method with dc current in the temperature range from 4.2 to 500 K.

## III. Experimental Results

The magnetic susceptibility of $(Co_{1-x}Mn_x)_2B$ amorphous alloys was measured in the temperature range from room temperature to 680 K. Its reciprocal magnetic susceptibility $X_g^{-1}$ is shown in Fig.1. $X_g^{-1}$ depending

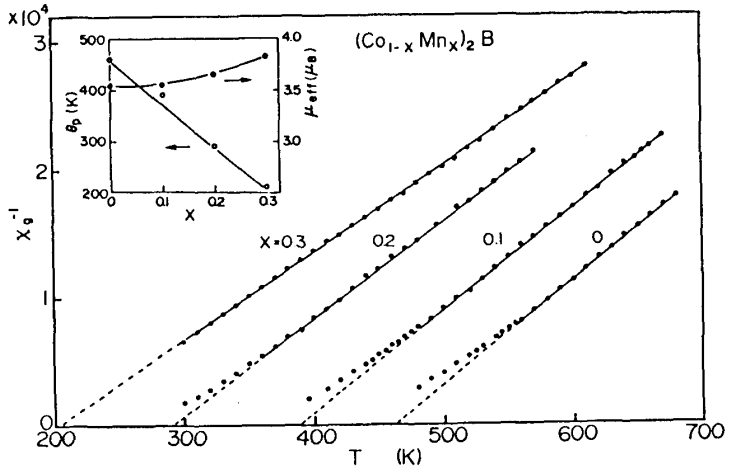


Fig.1 Temperature dependence for $(Co,Mn)_2B$ amorphous alloys. The inset shows the paramagnetic Curie temperature($\theta$) and effective magnetic moment per transtion atom ( $\mu$eff).

on the temperature varies linearly as following the Curie-Weiss law. The paramagnetic Curie temperature $\theta_p$ and effective magnetic moment per transition metal atom $\mu_{eff}$ depend on the manganese concentration x as shown in the inset of the figure. The values of $\theta_p$ and $u_{eff}$ at x=0 obtained to be 460 K, 3.6 and the variation for $\theta_p$ decreases linearly and for $\mu_{eff}$ increases monotonously with increasing x. The permiability for x =0 and 0.2 depending on the temperature under pressure is shown in Fig. 2. The Curie temperature Tc was defined with intersection point of two extrapolated lines of the permiability vs. temperature curve as shown in the figure. This determination is very effective since the magnetization

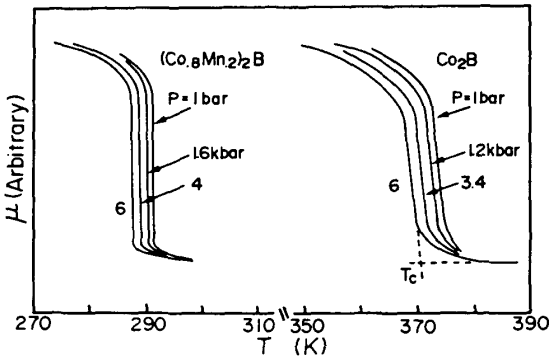


Fig.2 Temperature dependence of the initial susceptibility under hydrostatic pressure for the amorphous $Co_2B$ and $(Co_{0.8}Mn_{0.2})_2B$ alloys.

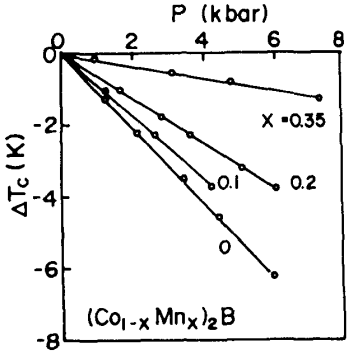


Fig.3 Curie temperature shift by compression for $(Co,Mn)_2B$ amorphous alloys.

vs. temperature curve is not shown the sharp variation in the vicinity of the Curie temperature. The Tc of all the present amorphous alloys shifts to the lower temperature side by compression. This Curie temperature shift $\Delta$Tc as a function of pressure was plotted in Fig.3 as the manganese concentration x is a parameter. The slope of the $\Delta$Tc vs.

pressure (P) curve (dTc/dP) increases with increasing x and this x depdndent dTc/dP curve is shown in Fig.4 together with that of the crystalline compounds by Kadomatsu et al.[3]. The sign of the grade of the

