Temperature dependence of transport spin polarization in $NdNi_5$ from point-contact Andreev reflection

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We report a study in which point-contact Andreev reflection (PCAR) spectroscopy using a superconducting Nb tip has been carried out on $NdNi_5$, a ferromagnet with a Curie temperature of $T_C \sim 7.7$ K. The measurements were performed over a temperature range of 2–9 K, which spans across the ferromagnetic transition temperature. From an analysis of the spectra, we show that (i) the temperature dependence of the extracted value of transport spin polarization closely follows the temperature dependence of the spontaneous magnetization; (ii) the superconducting quasiparticle lifetime shows a large decrease close to the Curie temperature of the ferromagnet. We attribute the latter to the presence of strong ferromagnetic spin fluctuations in the ferromagnet close to the ferromagnetic transition temperature.

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INTRODUCTION

With the emergence of "spintronics"[1] the ability to manipulate electronic spin as well as the charge in a solid has gained particular significance. Central to the progress of this field is the identification of ferromagnetic materials with a large degree of spin polarization, which can be used as a spin source in electronic devices, which will use both the charge and spin of the electrons. In this context, the experimental determination of the spin polarization (defined as $P=[N_\uparrow(E_F)-N_\downarrow(E_F)]/[N_\uparrow(E_F)+N_\downarrow(E_F)]$, where $N_\uparrow(E_F)$ and $N_\downarrow(E_F)$ are the spin-up and spin-down density of states at Fermi level) in ferromagnets has gained particular significance. Conventionally, two techniques have been widely used for the determination of spin polarization in ferromagnetic materials. The first one, pioneered by Meservey and Tedrow,[2] involves spin-dependent tunneling from a ferromagnet to a superconductor when the superconducting density of states is Zeeman split by the application of a magnetic field. This technique involves the tedious process of fabricating a tunnel junction and the need to apply a large perturbing magnetic field of several tesla. The second one, spin-resolved photoemission,[3] relies on the measurement of the spin of electrons that emerge from the surface of a ferromagnet close to the Fermi level. However, since the electrons measured in photoemission experiments emerge from a depth of a few angstroms to a few tens of angstroms from the surface, this technique is extremely surface sensitive. More recently, a third technique, namely, point-contact Andreev reflection (PCAR) spectroscopy[4] has emerged as arguably one of the most popular probes[5–12] for the measurement of transport spin polarization in ferromagnetic materials. This technique relies on the fact that the process of Andreev reflection,[13] through which an electron incident on a normal metal/superconductor (N/S) interface with energy less than the superconducting energy gap (Δ) gets reflected back as a hole in the opposite spin band of the metal, is strongly suppressed when the normal metal electrode is a ferromagnet. The transport spin polarization is thus determined from the analysis of the conductance $[G(V)]$ versus voltage (V) characteristics of a ballistic or diffusive point contact established between a superconducting tip and the ferromagnet. This technique has several advantages: It is simple to implement, does not require fabrication of tunnel junctions, and can be used for a wide variety of materials; it also does not require the application of a magnetic field.

One obvious limitation of the PCAR technique is that the temperature range of measurement is limited by the superconducting transition temperature of the superconducting tip. The determination of spin polarization of ferromagnets is thus typically carried out at temperatures that are two orders of magnitude lower than the ferromagnetic transition temperature (T_C) of most ferromagnets. Since much of the practical interest is in the value of spin polarization close to room temperature, the bulk spin polarization at elevated temperature is often estimated based on the assumption $P(T) \propto M_s(T)$, namely, that the spin polarization of the electrons close to the Fermi level is proportional to the spontaneous magnetization $[M_s(T)]$ of the ferromagnet.[14–17] Though the spin polarization as a function of temperature has been measured using spin-polarized photoemission for a few ferromagnets,[18,19] this simple relation has so far remained experimentally unverified even for relatively simple ferromagnets.[20] The experimental verification of this relation is important for two reasons. First, M_s is a bulk property that depends on the total number difference of the up- and down-spin electrons, whereas P is only sensitive to the electrons close to Fermi level; this intuitive relation is thus based on a simplistic picture, which is strictly valid only for free-electronlike parabolic bands. Second, both PCAR and the Meservey-Tedrow technique measure the spin polarization in the transport current[21] (P_t) rather than the spin polarization in the density of states, which depends on a weighted average of the density of states and Fermi velocity of the two spin bands. The transport spin polarization is given by[21] $P_t = [\langle N_\uparrow(k)(v_{k\uparrow})^n\rangle_{FS} - \langle N_\downarrow(k)(v_{k\downarrow})^n\rangle_{FS}] / [\langle N_\uparrow(k)(v_{k\uparrow})^n\rangle_{FS} + \langle N_\downarrow(k) \times (v_{k\downarrow})^n\rangle_{FS}]$, where $n=1$ for a ballistic point contact, and $n=2$ for diffusive point contact. It has been shown that the spin polarization measured from the Meservey-Tedrow technique is identical to the diffusive point contact. It is not obvious that the temperature dependence of the spin polarization extracted from these techniques should follow M_s. However, since this relation forms the basis of much of the experimental studies related to spin transport in a variety of spintronic materials, it is important to investigate if it holds good in a typical ferromagnet.

