Change in the superconducting transition temperature in the $Cu_{1.8-x}Ni_xMo_6S_8$ system

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The effect of a nickel impurity on the superconducting transition temperature T_c of $Cu_{1.8}Mo_6S_8$ in the $Cu_{1.8-x}Ni_xMo_6S_8$ system has been studied. In contrast with the behavior $T_c(x)$ for compounds of $SnMo_6S_8$ with Fe, the behavior $T_c(x)$ for $Cu_{1.8-x}Ni_xMo_6S_8$ is markedly at odds with the theoretical Abrikosov–Gor'kov behavior. The results are discussed.

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It has been reported previously[1] that ferromagnetic impurities lower the critica temperature T_c of superconducting sulfides of molybdenum having the Chevrel-phas structure. The effect of an iron impurity on T_c of $SnMo_6S_8$ was studied in Ref. 2. I was found that the behavior $T_c(x)$ for $Fe_xSnMo_6S_8$ can be described satisfactorily b

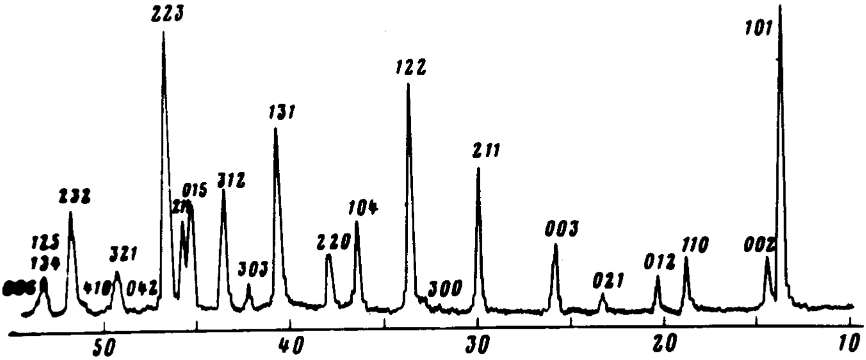

FIG. 1. Diffraction pattern of a $Cu_{1.1}Ni_{0.7}Mo_6S_8$ sample (CuK_α radiation, $\lambda = 1.541$ Å).

the Abrikosov–Gor'kov formula and that the critical density is $x_{cr} = 0.06$. When small amounts of iron were added, it was found that the pressure has an enhanced effect on T_c. The critical concentration found from experiments on the decrease in T_c with the pressure was also about $x_{cr} = 0.06$, and the behavior $T_c(P)$ was similar to $T_c(x)$.

We were interested in the changes in T_c upon the doping of Chevrel superconducting phases with nickel. To study these changes we selected the compound $Cu_{1.8}Mo_6S_8$, which forms a continuous series of solid solutions with $Ni_{1.8}Mo_6S_8$. We have previously studied the behavior $T_c(x)$ for $Fe_xCu_{1.8}Mo_6S_8$ (see Ref. 3, for example); in that case, the value of $(\partial/\partial X)(T_{cx}/T_{c0})$ is 5.0.

Samples were synthesized by the standard method which we have used previously (see Ref. 4, for example). After synthesis, the samples were annealed at 1000 °C. When the annealing temperature was 900 °C we found lines of MoS_2 in the x-ray diffraction pattern, while samples annealed at 1000 °C were essentially single-phase samples. Figure 1 shows a diffraction pattern from a sample with $x = 0.70$. This pattern was recorded in a Geigerflex diffractometer (CuK_α radiation, $\lambda = 1.541$ Å). The samples were either disks 9 mm and ~1 mm thick or parallelepipeds with dimensions of $0.5\times1.5\times4.5$ mm. We measured T_c by inductive and potentiometric methods; for these measurements the samples were mounted in a holder which was inserted into a transfer Dewar for working at temperatures above 4.2 K. For measurements below 1.4 K we used an apparatus, similar to that described in Ref. 5, which performed an adiabatic demagnetization of erbium-yttrium aluminum garnet. In this case the value of T_c was determined from the change in the resistance, while the temperature was determined from the magnetic susceptibility of the garnet.

Figure 2 shows the dependence of T_c on the nickel concentration x. We see that the behavior is different from the Abrikosov–Gor'kov prediction. On the other hand, we observed that this system exhibits a decrease in the hexagonal-cell volume V_H upon an increase in x, similar to that which has been observed elsewhere for $Cu_{1.8}Mo_6S_8$ samples synthesized in a pressure chamber.[6] It may be suggested that the dependence $T_c(x)$ is influenced by two factors: the scattering of Cooper pairs by nickel atoms and a

