PHYSICAL REVIEW B **85**, 094406 (2012)

Antiferromagnetism in the spin-gap system NaV_2O_5: Muon spin rotation measurements

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(Received 3 November 2011; revised manuscript received 18 January 2012; published 5 March 2012)

Muon spin rotation measurements have been carried out in a stoichiometric spin ladder compound NaV_2O_5 in the temperature range from 2 to 300 K, through the spin-gap transition at $T_c = 35$ K, in transverse magnetic fields from 0.3 to 7 T. Antiferromagnetic order with a local magnetic field at a muon site of about 0.17 T is detected coexisting with the spin-gap state below 15 K. Above 20 K, the signature of a spin-polaron state is observed, which persists to about 100 K.

DOI: 10.1103/PhysRevB.85.094406 PACS number(s): 75.45.+j, 71.38.Ht, 75.50.Ee, 76.75.+i

I. INTRODUCTION

Quasi-low-dimensional spin systems have attracted considerable attention due to the appearance of various nonmagnetic quantum states that can emerge when conventional long-range order is suppressed. In particular, in the quantum limit of spin $1/2$, an antiferromagnetic (AF) state often has strong competition from a dimerized state (DS) of singlet bonds to become the ground state. Such a DS is characterized by zero average on-site spin and formation of a spin gap Δ of up to several hundred Kelvin, which separates the spin-singlet ground state from the first excited spin triplet. The prototypical systems include the Haldane and spin-Peierls compounds, spin chains, or spin ladders.[1–3]

The absence of spectral weight at the Fermi level suggests that many of these systems are Mott insulators in which strong correlations are responsible for their insulating nature.[4] However, even very low ($\sim 10^{-2}$) doping or the application of pressure can cause a remarkable collapse of the spin gap with emerging AF order or even metallic behavior and superconductivity.[5] Impurities or defects in a spin-gap system may have profound effects on its magnetic state,[6] triggering long-range AF ordering.[7] A better-controlled way of altering the magnetic state of a quantum magnet from spin-singlet dimers to long-ranged AF order is achieved by the application of pressure altering the effect of spin fluctuations, thus, driving a quantum phase transition (QPT) between competing ground states.[8] Distinct magnetic Bragg peaks observed by neutron spectroscopy in *stoichiometric* $KCuCl_3$ and $TlCuCl_3$ indicate the emergence of the ordered AF moments.[9,10]

An even more delicate way to destroy a dimerized state in a *stoichiometric* spin-gap system is to drive it through a magnetic-field-induced QPT.[11] Typical examples include coupled spin ladders (such as $TlCuCl_3$ or $KCuCl_3$),[12] weakly coupled chains of $S = 1$ Ni atoms[13] or planes of Cu dimers.[14] In the presence of a magnetic field, the Zeeman energy reduces the gap to $\Delta(H) = \Delta - g\mu_B H$. At $T = 0$, a finite magnetization associated with AF order appears above the critical field $H_c = \Delta/g\mu_B$. The applied field, thus, acts like a chemical potential, and the Bose gas of triplets is populated above H_c.[15] For most known systems, a rather high value of Δ drives H_c well above 100 T. Nevertheless, an exceptionally low Δ on the order of several Kelvin in some quantum spin magnets indicated above makes it possible to observe magnetic-field-induced AF ordering above the corresponding $H_c = \Delta/g\mu_B$: In the canonical magnetic-field-induced QPT systems, TlCuCl3 and $KCuCl_3$ Δ of 7.5 and 30 K correspond to H_c of 6 and 23 T, respectively.[12]

In contrast to all of the ways to destroy a DS mentioned above, in this paper, we present spectroscopic evidence for AF ordering below $T = 15$ K at *ambient pressure* in the *stoichiometric* spin-gap system NaV_2O_5 with $\Delta \approx 100$ K (Ref. 16) in a magnetic field of, at least, 2 orders of magnitude lower than H_c expected for a material with such a large Δ.

