ac susceptibility and electrical resistivity in $Fe_{80-x}Ni_xCr_{20}$ ($21 \leq x \leq 30$) alloys

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(Received 29 May 1984; revised manuscript received 7 September 1984)

ac susceptibility and electrical resistivity studies on polycrystalline $Fe_{80-x}Ni_xCr_{20}$ ($21 \leq x \leq 30$) alloys, with $x=21$, 23, 26, and 30, between 4.2 and 80 K, are reported. A previous dc magnetization study indicated the presence of ferro–spin-glass mixed-phase behavior in $x=23$ and 26 alloys while the alloys with $x=21$ and 30 were found to be spin-glass and ferromagnetic, respectively. The present ac susceptibility results support the above picture. In the electrical resistivity study, a low-temperature minimum in the resistivity-temperature curve is observed in all the alloys except the ferromagnetic one.

I. INTRODUCTION

It is commonly observed that in magnetic alloys, dilution causes the lowering of the transition temperature T_c with concentration c. At some concentration C_p—the percolation concentation—$T_c(C_p)$ reaches 0 K and at this special point the critical behavior is expected to change. The concentration region around this special point has drawn a lot of attention and in the last few years a lot of work has been reported both in the crystalline systems like *Au*Fe,[1] *Cr*Fe,[2] PdFeMn,[1] FeNiMn,[3] *Pt*Mn,[4] *Ni* Mn,[5] Co_2TiO_4,[6] $Eu_xSr_{1-x}S$,[7] and in amorphous alloys like the Fe-Ni (Refs. 8 and 9) and Fe-Mn (Ref. 10) series. In these systems with the variation of concentration of one or more magnetic component, one passes from a spin-glass to a ferromagnet through a new low-temperature magnetic state. There is a lot of controversy about the exact nature of this new magnetic phase. Originally it was thought to be a reentrant spin-glass with no spontaneous magnetization, as was suggested by Sherrington and Kirkpatrick.[11] But recent theories[12–14] suggest that the new magnetic state consists of long-range order along the direction of broken symmetry and spin-glass-like freezing in the perpendicular plane—a mixed phase with the coexistence of spin-glass and fertism.

Majumdar and Blanckenhagen[15] (MB) have made detailed dc magnetic measurements on polycrystalline $Fe_{80-x}Ni_xCr_{20}$ alloys with $10 \leq x \leq 30$. They found that two of the alloys with $x=23$ and 26 make transitions at T_{SG} from a higher temperature ferromagnetic to a lower temperature ferro–spin-glass mixed phase. They observed spontaneous magnetization below T_{SG} down to the lowest temperature of 2 K, in those two alloys. The magnetization, apart from showing the usual paramagnetic-ferromagnetic transition at a temperature T_c, again drops off at a lower temperature T_{SG}. This is the typical signature of a ferromagnetic to a mixed-phase transition.[1] Here, we shall report the results of study of the ac susceptibility and electrical resistivity of these two alloys from 4.2 to 80 K as well as on two other alloys of the same series with $x=21$ and 30 which were found to be pure spin-glass and ferromagnetic, respectively, from neutron diffraction and dc-magnetization measurements.[15] The motivation behind the present work is to check whether or not the double transition shows up in the ac susceptibility and resistivity measurements as well.

II. EXPERIMENT

The alloys were prepared by induction melting in an argon atmosphere.[15] For resistivity measurements, thin rectangular strips, obtained after cold rolling the master ingots, were used. For susceptibility measurements we used thin needle-like samples. This was done in order to minimize the demagnetization factor. To avoid strains, etc., introduced during the cold rolling, the samples were annealed in argon atmosphere at 1050 K for 24 h and then quenched in water.

For susceptibility measurements a mutual inductance bridge has been used. The bridge operates at a frequency of 117 Hz, which is sufficiently removed from the main's frequency. The susceptibility is related to the change in mutual inductance produced by the introduction of the sample into a coil system involving a primary and two secondaries. The secondaries are wound in opposite senses and the sample is placed in one of the secondaries. The change in susceptibility is measured using a bridge on the lines of the vacuum tube version of Pillinger, Jastram, and Daunt[16] and the solid-state version of Whitmore and Ryan.[17] A PAR lock-in amplifier (model 5204) was used as a null detector for the bridge. A voltage divider (Kelvin Varley) and a decade resistor was used to balance, independently, the real and the imaginary part of the susceptibility, respectively.