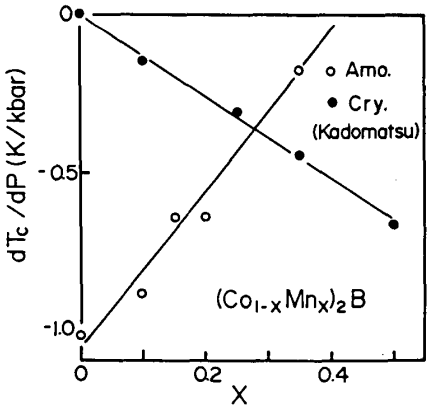


Fig.4 Mn concentration x dependence of the Curie temperature shift vs. pressure curve for $(Co,Mn)_2B$ amorphous alloys and crystalline compounds after Kadomatsu et al.(ref.2).

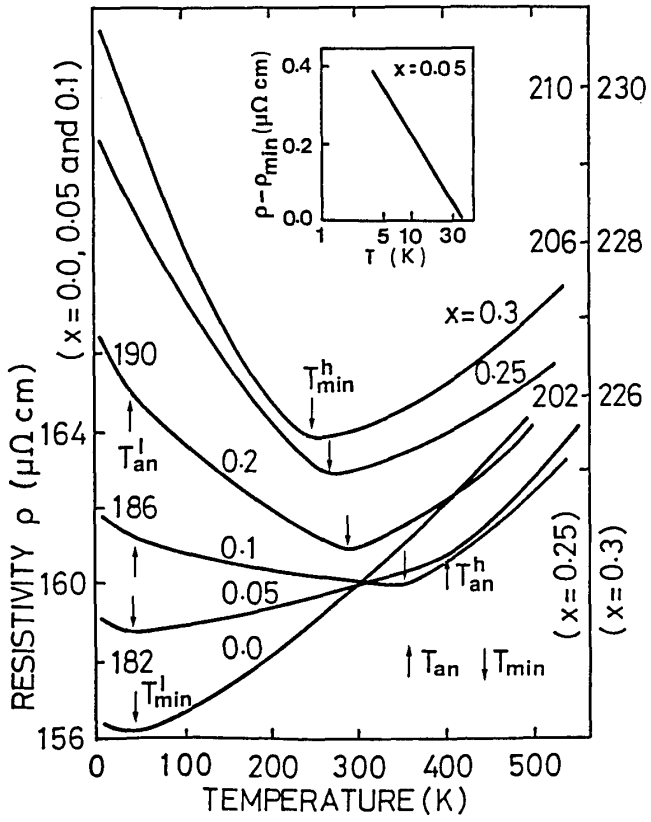


Fig.5 Temperature dependence of the elecric resistivity ρ for $(Co,Mn)_2B$ amorphous alloys; $T^l_{min}$ and $T^l_{an}$ stand for the temperature of the resistivity minimum and anomaly at low temperatures respectively, and $T^h_{min}$ and $T^h_{an}$ stand for those at high temperatures respectively.

curve for $(Co_{1-x}Mn_x)_2B$ crystalline specimens is opposite to that for the amorphous alloys. That is, the value of dTc/dP for the amorphous alloys is 1.06 K/kbar at x=0 and decreases linearly and becomes zero K/bar at 0.4 when x increases. However, that for the compounds is zero K/kbar at x=0 and increases linearly and 0.5.

Fir.5 shows the resistivity vs. temperature curves for the $(Co_{1-x}Mn_x)_2B$ amorphous alloys. The curves for x=0 and 0.05 show a minimum at low temperatures and those for x=0.1 and 0.2 show an anomaly at low temperatures. On the other hand, another minimum appears also near the Curie temperature for $x \geq 0.1$ in addition to the low temperature resistivity-minimum. The anomaly,however,appears near the Curie point for x= 0.05.The resistivity for the crystalline compounds also was measured in