II. BACKGROUND

Highly anisotropic NaV_2O_5 has an orthorhombic structure (P_{mmn}) at room temperature.[17] In the high-temperature phase, its magnetic susceptibility, χ, behaves similar to that expected for Heisenberg spin-1/2 chains. Below $T_c = 34$ K, a gap of $\Delta \sim 100$ K opens up in the spectrum of its magnetic excitations accompanied by a sharp reduction in χ due to dimerization and doubling of the lattice constants, characteristic of spin-Peierls systems.[16] However, the strong suppression of T_c by a magnetic field inherent to spin-Peierls systems does not occur, whereas, $2\Delta/T_c$ is almost two times higher than that in genuine spin-Peierls systems.[18] These facts, along with a very high jump in entropy at T_c, indicate that the driving force for the phase transition that results in an opening in a spin gap between the spin-singlet ground and spin-triplet excited states in NaV_2O_5 is the *charge ordering* of electrons in the quarter-filled vanadium ladders. Such a phase transition transforms a mixed-valence $V^{+4.5}$ state with one electron shared between two vanadium positions on a V-O-V rung above T_c to localized d electrons below T_c,[17] corresponding to charge ordering as revealed by x-ray diffraction,[19] NMR,[20] and dielectric[21] studies. It is suggested that a spin-singlet pair (dimer) is formed on adjacent rungs in a charge-ordered ladder.[22–24] To the best of our knowledge, no AF phase transition has been reported in NaV_2O_5 down to 77 mK.[6]

Although a controversy over the number of inequivalent vanadium positions and their valences (V^{+4}, $V^{+4.5}$, and V^{+5})

has resulted in two theoretical models in which charge ordering occurs either in every vanadium ladder[22] or in every other ladder,[23,24] those models do explain most of the electronic and magnetic properties of NaV_2O_5. However, several experiments, which reveal distinct anomalies well below T_c in the 10–15-K range, still require explanation. Those are an enormous increase in thermal conductivity peaked at about 15 K,[25] a steep increase in the electron spin resonance linewidth below about 15 K[6,26] and static spin freezing around 11 K found by muon spin relaxation (μ^+SR)[27] all of which disappear upon introduction of about 1% Na vacancies. Another μ^+SR paper,[28] using samples from a different source, also shows increasing relaxation of muon spins with decreasing temperature characteristic of slowing spin fluctuations and suggests the possibility of magnetic ordering near 15 K, well below the spin-gap transition. These facts indicate that all of those effects are rather intrinsic and possibly reflect a magnetic phase transition unrevealed so far.

The muon experiments[27,28] attract particular attention as, although having an unparalleled sensitivity to local magnetism, muons do not notice any sign of the spin-gap transition at T_c, clearly detected by many other techniques.[16,19–21] In particular, the muon relaxation rate[27,28] does not follow the sharp reduction in χ at T_c (Ref. 16), which clearly implies that the muon does not act as a local magnetometer in this temperature range. This fact may indicate that the local magnetic environment around the muon is fundamentally different from the rest of the host and that this local environment does not change around T_c. On the other hand, spin polarons (SPs) (which may form a local magnetic environment around the muon fundamentally different from that of the host) have long been predicted to persist around a magnetic transition.[29]

III. THE EXPERIMENT

Time-differential muon spin rotation experiments,[30] using positive muons 100% spin-polarized transverse to the applied magnetic field and the c axis of single crystals of NaV_2O_5 (from the same source as those used in Ref. 27), were carried out on the M15 muon channel at TRIUMF using the *HiTime* spectrometer in magnetic fields up to 7 T and temperatures from 300 down to 2 K. X-ray diffraction and magnetic susceptibility (Fig. 1) measurements on these crystals and a polycrystalline pressed powder pellet of NaV_2O_5 (also examined) produce results consistent with the literature data.

