Pressure dependence of superconductivity in amorphous Zr_xNi_{100-x} alloys

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The pressure dependence of superconducting transition temperature T_c of amorphous Zr_xNi_{100-x} alloys ($x = 80$, 71, 67, 63.5, 60, 55) has been studied under quasihydrostatic pressure to 8 GPa. For amorphous samples having Zr concentration $x > 60$, $\partial T_c/\partial P$ is positive in sign and it decreases nonlinearly with increasing x, whereas $\partial T_c/\partial P$ is negative in sign for 55 at.% Zr concentration. These results were explained in terms of character change of the conduction carriers at the Fermi surface. In addition, for $Zr_{80}Ni_{20}$ we observed the formation of the ω-Zr phase for pressures greater than 2 GPa.

Zr forms amorphous alloys with a variety of transition elements such as Ni, Cu, Fe, Co, and Pd.[1,2] The electronic properties of these alloys at low temperatures vary from ferromagnetic to superconducting, depending on the alloy composition. The Ni-Zr alloy is an attractive system for studies of fundamental properties of amorphous materials as it has the widest range of composition for glass formation. This makes it possible to examine systematic changes within one alloy system. Zr_xNi_{100-x} ($80 < x < 30$) amorphous alloys show superconductivity at low temperatures where the superconducting transition temperature (T_c) decreases as a function of increasing Ni concentration.[1] Batalla, Altounian, and Strom-Olsen[3] calculated the spin-fluctuation (SF) mass enhancement λ_{SF} of Zr_xNi_{100-x} amorphous alloys using the corrected McMillan equation.[4,5] They found that λ_{SF} is less than 0.01 for $Zr_{80}Ni_{20}$ while it is about 0.12 for $Zr_{36}Ni_{64}$. We set up experiments to determine the effect of high pressures on $T_c(P)$ of several amorphous Zr_xNi_{100-x} ($x = 80$, 71, 67, 63.5, 60, 58) alloys. In this study we attempt to determine how the high pressures affect T_c as well as the possible role of the spin-fluctuation mass enhancement λ_{SF} on $T_c(P)$.

Samples of amorphous ribbon Zr_xNi_{100-x} alloys were prepared by melt spinning onto a copper wheel; they were characterized by differential scanning calorimetry and x-ray diffraction. Details are given in Ref. 1. The pressure dependence of $T_c(P)$ for alloys with different concentrations of Ni were measured under quasihydrostatic pressure up to 8 GPa (1 GPa=10 kbar). The quasihydrostatic pressure cell consisted of a pyrophyllite gasket with stealite disk as the pressure transmitting medium.[6] The T_c of Pb was used as an internal pressure manometer where the Pb manometer is situated near the sample and the T_c of both measured simultaneously. The temperature was monitored by a calibrated carbon-glass thermometer. A four-point dc resistivity technique was employed to measure T_c in the quasihydrostatic cell. The value of T_c was taken at the midpoint of the resistive transition.

$T_c(P)$ for six Zr_xNi_{100-x} samples ($x = 80$, 71, 67, 63.5, 60, 55) was measured up to quasihydrostatic pressure of 8 GPa. Figure 1 shows relative resistivity as a function of temperature and pressure for a $Zr_{80}Ni_{20}$ amorphous alloy. A sharp superconducting transition with a transition width less than 0.05 K was observed at 1.8 ± 0.2 GPa. However, upon increasing the pressure, the transition width increases and resistivity data clearly shows two transition temperatures. This indicates the existence of two regions within the sample induced by pressure. The T_c of one region increases faster with pressure compared to the second region. Figure 2 shows the T_c of these two regions as a function of pressure. In both cases dT_c/dP is positive. The least-squares fit of T_c's vs pressure intercepts the $p = 0$ axis at temperatures 3.82 ± 0.02 K, and 3.72 ± 0.02 K, where the higher temperature within the experimental error is the T_c of $Zr_{80}Ni_{20}$ at ambient pressure. We carried out an additional high-pressure experiment on $Zr_{80}Ni_{20}$ and we observed that the above results are reproducible.

Samples of Zr_xNi_{100-x} with the $x < 80$ did not show two regions upon increase of pressure. In all samples we observed a sharp transition at T_c with $\Delta T_c < 0.1$ K. In all but one case dT_c/dP is positive and decreases as the concentration of Ni increases. Table I shows the ambient pressure value $T_c(0)$,[1] the extrapolated $T_c(0)$, and pressure derivative of superconducting transition temperature of amorphous Zr_xNi_{100-x} alloys.

We should note here that upon releasing the pressure in the quasihydrostatic cell we observed hysteresis in $T_c(P)$ (about 0.2 to 0.4 K) which is due to the flow of the stealite disk upon releasing of the pressure which produces a large strain in the sample.