In this paper, we report the PCAR studies carried out on $NdNi_5$, a ferromagnet[22–24] with a $T_C \sim 7.7$ K. Like most RNi_5 (R=rare earth) compounds, the moment primarily resides on the Nd sites inducing a small moment on the Ni sites. The low T_C of this material enables us to extract the transport spin polarization (P_t) all the way up to T_C by carrying out the measurements using a finely cut superconducting Nb tip (superconducting transition temperature, T_c=9.2 K). The reproducibility of our results was confirmed by carrying out measurements on two different samples on $NdNi_5$. In this paper we will primarily concentrate on the measurements carried out on one of these samples.[25] The central result of this paper is that the temperature dependence of P_t in $NdNi_5$ closely follows the temperature dependence of M_s, validating the relation $P_t(T) \propto M_s(T)$. In addition, we observe a decrease of the superconducting quasiparticle lifetime close to the ferromagnetic transition temperature. We attribute this effect to the large spin fluctuation in the $NdNi_5$ close to the critical temperature. This hypothesis is further supported by PCAR measurements on two other classes of materials: (i) The ferromagnetic metal Fe for which the T_C is much higher than the temperature range of measurements and therefore the effect of spin fluctuations are unimportant; and (ii) the isostructural filled skutterudites KFe_4Sb_{12}, $YbFe_4Sb_{12}$, and $CaFe_4Sb_{12}$. Of these three compounds KFe_4Sb_{12} is a ferromagnet[26] with $T_c \sim 85$ K, whereas $YbFe_4Sb_{12}$ and $CaFe_4Sb_{12}$ are nonferromagnetic metals,[27] where detailed magnetic studies show the evidence of large ferromagnetic spin fluctuations at low temperatures.

EXPERIMENTAL DETAILS

A polycrystalline sample $NdNi_5$ was prepared by repeated arc melting of the stoichiometric amounts of the constituent elements on water-cooled copper hearth in a purified argon atmosphere. The button was flipped and remelted several times to ensure the homogeneity. A titanium button was used as an oxygen getter. The total weight loss during the arc melting was less than 0.5% and hence the alloy compositions were assumed to remain unchanged from the original stoichiometric ratios. A room-temperature powder x-ray-diffraction pattern of the sample was obtained using a panalytical x-ray diffractometer equipped with Cu $K\alpha$ radiation. In order to obtain the lattice parameters of the compound and confirm its homogeneity to the accuracy of the x-ray pattern, a Rietveld refinement using the FULLPROF program of the obtained x-ray-diffraction (XRD) pattern was done. The resistivity (ρ) and magnetization (M) of the sample was measured in the temperature range 3–300 K using a homemade resistivity setup and Quantum Design SQUID magnetometer, respectively. PCAR measurements were performed in the temperature range 2.4 to 9 K in a continuous-flow He4 cryostat. The sample was polished to a mirror finish and loaded

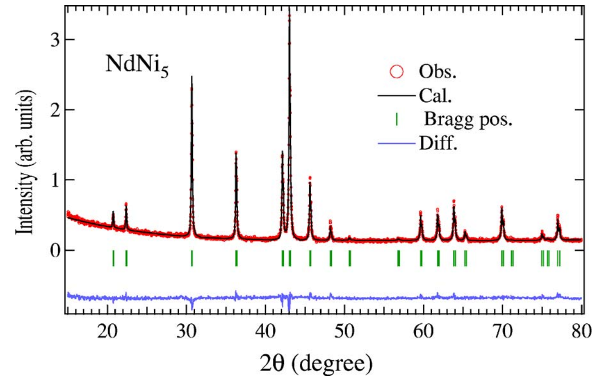

FIG. 1. (Color online) The observed and calculated x-ray-diffraction pattern of $NdNi_5$ along with their difference. The vertical bars indicate Bragg reflections.

