Study of spin fluctuations in $Ni_{3\pm x}Al_{1\mp x}$ using point contact Andreev reflection spectroscopy

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We report point contact Andreev reflection (PCAR) spectroscopy studies on $Ni_{3\pm x}Al_{1\mp x}$ with composition range spanning the ferromagnetic to paramagnetic phase boundary. PCAR studies performed using Nb tip as counterelectrode reveal that the superconducting quasiparticle lifetime (τ) and superconducting energy gap (Δ) decrease with increasing spin fluctuation in the normal metal electrode. Our study reveals that PCAR could be a useful probe to study spin fluctuations in systems that are on the verge of magnetic instability.
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Point contact spectroscopy[1] has been used for many years to investigate the static and dynamic spin dependent properties of magnetic systems at the Fermi level (E_F), viz., electron-magnon interaction,[2] Kondo scattering,[3] etc. In recent years, a variant of this technique, namely, point contact Andreev reflection (PCAR), has emerged as a popular technique to measure the spin polarization at E_F in itinerant ferromagnets.[4] In this technique a ballistic contact is established between a sharp superconducting tip and a normal metal electrode. In such a contact the electrical transport is governed by the process of Andreev reflection where an electron incident from the normal metal electrode on the normal-metal/superconductor interface is reflected as a hole in the opposite spin band and a Cooper pair propagates in the superconductor. This causes a doubling of the differential conductance $[G(V)= dI/dV|_V]$ of the junction for bias voltages lower than the superconducting energy gap (Δ). In a ferromagnet, the Andreev reflection process and the resulting increase in $G(V)$ are suppressed due to the unequal density of states of up and down spins. The analysis of PCAR conductance spectra thus provides a method to measure transport spin polarization[5] (P_t) at E_F. Recently[6] we have shown that in addition to transport spin polarization, PCAR can also be utilized to detect spin fluctuations in systems that are close to a magnetic instability. The proximity effect between a spin fluctuating magnetic electrode and the superconductor causes a decrease in the superconducting quasiparticle lifetime (τ), thereby broadening[7,8] the quasiparticle density of states. The broadened density of states is given[9] by $N(E)=\mathrm{Re}\{(E+i\Gamma)/[\sqrt{(E+i\Gamma)^2-\Delta^z}]\}$, where the broadening parameter (Γ) is given by $\Gamma=h/\tau$. From the magnitude of Γ, we get an estimate of the degree of spin fluctuation present in the system.[6]

In this paper we report PCAR spectroscopy on the itinerant magnetic system $Ni_{3\pm x}Al_{1\mp x}$, which has been widely studied[10–12] due to its unusual compositional phase boundary around 26 at. % Al. The parent alloys Ni_3Al and its derivatives, crystallize with a simple cubic (of the form Cu_3Au) structure in the composition range 23–27.5 at. % Al. Between 23–26 at. % Al, $Ni_{3\pm x}Al_{1\mp x}$ exhibits ferromagnetism with moderate Curie temperature (T_C) in the range $\sim$40–80 K; for Al concentrations >26 at. %, it becomes a paramagnet with large spin fluctuation. In this paper, we study the effect of spin fluctuation on the PCAR spectra of $Ni_{3\pm x}Al_{1\mp x}$, in three samples with compositions $Ni_{76}Al_{24}$, $Ni_{74}Al_{26}$, and $Ni_{73}Al_{27}$, which span the ferromagnet to paramagnet compositional phase boundary.

Polycrystalline samples of $Ni_{3\pm x}Al_{1\mp x}$ were grown through arc melting. Details of $Ni_{3\pm x}Al_{1\mp x}$ sample preparation, chemical characterization, and compositional analysis have been described elsewhere.[10] To characterize the sample, magnetization measurements are carried out using a vibrating sample magnetometer in the temperature range 2–300 K. Magnetoresistance (MR) measurements are carried out up to a field of 12 T using a Quantum Design physical property measurement system. Point contact measurements are performed with a superconducting Nb tip in a liquid ^{4}He continuous flow cryostat by standard four-probe modulation technique using a lock-in amplifier.