Figure 1 clearly indicates the existence of two phases upon applying pressure to a $Zr_{80}Ni_{20}$ amorphous alloy. We carried out an x-ray study of a pressurized sample. The x-ray pattern indicated the existence of several lines apart from the amorphous x-ray pattern. These x-ray lines were matched to the lines due to a crystalline ω-Zr phase with particle size of approximately 180 Å in diameter (this was estimated from broadening of x-ray lines). According to Olinger and Jamieson[7] at high pressure one can form ω-Zr which retains its form when the pressure is
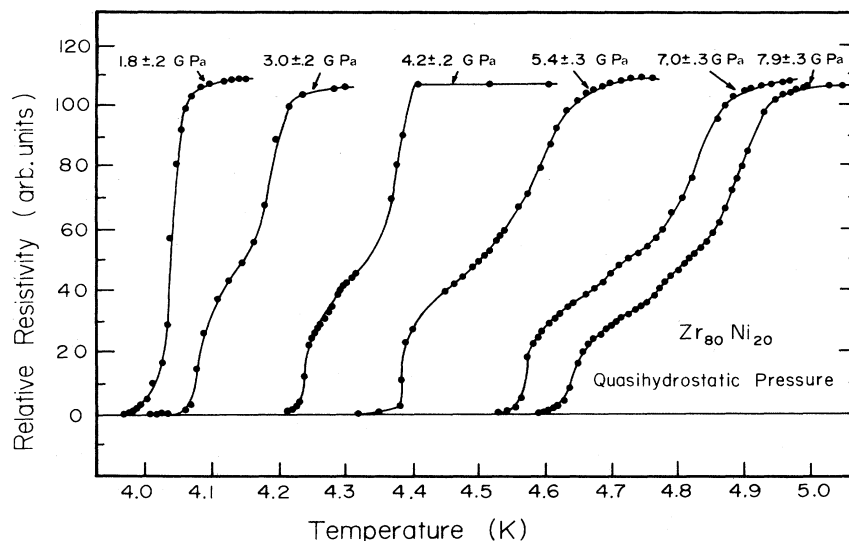
FIG. 1. Normalized resistivity near superconducting transition temperature T_c as a function of pressure for $Zr_{80}Ni_{20}$.

released. It is possible that, in the immediate neighborhood of these ω-Zr crystals, the composition of the glass is slightly richer in Ni content (> 20 at.%) than the rest of the sample; thus it has a lower T_c as well as a lower dT_c/dP in agreement with the data shown in Fig. 2. The estimated concentration of these regions is $\sim Zr_{74}Ni_{26}$. Here we should point out that with our present high-pressure resistivity setup it was not possible to observe the T_c of ω-Zr below the superconducting transition of $Zr_{80}Ni_{20}$ alloy (Zr becomes a superconductor below 0.6 K).

In Fig. 3 we plotted $(1/T_c)\partial T_c/\partial P$ (normalized pressure dependence of T_c) as a function of concentration. The results clearly show that $(1/T_c)\partial T_c/\partial P$ decreases as we move towards a higher Ni concentration and changes sign above 40 at.% Ni concentration.

To understand the above results we discuss qualitatively the effect of pressure on T_c derived from the corrected

McMillan equation[4,5] in which the effect of spin fluctuation is included,

$$T_c = \frac{\Theta_D}{1.45} \exp\left[-\frac{(1+\lambda_{e\text{-ph}}+\lambda_{SF})}{\lambda_{e\text{-ph}}-\lambda_{SF}+\mu^*} \right],$$

where Θ_D is Debye temperature, $\lambda_{e\text{-ph}}$ is the electron-phonon mass enhancement factor, λ_{SF} is the electron mass enhancement parameter due to the spin fluctuation, μ^* is the Coulomb pseudopotential.

If we attribute the change of T_c as a function of concentration at ambient pressure which is mainly due to the effect of spin fluctuation, then the spin fluctuation effects are expected to be reduced by pressure in almost all cases, on the basis of either localized or band models.[8] Thus, according to the corrected McMillan equation, we expect that T_c increases with pressure. However, our results show otherwise, i.e., as we move towards a higher Ni concentration (larger spin fluctuation λ_{SF}) the $(1/T_c)\partial T_c/\partial P$ decreases and even changes its sign to negative for 45 at.% Ni concentration. Thus, the effect of spin fluctuation mass enhancement in Ni-Zr amorphous alloys must be negligible.
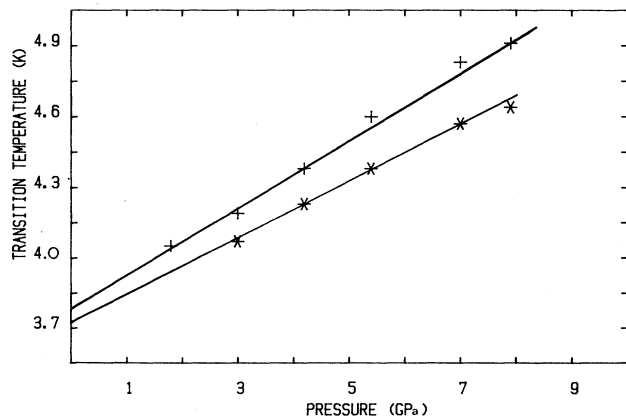

FIG. 2. Quasihydrostatic pressure dependence of T_c for $Zr_{80}Ni_{20}$ upper transition ($+$) and lower transition ($*$) of Fig. 1.