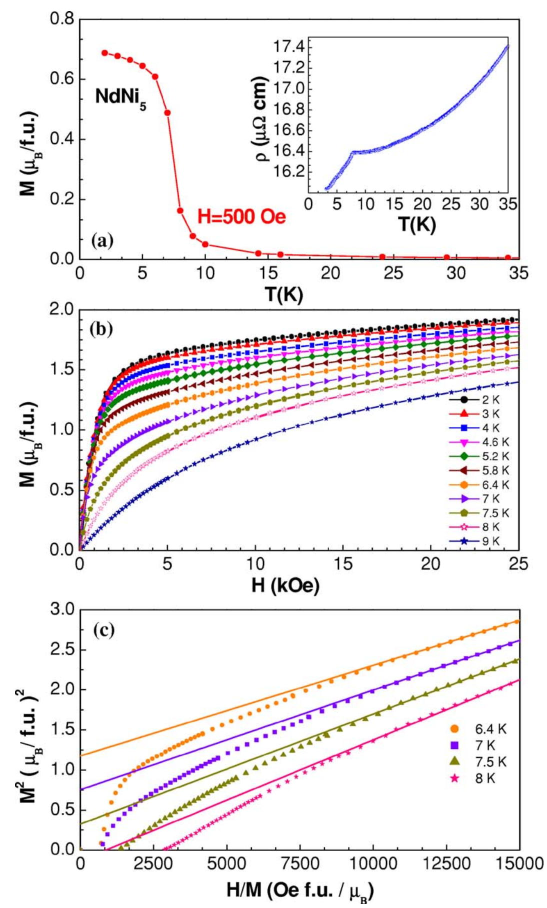

FIG. 2. (Color online) (a) Magnetization vs temperature of $NdNi_5$ measured in 500 Oe. The inset shows the resistivity as a function of temperature. (b) Isothermal M-H curves at various temperatures. (c) Arott plots (M^2 vs H/M) at four temperatures close to T_C.

immediately for experiment to avoid surface degradation. A mechanically cut sharp Nb tip was brought in contact with the sample at low temperatures using a differential screw arrangement and the conductance versus voltage characteristics of the contact was measured using a four-probe current-modulation technique. Typical contact resistance in these measurements ranged between 10 and 20 Ω.

RESULTS AND DISCUSSION

Figure 1 shows the Rietveld refinement of the x-ray pattern of $NdNi_5$, which forms in a $CaCu_5$-type hexagonal structure with a space group $P6/mmm$. The excellent agreement with the experimental diffraction pattern (deduced from the near-zero difference plot) confirms that the material is single phase. The obtained lattice parameters $a=4.953$ Å and $c=3.967$ Å are in agreement with the published reports.[22]

The magnetization versus temperature of the $NdNi_5$ sample [shown in Fig. 2(a)] measured at 500 Oe reveals a sharp ferromagnetic transition with $T_C \sim 7.7$ K. The resistivity [the inset of Figure 2(a)] also shows a pronounced anomaly at the same temperature. Figure 2(b) shows the isothermal M-H curves recorded at various temperatures. The M-H curve does not saturate up to 2.5 T due to the large magnetocrystalline anisotropy in this material. At low temperatures, M_s was estimated by linearly extrapolating the high-field slope of the M-H curve. The value of M_s at 2 K ($\sim 1.68 \mu_B$/f.u.) is much lower than the expected saturation moment of $3.28\mu_B$ for the free Nd^{3+} ion. This is due to crystal-field spitting of the $4f$ energy levels in Nd as has been shown in numerous previous studies.[28,29] Above 6.4 K the high-field M-H curve was no longer linear. In this temperature range M_s was estimated from the Arott plots (M^2 vs H/M) shown in Fig. 2(c).

