controlled more carefully than stochastic noise.

Whether resilient quantum computation can be implemented in practice remains to be seen. However, the results obtained here show that, in principle, noise of a level below the error threshold is not an obstacle for quantum computation.

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37. We thank the National Security Agency for support.

15 May 1997; accepted 21 November 1997

Absence of a Spin Gap in the Superconducting Ladder Compound $Sr_2Ca_{12}Cu_{24}O_{41}$

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Transport and copper-63 nuclear magnetic resonance measurements of the Knight shift and relaxation time T_1 performed on the two-leg spin ladders of $Sr_2Ca_{12}Cu_{24}O_{41}$ single crystals as a function of pressure show a collapse of the gap in ladder spin excitations when superconductivity is stabilized at 31 kilobars. This result suggests that the superconducting phase in these materials may be connected to this transition and the collapse of the spin gap, and support the prediction made with exact diagonalization techniques in two-leg isotropic $t - J$ ladder models of a transition between a low-doping spin gap phase and a gapless regime.

The existence of superconductivity in two families of materials where this property was not expected at first sight [the low-dimensional organic conductors and the high–transition temperature (T_c) cuprates] has been a major achievement of condensed matter research of the last two decades. The mechanism of superconductivity for both classes of compounds is still under intense debate, but there is already a consensus about their low-dimensional electronic structure that may be the clue governing superconducting (SC) pairing correlations. The recent finding of new SC copper oxide structures (*1*) exhibiting one-dimensional (1D) features with both isolated CuO_2 chains and Cu_2O_3 ladders—that is, pairs of CuO_2 chains linked by oxygen atoms between the coppers—has profoundly revived the interest for superconductivity in cuprates and 1D materials.

We discuss the ladder compound $Sr_2Ca_{12}Cu_{24}O_{41}$, which derives from the parent compound $Sr_{14}Cu_{24}O_{41}$ through Ca substitution. The structure of $Sr_{14}Cu_{24}O_{41}$ displays CuO_2 chains and Cu_2O_3 two-leg ladders parallel to the *c* axis of the structure (*2*); other insulating 1D materials like $SrCu_2O_3$ contain only Cu_2O_3 ladders and no chains. In contrast, the undoped parent compound for the high-T_c cuprates exhibits a 2D CuO_2 layer structure. In both systems, all copper sites belonging to the ladders or to the planes are occupied by a spin 1/2 Cu^{2+} ion. However, although long-range antiferromagnetism is stabilized at low T in the 2D spin system, the properties of the spin ladder materials can be drastically different. In two-leg ladder systems, dominant antiferromagnetic (AF) coupling J between the copper spins on the same rung leads to the formation of a spin singlet on each rung. Consequently, the ground state of the whole ladder is a singlet spin state, and a finite energy is needed to excite a rung spin singlet to a spin triplet state. A spin gap situation is obtained with a characteristic exponential drop of the spin susceptibility upon cooling down.

The existence of a spin gap in a spin-ladder structure has been first proposed theoretically (*3*) and found experimentally in several even-leg ladder copper oxide systems [$SrCu_2O_3$ (*4*, *5*), $LaCuO_{2.5}$ (*6*)] or organic materials (*7*). The spin gap is expected to be quite robust to various perturbations. For example, it is predicted to be stable up to arbitrary small magnetic coupling along the rungs of the ladders (*8*) or in the presence of a small interladder coupling (*9*).

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The persistence of the spin gap upon light hole doping has been established by various techniques [see, for example, (*3*, *10–12*)]. Even more exciting was the suggestion that the spin gap leads to an attractive interaction (*3*, *10*) of holes on the same rung, hence providing dominant *d*-wave–like SC pairing correlations (*13*, *14*) that could possibly materialize into a 3D SC state at low *T*. In addition, the numerical investigation of the complete phase diagram (*15*) of the two-leg isotropic $t - J$ ladder (in this model the motion of the Zhang-Rice singlet holes are described by a hopping matrix element t) suggests the possibility of a transition from the low-doping spin-gap phase (with the concomitant formation of hole pairs) toward a gapless regime. Although the existence of such a transition is clear at sufficiently large doping, the existence of a gapless phase at small doping and small J/t ratio is still controversial.