Figures 1(a)–1(c) show the temperature dependence of magnetic moment $[M(T)]$ and inverse susceptibility $(1/\chi = H/M)$ down to 2 K. $Ni_{76}Al_{24}$ shows a clear $T_C \sim 72$ K

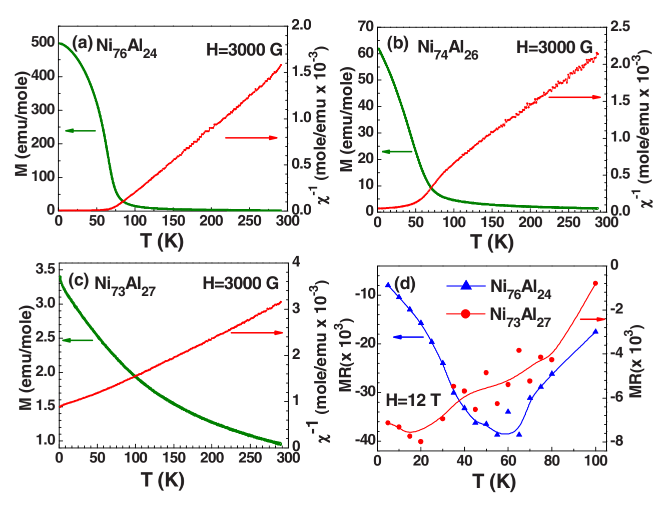

FIG. 1. (Color online) (a)–(c) shows the temperature (T) variation of magnetization (M) and inverse susceptibility (χ^{-1}) for $Ni_{76}Al_{24}$, $Ni_{74}Al_{26}$, and $Ni_{73}Al_{27}$, respectively. (d) MR as a function of temperature (T) for $Ni_{76}Al_{24}$ and $Ni_{73}Al_{27}$, respectively. Solid lines are guide to the eyes.

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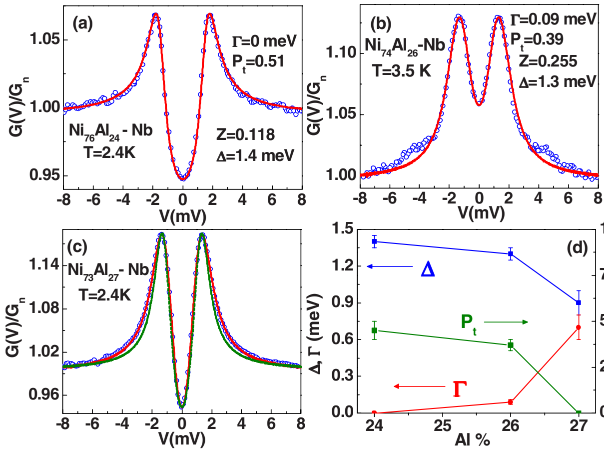

FIG. 2. (Color online) [(a) and (b)] PCAR [$G(V)/G_n$ vs V] spectra for $Ni_{76}Al_{24}$ and $Ni_{74}Al_{26}$ with mBTK fits (solid line). (c) PCAR spectra for $Ni_{73}Al_{27}$. Light gray (red) line is the mBTK fit to data with best fit parameters $P_t=0$, $\Gamma=0.44$ meV, $\Delta=1.18$ meV, and $Z=0.72$. Dark gray (green) line is simulated spectra for the same with $\Gamma=0$, $\Delta=1.28$ meV, $Z=0.32$, and $P_t=0.39$. G_n corresponds to the conductance value at 8 meV. (d) Variation in Δ, Γ, and P_t with Al concentration.

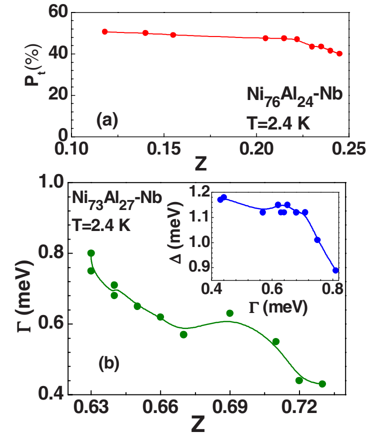

FIG. 3. (Color online) (a) Variation in P_t with Z at 2.4 K for different $Ni_{76}Al_{24}$–Nb contacts. (b) Variation in the extracted value of Γ with Z at 2.4 K for different $Ni_{73}Al_{27}$–Nb contacts. Inset shows variation in Δ with Γ. Solid lines are guide to eyes.

