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Near zero temperature coefficient of resistivity in antiperovskite $Mn_3Ni_{1-x}Cu_xN$

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The near zero temperature coefficient of resistivity (NZ-TCR) in Mn-based antiperovskite $Mn_3Ni_{1-x}Cu_xN$ is reported. The temperature range of NZ-TCR is controllable by changing Cu content. Further, the TCR value of 0.09 ppm K^{-1} was obtained in $Mn_3Ni_{0.5}Cu_{0.5}N$ over a broad temperature range around room temperature. The anomalous resistivity change of $Mn_3Ni_{1-x}Cu_xN$ from metal-like to NZ-TCR behavioris apparently due to a magnetic transition. The possible reason for the formation of NZ-TCR is interpreted on the basis of spin-disorder scattering. © *2011 American Institute of Physics*. [doi:10.1063/1.3671183]

Manganese antiperovskite materials Mn_3AX (A = transition metals and Ga, Ge, Ag, and, Sn; X = N or C) have exhibited a wide range of interesting physical properties, such as negative thermal expansion (NTE),[1–4] giant magnetoresistance (GMR),[5] piezomagnetic effect,[6] magnetocaloric effect (MCE),[7] and near zero temperature coefficient of resistivity (NZ-TCR),[8] due to strong correlation among lattice, spin, and charge.[9] Early reports suggested that the physical properties of these materials are mainly originated from the complex magnetostructure and the induced variable band structure.[10–12] The ability to control and tune the electronic transport, magnetic transition, and abnormal thermal expansion behaviors of these materials makes it worthy to be studied for fundamental research as well as for potential applications.

Electrical resistivity (a physical property of enormous importance) displays increasing or decreasing normally as a function of temperature. A very limited number of materials, however, exhibit invariant electrical resistivity with the changing temperature and show an NZ-TCR behavior. These materials are used in many fields such as resistors in high precision electronic measuring systems, automobile temperature sensing circuitry, and in thermoelectric devices.[14,15] Currently, for example, the characteristic application is surface mounted device (SMD) resistor,[13] which is well known for remarkable high precision and high stability. It is popularly applied in high precision electronic systems such as communication device, Global Positioning System (GPS), Personal Digital Assistant (PDA), and so on. From a practical viewpoint, however, SMD resistors are very expensive possibly because of their complicated preparation routes and the cost of raw materials. To promote a wide range of applications, it is desired to find low-cost substitutes for the current resistors. Up to now, it has been found that Mn_3CuN, Mn_3AgN, and Mn_3NiN compounds exhibit the NZ-TCR behavior when they are in pure-form.[8,16,17] As reported by Chi *et al.*,[8] the NZ-TCR behavior was discovered in Mn_3CuN above 150 K, and the temperature coefficient of resistivity (TCR) value was up to 46 ppm K^{-1}. We have recently reported the low TCR in the Mn_3NiN compound, in which the electrical resistivity remained almost constant above 250 K.[17] The Mn_3AX compounds, as promising NZ-TCR candidate, possess the advantages of high electronic and thermal conduction. It has been difficult to achieve NZ-TCR in a controllable temperature range. In the Mn_3AX, the metal atoms on A site have itinerant electrons at the Fermi level, and different elements display different properties in magnetism and electronic transport.[18] By studying the doping action in antiperovskite material Mn_3NiN, we want to test the feasibility to tune the temperature range for NZ-TCR and decrease the TCR value in these materials.

In the work reported in this article, we investigated the electronic transport and magnetic transition of $Mn_3Ni_{1-x}Cu_xN$. All of the compounds show an NZ-TCR behavior with a controllable temperature range for NZ-TCR. The small percent of Cu dopant at the Ni site of Mn_3NiN

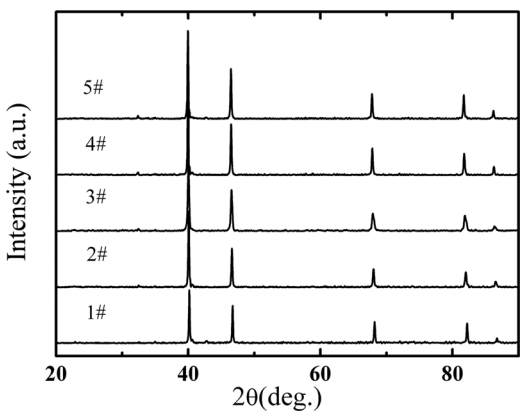

FIG. 1. X-ray diffraction patterns of $Mn_3Ni_{1-x}Cu_xN$ samples in the 2θ range of 20° to 90°: (1) Mn_3NiN, (2) $Mn_3Ni_{0.7}Cu_{0.3}N$, (3) $Mn_3Ni_{0.5}Cu_{0.5}N$, (4) $Mn_3Ni_{0.3}Cu_{0.7}N$, and (5) Mn_3CuN.