In Fig. 3(a) we show the PCAR $G(V)$-V spectra (normalized with respect to the conductance values at large bias, G_n) for $NdNi_5$ recorded at various temperatures. The normalized conductance spectra are fitted with a modified Blonder-Tinkham-Klapwizk[30] (BTK) theory, which takes into account the spin polarization of the ferromagnet.[31] Within this model the current through the point contact consists of a fully polarized (I_p) and an unpolarized (I_u) component such that the total current in terms of the transport spin polarization is given by $I=P_tI_p+(1-P_t)I_u$. The unpolarized component of the current undergoes Andreev reflection in the same way as in an interface between a nonmagnetic metal and a superconductor. For the polarized component, on the other hand, the Andreev reflected hole cannot propagate and decays as an evanescent wave close to the N/S interface. I_u and I_p are thus calculated by using the BTK expression for the current,

$$I_{u,p}(V) \propto \int_{-\infty}^{\infty} [f(E-eV) - f(E)][1 + A_{u,p}(E) - B_{u,p}(E)]dE, \tag{1}$$

where $f(E)$ is the Fermi function and $A_u(E)$ [$B_u(E)$] and $A_p(E)$ [$B_p(E)$] are the Andreev reflection and normal reflection coefficients, calculated by solving the Bogoliubov–de

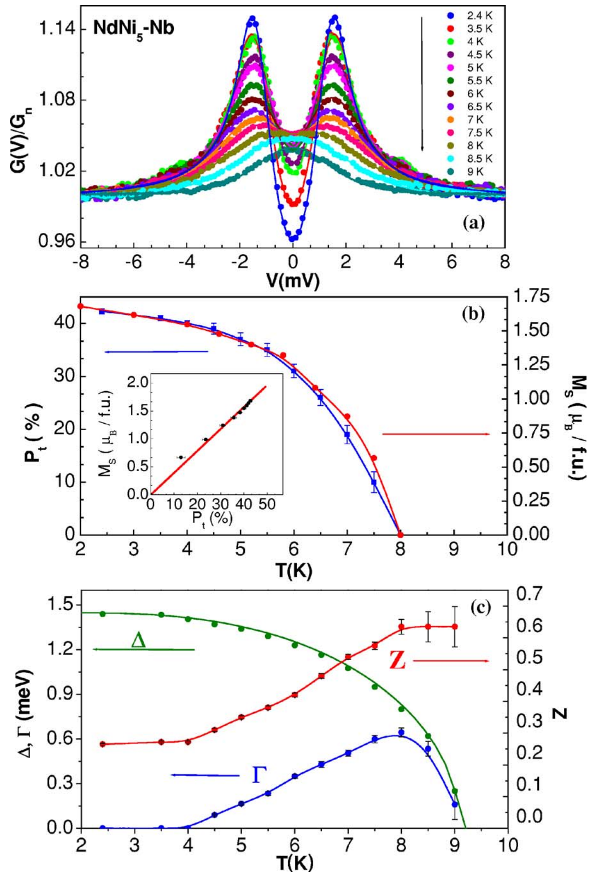

FIG. 3. (Color online) (a) Normalized PCAR spectra of $NdNi_5$-Nb point contact at different temperatures. The solid lines show the fits to the spectra using the modified BTK model. (b) Temperature variation of the P_t (squares) and M_s (circles). The solid lines are a guide to the eye. The inset shows the linear variation of M_s with P_t. (c) Temperature dependence of the superconducting energy gap (Δ), the barrier parameter (Z), and the broadening parameter (Γ). The solid line passing through Δ shows the expected temperature variation from BCS theory. The solid lines passing through Z and Γ are a guide to the eye.

