

CeMnNi₄: A soft ferromagnet with a high degree of transport spin polarization

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In this letter we introduce a soft ferromagnetic compound, i.e., CeMnNi₄, which exhibits a large moment ($\sim 4.95 \mu_B/\text{Mn}$) and high degree of spin polarization. The system has a ferromagnetic transition temperature of 140 K. Isothermal magnetization measurements at 5 K reveal that the material is a soft ferromagnet with a magnetization saturating at about 1500 Oe and a coercive field of < 5 Oe. We determine the transport spin polarization of this material from point contact Andreev reflection measurements to be 66% thereby making this material potentially important for spintronic applications. © 2006 American Institute of Physics. [DOI: 10.1063/1.2162798]

With the advent of “spintronics,” realizing the ability to inject and manipulate spin polarized electrons in metals and semiconductors has gained particular relevance.¹ The basic philosophy behind this emerging area of research is to be able to inject spin polarized electrons and manipulate the spin as well as the charge to realize novel device schemes and to enhance functionality of existing devices. In this context the search for ferromagnets with a high degree of spin polarization, which can be used as source of spin polarized electrons, has gained particular importance. The forerunner in this class of materials² is CrO₂ which exhibits a near complete spin polarization (96%) at the Fermi level. However, difficulties in synthesizing high quality CrO₂ films through conventional deposition techniques have so far hindered the use of this material for spin injection in devices. Though several other compounds such as CoS₂,³ the Heusler alloy NiMnSb (Ref. 4), and Co₂MnSi (Ref. 5), and the doped rare-earth manganite La_{0.7}Sr_{0.3}MnO₃ (Ref. 6) have been predicted to have a large degree of spin polarization at the Fermi level from electronic structure calculations, experimental observations of spin polarization exceeding 60% have been very few. Several detrimental factors, such as depolarization of the electrons at a disordered interface,⁷ spin orbit coupling and spin fluctuation effects are believed to be responsible for the reduced spin polarization observed in these compounds. It is also important to note that though the degree of spin polarization is one of the most important criterion for a spin source, several other factors are also crucial to realize efficient spin injection: These include, the Fermi velocity mismatch between the source and the material in which the electrons are injected, the ease to fabricate the material in thin film form, the robustness of the spin polarization against detrimental effects of disorder and the magnetic properties of the material. Since these requirements are expected to be different for different applications, identifying ferromagnets with a large degree of spin polarization is crucial for the progress of spintronics.

In this letter we report the magnetic and transport properties of CeMnNi₄, a soft ferromagnet with a $T_c \sim 140$ K which exhibits a large degree of transport spin polarization.⁸ This material is derived from the Pauli paramagnet CeNi₅,⁹

by substituting one Ni site with Mn in the parent compound. Magnetic measurements on CeMnNi₄ show that this material is a soft ferromagnet with a coercive field of less than 5 Oe. Point contact Andreev reflection (PCAR) measurements on this compound reveal that the material has a large degree of transport spin polarization.

The alloy CeMnNi₄ was prepared by arc melting the constituents in several steps. In the first step, stoichiometric amounts of Ce and Ni were melted to form Ce+4Ni alloy. Subsequently, small amounts of Mn (~ 20 mg) are added to the alloy obtained from the previous melt and the alloy is remelted until the required amount of Mn is reacted to obtain CeMnNi₄. In each step the weight of the alloy is monitored and any weight loss due to the volatile Mn is compensated by taking extra Mn in the subsequent melting. The alloy thus obtained is then remelted several times to ensure proper homogenization. The as cast button was then sealed in an evacuated quartz tube and heated to 700 °C at 1.5 °C/min and kept at 700 °C for 2 days. The temperature was then gently raised to 800 °C and maintained for 8 days followed by quenching in liquid nitrogen. The sample was characterized by x-ray diffraction using Cu K_α radiation. Magnetic measurements were carried out on a superconducting quantum interference device magnetometer. The transport spin polarization was measured using a home built PCAR setup operating down to 2.8 K in a continuous flow cryostat. Prior to the measurements the sample surface was polished with fine emory paper to a mirror finish and immediately loaded for measurements to avoid surface degradation. Mechanically cut fine tips of Nb were engaged on the sample surface at low temperature using a differential screw arrangement to establish a point contact. The differential conductance [$G(V)$] of the contact was recorded as a function of voltage (V) using a four-probe modulation technique operating at 372 Hz. The resistance of all the point contacts analyzed in this paper was between 10 and 20 Ω .