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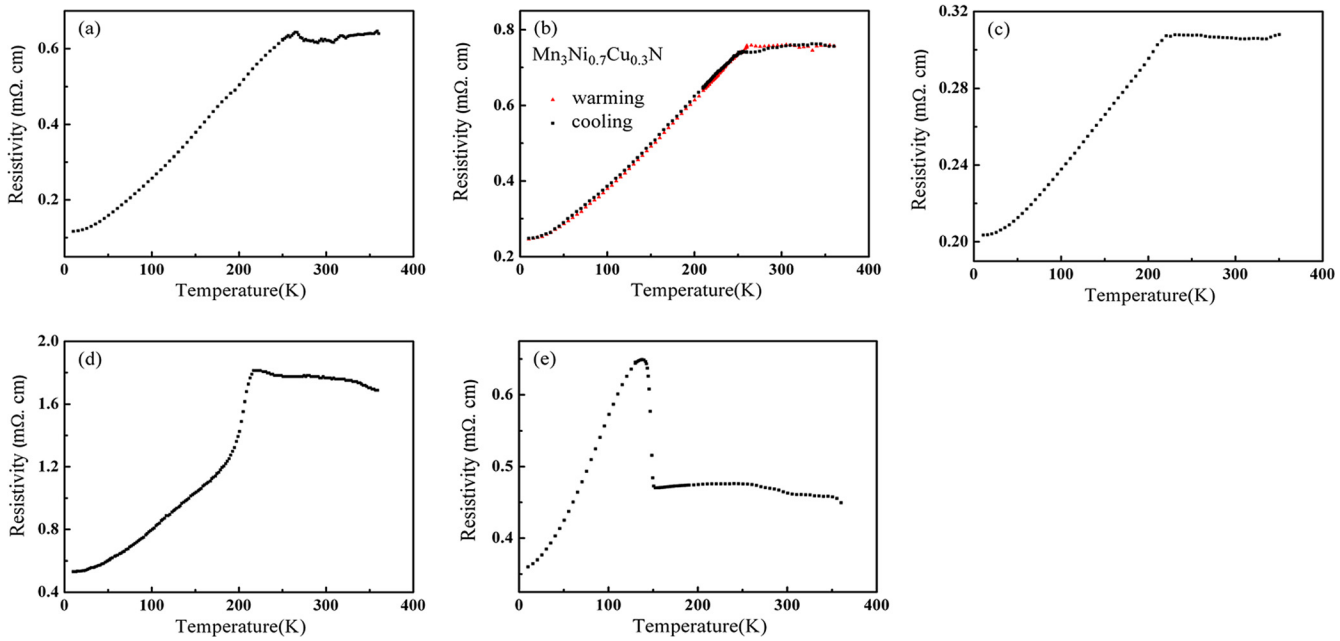

FIG. 2. (Color online) Temperature (in unit of K) dependence of the electrical resistivities measured during a warming process from 10 K to 360 K in $Mn_3Ni_{1-x}Cu_xN$ materials: (a) Mn_3NiN, (b) $Mn_3Ni_{0.7}Cu_{0.3}N$ (during the cooling and warming processes), (c) $Mn_3Ni_{0.5}Cu_{0.5}N$, (d) $Mn_3Ni_{0.3}Cu_{0.7}N$, and (e) Mn_3CuN.

effectively changes the TCR value of $Mn_3Ni_{1-x}Cu_xN$, and we obtained the lowest TCR value of 0.09 ppm K^{-1} in anti-perovskite materials.

Polycrystalline samples $Mn_3Ni_{1-x}Cu_xN$ ($x = 0$, 0.3, 0.5, 0.7, and 1) were synthesized by solid-state reaction, as we have reported.[4,17,19] X-ray diffraction (XRD) patterns of the resulting samples were obtained from an X'Pert PRO powder diffractometer using Cu k_α radiation at room temperature. Structural characterization showed that the samples crystallize in the cubic system with $Pm\bar{3}m$ space group. The resistivity measurements were done by standard four-probe technique from 10 K to 360 K using a physical property measurement system (PPMS). All the M (T) curves from 10 K to 360 K were measured with magnetic property measurement system (MPMS) with an external magnetic field of 500 *Oe*, with a temperature stability of $\pm 0.5\%$, and temperature accuracy of less than 1%.

Figure 1 displays the XRD patterns for $Mn_3Ni_{1-x}Cu_xN$. The crystallographic data were calculated using the Powder X software.[20] It was found that the lattice constant increases with increasing Cu contents, indicating that the Cu atoms were substituting Ni atoms in these materials.