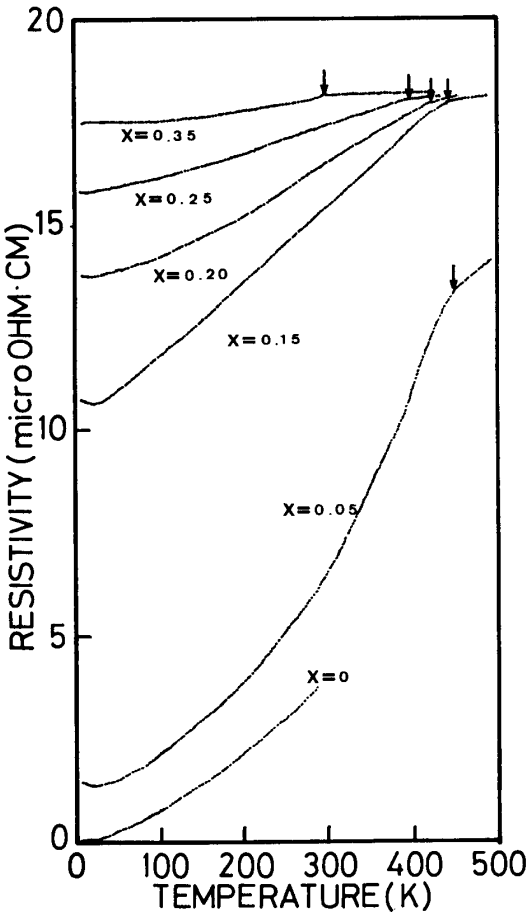


Fig.6 Temperature dependence of the resistivity for $(Co_{1-x}Mn_x)_2B$ crystalline compounds. Allows show the Curie temperature.

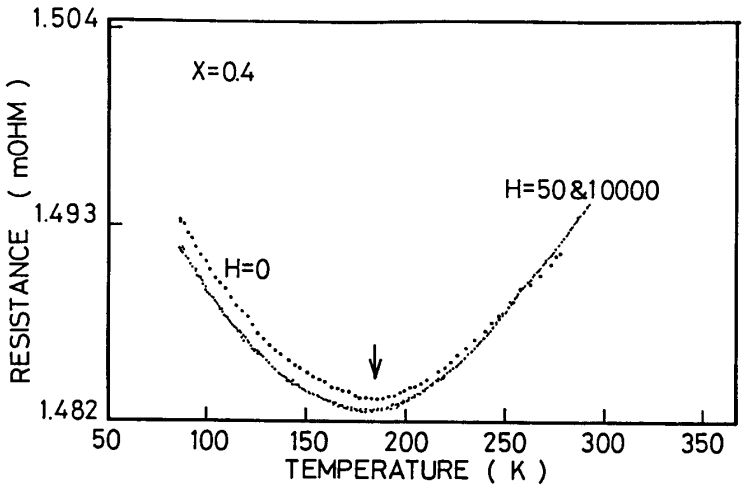


Fig.7 Temperature dependence of the resistance at magnetic field H= 0, 50 and 10000 Oe . A allow shows the resistance minimum.

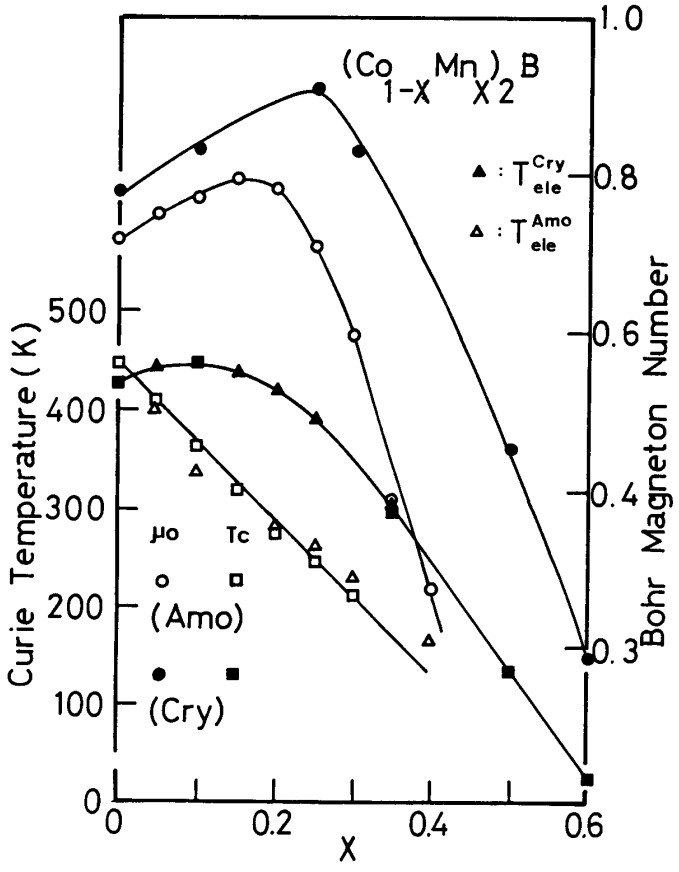


Fig.8 Manganese concentration x dependence of the average magnetic moment per magnetic atom and the Curie temperature for $(Co_{1-x}Mn_x)_2B$ amorphous alloys and crystalline compounds. △ and ▲ : Tc determined from the electric resistivity measurement.