Some similarities between the superconductivity in quasi-1D organic conductors (*16*) and in ladder copper oxides have been noted (*17*) because, for both cases, superconductivity arises once a charge localized state is suppressed above a critical pressure (*P*). The resemblance is not complete, however, because the band is a half or a quarter filled in organic conductors, whereas it is very likely incommensurate and possibly *P*-dependent in the case of ladders. Furthermore, the localization process occurs through a spin density wave state in organics with the concomitant opening of an exchange gap at the Fermi level (*16*), whereas it can be attributed to a charge density wave state in ladder compounds.

Optical conductivity measurements of $Sr_{14-x}Ca_xCu_{24}O_{41}$ have shown that holes are transferred to the ladders from the chains upon increasing the Ca concentration (*18*). A copper valency of ~2.2 is therefore reached in $Sr_2Ca_{12}Cu_{24}O_{41}$ (hole density of 0.2 per ladder-Cu). Increasing *P* might further increase the density of holes in Cu-ladders or possibly decrease the ratio J/t, or both, and trigger the transition from the spin gap regime to a gapless phase with low-lying spin excitations.

The prediction of a coexistence, in the lightly doped regime, of the spin gap with divergent SC correlations (*13*) is very suggestive for experimentalists. Therefore, it is a crucial experimental test to investigate whether the finite spin gap persists in spin excitations of $Sr_{14-x}Ca_xCu_{24}O_{41}$ when superconductivity of Ca-substituted compounds is stabilized under *P*. We present the preliminary results of transport and ^{63}Cu–nuclear magnetic resonance (NMR) studies performed on a $Sr_2Ca_{12}Cu_{24}O_{41}$ single crystal under *P*. We show, at 31 kbar, the simultaneous onset of superconductivity below 5 K and the existence of low-lying spin excitations from the *T* dependence of the ^{63}Cu Knight shift belonging to the ladders.

Experimental results. Two samples (0.75 mm by 0.65 mm by 1.18 mm for transport experiments and 2.12 mm by 0.9 mm by 1.12 mm for NMR, along the *a*, *b*, and *c* axes, respectively) were cut out from the same slice of a monodomain single crystal, several centimeters long, grown by the traveling solvent floating zone method in an infrared image furnace under an oxygen *P* of 13 bar (*19*).

Transport data were obtained by a conventional four-contact ac lock-in technique with a nonmagnetic high-*P* clamped cell. *P* was calibrated at room *T* via a manganine gauge. The increase of the *c* axis conductivity under pressure is in agreement with values reported (*20*, *21*) for single crystals. At 26 kbar, ρ_c reveals a metal-like *T* dependence, with a charge localization beginning around 30 K but no sign of superconductivity above 1.8 K, which was our lowest *T*. The metal-like behavior of ρ_c is similar under 31 kbar (Fig. 1), but the weak localization at low *T* is stopped at 5 K by a sudden drop of the resistance, which compares well with the *P* data obtained on a sample that was slightly less hole-doped (*21*). We believe that the resistance drop can be confidently ascribed to the onset of a *P*-induced SC ground state.

The ^{63}Cu-NMR experiments were performed in the same high-*P* cell at an NMR frequency of 87.05 MHz. We obtained ^{63}Cu-NMR signals by recording the spin-echo amplitude of the central transition (1/2, −1/2) of the copper nuclei pertaining to the ladders. A narrow ^{63}Cu-NMR signal was also obtained by the usual Fourier transform method. It has been attributed to ^{63}Cu nuclei in the *P* cell surrounding the sample. Because the Knight shift of copper metal is insensitive to *P* on the scale of Knight shift variations to be discussed below (*22*), the narrow signal has been used as a sensitive in situ magnetic field marker.