TABLE I. Ambient pressure values and extrapolated values as well as pressure derivative of superconducting transition temperature T_c of Zr_xNi_{100-x} amorphous alloys.

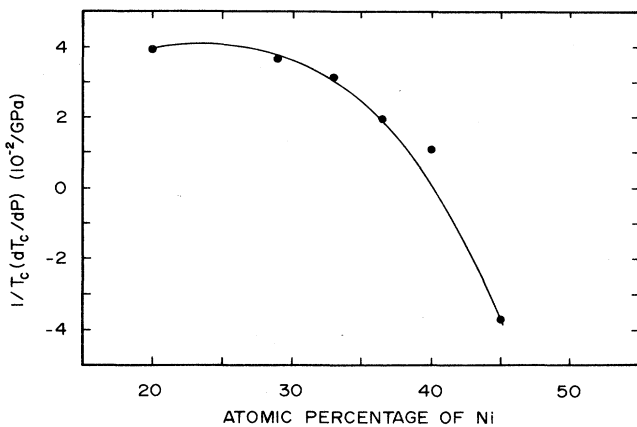| x (at.% Zr)	Ref. 1 $T_c(0)$ (K)	Extrapolated $T_c(0)$ (K)	$(\partial T_c/\partial P)_{p=0} \pm 12\%$ (10^{-2} K/GPa)
80	3.97	3.77	+14.8
71	3.24	2.92	+10.7
67	2.88	2.6	+8.1
63.5	2.56	2.44	+4.7
60	2.35	2.31	+2.45
55	1.83	2	−7.5

FIG. 3. Normalized pressure derivative of superconducting transition temperature $(1/T_c)\partial T_c/\partial P$ as a function of Zr concentration for $Zr_x Ni_{100-x}$ amorphous alloys.

Since there exists no data on the pressure dependence of Θ_D, we estimated the change in Debye temperature as a function of pressure using the compressibility results[9] and a Grüneisen parameter of about 1.5 estimated from other Zr-amorphous alloys.[10] Our estimate for the change in Debye temperature as a function of pressure is of the order of 20% to 16% for $Zr_{80}Ni_{20}$ and $Zr_{55}Ni_{45}$, respectively. Thus, this change in Debye temperature alone cannot account for the observed 75% change in $(1/T_c)\partial T_c/\partial P$ from 80% Zr concentration to 60% Zr concentration as well as a negative $(1/T_c)\partial T_c/\partial P$ for $Zr_{55}Ni_{45}$.

The observed pressure dependence of T_c can be attributed mainly to the change in the density of state which is directly proportional to $\lambda_{e\text{-ph}}$. Figure 3 shows that $(1/T_c)\partial T_c/\partial P$ changes sign at 60 at.% of Zr. Similar behavior was also observed by Cochrane, Destry, and Tudeau[11] in the Hall coefficient measurements of Zr_x-

Ni_{100-x} alloys. Their measurements showed that the Hall coefficient is positive for Zr concentrations above 60 at.% and negative below this concentration. Several band-structure[12-14] calculations on amorphous $Zr_x Ni_{100-x}$ alloys showed that the character of the eigenstates of electron at Fermi energy changes from Zr $4d$ to Ni $3d$ as x decreases. To account for the change of the sign of Hall coefficient using the band-structure results, Cochrane et al.[11] argue that the s-d hybridization and the broader Zr-$4d$ band at the Fermi energy E_F is principally responsible for the positive contribution to R_H. On the other hand, on the Ni-rich side of $Zr_x Ni_{100-x}$ $(x < 60)$ the $3d$ band of Ni at E_F is responsible for negative R_H. Our pressure results agree qualitatively with this picture. For $x > 60$, the application of pressure increases the s-d hybridization. Consequently, the density of states $N(E_F)$ and hence $\lambda_{e\text{-ph}}$ may be enhanced with pressure, i.e., T_c increases with pressure as we observed experimentally. As we move towards the Ni-rich side, the amount of s-d hybridization is reduced by the $3d$ character of Ni; consequently, the T_c changes are slower with the application of pressure. For $Zr_{55}Ni_{45}$ the density of states at the Fermi surface has a $3d$ character[11] and the application of pressure causes the broadening of the $3d$ band. This may reduce the $N(E_F)$ which, consequently, reduces $\lambda_{e\text{-ph}}$ and, thus, T_c decreases as a function of pressure as observed experimentally.

In conclusion, based on the character of the conduction carriers at the Fermi surface we were able to explain the change of T_c as a function of pressures for amorphous $Zr_x Ni_{100-x}$ alloys. However, we should point out that in our arguments we consider that the Coulomb pseudopotential μ^* is pressure independent.

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