Coexistence of ferromagnetism and superconductivity in the hybrid ruthenate-cuprate compound RuSr$_2$GdCu$_2$O$_8$ studied by muon spin rotation ($\mu$SR) and DC-magnetization

C. Bernhard$^1$, J.L. Tallon$^2$, Ch. Niedermayer$^3$, Th. Blasius$^3$, A. Golnik$^1$, E. Brücher$^1$, R.K. Kremer$^1$, D.R. Noakes$^4$, C.E. Stronach$^4$ and E.J. Ansaldo$^5$

1) Max-Planck-Institut für Festkörperforschung Heisenbergstrasse 1, D-70569 Stuttgart, Germany
2) Industrial Research Ltd., P.O. Box 31310, Lower Hutt, New Zealand
3) Universität Konstanz, Fakultät für Physik, D-78434 Konstanz, Germany
4) Department of Physics, Virginia State University, Petersburg, Virginia 23806, USA
5)University of Saskatchewan, Saskatoon S7N OWO, Canada (26.1.99)

We have investigated the magnetic and the superconducting properties of the hybrid ruthenate-cuprate compound RuSr$_2$GdCu$_2$O$_8$ by means of zero-field muon spin rotation- (ZF-$\mu$SR) and DC magnetization measurements. The DC-magnetisation data establish that this material exhibits ferromagnetic order of the Ru-moments ($\mu$(Ru) $\approx$ 1 $\mu_B$) below $T_{Curie} = 133$ K and becomes superconducting at a much lower temperature $T_c = 16$ K. The ZF-$\mu$SR experiments indicate that the ferromagnetic phase is homogeneous on a microscopic scale and accounts for most of the sample volume. They also suggest that the magnetic order is not significantly modified at the onset of superconductivity.

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I. INTRODUCTION

Since the discovery of superconductivity in the cuprate system La$_{2-x}$Ba$_x$CuO$_4$ in 1986 an ever growing variety of high-$T_c$ superconducting cuprate compounds has been synthesized all of which contain CuO$_2$ planes (some also contain CuO chains) as their essential structural elements which host the superconducting charge carriers.

Between the CuO$_2$ planes are various kinds of layers, typically NaCl-type, which are insulating and act merely as a charge reservoir. To date, the ruthenate compound Sr$_2$RuO$_4$ is the only known layered perovskite-like system which becomes superconducting even though it does not contain any CuO$_2$ planes or CuO chains. Despite its rather low transition temperature $T_c = 1.5$ K the study of its electronic and magnetic properties has become a very rich and active field of research. In parallel, the electronic and magnetic properties of the related ruthenate compounds, such as for example the SrRuO$_3$ system which is an itinerant 4d-band ferromagnet with $T_{Curie} \approx 165$ K, have attracted a great deal of interest.

Another potentially promising and exciting direction of research has been promoted by the circumstance that the RuO$_2$ layers share the same square-planar coordination and a rather similar bond length with their CuO$_2$ counterparts. A whole new family of hybrid ruthenate-cuprate compounds may therefore be constructed whose members consist of different sequences of alternating RuO$_2$- and CuO$_2$ layers. Recently, one such a hybrid ruthenate-cuprate compound, the 1212-type RuSr$_2$GdCu$_2$O$_8$ system comprising CuO$_2$ bilayers and RuO$_2$ monolayers, has been synthesized as single-phase material. A subsequent study of its electronic and magnetic properties has revealed that this material exhibits ferromagnetic order at a rather high Curie temperature $T_{Curie} = 133-136$ K and becomes superconducting at a significantly lower critical temperature $T_c = 15-40$ K (depending on the condition of preparation and annealing) [6-8]. The most surprising observation, however, is that the ferromagnetic order does not vanish when superconductivity sets in at $T_c$. Instead, it appears that the ferromagnetic state remains largely unchanged and coexists with superconductivity. This finding implies that the interaction between the superconducting- and the ferromagnetic order parameters is very weak and it raises the question of whether both order parameters coexist on a truly microscopic scale. Since the early investigations of Ginzburg in 1957 the prevailing view is that the coexistence of a superconducting- (with singlet Cooper pairs) and a ferromagnetic order parameter is not possible on a microscopic scale since the electromagnetic interaction and the exchange coupling cause strong pairbreaking. Indeed, merely based on magnetisation and transport measurements one cannot exclude the possibility that the RuSr$_2$GdCu$_2$O$_8$ samples may be spatially inhomogeneous with some domains exhibiting ferromagnetic order and others superconducting order. We note that unambiguous evidence for the occurrence of bulk superconductivity in RuSr$_2$GdCu$_2$O$_8$ has recently been obtained from specific heat measurements which reveal a sizeable jump
at $T_c$ of $\Delta \gamma \equiv C_p/T \approx 0.35 \text{ mJ/g K}^2$ characteristic of a strongly underdoped cuprate superconductor. In the following we report on muon-spin rotation ($\mu$SR) measurements which establish that the ferromagnetic order is uniform and homogeneous even on a microscopic scale.