Figure 2 presents the *T* dependence of the ^{63}Cu-NMR line position with $H//b$ axis at 32.2 kbar and also, in the same *P* cell, after releasing the pressure. The central line position is affected by magnetic and second-order quadrupolar shifts. We here estimate the latter to be about 50 kHz following the determination of the nuclear quadrupole tensor in Ca-substituted samples (*23*). Furthermore, this tensor is not expected to be strongly affected by a lattice contraction (going to low *T* or under *P*), and an overall variation exceeding 10% (that is, 5 kHz) seems very unlikely (*24*, *25*). Such a small shift of the line position (50 parts per million) can admittedly be neglected on the scale of the *T* dependence of the Knight shift. A line-broadening even larger than at ambient *P* is observed at low *T* under *P*. Its origin remains to be understood. The magnetic shift consists usually of a *T*-independent orbital contribution, plus the spin part that follows the uniform susceptibility, the latter being possibly *T*- and *P*-dependent. Because our lowest *T* for the NMR experiments was 5 K, the data were taken in the normal state only. The *T* dependence of the line position at ambient *P* leads to an orbital contribution of $K_{orb} \sim 1.33\%$, which is in agreement with other data at ambient *P* (*23*). The resulting *T*-dependent Knight shift is plotted in Fig. 2 and leads to $K_{orb} \sim 1.33\%$ and a spin gap $\Delta_{sp} \sim 250$ K, given the expression for the *T*-dependent part (*26*): $K_s(T) \sim (1/\sqrt{T}\ \exp(-\Delta'/kT)$ at $kT \ll \Delta_{sp}$ (*k* is Boltzmann's constant). Making the reasonable assumption of a *P*-independent orbital shift and a negligible *T* dependence of the quadrupolar contribution, the 32.2 kbar data in Fig. 2 show only a weak *P* dependence of the spin part at $T > 150$

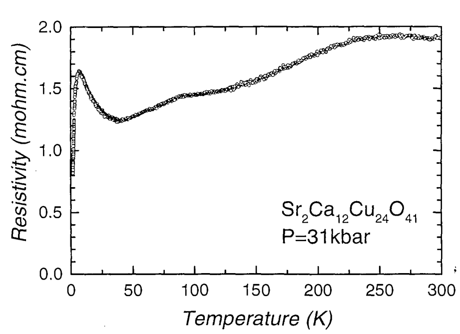

Fig. 1. Temperature dependence of the resistivity along the ladders under 31 kbar. The onset of superconductivity is at 5 K.

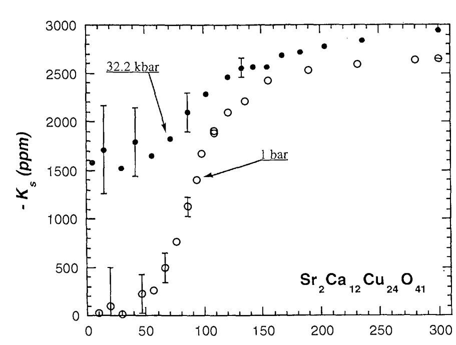

Fig. 2. Temperature dependence of the ^{63}Cu-ladder nuclei Knight shift. A spin gap $\Delta_{sp} \sim 250$ K is obtained from an activation plot below 150 K.

K, possibly ascribed to the P dependence of the quadrupolar shift, which is at variance with the drastic P dependence of the spin part observed at low T. Figure 2 reveals the emergence of a zero-T susceptibility with a loss in cooling that amounts to no more than half the loss observed at ambient P. Our data at ambient P confirm the depression of the spin gap upon doping, as previously announced in the Knight shift study of a $Sr_5Ca_9Cu_{24}O_{41}$ single crystal (*23*) and also from T_1 data in the whole series corresponding to $0 \leq x \leq 9$ (*27*). However, what is most remarkable in Fig. 2 is the existence of low-lying spin excitations in the compound $Sr_2Ca_{12}Cu_{24}O_{41}$ at P = 32.2 kbar, which is large enough to evidence the onset of superconductivity at 5 K. The fact that superconductivity arises when the spin gap on the ladders has collapsed is an argument in favor of superconductivity taking place on the ladders.