the temperature range from 4.2 K to 450 K. The obtained result is shown in Fig.6. The curve for $x \geq 0.05$ has a minimum at low temperature and show a similar anomaly at the Curie point as those for the ordinary ferromagnetic metals have. The curve for x=0 does not have a minimum at low temperature. Fig.7 shows the experimental result in the case of applying the magnetic field in a perpendicular to the surface of the amorphous ribbon specimen. The temperature of the minimum at H=0 is 180 K and this value was plotted in Fig.8 as Curie temperature. This minimum temperature is not depend on the magnetic field. The values of the resistance for H=50 Oe and 10000 Oe is not different in the temperature range of 77 to 273 K. The resistance value at H=5 Oe is smaller than that at H=0 till 240 K.

The average magnetic moment per transition metal atom at 0 K ($\mu o$) and Curie temperature of $(Co_{1-x}Mn_x)_2B$ amorphous alloys are shown as a function of x in Fig.8 together with those of the compounds with the same composition. The closed marksin Fig.8 are for the crystalline specimens where the circles and squares were quted from the data by Kadomatsu et al.[3] and the triangles were determined by the electric resistance measurement. The open marks mean for the amorphous alloys and the circles are on the magnetic moment and the squers are on the Curie point. The curves on the moment for both the crystalline and noncrystalline $(Co_{1-x}Mn_x)_2B$ have a maximum at x=0.25 and at x=0.2 respectively and after that decrease sharply to zero around x=0.5 for the amorphous alloys. The curve on the Curie point for the crystalline compounds has a maximum at x=0.1. The curve, however, for the amorphous alloys decreases monotonously with increasing x.

We found the discrepancy and agreement between the experimental results for $(Co_{1-x}Mn_x)_2B$ amorphous alloys and crystalline compounds. In the next section, these results are discussed with the pair interaction model and local enviroment effect.

## IV. Discussins and Conclusions

The density was measured to obtain the information of a mean distance between metal-metal atoms in $(Co_{1-x}Mn_x)_2B$ amorphous alloys. The slope of the observed density D vs. manganese concentration x curve ( dD/dx ) was determined to be -1.6 $g/cm^3$. The mean distance $d_m$ between the nearest neighbor atoms in $Co_2B$ amorphous alloys is estimated to be about 2.53 A from the interporation of the experimental values 2.5 A for x=0.185[10] and 2.57 for x=0.4[11] determined by X-ray diffraction. Assuming that the mean distance is proportional to a length of a cube

having the average volume for a cobalt atom which is defined from deviding the whole volume by the number of cobalt atoms, the mean distance can be calculated from the density by taking the proportional constant K=1.067 for $Co_2B$ amorphous alloys which was obtained from deviding the mean distance ($d_m$=2.53 A) with the length of a side of the average cube(2.387) . Therefore the $d_m$ for x=0.3 is estimated to be 2.58 A. This assumption is proper if the packing on neighbor atoms is hold symmetrically, for example the K value is 1.122 in the case of fcc and 1.091 in the case of bcc packing.

The reciprocal magnetic susceptibility shown in Fig.1 follows the Curie-Weiss law so that it may be allowed to use the locarized moment model. AS the Curie temperature is a function of manganese concentration x , let it denote $T_c(x)$. According to the pair interation model , Tc(x) is expressed as follow[3];