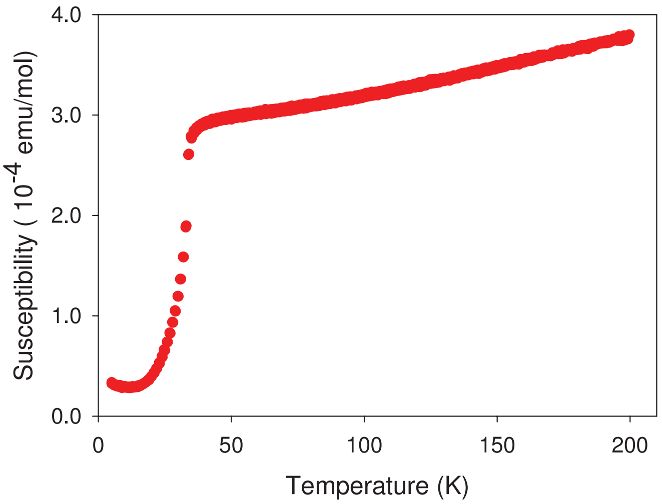

FIG. 1. (Color online) Temperature dependence of the magnetic susceptibility of NaV_2O_5 single crystals in magnetic fields at $H = 0.1$ T.

IV. RESULTS AND DISCUSSION

At low temperatures, in magnetic fields transverse to the initial muon polarization, Fourier transforms of the μ^+SR time spectra consist primarily of two satellite lines positioned symmetrically on either side of the central narrow line (Fig. 2), which appears precisely at the bare-muon Larmor frequency of $\nu_\mu = \gamma_\mu B/2\pi$ (where $\gamma_\mu = 2\pi \times 135.538\,79$ MHz/T is the muon gyromagnetic ratio and B is the magnetic field). The position of this central line is temperature independent and coincides with the single peak observed in a reference sample ($CaCO_3$); thus, it constitutes the signal from those muons whose immediate environments are *nonmagnetic*. Satellite lines represent signals from those muons that have different magnetic environments. Positions of the satellites with respect to the central line do not depend on a magnetic field and correspond to two characteristic local magnetic fields at

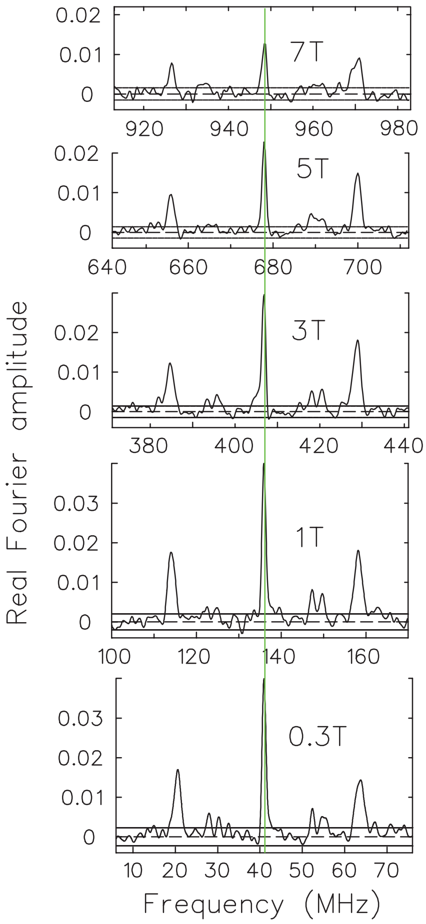

FIG. 2. (Color online) Frequency spectra of muon spin precession in NaV_2O_5 in different magnetic fields at $T = 2$ K. Each spectrum is offset horizontally to place the bare-μ^+ frequency on the same vertical line (green online).

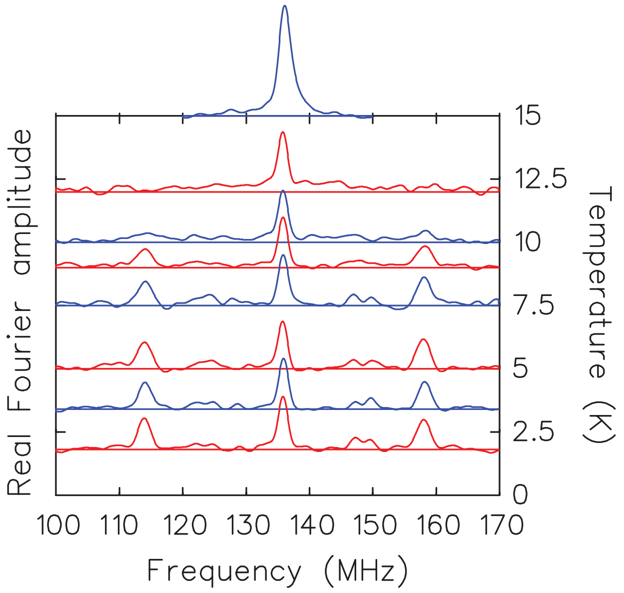

FIG. 3. (Color online) Frequency spectra of the muon spin precession signals in NaV_2O_5 in a magnetic field of 1 T at different temperatures. Characteristic AF lines disappear above 10 K.