By comparing the 1 bar and 31 kbar resistivity (ρ) data, we can infer from the smaller increase of the ρ_a/ρ_c anisotropy on cooling at 31 kbar that the confinement of the carriers along the ladders could correlate with the size of the spin gap (*28*). This finding suggests a picture of hole pairs being responsible for the conduction within the ladders as long as the magnetic forces can provide the binding of two holes on the same rung (*3*). The vanishing of the spin gap could thus be responsible for the dissociation of the pairs, making in turn the hopping of the transverse single particle easier. Second, the behavior of the susceptibility under 32.2 kbar with low-lying spin excitations is at first sight reminiscent of the situation that prevails in quasi-1D organic conductors where the susceptibility is T-dependent below 300 K, but finite and noticeably independent of the behavior of the charge degrees of freedom (spin-charge separation in a 1D chain), which have been understood in terms of a correlated Tomonaga-Luttinger (TL) liquid (*16*).

The nuclear spin lattice relaxation has also been measured because this quantity is a very sensitive probe for the low-lying spin excitations in cuprates (*29*) and 1D organic conductors (*30*). The spin-lattice relaxation with $H//b$ was measured by the saturation-recovery technique, and T_1 was determined from the single time constant governing the magnetic relaxation in high-field NMR of a quadrupolar nucleus such as ^{63}Cu (spin = 3/2) (*31*). The exponential fit with a single time constant is excellent down to 70 K but is not as good at low T and leads to a poorer T_1 determination as indicated by the error bars in Fig. 3. We take the single exponential recovery of the magnetization as a serious argument supporting the magnetic origin of the spin-lattice relaxation. In case of the compound $Sr_{14}Cu_{24}O_{41}$, the single exponential fit is no longer valid below 200 K (*25*). Moreover, the relaxation rate increases by about a factor of 10 in the T regime 30 to 100 K going from $Sr_{14}Cu_{24}O_{41}$ to $Sr_{2.5}Ca_{11.5}Cu_{24}O_{41}$ (*23*), and another factor of 5 is gained at 32.2 kbar. A quadrupolar contribution to the relaxation at low T seems unlikely at 32.2 kbar but cannot be completely excluded until detailed experiments on the P dependence of the quadrupolar coupling are performed. The 1 bar and 32.2 kbar data for T_1^{-1} versus T in logarithmic scales are shown in Fig. 3. From 300 to 200 K, T_1 is both T- and P-independent. At 1 bar, T_1 becomes activated below 120 K with an activation energy Δ' = 350 K. This behavior is in fair agreement with other experiments on single crystals (*23*). This activated behavior breaks down below 40 K. At 32.2 kbar, the relaxation displays quite a different T dependence. We can attribute the nonactivated T dependence to the collapse of the spin gap and the persistence even at low T of populated spin excitations modes contributing to the relaxation. At high T, however, the absence of significant T dependence suggests a relaxation induced by AF fluctuations in Heisenberg chains. In this T range, such a relaxation channel should not be sensitive to the presence or the absence of a spin gap.