$$Tc(x) = T_{CoCo}(1-x)^2 + 2T_{CoMn}x(1-x) + T_{MnMn}x^2 , \quad (1)$$

where $T_{CoCo}$,$T_{CoMn}$ and $T_{MnMn}$ stand for the interaction energy in the temperature unit with respect to Co-Co, Co-Mn and Mn-Mn pair respectively. The original pair interation model was proposed by Kouvel[4] . In many cases, however, this model was modefied the simpler one assuming that the cobalt and mangnese atoms have a constant magnetic moment independing of manganese concentration x and that a cobalt atom or a manganese atom as a center atom loses the moment or is coupled antiparallel with the nearest neighbor atom moments when a atom is surrounded by the more than a critical number no of manganese atoms. The values of $T_{CoCo}$, $T_{CoMn}$ and $T_{MnMn}$ can be determined as reproducing the experimental Tc vs. x curve in Fig. 8. The result is $T_{CoCo}$=448 K, $T_{CoMn}$=48 K and $T_{MnMn}$=-352 K. On the other hand, these values for the crystalline specmences reported by Kadomatsu et al.[3] are $T_{CoCo}$=426 K, $T_{CoMn}$596 K and $T_{MnMn}$=-934 K.The average magnetic moment $\mu o$ vs. x curve in Fig.9 is specified as follow: when the Co atoms are replaced with a small amount of Mn the average moment increases since the manganese magnetic moment (1.35 $\mu o$) is larger than the cobalt one (0.715 $\mu o$). A cobalt or a Mn atom will be surrounded with more Mn atoms as Mn concentration increases. According to the local enviroment effect, center atoms (Co or Mn atoms) lose the magnetic moment or couple antiferromagnetically with the neighbor atoms when the number of manganese atom is greater than the critical no. and then the average magnetic moment has a maximum value and is decreasing with increasing x. Since the cobalt moment is zero in the range of $x \geq 0.3$ it is expected that the mictomagnetism takes place by the antiferromagnetic interaction of Mn-Mn pairs. Obi et al.[5] have found the

anomarous behavior of the magnetization at low temperatures. It suggests that there could exist the inhomogeneous magnetic state below the anomous temperature. The critical number of no is an important parameter when the enviromental effect is discussed. The experimental result on the magnetic moment in the amorphous alloys can be explained well with the critical number no=4 and nearest neighbor atom number N=10.[12] The no for Co-Mn alloys is estimated to be 4[13], but one for the crystalline $(Co_{1-x}Mn_x)_2B$ compound is given by Kadomatsu et al.[3] as 6. The effect that the value of the magnetic moment depends on the nearest neighor distance has been neglected in the local enviroment effect. The nearest neighbor distances for Co-Mn alloys and for $(Co_{1-x}Mn_x)_2B$ amorphous alloys are almost same values(2.5 A) and that for the compounds is about 2.75 A. Therefore it seems that the distance effect can not be neglected. In fact, the maximum on the moment for the compounds in Fig. 8 shifts to the higher concentration of x comparing with that for the amorphous alloys in order that the effect reducing the average magnetic moment in the crystalline compounds is weeker than in the amorphous alloys. As Tc(x) is differentiated with restect to pressure P, the following equation is obtained as;

$$\frac{dTc}{dP} = x^2\left(\frac{dT_{CoCo}}{dP} + \frac{dT_{MnMn}}{dP} - 2\frac{dT_{CoMn}}{dP}\right) + 2x\left(\frac{dT_{CoMn}}{dP} - \frac{dT_{CoCo}}{dP}\right) + \frac{dT_{CoCo}}{dP} \quad . \qquad (2)$$

The value of the differentiation of $T_{ij}$ (i,j = Co or Mn) with respect to P are determined with the experimental values in Fig.4. That is; $dT_{CoMn}/dP$ = -1.06 K/kbar, $dT_{CoMn}/dP$ = 0.26 K/kbar and $dT_{MnMn}/dP$ = 1.16 K/kbar. The values for the crystalline $(Co_{1-x}Mn_x)_2B$ obtained by Kadomatsu et al.[3] are $dT_{CoCo}/dP$ = 0, $dT_{CoMn}/dP$ = -0.65 and $dT_{MnMn}/dP$ = -1.3 K/kbar. Supposing that the pressure effect in the magnetic moment is neglected the interaction between Co-Co atoms or between Mn-Mn atoms for the amorphous alloys is weakened ferrommagnetically or antiferromagnetically with increasing pressure. The ferromagnetic interaction of Co-Mn distance. Accoring to the interaction curve for the manganese atoms shown in Fig. 9 which was proposed by Yamada[14] the antiferromagnetic interaction at 2.77 A will be stronger when the Mn-Mn distance decrease. Therefore $dT_{MnMn}/dP$ for the compounds has a negative value. Allows in the figure show the neighbor atoms positions and the numerical number stand for the number of the atoms. On the other hand, the mean distance for the amorphous alloys is from 2.53 to 2.58 so that $dT_{MnMn}/dP$ has an positive value.