Figure 1 shows the powder x-ray diffraction pattern of CeMnNi₄. All the peaks could be indexed on the basis of cubic AuBe₅ (C15b)-type structure with a lattice parameter $a = 6.987$ Å. Our result is in conformity with the earlier observation by Kalychak *et al.*¹⁰ who had found a change of structure in CeMn _{x} Ni_{5- x} from hexagonal CaCu₅ ($x \leq 0.9$) to cubic AuBe₅ type at higher Mn concentrations; i.e., $0.9 \leq x$

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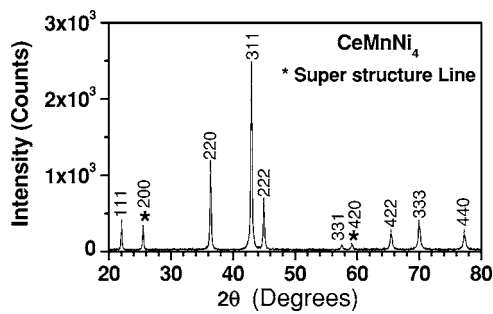


FIG. 1. Powder x-ray diffraction spectrum of CeMnNi_4 . The peaks are indexed on the basis of C15b structure. The lines marked "*" are the superstructure lines arising from the ordering of Ce and Mn.

≤ 2.1 . In analogy with UMNi_4 and ZrMnNi_4 (Ref. 11) ($M = \text{In, Zn and Sn}$), a possible superlattice cubic structure of MgSnCu_4 type for CeMnNi_4 is inferred from the presence of superstructure lines (200) and (420) in the x-ray diffraction spectrum. The superstructure arises when the Mn atoms selectively substitute for the Ni atoms at the 4(c) site of the cubic AuBe_5 -type unit cell.

Magnetization versus temperature for the CeMnNi_4 sample recorded in a field of 100 Oe [Fig. 2(a)] reveals a sharp ferromagnetic transition with $T_c \sim 140$ K. The signature of the ferromagnetic transition is also seen as a pronounced knee in the resistivity versus temperature (ρ - T) at the same temperature [inset Fig. 2(a)].¹² A saturation magnetization of $4.94 \mu_B/\text{f.u.}$ is obtained from isothermal magnetization versus field (M - H) scan recorded at 5 K [Fig. 2(b)]. This value is in remarkable agreement with moment calculated from recent electronic structure calculations.¹³ While in-principle, Ce, Mn, and Ni can all possess magnetic moments, the parent compound CeNi_5 is a Pauli paramagnet where Ce and Ni do not carry any moment.¹⁴ We therefore believe that the ferromagnetic transition at 140 K arises due to the ordering of the Mn moments, which in-turn implies

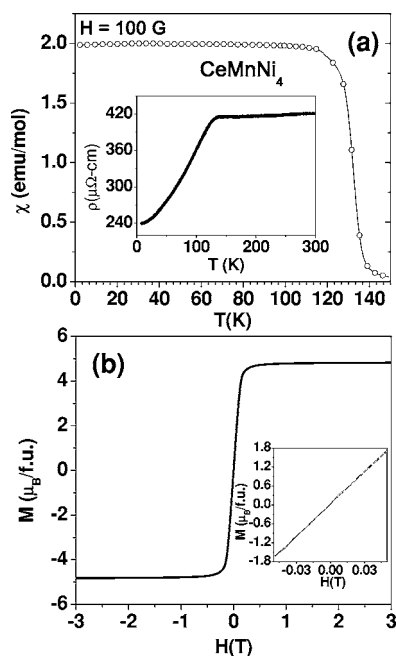


FIG. 2. (a) Magnetization vs temperature of CeMnNi_4 measured in a field of 100 Oe; the inset shows the resistivity as a function of temperature. (b) M - H measured at 5 K; the inset shows a blow up of the M - H loop close to zero field.

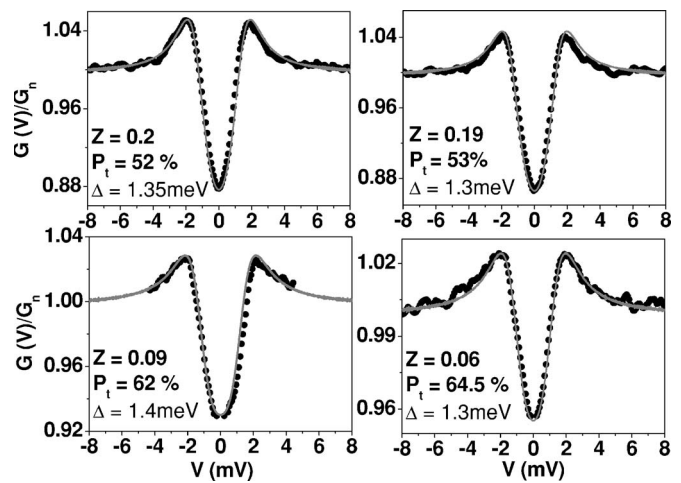


FIG. 3. Representative PCAR, G - V spectra measured at 3.4 K using a Nb tip. The filled circles show the experimental data and the solid lines show the best fit to the modified BTK model. The best fit values of P_t , Z , and Δ are shown in the figure. The values of Δ extracted from the best fit curves of all the spectra range between 1.3 and 1.4 meV.

that manganese in this compound is in a high spin Mn^{2+} valence state. The four quadrant M - H scan also reveals that the system has very low hysteresis [inset Fig. 2(b)] with a coercive field $H_c < 5$ Oe and a very low saturation field ~ 1500 Oe. The observation of such a small hysteresis and saturating field for a strong ferromagnet is intriguing from the point of view of application since the magnetization can be reversed by the application of small magnetic field. The compound shows a metallic behavior in the entire temperature range [inset Fig. 2(a)] with a residual resistivity, $\rho_0 \sim 240 \mu\Omega \text{ cm}$.