Discussion. We now discuss the above experimental data in the context of existing theoretical works. As stated above, such materials are expected to be accurately described by a two-leg $t - J$ ladder that accounts for the strong nearest-neighbor AF coupling J between the spins and the hole delocalization t. It is believed that, in these materials, interchain and intrachain couplings are similar (isotropic case) and comparable in magnitude to their values in the 2D copper oxide materials (typically $J/t \simeq 0.3 \ldots 0.4$ corresponding to the strong coupling limit). Exact diagonalization techniques applied to the two-leg $t - J$ ladder have proven, in the isotropic case, the formation of hole pairs at low doping with the concomitant formation of a spin gap (*5*, *10*, *11*), as initially suggested in the anisotropic limit ($J_\perp \gg J_\parallel$). A phase diagram for the isotropic $t - J$ ladder as a function of J/t and doping parameters has been established (*15*) and is schematically shown in Fig. 4. Near the half-filled band situation, that is, with a small hole concentration, the spin gapped phase (with a zero energy charge mode) is recovered while a phase of the TL universality class, that is, with a single gapless spin and a single gapless charge mode, becomes stable under larger hole doping (*11*). Physically, the existence of such a transition can easily be understood from a weakly interacting band picture (*32*). Indeed, for hole density $n_h \geq 0.5$, the higher energy antibonding band becomes unoccupied, and one recovers a single band picture analogous to the single-chain case. However, the line $n_h = 0.5$ is expected to be quite singular because Umklapp scattering characteristic of a half-filled band likely leads to an insulating state (*33*). More interestingly, the pos-

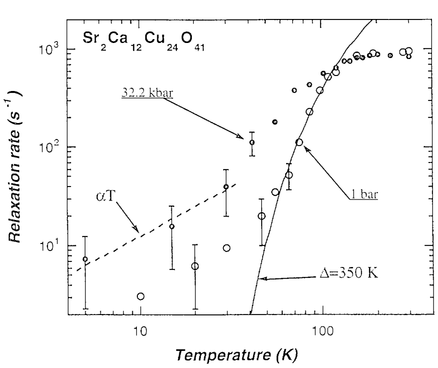

Fig. 3. Temperature dependence of the ^{63}Cu-ladder relaxation rate T_1^{-1} at 1 bar and under 32.2 kbar.

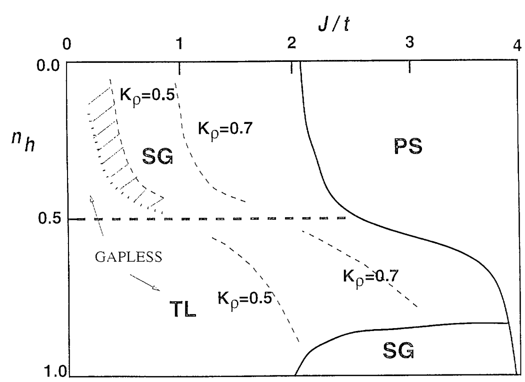

Fig. 4. Schematic phase diagram of the two-leg $t - J$ ladder as a function of J/t and hole density n_h based on exact diagonalizations of small clusters [see (*15*)]. The thick dashed and dotted lines separate the spin gapped (SG) and gapless phase of the Tomonaga-Luttinger (TL) type. At large unphysical J/t ratios, phase separation (PS) occurs. Lines of constant K_ρ (see text) are also indicated (thin dashed lines). The hatched region of the SG phase corresponds to dominant CDW correlations.

sibility of a similar crossover between the spin gapped phase toward a gapless phase also exists at low doping for a sufficiently small J/t ratio, although it is difficult to estimate numerically the transition line (*33*) (indicated tentatively in Fig. 4 by a dotted line). Whether this gapless phase is also of the TL type remains unclear.

We shall argue here that the experimental data of T_1, together with the loss of susceptibility by a factor of about 2, could possibly support the existence of such a crossover in the $Sr_{14-x}Ca_xCu_{24}O_{41}$ materials, the 1 bar and 32.2 kbar phases being ascribed to the spin gapped and gapless phases, respectively. From simple symmetry considerations (*11*), in a ladder system one expects two spin excitations branches that could independently acquire a gap or be gapless. The exponential decrease of the spin susceptibility at 1 bar (Fig. 2) is characteristic of a full gap in the spin sector. However, the gapless phase of the TL type would exhibits one gapless mode and one mode with a gap. The susceptibility data at 32.2 kbar are consistent with this scenario. At sufficiently high Ts, both modes are expected to contribute. However, the gapped spin mode is depopulated on decreasing T. At the lowest Ts, half of the spin degrees of freedom contribute to the static susceptibility as portrayed by the T variation of K_s in Fig. 2.