Gennes (BdG) equations for a nonmagnetic metal/superconductor and a fully polarized ferromagnet/superconductor, respectively. To simulate a realistic interface, a delta-function potential of the form $V_0\delta(x)$ is assumed at the interface. This delta-function potential, parametrized within the model as a dimensionless parameter, $Z=V_0/\hbar v_F$, takes into account multiple effects: First, it takes into account the effect of any oxide barrier that may be present at the interface; second, Z also accounts for an effective barrier arising from the Fermi velocity mismatch between the normal metal and the superconductor. The lifetime (τ) of the superconducting quasiparticle is incorporated in this model by including a broadening parameter[32] $\Gamma(=\hbar/\tau)$, while solving the BdG equations. We have thus four fitting parameters: P_t, Δ, Γ, and Z. In order to reduce the number of free parameters, we restrict Δ to within 5% of its BCS value for Nb at all temperatures. The resulting fit of the spectra at various temperatures is shown through the solid lines in Fig. 3(a).

Figure 3(b) shows the extracted values of P_t of $NdNi_5$ as a function of temperature. While, in the absence of a detailed estimate of the elastic and inelastic mean-free paths, we cannot ascertain whether the contacts are in the ballistic or diffusive limit, the latter is more likely since our sample has a relatively small residual resistivity ratio; i.e., $\rho(300\ \text{K})/\rho(3\ \text{K})=3.59$. In the same graph we also show the temperature variation of M_s. It can be easily seen that the temperature variation of the two quantities is similar. To further illustrate this point, in the inset we plot P_t as a function of M_s. Barring temperatures very close to T_C, where we see a small deviation, the points fall on a straight line with zero intercept showing that the transport spin polarization $P_t(T) \propto M_s(T)$. We would, however, like to note that this does not necessarily imply that the spin polarization at Fermi level $P(T) \propto M_s(T)$, unless the Fermi velocities of the up-spin band and the down-spin bands are almost equal.

We now focus our attention on the temperature dependence of the other quantities, namely, Δ, Γ, and Z. The temperature variation of these three quantities extracted from the fits in Fig. 3(a) is shown in Fig. 3(c). We would like to note that all the spectra could be fitted very well with the constraint on Δ stated earlier, with $\Delta(T=0)=1.45$ meV. For temperatures in the range 2.4–4 K the spectra can be fitted without incorporating any broadening parameter ($\Gamma=0$). Above 4 K, Γ gradually increases and reaches a maximum value of $\Gamma=0.65$ meV at 8 K, which coincides with the T_C of $NdNi_5$. Above 8 K, Γ decreases reaching a value of $\Gamma=0.16$ meV at 9 K. The barrier parameter Z, on the other hand, remains constant in the range 2.4–4 K with a value of $Z=0.27$. Above 4 K, Z increases monotonically up to 8 K and tends to saturate to a value of $Z=0.635$.

Before discussing the implications of these results, we would first like to comment on the reliability of the fits of the PCAR spectra, particularly at elevated temperatures. With increasing temperature, PCAR spectra get gradually thermally smeared. At temperatures greater than 8 K the most dominant feature of the spectra, namely, the two peaks in the conductance spectra associated with the superconducting energy gap, gets smeared into one broad peak. Since the saturation in the value of Z happens in this temperature range, the fit of the spectra for $T>8$ K needs careful attention. To cross-check the reliability of our fits in Fig. 4, we show two fits of the same spectra taken at 8.5 K: One fit with the parameters shown in Figs. 3(b) and 3(c) (solid line) and the second one (dashed line) where Z is deliberately reduced and Γ is adjusted to obtain the best possible fit. Though the parameters can be adjusted to reproduce the peak value in the normalized $G(V)$ vs V curves in both cases, the latter does not reproduce the width of the curve close to zero bias (the inset of Fig. 4). Nevertheless, above 7.5 K the uncertainty in the value of Z and Γ significantly increases as shown in Fig. 3(c).