The $\mu$SR technique is ideally suited for such a purpose since it provides an extremely sensitive local magnetic probe and, furthermore, allows one to reliably obtain the volume fraction of the magnetically-ordered phase. Here we present the result of a zero-field muon-spin rotation (ZF-$\mu$SR) study of a RuSr$_2$GdCu$_2$O$_8$ sample with $T_c=16$ K and $T_{\text{Curie}}=133$ K which provides evidence that the magnetic order parameter is spatially homogeneous and accounts for most of the sample volume. Furthermore, the ZF-$\mu$SR data establish that the ferromagnetic order is hardly affected by the onset of superconductivity and persists to the lowest available temperature of the experiment $T=2.2$ K. The ZF-$\mu$SR data can be complemented by DC-magnetisation measurements which establish the presence of ferromagnetic order from the observation of a spontaneous magnetization at $T_{\text{Curie}} = 133$ K and of hysteretic isothermal magnetic behavior with a remanent magnetization. It is shown that the ferromagnetic ordering involves the Ru magnetic moments with $\mu(\text{Ru}) \approx 1.05(5) \mu_B$, while the larger Gd-moments with $\mu(\text{Gd}^{3+}) \approx 7.4(1) \mu_B$ remain paramagnetic down to very low temperatures. In addition, the magnetisation measurements indicate an almost complete diamagnetic shielding effect below $T_c$.

II. EXPERIMENT

A. sample preparation and characterization

Polycrystalline samples of the 1212-type system RuSr$_2$GdCu$_2$O$_8$ have been synthesized as previously described by solid state reaction of RuO$_2$, SrCO$_3$, Gd$_2$O$_3$ and CuO powders. The mixture was first decomposed at 960 °C in air. It was then ground, milled and die-pressed into pellets. The first sintering step took place in flowing nitrogen atmosphere at 1010 °C. This step results in the formation of a mixture of the precursor material Sr$_2$GdRuO$_6$ and Cu$_2$O and is directed towards minimizing the formation of SrRuO$_3$. The material was then reground before it was reacted in flowing oxygen for 10 hours at 1050 °C. This sintering step was repeated twice with intermediate grinding and milling. Each reaction step was carried out on a MgO single crystal substrate to prevent reaction with the alumina crucible. Finally the samples were cooled slowly to room temperature in flowing oxygen. Following this procedure we have also made a Zn-substituted RuSr$_2$GdCu$_1.91$Zn$_{0.09}$O$_8$ sample and a Y $\leftrightarrow$ Gd cosubstituted sample RuSr$_2$Gd$_{0.9}$Y$_{0.1}$Cu$_2$O$_8$. X-ray diffraction (XRD) measurements indicate that all samples are single phase 1212-type material and give no indication for traces of the ferromagnetic phase SrRuO$_3$. Figure 1(b) displays a representative XRD-spectrum of RuSr$_2$GdCu$_2$O$_8$, the plus signs show the raw data and the solid line shows the result of the Rietveld refinement. The related structure of RuSr$_2$GdCu$_2$O$_8$ is shown in Fig. 1(a).
The electronic properties of RuSr$_2$GdCu$_2$O$_8$ have been characterized by measurements of the temperature-dependent resistivity and thermo-electric power. Representative results are shown in Fig. 2(a) and 2(b) respectively (see also Ref. 8).

The temperature dependence of the resistivity, $\rho$, of RuSr$_2$GdCu$_2$O$_{8-\delta}$. (b) The temperature-dependent thermo-electric power $S(T)$. 

The temperature dependence of the thermo-electric power $S(T)$ and, in particular, its normal state value of $S(300 \text{ K}) \approx 75 \mu \text{V/K}$ is rather typical for a strongly underdoped cuprate superconductor with $T_c << T_{c,max}$, consistent with a hole content of $\rho \approx 0.07$ holes per CuO$_2$ planes and a value of $T_{c,max}$ of the order of 100 K [8,12]. The resistivity measurements indicate that the RuSr$_2$GdCu$_2$O$_8$ sample exhibits zero resistivity at a critical temperature of $T_c = 16 \text{ K}$. The precise value of $T_c$ varies between 12 and 24 K, depending on synthesis conditions, and may be raised to 40 K by long-term annealing. The temperature dependence of the normal-state resistivity is again characteristic of a strongly underdoped superconducting cuprate compound. The ferromagnetic transition at $T_{Curie}=133 \text{ K}$ causes only a small yet noticeable drop in the resistivity indicating that the RuO$_2$ layer is almost insulating above $T_{Curie}$ while being poorly conducting in the ferromagnetic state.