For the measurement of resistivity we have made use of the four-probe ac technique. In our case the constant current (alternating) which is made to pass through the sample, also passes through a standard resistor. A part of the voltage developed across the standard resistor is compared with the voltage developed across the sample. This comparison is done using the same PAR lock-in amplifier as in the susceptibility measurements. The output of the lock-in amplifier is made zero by tapping out a voltage (equal to the voltage developed across the sample) from the standard resistor by a highly stable voltage divider (Kelvin Varley). In this method a resistance change of 1 part in 10^5 or better can be detected.

The same cryostat is used to measure both the resistivity and the susceptibility as a function of temperature. The design of the cryostat was such that the sample zone could be directly immersed in a commercial liquid-helium Dewar. The temperature of the sample zone could be changed by varying the position of the sample zone from the liquid-helium level. An Allen Bradley carbon resistor was used as a thermometer. With this arrangement the accuracy of the temperature measurement was ±0.25 K between 4.2 and 40 K and ±1 K between 40 and 80 K.

III. RESULTS AND DISCUSSION

A. ac susceptibility

Figures 1(a)–1(c) show the zero-field-cooled ac susceptibility χ vs T for the alloys with $x = 21$, 23, and 26. The pure spin-glass sample ($x = 21$) shows a sharp peak at a temperature $T_{SG} = 17$ K. The previous dc magnetization experiments[15] showed that the spin-glass transition temperature for this alloy was 10 K. For the two mixed-phase samples there are high peaks at $T = T_c$ (23 and 40 K for $x = 23$ and 26, respectively) indicating a paramagnetic-ferro-