We now come to the significance of the temperature variation of Γ and Z. First we focus on the temperature dependence of Γ. The increase in Γ, which peaks close to the critical point of the ferromagnet, signifies a corresponding decrease in quasiparticle lifetime at the same temperature. It is known that ferromagnetic spin fluctuation in an s-wave superconductor increases the singlet-state repulsion.[33] Since ferromagnetic spin fluctuation in $NdNi_5$ is maximum at temperatures close to T_C, it would be natural to attribute the decrease in the superconducting quasiparticle lifetime to the proximity of the superconductor to strong ferromagnetic spin fluctuation. In addition, we would like to note that at temperatures close to the superconducting transition temperature, additional broadening is likely to arise from the intrinsic decrease in the quasiparticle lifetime of the superconductor. Therefore, for temperatures $T \geq 8$ K, Γ could also have a contribution from intrinsic origin. While this effect might change the temperature variation of the Γ arising from the ferromagnetic spin fluctuation to a small extent, the basic feature would remain unchanged. We would also like to note that a self-consistent solution of this problem should, however, also take into account the corresponding decrease in Δ. At present we do not have a self-consistent model to incorporate this effect into our analysis. The temperature variation of Z, on the other hand, is more complex to understand. It has been pointed out by several authors that in the analysis of a ferromagnet/superconductor interface, Z implicitly incorporates much more physics than a simple potential barrier at the interface. For a nonmagnetic metal/superconductor interface, Z is given by[34] $Z=Z_b+[(r-1)^2]/4r=Z_b+Z_i$, where r is the ratio of the Fermi velocity in the normal metal and the superconductor. The first term (Z_b) arises from a physical barrier arising from imperfect interface and oxide barrier and the second term (Z_i) incorporates the effect of Fermi velocity mismatch between the normal metal and the superconductor. In the case where the normal metal is a ferromagnet with different Fermi velocity of the up- and down-spin bands, the derivation of the second term is not straightforward. In this case it is expected that Z_i would be governed by a weighted average of two contributions: (i) The mismatch between the spin-up-band Fermi velocity and the Fermi velocity in the superconductor and (ii) the mismatch between the spin-down-band Fermi velocity and the Fermi velocity in the superconductor. The dominant contribution will, however, come from the mismatch between the Fermi velocity of the majority spin subband and the superconductor. At temperature $T \ll T_c$, where the exchange splitting is roughly temperature independent, Z_i will be constant as a function of temperature. Again, at tempera-

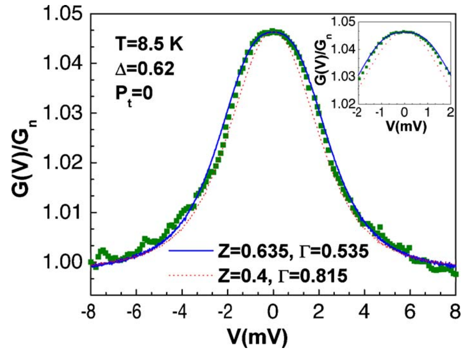

FIG. 4. (Color online) Comparison of the fits of PCAR spectra for Nb-$NdNi_5$ contact at 8.5 K with two sets of fitting parameters shown in solid and dashed lines. The fit parameters for the two fits are shown in the figure. Δ and Γ are in meV. The inset is an enlarged view for the low-bias region.

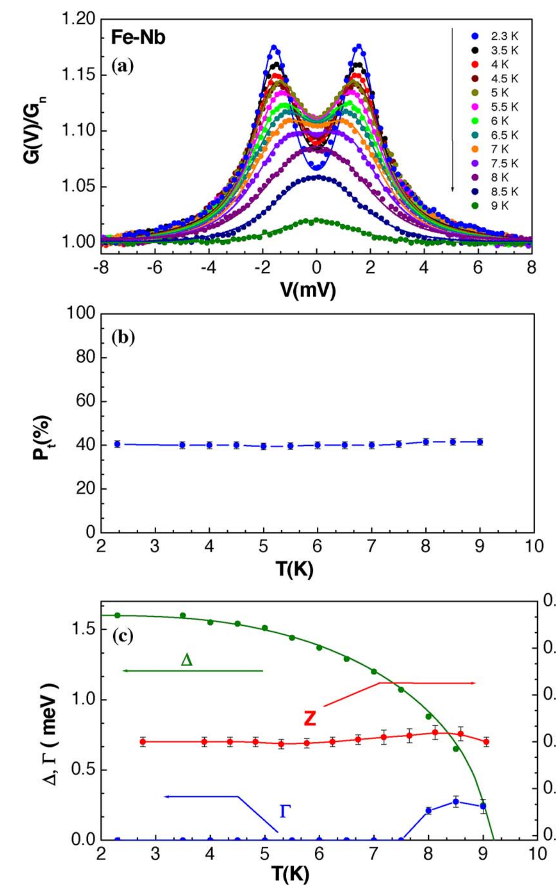

FIG. 5. (Color online) (a) Normalized PCAR spectra of Fe-Nb point contact at different temperatures. The solid lines show the fits to the spectra using the modified BTK model. (b) Temperature variation of the transport spin polarization (P_t) in the temperature range 2.3–9 K. The solid line is a guide to the eye. (c) Temperature dependence of the superconducting energy gap (Δ), the barrier parameter (Z), and the broadening parameter (Γ). The solid line passing through Δ shows the expected temperature variation from BCS theory. The solid lines passing through Z and Γ are guides to the eye.