The electrical resistivities of specimens of $Mn_3Ni_{1-x}Cu_xN$ are shown as a function of temperature in Fig. 2. From the $\rho - T$ curves, it is clear that the electrical resistivity is temperature independent in certain range of temperature. This is usually called NZ-TCR, and the critical temperature point of NZ-TCR is indicated as T_{NZ}. The samples show an anomalous variation of resistivity behavior, i.e., the samples exhibit a metallic conduction behavior below T_{NZ}, and the resistivity becomes essentially temperature independent above T_{NZ} It is noteworthy that the critical temperature point changed with the introduction of dopant. With increasing Cu-doped contents, T_{NZ} decreases (the T_{NZ} of $Mn_3Ni_{1-x}Cu_xN$ are 261 K, 260 K, 220 K, 216 K, and 150 K for $x = 0$, 0.3, 0.5, 0.7, and 1.0, respectively). In the measured temperature range ($10\,K < T < 360\,K$), values of ΔT_{NZ} displaying NZ-TCR behavior are 85 K, 100 K, 140 K, 144 K, and 210 K for Mn_3NiN, $Mn_3Ni_{0.7}Cu_{0.3}N$, $Mn_3Ni_{0.5}Cu_{0.5}N$, $Mn_3Ni_{0.3}Cu_{0.7}N$, and Mn_3CuN, respectively, showing that we can obtain an NZ-TCR in controllable temperature region.

In order to get the TCR value, the $\rho - T$ curve is linearly fitted for each sample. The fitting results are listed in Table I. For $Mn_3Ni_{0.7}Cu_{0.3}N$, the value of $d\rho/dT$ is determined as 1.67×10^{-8} Ω. cm K^{-1}, and the TCR value is obtained using the equation

$$\mathrm{TCR} = \rho_0^{-1} d\rho/dT, \tag{1}$$

where ρ_0 is the resistivity at 273 K. For the $Mn_3Ni_{0.7}Cu_{0.3}N$ compound, the TCR is 22 ppm K^{-1} ($260\,K < T < 360\,K$). Remarkably, the TCR value is close to the value for zero TCR materials, which was below 25 ppm K^{-1}.[8] It is nearly one-half of the TCR value of 46 ppm K^{-1} observed by Chi *et al.* in Mn_3CuN.[8] Furthermore, the values of $d\rho/dT$ and TCR for all of the compounds are shown in Table I. For $Mn_3Ni_{0.5}Cu_{0.5}N$, the TCR is about 0.09×10^{-6} K^{-1} (0.09 ppm K^{-1}) between 300 K and 330 K, which is near

TABLE I. The fitting parameters $d\rho/dT$ and the calculated results of TCR value for $Mn_3Ni_{1-x}Cu_xN$ samples.

Samples	$x = 0$	$x = 0.3$	$x = 0.5$	$x = 0.7$	$x = 1.0$
$d\rho/dT$ (Ω cm K^{-1})	1.86×10^{-7}	1.67×10^{-8}	1.15×10^{-8}	6.79×10^{-7}	6.88×10^{-8}
TCR (ppm K^{-1})	288	22	37	346	146

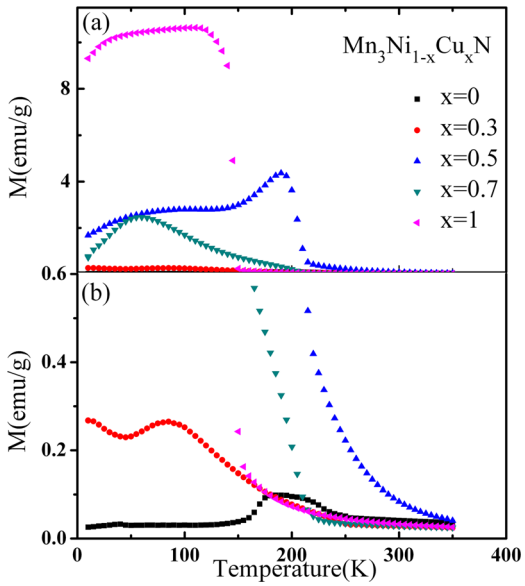

FIG. 3. (Color online) The temperature-dependent magnetization M (T) of $Mn_3Ni_{1-x}Cu_xN$ measured during a warming process in an applied magnetic field $\mu_0 H = 0.5$ T. Note: (b) shows the magnified *M-T* curves of (a).