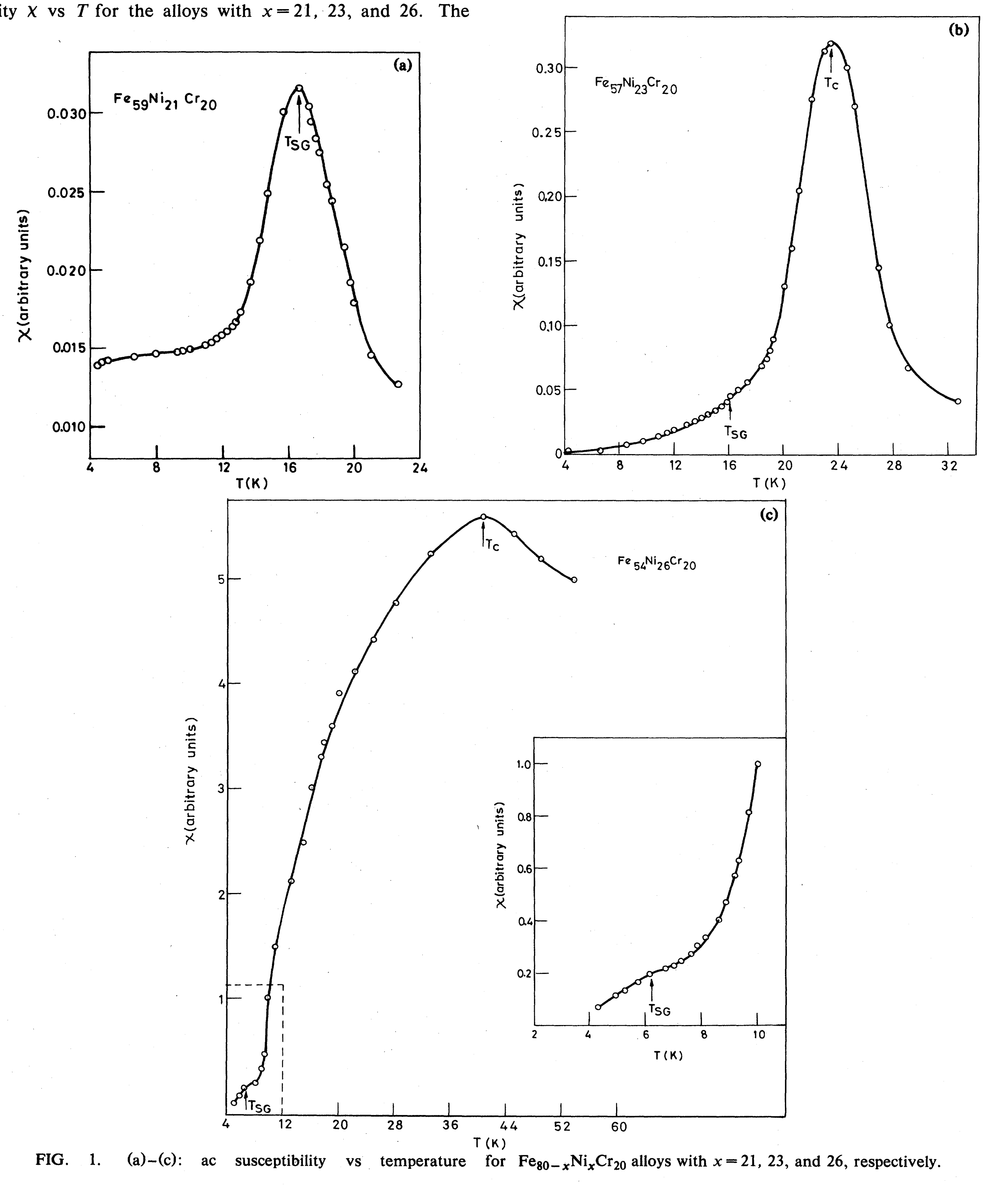

FIG. 1. (a)–(c): ac susceptibility vs temperature for $Fe_{80-x}Ni_xCr_{20}$ alloys with $x = 21$, 23, and 26, respectively.

magnetic transition, with a change in slope at a lower temperature T_{SG} (16 and 6 K for $x=23$ and 26, respectively). This behavior of ac susceptibility is similar to that observed in the mixed-phase region of AuFe.[18] The change in slope of the susceptibility curve is taken as an indication of the onset of a spin-glass-like phase at the lower temperatures. The T_c and T_{SG} obtained from the present measurements are slightly different from those observed in dc magnetization.[15] The ratio of $\chi(\text{peak})/\chi(4.2\text{ K})$ is about 200 times higher for the two mixed-phase alloys than the corresponding value in the spin-glass alloy showing that the former two peaks are at T_c while the latter is at T_{SG}.

B. Resistivity

The resistivity $\rho(T)/\rho(4.2\text{ K})$ vs T plot is shown in Fig. 2. The two mixed-phase alloys with $x=23$, 26 show a change in slope at $T_c=39$ and 63 K, respectively. But these temperatures are shifted from what have been observed through present magnetic measurements as well as those of MB.[15] At lower temperatures these two alloys show broad minima around 10 and 15 K, respectively, as shown in Fig. 2. For the other two alloys, the ferromagnetic one shows no minimum but the spin-glass alloy again shows a broad low-temperature minimum around 14 K.

It is now well known that very dilute alloys of Cu, Ag, Au, Mg, Zn with Cr, Mn, and Fe, show a low-temperature resistivity minima—this is the famous Kondo effect. But one hesitates to say that the observed concentrated region minima as in the present case and in the other concentrated magnetic systems (which we shall discuss subsequently) are of the same Kondo-type origin. Electrical resistivity measured in Au-Ni alloys containing 0–60 at. % Ni in the range 1.5–300 K, show resistivity minima between 30 and 42 at. % Ni while the alloys below 20 at. % Ni behave as normal metals.[19] Above 42–45 at. % Ni, long-range ferromagnetic ordering sets in. The concentration region where the minima have been observed is sandwiched between a normal paramagnetic region and a ferromagnetic region. This fact leads to a belief of magnetic origin of the observed minima. Shiozaki and co-workers[20] found an electrical resistivity minimum in highly concentrated *Au*Cr alloys with 22.2 and 28.3 at. % Cr. The resistivity minimum was observed only for samples with a long-range antiferromagnetic order. The anomalous part of the resistivity can be fitted to a $\ln T$ expression and this tends to suggest a Kondo-like behavior. But the authors (Shiozaki and co-workers) argued that the magnetic moment of a Cr atom, forced to be ordered could not produce such an effect. They also checked that changes in the purity of the starting materials for alloy preparation has no influence on resistivity and hence another magnetic dilute impurity with a low Kondo temperature could not be responsible for the origin of this minimum. Resistivity minimum has also been observed in *Au*Fe(6 at. % Fe) film,[21] NiCu,[22] *Ni*V,[23] Pt-Co,[24] Pt-Cr,[25] Pt-Mn,[26] PdAgFe,[27] FeSi (Ref. 28) at low temperatures. Such resistivity minima are well known in amorphous alloys both magnetic and nonmagnetic.[29] There is a lot of controversy whether such a minimum, in amorphous alloys, is of magnetic or structural origin.[29]