tures $T \geqslant T_c$ the exchange splitting disappears and Z_i is solely determined by the Fermi velocity in the paramagnetic state. At intermediate temperatures the Z will either increase or decrease depending on the relative values of the mismatch between the Fermi velocities of the majority spin subband and the superconductor in the ferromagnetic state, and the mismatch between the Fermi velocities in the paramagnetic state and the superconductor. As the temperature of the ferromagnet is raised towards the ferromagnetic transition, the Fermi velocities of the up- ($v_F\uparrow$) and down- ($v_F\downarrow$) spin bands gradually change due to the reduction in exchange splitting and eventually become equal at T_C. The gradual change in Z from 4 to 8 K and the leveling off to a constant value above 8 K suggests that this evolution in $v_F\uparrow$ and $v_F\downarrow$ is reflected in the temperature dependence of Z. We would like to note that the temperature dependence of Z was similar in both the samples that we studied.[25] It is also expected that the ferromagnetic spin fluctuations will have additional effects on Z. This issue is currently beyond the scope of our paper and needs to be explored theoretically.

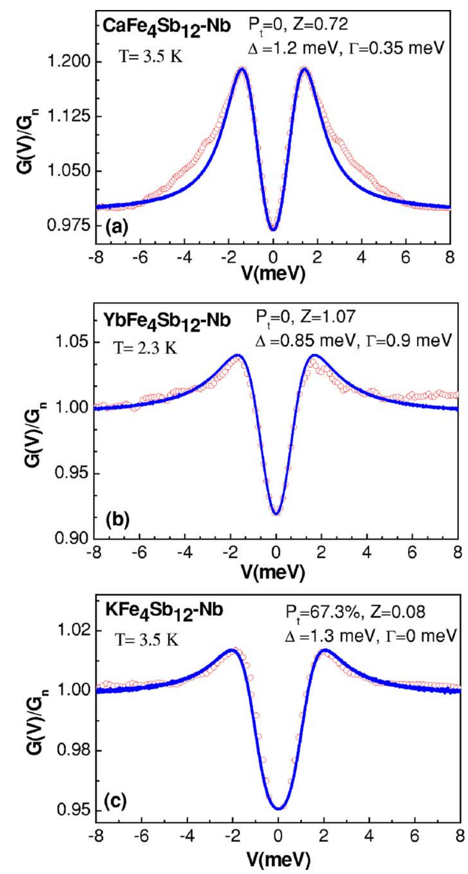

FIG. 6. (Color online) Normalized PCAR spectra for (a) $CaFe_4Sb_{12}$-Nb point contact at 3.5 K; (b) $YbFe_4Sb_{12}$-Nb point contact at 2.3 K; (c) KFe_4Sb_{12}-Nb point contact at 3.5 K. The solid lines are the best fits of the spectra to the modified BTK model. The best-fit parameters are also shown in the figures.