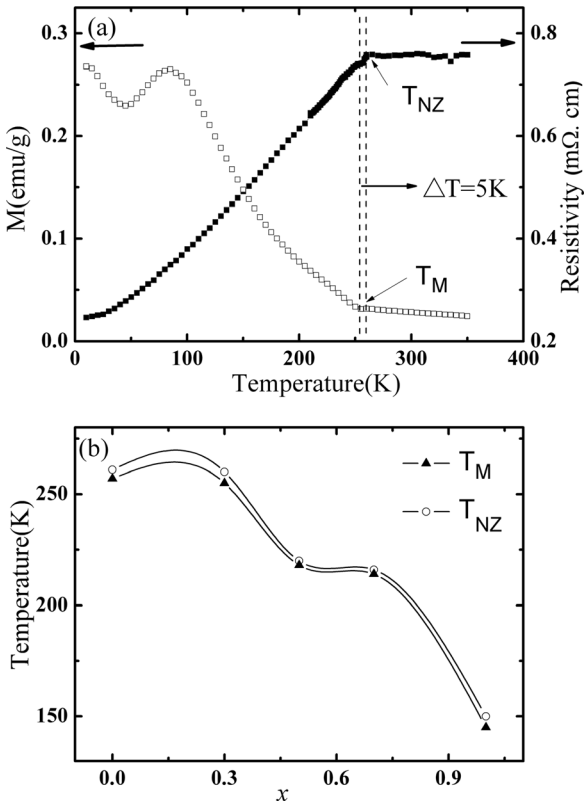

FIG. 4. (a) Temperature dependence of the resistivity and the magnetization for the compound $Mn_3Ni_{0.7}Cu_{0.3}N$. In addition, the relationship ($\Delta T = 5$ K) between the T_M and T_{NZ} for $Mn_3Ni_{0.7}Cu_{0.3}N$ is also shown. (b) Relationship between the temperature of magnetic transition (triangles) and the temperature of NZ-TCR (circles) in the $Mn_3Ni_{1-x}Cu_xN$ compounds (x = 0, 0.3, 0.5, 0.7, and 1.0).

room temperature. This value of TCR is much smaller than the previously reported in $Mn_3Ag_{1-x}Cu_xN$.[16] In order to verify the stability and reproducibility of the materials and their NZ-TCR behavior, the $\rho - T$ curve of the $Mn_3Ni_{0.7}Cu_{0.3}N$ compound was measured in the warming and cooling processes. As indicated by Fig. 2(b), the material is stable and the $\rho - T$ curves during the thermal cycling are almost overlapped.

Previous investigations show that the antiperovskite compounds have intriguing magnetic structure with strong correlation between the magnetic transition and abnormal electronic transport. The zero-field-cooled (ZFC) magnetization curves M(T) of $Mn_3Ni_{1-x}Cu_xN$ from 10 K to 350 K at an external field of 500 *Oe* are shown in Fig. 3. We indicated the initial temperature of spin-ordering as TM for all of the compounds. Analysis of the M(T) indicates that the T_M temperatures are 257 K, 255 K, 218 K, 214 K, and 145 K for x = 0, 0.3, 0.5, 0.7, and 1.0, respectively. To show the correlation between magnetism and electronic transport properties, the temperature dependences of the magnetization and the resistivity for the $Mn_3Ni_{0.7}Cu_{0.3}N$ compound are shown in Fig. 4(a), showing that the gap between the T_M and T_{NZ} for $Mn_3Ni_{0.7}Cu_{0.3}N$ is $\Delta T = 5$ K. Then, the correlation between T_M and T_{NZ} as a function of Cu-doped content is shown in Fig. 4(b) for all the samples. The figure indicates the close correlation between electronic transport and magnetic transition and implies that the magnetic transition plays a critical role in engendering NZ-TCR behavior.

Disorder among the ionic moments enhances with increasing temperature, producing a corresponding increase in the spin-disorder contribution to the resistance until the transition temperature is reached. Above the transition temperature, long-range order is completely destroyed, but the short-range order remains effective. De Gennes and Friedel have reported that short-range effects can cause the resistivity not only to be temperature dependent above the transition point but actually to exhibit a negative temperature coefficient.[21] The short-range effects probably balance the phonon effect, producing a constant resistivity, in some cases producing a shallow minimum before achieving a constant value of resistivity.[22,23]

In summary, we synthesized a series of materials of $Mn_3Ni_{1-x}Cu_xN$ (x = 0, 0.3, 0.5, 0.7, and 1) by solid-state reaction in vacuum, and all compounds exhibited an NZ-TCR behavior. In particular, it was found that the temperature range with NZ-TCR behavior is controllable by changing composition. Furthermore, the lowest value of TCR was found to be 0.09 ppm K^{-1}. The anomalous change in electronic transport from metal-like to NZ-TCR behavior is corresponding well to the magnetic transition. The general explanation of the anomalous resistivity was discussed based on the spin-disorder scattering picture. The present material may have a wide range of applications by virtue of the controllable temperature range of NZ-TCR.

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