It is worth looking at the T profile of the relaxation rate that would be predicted in the above scenario of the $t-J$ ladder model. First, for $T > 150$ K, T_1^{-1} is T independent, indicating that AF chainlike spin correlations dominate the relaxation. This suggests that the contribution coming from uniform spin correlations to T_1^{-1} is sufficiently small to be safely ignored at all Ts and Ps of interest (*30*). In the spin-gap phase at 1 bar, where both branches of spin excitations are frozen, T_1^{-1} and K_s are thermally activated with slightly different activation energies (*23*). At high P, however, the restoration of a phase with a gapless spin and a gapless charge modes will promote a new channel of relaxation of the TL type. This is well known to introduce a power law component in the T dependence of T_1^{-1} that will read

$$T_1^{-1} = C_1 \exp(-\Delta'/k_B T) + C_2 T^{2K_\rho}$$

in the low-T domain, where $C_{1,2}$ are positive constants. Here, K_ρ stands as the power law exponent of the AF response function ($\chi \sim T^{-1+2K_\rho}$) (*15*). In the metallic phase that is the precursor to superconductivity, $2K_\rho$ should be close to unity, so it follows that a nonthermally activated component should emerge for T_1^{-1} at sufficiently low T. This prediction seems to be in qualitative agreement with the T_1^{-1} data given in Fig. 3. Indeed, one finds no thermal activation for the relaxation and this, especially below 50 K where K_s becomes metallic and T-independent.

We conclude with a comment on the role of impurity scattering in the framework of the $t-J$ model. The SG phase is characterized by competing 1D SC and charge density wave (CDW) fluctuations. The line $K_\rho = 0.5$ in Fig. 4 separates the regions of dominant CDW fluctuations (hatched region) and of dominant SC fluctuations at larger J/T ratios (*15*). Orignac and Giamarchi (*34*) have shown that impurity scattering becomes strongly relevant in the case of dominant CDW fluctuations leading to localization and possibly explaining the insulating behavior seen at low T in resistivity measurements.

Transport and ^{63}Cu-NMR measurements on the prototype $Sr_2Ca_{12}Cu_{24}O_{41}$-doped spin ladder under P have been reported and analyzed in the context of the $t-J$ ladder model. The drastically different behaviors observed at 1 bar and 31 kbar of the T dependence of the ladder static susceptibility and of the relaxation rate $1/T_1$ are attributed to the appearance of a zero energy spin mode at 31 kbar. We argue that this phase can be identified to the gapless TL metallic phase of the $t-J$ ladder. Because, in the 31 kbar phase, superconductivity sets in at low T, this suggests that superconductivity itself might be connected to such a transition and to the collapse of the spin gap. We emphasize that it might also be possible to understand the relaxation data at 32.2 kbar in terms of an activated behavior between 30 and 150 K, with a gap of 120 K coming from a single gapped spin mode in a TL regime that would thus be stable even in the low-doping region of the phase diagram (Fig. 4). This interpretation would not rule out the crucial result of this work, namely, the existence of low-lying spin excitations under P as seen from the susceptibility data. Because a small interladder coupling is expected to be far more effective in this phase than in the SG phase, this scenario would, in addition, explain the drastic decrease of the anisotropy of the resistivity seen in experiments (*21*, *28*). Further experimental and theoretical work is needed to clarify this important issue.

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35. H.M. thanks the Direction de la Recherche et de la Technologie (DRET) for financial support. We acknowledge illuminating discussions with T. Giamarchi. U.A. acknowledges support from Deutscher Akademischer Austausch Dienst (DAAD) in the frame of the Procope program and the graduiertenkolleg of the Deutsche Forschungsgemeinschaft.

29 September 1997; accepted 9 December 1997