In order to cross-check the conjecture that spin fluctuations in the ferromagnet cause the decrease in the superconducting quasiparticle lifetime, we have performed PCAR measurements on two different kinds of systems, which are in two extremes in terms of spin fluctuations. The first measurement was on Fe (using a Nb tip), for which the $T_C \sim 1043$ K is two orders of magnitude larger than the temperature range over which the measurement is carried out. Thus for Fe both ferromagnetic spin fluctuations as well as the decrease in exchange splitting is likely to be insignificant. The second class of systems is the isostructural-filled skuterrudites[27] KFe_4Sb_{12}, $CaFe_4Sb_{12}$, and $YbFe_4Sb_{12}$. The first of these compounds is a ferromagnetic metal with $T_c \sim 80$ K, whereas the latter two are nearly ferromagnetic metals for which large spin fluctuations are expected to be present even at the lowest temperatures. Figures 5(a)–5(c) show the temperature dependence of the PCAR spectra and best-fit parameters for the Fe-Nb point contact. As expected within experimental errors, the transport spin polarization [Fig. 5(b)] of Fe is constant over the entire temperature range of measurement with $P_t \sim 40\%$. Figure 5(c) shows the temperature variation of Z and Γ. Z is constant over the entire temperature range. Γ, on the other hand, remains zero except at temperatures very close to the transition temperature of the superconductor, where it shows a slight increase. This slight increase is in qualitative agreement with the intrinsic decrease of the quasiparticle lifetime of the superconductor as theoretically predicted,[35] and experimentally observed in strong-coupling superconductors.[36,37] Figure 6(a) shows the PCAR spectra on the $CaFe_4Sb_{12}$. In this case the PCAR spectra recorded at 3.5 K can be fitted only by incorporating a finite value of Γ(=0.35 meV). The situation is similar for $YbFe_4Sb_{12}$ [Fig. 6(b)], where the PCAR spectra recorded at 2.3 K can only be fitted incorporating Γ=0.9 meV. On the other hand, the PCAR data[12] on the ferromagnetic compound, KFe_4Sb_{12} [Fig. 6(c)] can be fitted as expected with Γ=0 since the measurements are performed at temperatures much below the ferromagnetic transition temperature.[38,39] Our results are also consistent with earlier reports on $CaFe_4Sb_{12}$ and $YbFe_4Sb_{12}$, where it has been shown from magnetization and heat capacity that spin fluctuations are larger in the latter compound. Concomitant with the large increase in Γ, the superconducting energy gap is reduced from the bulk value for $CaFe_4Sb_{12}$ and $YbFe_4Sb_{12}$: Δ =1.2 meV for the Nb-$CaFe_4Sb_{12}$ contact and Δ=0.85 meV for the Nb-$YbFe_4Sb_{12}$ contact. While it is tempting to attribute this decrease in Δ to the effect of finite lifetime of the superconducting quasiparticle, a similar effect is not seen in Nb-$NdNi_5$ spectra close to the ferromagnetic transition temperature. We believe that this issue will have to be addressed further through experimental and theoretical studies.

CONCLUSIONS

In summary, we have shown that in $NdNi_5$, the temperature dependence of the transport spin polarization extracted from PCAR spectroscopy closely follows the temperature variation of the spontaneous magnetization M_s. We have also shown that the superconducting quasiparticle lifetime extracted by fitting the PCAR spectra shows a minimum close to the ferromagnetic transition temperature of $NdNi_5$. Through a detailed comparison with measurements carried out on the ferromagnet Fe and the nearly ferromagnetic compounds $CaFe_4Sb_{12}$ and $YbFe_4Sb_{12}$, we attribute this decrease in the quasiparticle lifetime to the effect of large spin fluctuations close to the critical temperature of the ferromagnet. We believe that a detailed theoretical understanding of the role of spin fluctuations could establish PCAR as an alternative technique to probe to investigate ferromagnetic spin fluctuations in nearly ferromagnetic metals.

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[37] A slight anomaly is observed in the fact that the Γ at 9 K is marginally smaller than the Γ at 8.5 K. However, since it is difficult to obtain an accurate value of Γ from PCAR at temperatures very close to the superconducting transition temperature, we would not like to ascribe particular significance to this decrease.

[38] While a large value of Γ is sometimes observed due to grain-boundary scattering in polycrystalline samples, it is difficult to attribute the Γ observed in $CaFe_4Sb_{12}$ and $YbFe_4Sb_{12}$ to this effect for two reasons: (i) All the three samples consist of large polycrystalline grains varying between 1–5 μm (Ref. 39), which is much larger than point contact formed with the tip; and (ii) KFe_4Sb_{12}, $CaFe_4Sb_{12}$, and $YbFe_4Sb_{12}$ have very similar microstructures and electrical resistivity, and therefore grain-boundary scattering is unlikely to be significantly different in the three compounds.

[39] For the microstructures of the KFe_4Sb_{12}, $CaFe_4Sb_{12}$, and $YbFe_4Sb_{12}$, see Fig. 2 in the EPAPS (Ref. 25).