$B_\pm = B \pm 0.17$ T. The local magnetic field of 0.17 T is typical for a muon at an interstitial position in various magnetic materials.[30]

Figure 3 presents the evolution of μ^+SR spectra with temperature. Satellite lines disappear above 10 K. Below 10 K, spectral weights of the satellite lines are identical at the lower fields, and each amounts to about 1/2 the spectral weight of the central line. Above 15 K, the spectral weight of the central line doubles at the expense of the satellite signals. We claim that such an evolution reflects an AF phase transition at about 15 K, which results in three magnetically inequivalent muon positions in the AF phase: 50% of muons in the *nonmagnetic* environment and the other 50% residing equally in the *magnetic* environment of the *two AF sublattices*. The three main lines can result from a single type of crystallographic muon site; the total spectral weight of other features in the frequency spectra do not exceed 10% of the overall spectral weight. Satellite lines are not seen in the polycrystalline sample, which is evidence for strong anisotropy of the local magnetic field. This latter fact and the results presented in Figs. 2 and 3 suggest that the local magnetic field is (anti)parallel to the c axis, which also is supported by the absence of muon spin precession in Ref. 27. Thus, the geometry chosen in the current experiment is the most convenient for the determination of the local magnetic field.

In NaV_2O_5, the exchange interaction between vanadium ladders at high temperature $J_\perp \approx 35$ K is rather high,[31] which might result in three-dimensional AF ordering. Nevertheless, the spin-gap state persists below T_c up to the highest applied field of 33 T as $\Delta \approx 100$ K is larger than $J_\perp$.[32] However, formation of the spin-gap state in NaV_2O_5 is intimately connected to the charge ordering on the Trellis lattice (two-dimensional frustrated coupled ladders), which does not allow full charge ordering due to frustration.[24] This fact makes up the core of the model, which suggests charge ordering in every other vanadium ladder.[24] This results in differentiation of the spin subsystem into magnetic and nonmagnetic ladders equally populated in NaV_2O_5 which is fully consistent with our experiment: Those muons, which reside within charge-ordered ladders, find themselves in the nonmagnetic environment of a spin-gap state, whereas, those muons that rest within charge-disordered ladders experience an AF transition at about $T_N \sim 15$ K as a result of a weak exchange interaction $J_\perp$ between disordered ladders. At low temperatures, $J_\perp$ is reduced with respect to its high-temperature value due to the intervening charge-ordered ladders and lattice doubling at T_c. According to the mean-field theory for a quasi-one-dimensional quantum AF system,[33] $J_\perp$ can be estimated as $J_\perp \approx k_B T_N/[1.28\sqrt{\ln(5.8J/k_B T_N)}] \approx 5$ K, where k_B is the Boltzmann constant and $J \approx 560$ K is the exchange interaction within the ladder.[16] This value is consistent with $J_\perp \approx$ 2–4 K calculated for the low-temperature phase of NaV_2O_5.[24]

Thus, we find coexistence of spin-gap and AF phases in stoichiometric NaV_2O_5 below 15 K. As the muon is a local magnetic probe, we cannot determine if this AF ordering is a long-range transition or a local AF cluster formation. However, the experiments of Refs. 25–27 indicate a cooperative phenomenon rather than local clustering.