The electric resistivity vs. temperature curves for Fe-B, Co-B

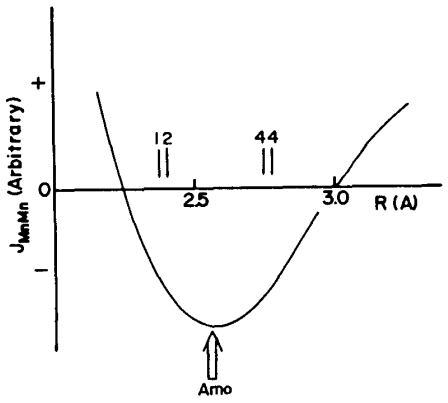


Fig.9 Interaction curve on $J_{MnMn}$ ($\propto T_{MnMn}$) as a function of the distance R(in A unit) between Mn-Mn atoms in α-Mn by Yamada(ref.14). Small allows show the respective location of eleven neighbor Mn atoms in $(Co,Mn)_2B$ crystalline compounds. A bold arrow shows the average distance between Mn-Mn atoms in $(Co,Mn)_2B$ amorphous alloys.

and Fe-P amorphous alloys have a minimum at low temprature.[15] There also appears such a minimum in the case of $(Co_{1-x}Mn_x)_2B$ ( $x<0.2$ ) amorphous alloys.[16] When the x is more than 0.2 the resistivity vs. temperature curve has an anomal instead of the minimum. The origin of the minimum has been considered to be the Kondo effect or the two level model. The resistivity for the $(Co_{1-x}Mn_x)_2B$ crystalline compounds in Fig. 6 also show the minimum near 20 K. This minimum may be attributed to the Kondo effect which is expected to appear even in the ferromagnetic materials if there are a few of manganese atoms being in extreme weak internal field. In fact, it is shown from the analysis with the pair interaction model that when a manganese atom is surrounded by Co atoms and Mn atoms it is possible that the gross interaction for the center Mn atom becomes to be zero by cancellation with the Co-Mn ferromagnetic and Mn-Mn antiferromagnetic interaction. The resistivity vs. temperature curve for the crytalline specimens shows a kink at the Curie temperature. The curves for the amorphous alloys ($x\geq 0.2$), however, show the minimum near the Curie points. The close correlation between the minimum and the Curie temperature is concluded from the fact that the minimum temperature are on the Curie temperature vs. Mn concentration curve as shown in Fig.8.

The temperature dependent electric resistance curves for x=0.4 on $(Co,Mn)_2B$ amorphous alloys at the applied magnetic field H=50 and 10000 Oe in Fig. 7 are almost coinsident each other so that the magnetization should be saturated at 50 Oe. Below the Curie temperature, the resistance decreases with applying magnetic field such a phenomena found in the pure nickel and is known as the transverse effect. The difference between the resistance at 0 and 50 Oe is almost constant till the temperature of the minimum and become small above 180K. It means that there exist Curie ponit in the vicinity of the resistance minimum and then the strong correlation between the Curie temperature and minimum. Obi et al.[5] pointed out that the minimum is not intrinsic since the resistance vs.

temperature curve showed the hysteresis near the Curie temperature. In the present case, however, the curve showed a reversible variation. This discripancy is in order that their amorphous alloys specimens have higher Curie temperature near which there is a crystallization point and then it is possible to crystallize partially in the specimens once they are heated up over the Curie temperature.

The concept of the pair interaction model and local enviroment effect is very useful for analyzing our experimental results. The Co atom in $(Co_{1-x}Mn_x)_2B$ amorphous alloys loses the magnetic moment at smaller Mn concentration than that for the compounds. The strength of the Co-Mn interaction becomes weaker with increasing the number of Co atoms having no magnetic moment. This thing causes the phenomenon that the Tc vs. x curve for the amorphous alloys showes the monotonous decrease with increasing Mn concentration, nevertheless that for the compounds have a minimum. Furthermore, it is predicted that there may arise the mictomagnetism for $(Co_{1-x}Mn_x)_2B$ amorphous alloys ($x \geqq 0.4$) by Mn-Mn pair antiferromagnetic interaction. The other hand, it is pointed out from the electric resistivity measurement that the existence of the Mn atom which is in a local paramagnetic state under the ferromagnetic state is possible for both the amorphous alloys and crystalline compounds. The value of the effective magnetic moment for CoB amorphous alloys obtained from the reciprocal susceptibility is about four times larger than the average magnetic moment. It means that the variation of the magnetic moment in the pair interaction model and local enviroment effect cannot be neglected. It is important how to understand that the Co or Mn magnetic moment is annihilated by the manganese atoms neighbor. The investigation with FMR and the pressure dependence of the magnetization will be performed in the future in order to make the electronic state of the magnetic atoms clear.

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