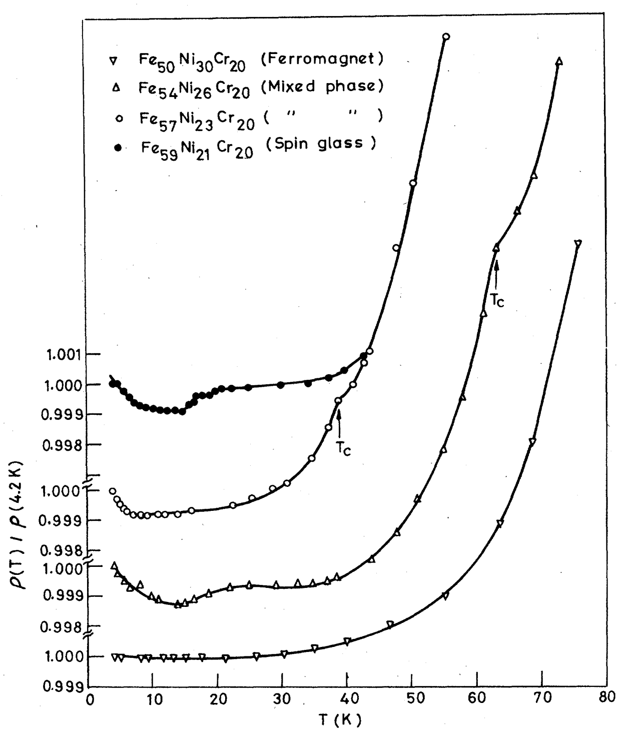

FIG. 2. $\rho(T)/\rho(4.2\text{ K})$ vs temperature for $Fe_{80-x}Ni_xCr_{20}$ alloys with $21 \leq x \leq 30$.

In our present study we have gathered from different isolated works (as mentioned above) that in many of the systems the resistivity minimum occurs in a concentration region, where long-range magnetic order sets in. This behavior suggests a correlation between the observed resistivity minimum and the magnetic order in that concentration region. Magnetic studies on Au-Ni (Ref. 30) and Ni-Cu (Ref. 31) revealed a low-temperature ferro–spin-glass mixed phase in the concentration region just prior to the onset of long-range magnetic order. Our present investigation is interesting in the fact that in all the alloys, which show the resistivity minimum, a spin-glass or ferro–spin-glass mixed phase exists at low temperatures. We observed no such minimum in the pure ferromagnetic alloy. This shows that the spin-glass-like ordering at low temperatures certainly plays a role in the resistivity minimum phenomenon.

To our knowledge there is no general theory which takes into account of the resistivity minimum in concentrated crystalline magnetic alloys. But Rivier[32] and Fischer,[33] using two different arguments, have discussed the possibility of a resistivity minimum in spin-glasses such as *Pt*Mn. A preliminary theoretical calculation[34] shows that partial spin freezing in the ferro–spin-glass mixed-phase region may increase the resistivity. Levin and Mills[35] tried to explain the anomalous temperature dependence of the resistivity in Ni-Cu alloys near the critical concentration for ferromagnetism, in terms of Kondo minimum arising from spin clusters in that region. In the amorphous front it is still controversial whether the observed resistivity minimum is of structural or magnetic origin.[36] But the trend nowadays seems to be in favor of explaining the resistivity minimum by magnetic effect rather than by nonmagnetic mechanism.[37] Support in this regard has been obtained from systematic studies on amorphous FeNi alloys[38–40] which have shown definite correlations between the resistivity anomalies and the magnetic properties. Several mechanisms like the modified Kondo effect,[41] conduction electron scattering from correlated magnetic clouds,[42] and spin couples[43–46] have been suggested as the cause of the resistivity anomaly in amorphous magnetic alloys.

In conclusion, we point out that our ac susceptibility measurements, which is the first on the present series of alloys gives further support to the existence of ferro–spin-glass mixed phase, revealed only by the previous dc magnetization measurement.[15] Furthermore, resistivity study shows a low-temperature anomaly in the form of a resistivity minimum. We suggest here that this anomaly in the present system as well as in others cited above has some correlation with the complicated magnetic ordering in these alloys. At present we are trying to study the magnetoresistance of these four $Fe_{80-x}Ni_xCr_{20}$ alloys. This, we hope, will throw more light on the exact nature of the low-temperature mixed phase and hence to the observed low-temperature anomalies.

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