While staying bare and acting as a local magnetometer at low temperatures, the muon does not stay bare at a higher temperature: At temperatures above the AF transition, we observe the spectroscopic signature of spin polarons in NaV_2O_5. As shown in Fig. 4 as a function of magnetic field at $T = 29$ K, the μ^+SR spectra exhibit the characteristic two-frequency precession (doublet) in high magnetic fields[34,35] (the low-frequency line is a background signal whose frequency coincides with that in $CaCO_3$). Such a doublet corresponds to two muon spin-flip transitions between states with fixed electron spin orientation within the SP.[30,34,35] Amplitudes of the SP lines decrease very rapidly above 100 K implying significantly reduced SP formation probability in that temperature region. The characteristic widths of the polaron lines are consistent with the muon relaxation rate measured in a longitudinal magnetic field and are assigned to spin fluctuations associated with the spin gap.[27]

In insulators and semiconductors, the positive muon can bind an electron to form a muonium (Mu) atom analogous to a hydrogen atom in which the proton is replaced by a muon.[30,36] In the μ^+SR experiments on insulators[37,38] and semiconductors,[39–42] each incoming 4-MeV muon injects a very low ($\sim 10^6$) concentration of free carriers liberated during its thermalization into the empty conduction band; one of those electrons can be captured by the muon. A positive muon, thus, acts as an attractive Coulomb center for electron localization.[43]

In a magnetic system, the exchange interaction, I, between free electrons and localized spins creates yet another channel for electron localization—the charge carrier localizes into a ferromagnetic (FM) ‘droplet’ on the scale of the lattice spacing in a paramagnetic (PM) or AF ‘sea’—a spin polaron.[44] In this case, the long-range Coulomb interaction ensures initial electron capture, whereas, the short-range exchange interaction provides further localization into a bound SP (BSP) to the muon. Formation of a BSP around a positive muon was demonstrated recently in PM (Refs. 34 and 35) and AF (Ref. 45) hosts.

If a BSP were to form in a DS, the increase in the electron kinetic energy due to localization would have to be compensated by the on-site exchange interaction $IS/2$ of

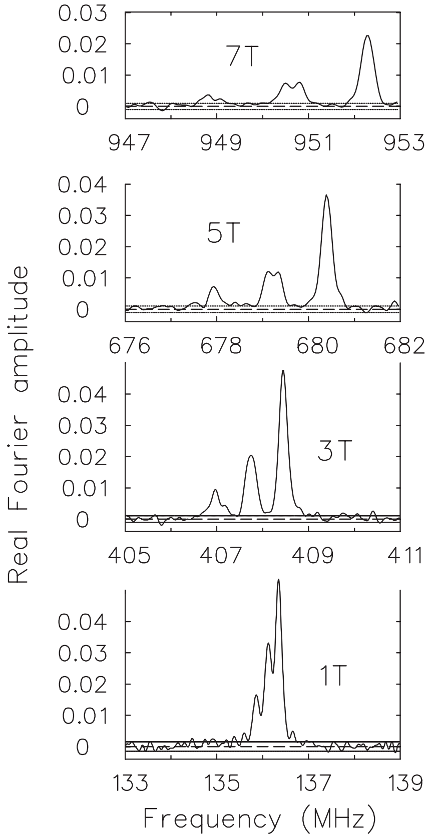

FIG. 4. Fourier transforms of the muon spin precession signal in NaV_2O_5 in different magnetic fields at $T = 29$ K. The low-frequency line of each spectrum is a background signal; two other lines represent two characteristic muon transitions (doublet) within a SP.

the electron with local spin S combined with the Coulomb potential of the muon versus the energy NJS^2 required to flip N local spins S with an effective exchange energy J to produce a FM droplet within the radius R plus the entropy change ΔW due to ordering within the SP so that the change in the free energy,

$$\Delta F = \frac{\hbar^2}{2mR^2} - I\frac{S}{2} - \frac{e^2}{\varepsilon R} + NJS^2 + T\,\Delta W \qquad (1)$$

has a minimum as a function of R—the radius of the electron's confinement.[45]

The probability of SP formation around the muon depends on the last two terms of Eq. (1): In a DS with low J, the SP is expected to form, whereas, at higher J, the muon is expected to stay bare and, therefore, may be used as a local magnetic probe. In NaV_2O_5, a rather high value of $J \approx 560$ K precludes SP formation in a fully developed DS. Instead, at low temperatures, the muon stays bare and sees either an AF or a DS environment (Figs. 2 and 3). By contrast, a SP bound to the muon is formed in NaV_2O_5 below about 100 K in the PM state and remains present through T_c down to 20 K, which is identified as the maximum temperature for a fully developed DS.[21] Above 100 K, the increasing entropy within the SP reduces its stability and suppresses its formation, and the muon stays bare. Polaron spectra in a polycrystal are almost the same as in the single crystal, which indicates the 1*s* isotropic nature of the BSP electron.[34,35]