(determined from the maximum in the double derivative of the M-T curve). $Ni_{74}Al_{26}$ also shows $T_C\sim 60$ K, but the moment is considerably smaller than that of $Ni_{76}Al_{24}$ and the magnetization does not saturate down to the lowest temperature indicating the presence of spin fluctuations even below T_C. $Ni_{73}Al_{27}$ does not exhibit any ordering down to the lowest temperature. However, unlike conventional Pauli paramagnets χ is temperature dependent and shows a Curie–Weiss-like behavior, showing the presence of large spin fluctuation in the paramagnetic state. The presence of spin fluctuations is further confirmed from the temperature variation of MR $(\mathrm{MR}=\Delta\rho/\rho=[\rho(H,T)-\rho(0,T)]/\rho(0,T))$ at $H=12$ T for the two extreme compositions [Fig. 1(d)]. The MR is negative over the entire temperature range for both samples. For $Ni_{76}Al_{24}$, the absolute value of the MR (|MR|) is low for $T<T_C$ and shows a maximum close to the T_C due to critical spin fluctuations. On the other hand, in $Ni_{73}Al_{27}$, the |MR| keeps increasing with decreasing temperature showing the presence of strong spin fluctuations down to the lowest temperature.

Figures 2(a)–2(c) show representative PCAR spectra for $Ni_{76}Al_{24}$, $Ni_{74}Al_{26}$, and $Ni_{73}Al_{27}$, respectively. These spectra are analyzed within the modified Blonder–Tinkham–Klapwijk (mBTK) formalism[13] using Δ, P_t, Γ, and an effective barrier potential Z as fitting parameters. The barrier potential takes into account[14] the potential barrier at the interface arising from both possible oxide layer as well as the mismatch between Fermi velocities of the normal metal and superconducting electrode. It is modeled[14] as a delta function, $V(x)=V_0\delta(x)$ at the interface and parametrized through the dimensionless quantity $Z=V_0/\hbar v_F$, where v_F is the Fermi velocity. The values of P_t, Δ, and Γ extracted from the mBTK fits are shown as a function of at. % Al in Fig. 2(d). With increasing Al, the spin polarization decreases and as expected is zero for the paramagnetic compound $Ni_{73}Al_{27}$. The broadening parameter Γ is zero for the ferromagnetic $Ni_{76}Al_{24}$ sample and increases gradually with Al content. Δ on the other hand decreases with increasing Al content as one goes from the ferromagnetic to the spin fluctuating regime. This is consistent with our earlier results[6] and confirms that the proximity of a superconductor to large spin fluctuations drastically decreases the superconducting τ and Δ. To cross check the uniqueness of the fit and the influence of individual parameters on it, the PCAR data for $Ni_{73}Al_{27}$-Nb contact are also fitted assuming $\Gamma=0$ and a finite P_t [dashed line in Fig. 2(c)]. The fit is considerably poorer particularly at voltage values above the superconducting energy gap.

To further explore the effect of spin fluctuations on Γ, we have done detailed PCAR studies on the two extreme compositions, viz., the ferromagnetic $Ni_{76}Al_{24}$ and the spin fluctuating $Ni_{73}Al_{27}$. Different PCAR spectra are recorded by engaging the Nb tip several times at different places. These spectra correspond to the same sample tip combinations but have statistically different values of Z. Figure 3(a) shows the variation in P_t with the barrier potential Z for the ferromagnetic $Ni_{76}Al_{24}$. As has been shown before,[15] the value of P_t decreases with increasing Z due to the presence of magnetic dead layer in the F-S interface. Similar studies on $Ni_{73}Al_{27}$ reveal a systematic variation between Z and Γ. With increasing Z, Γ decreases [Fig. 3(b)]. This is expected since a larger barrier parameter at the interface implies that the two electrodes are less strongly coupled to each other. Therefore, the influence of spin fluctuation would be less on the superconducting electrode. Consequently, we observe an inverse correlation between Δ and Γ extracted for contact with different Z [inset of Fig. 3(b)]. This observation provides a valuable consistency check of intrinsic nature of proximity effect between a superconductor and a spin fluctuating metal.

In summary, we have investigated the spin fluctuations in $Ni_{3\pm x}Al_{1\mp x}$ where the ground state evolves from a ferromagnet to a spin fluctuating paramagnet with increase in Al. The central observation of this paper is that while a static moment in the normal metal electrode has negligible effect on Γ and Δ extracted from PCAR spectra, spin fluctuations decrease both these quantities. This study shows that PCAR can be a valuable tool to explore spin fluctuations in itinerant systems and their effect on superconductivity.

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