In order to explore the transport spin polarization at the Fermi level we carried out PCAR measurements using a superconducting Nb tip. Figure 3 shows the representative PCAR $G(V)$ - V spectra recorded at 2.8 K. The spectra are fitted with the Blonder-Tinkham-Klapwizk (BTK) theory,¹⁵ modified to take into account the effect of spin polarization in the ferromagnet.¹⁶ All the spectra were fitted using three fitting parameters: The superconducting energy gap Δ , the transport spin polarization P_t , and the barrier parameter Z , which characterizes the potential barrier at the interface. The extracted value of P_t shows a decrease with increasing value of Z [Fig. 4], a feature which has been observed for a wide variety of ferromagnets. This decrease is believed to arise from the spin depolarization at the magnetically disordered scattering barrier formed at the interface.^{17,18} The intrinsic value of P_t is therefore extracted by linearly extrapolating the P_t vs Z curve to $Z=0$ (Fig. 4). The intrinsic value of the transport spin polarization extracted in this way is $P_t \sim 66\%$.

It is important to note that unlike spin polarized photoemission which measures the spin polarization in the density of states (DOS), i.e., $P = (N_{\uparrow} - N_{\downarrow}) / (N_{\uparrow} + N_{\downarrow})$, PCAR measures the spin polarization in the transport current. This quantity varies depending on whether the contact is in the ballistic regime where the contact diameter (d) is smaller than both the elastic (l_e) and the inelastic (l_i) mean free paths, or the diffusive regime where $l_i < d < l_e$. The transport spin polarization is given by $P_t = (\langle N_{\uparrow} v_{F\uparrow} \rangle_{FS} - \langle N_{\downarrow} v_{F\downarrow} \rangle_{FS}) / (\langle N_{\uparrow} v_{F\uparrow} \rangle_{FS} + \langle N_{\downarrow} v_{F\downarrow} \rangle_{FS})$ for a ballistic contact and $P_t = (\langle N_{\uparrow} v_{F\uparrow}^2 \rangle_{FS} - \langle N_{\downarrow} v_{F\downarrow}^2 \rangle_{FS}) / (\langle N_{\uparrow} v_{F\uparrow}^2 \rangle_{FS} + \langle N_{\downarrow} v_{F\downarrow}^2 \rangle_{FS})$ for a diffusive contact,

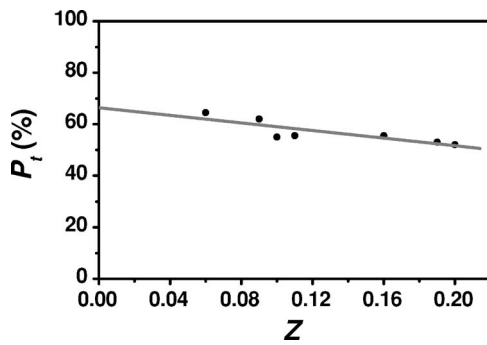


FIG. 4. Variation of the extracted value of P_t as a function of barrier height parameter Z . The solid line shows the linear extrapolation giving an intrinsic transport spin polarization $P_t \approx 66.5\%$.

where N_{\uparrow} and N_{\downarrow} are the DOS of the up and down spin channels at Fermi level and $v_{F\uparrow}$ and $v_{F\downarrow}$ are the respective Fermi velocities and the average is taken over the entire Fermi surface.¹⁹ The latter characterizes the spin polarization of a bulk current in the ferromagnet as well as the current through a low transparency barrier. Though, at present, we do not have an estimate of l_i and l_e in this material, from the relatively large residual resistivity ($240 \mu\Omega \text{ cm}$) it is likely that the contact is in the diffusive limit. The large value of transport spin polarization in this material seems to be in apparent disagreement with electronic structure calculations¹³ which predict a much smaller value for the transport spin polarization both in the diffusive and ballistic limit. At present we do not know the origin of this discrepancy which requires further experimental and theoretical studies.

In summary, we have synthesized a soft ferromagnet with very low hysteresis exhibiting a large moment and a high value of transport spin polarization by doping the Pauli paramagnet CeNi_5 with manganese. This material could be potentially important in devices where switching of the spin at low magnetic field is desired. We believe that further experimental and theoretical studies will shed more light on the magnetic and electronic properties of this interesting compound.

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