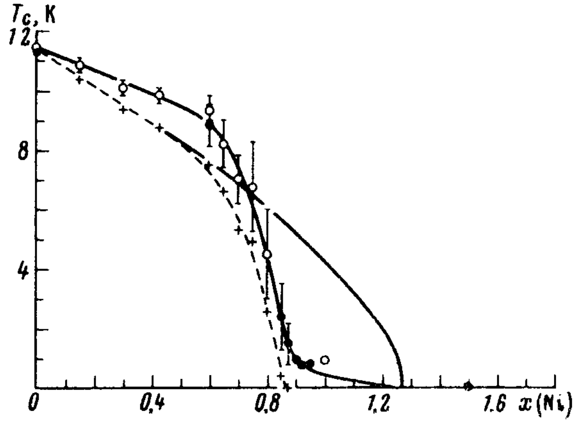

FIG. 2. Superconducting transition temperature T_c vs the nickel concentration. 1—Experimental curve (the vertical bars show the width of the superconducting transition); 2—$T_c(x)$ with allowance for the correlation between T_c and the hexagonal-cell volume V_H; 3—theoretical Abrikosov–Gor'kov formula.

decrease in V_H. If we assume that these two factors are working independently, we can write T_c as $T_c = f(X) + f(V_H)$. Estimating the effect of V_H on T_c from the data of Ref. 6 on the dependence $T_c(V_H)$ for $Cu_{1.8}Mo_6S_8$, we can subtract from the experimental $T_c(x)$ curve for the increase in T_c which might have resulted from a decrease in V_H; then $T_c(x)$ can be approximated by curve 2 in Fig. 2. This curve lies slightly below the Abrikosov–Gor'kov curve, however, and in this case it is very different from the latter. A study of the temperature dependence of the resistance revealed that at a concentration of only $x = 0.15$ the peak on the $\rho(T)$ curve associated with the structural transition of $Cu_{1.8}Mo_6S_8$ shifts toward lower temperatures. It does not disappear, however, until x nears its maximum value, i.e., $x = 1.8$.

From curve 1 in Fig. 2 we see that in the region $x = 0.6$–0.8 the width of the curves of the transition to the superconducting state is ≈ 2 K, i.e., considerably greater than this width at $x < 0.3$ or $x > 0.8$, where it is generally less than 0.1 K.

It should be noted that the broadening of the transition curves in this concentration range was observed in three lots of samples. On the basis of the diffraction patterns of these samples it may be suggested that changes of some sort are occurring in the lattices of these systems at concentrations $x = 0.7$–0.9 (the lines are quite narrow and the widths of the diffraction lines are not correlated with the transition width). As the concentration is raised above 0.9, the widths of the transition curves recorded at ultralow temperatures by the adiabatic-demagnetization method[5] again decrease. Unfortunately, we cannot at this point offer an unambiguous explanation for this behavior of the samples in this concentration range ($x = 0.7$–0.9).

Comparing the behavior $T_c(x)$ found in the present experiments with the corresponding behavior found for samples of $SnMo_6S_8$ and $Cu_{1.8}Mo_6S_8$ with an iron impurity,[2,3] we note that in the case of an iron impurity in $SnMo_6S_8$ and $Cu_{1.8}Mo_6S_8$ the derivative $\partial T_c/\partial X$ is extremely large,[1)] while it is almost an order of magnitude smaller at concentrations $x < 0.6$ in the case of nickel in $Cu_{1.8}Mo_6S_8$. As mentioned above, the dependence $T_c(x)$ in the case of an iron impurity in $SnMo_6S_8$ can be reconciled with the Abrikosov–Gor'kov formula quite well, while in the case of a nickel impurity in $Cu_{1.8}Mo_6S_8$ the $T_c(x)$ curve not only lies substantially higher on the iron concentration scale but also has a shape very different from that of the Abrikosov–Gor'kov curve.

We do not rule out the possibility that when $Cu_{1.8}Mo_6S_8$ is doped with nickel the changes in T_c are caused primarily by a nonmagnetic scattering (a scattering without a spin flip) and that the nickel impurity in this case behaves as do defects in films of certain compounds (see Ref. 7, for example). This interpretation is supported by the nature of the $T_c(\rho_{300}/\rho_{res})$ curve. On the other hand, we again cannot completely rule out the possibility that in the case of the nickel impurity a weak "magnetic" scattering[2)] might be intensified by a superposition on non-magnetic scattering. Such a non-magnetic scattering might, by reducing the mean free path, increase the effectiveness of the interaction with magnetic impurities. When we plot $\pi_{res}(x)$ we find it to be a very nonlinear function of x. In the case of $Cu_{1.8-x}Ni_xMo_6S_8$ we are dealing with relatively high impurity concentrations, in contrast with $SnMo_6S_8$ compounds with an iron impurity.

Further study will probably provide a more accurate explanation for the $T_c(x)$ behavior found for the $Cu_{1.8-x}Ni_xMo_6S_8$ system, especially in the concentration range ($x = 0.7$–0.9), where we find T_c falling off quite sharply with increasing x.

[1)]As mentioned in Ref. 3, the derivative $\partial T_c/\partial x$ for an iron impurity depends on the electron heat capacity, and for $Fe_xSnMo_6S_8$ the value of this derivative is nearly four times that for $Fe_xCu_{1.8}Mo_6S_8$.

[2)]A small fraction of the nickel atoms may retain their magnetic moment; the result would be only an insignificant increase in the susceptibility of the sample.

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