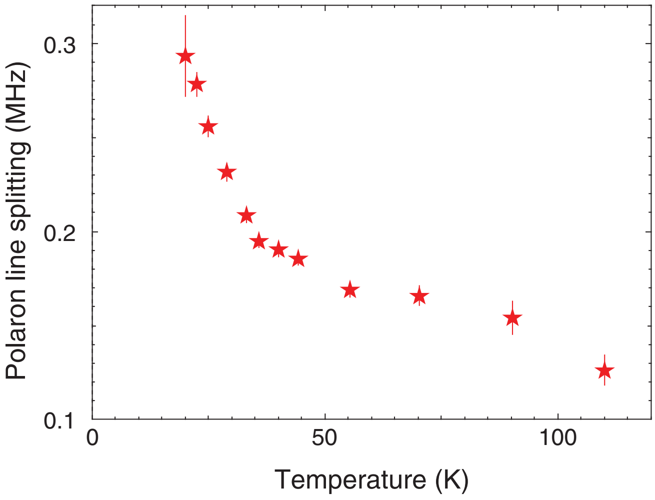

FIG. 5. (Color online) Temperature dependence of the SP lines splitting in a magnetic field of $H = 1$ T.

The observed splitting between the two SP lines in NaV_2O_5 is linear in H/T (Figs. 5 and 6), consistent with a Brillouin function in the small H/T regime and similar to observed dependences for SPs recently characterized in various other materials.[34,35,45–49]

An alternative interpretation of these spectra as arising from a simple Mu atom can safely be ruled out as spin exchange with the host's spins[36] would result in rapid spin fluctuations of the bound electron, averaging the muon-electron hyperfine interaction to zero, which would cause effective doublet disappearance. By contrast, when the electron spin is bound strongly into a SP, the local FM ordering holds the electron spin fixed, which manifests itself as a characteristic doublet.[34,35,45,46,48–50] Likewise, the insulating nature of NaV_2O_5 and a remarkable insensitivity to the spin-gap transition at T_c allows one to rule out possible Knight shifts within the bare-muon scenario. Finally, a strong shift in the SP lines to higher frequencies with respect to the background signal reflects the FM state within a SP.[34]

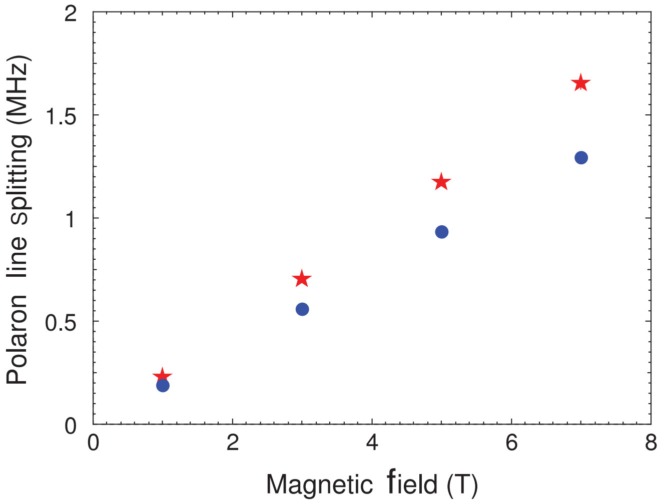

FIG. 6. (Color online) Magnetic-field dependences of the SP lines splitting at $T = 29$ K (stars, red online) and $T = 40$ K (circles, blue online).

V. SUMMARY

We have found a coexistence of spin-gap and AF states at low temperatures, $T \leqslant 15$ K, in NaV$_2$O$_5$. At higher temperatures, 20–100 K, we detected a SP bound to the muon.

ACKNOWLEDGMENTS

This work was supported by the NBIC Center of the Kurchatov Institute, NSERC of Canada, and the US DOE, Basic Energy Sciences (Grant No. DE-SC0001769).

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