B. The technique of muon spin rotation

The muon spin rotation ($\mu$SR) experiments have been performed at the M15 beamline of TRIUMF in Vancouver, Canada, which provides 100% spin polarized muons. The $\mu$SR technique is especially suited for the study of magnetic materials and allows one to study the homogeneity of the magnetic state on a microscopic scale and also to access its volume fraction. The $\mu$SR technique typically covers a time window of $10^{-6}$ to $10^{-9}$ seconds and allows one to detect internal magnetic fields over a wide range of 0.1G to several Tesla. The 100% spin-polarised 'surface muons' ($E_\mu \approx 4.2 \text{ MeV}$) are implanted into the bulk of the sample where they thermalize very rapidly ($\sim 10^{-12} \text{s}$) without any noticeable loss in their initial spin polarization. Each muon stops at a well-defined interstitial lattice site and, for the perovskite compounds, forms a muoxyl bond with one of the oxygen atoms. The whole ensemble of muons is randomly distributed throughout a layer of 100-200 $\mu$m thickness and therefore probes a representative part of the sample volume. Each muon spin precesses in its local magnetic field $B_u$ with a precession frequency of, $\nu_\mu = (\gamma_\mu/2\pi) \cdot B_u$, where $\gamma_\mu/2\pi = 135.5 \text{ MHz/T}$ is the gyromagnetic ratio of the positive muon. The muon decays with a mean life time of $\tau_\mu \approx 2.2 \mu\text{s}^{-1}$ into two neutrinos and a positron which is preferentially emitted along the direction of the muon spin at the instant of decay. The time evolution of the spin polarization $P(t)$ of the muon ensemble can therefore be obtained via the time-resolved detection of the spatial asymmetry of the decay positron emission rate. More details regarding the zero-field (ZF) $\mu$SR technique are given below.

III. EXPERIMENTAL RESULTS

A. DC magnetization

Before we discuss the result of the $\mu$SR experiments, we first present some DC-magnetization data which establish that the RuSr$_2$GdCu$_2$O$_8$ sample exhibits a spontaneous magnetization at a ferromagnetic transition of $T_{Curie} = 133 \text{ K}$ and becomes superconducting at a much lower temperature $T_c = 16 \text{ K}$. Figure 3(a) shows the temperature dependence of the volume susceptibility, $\chi_V$, which has been obtained after zero-field cooling the sample to $T = 2 \text{ K}$, then applying an external field of $H_{ext} = 5.5 \text{ Oe}$, and subsequently warming up to $T = 200 \text{ K}$. The density of the sample has been assumed to be $\rho = 6.7 \text{ g/cm}^3$ corresponding to stoichiometric RuSr$_2$GdCu$_2$O$_8$ with lattice parameters of $a = 3.84$ Å and $c = 11.57$ Å. The superconducting transition is evident in Fig 3(a) from the onset of a pronounced diamagnetic shift below $T_c = 16 \text{ K}$. The diamagnetic shift at the lowest available temperature of $T = 2 \text{ K}$ corresponds to an almost complete diamagnetic shielding of the sample volume, implying that at least the surface region of the sample is homogeneously superconducting. In fact, all pieces that have been cut from the pellet exhibit a similarly large diamagnetic shielding effect (small differences can be attributed to different demagnetization factors).
Nevertheless, the DC-magnetisation measurements cannot give unambiguous evidence for the presence of bulk superconductivity since an almost complete diamagnetic shielding may also be caused by a filamentary structure of superconducting material in a small fraction of the otherwise non-superconducting material.

The field-cooled molar magnetization, $M_{\text{mol}}$, for applied fields of $H$ (low) field-cooled molar magnetization measurements. Figure 3(b) displays the specific heat measurements have been performed on the RuSr$_2$GdCu$_2$O$_8$ at $T_c=16$ K and $T_{\text{Curie}}=133$ K, respectively. (b) The field-cooled molar magnetization, $M_{\text{mol}}$, for applied fields of $H=5.5, 10, 100$ Oe.

We note however, that unequivocal evidence for the occurrence of bulk superconductivity in RuSr$_2$GdCu$_2$O$_8$ has recently been obtained from specific heat measurements which reveal a sizeable jump of $\Delta\gamma \approx 0.35$ mJ/g at.K$^2$ at $T_c$, comparable to or greater than that seen in other underdoped cuprates. For comparison in strongly underdoped YBa$_2$Cu$_3$O$_{7-\delta}$ it is found that $\Delta\gamma \approx 0.2 - 0.3$ mJ/g at.K$^2$. We also note that the specific heat measurements have been performed on the same samples which have been studied by $\mu$SR- and DC-magnetisation measurements. Figure 3(b) displays the (low) field-cooled molar magnetization $M_m$ for applied fields of $H^{\text{ext}}=5.5, 50$ and 100 Oe. The ferromagnetic transition at $T_{\text{Curie}}=133$ K is evident from the sudden onset of a spontaneous magnetization. Evidently, the magnetic order parameter has at least a sizeable ferromagnetic component and it persists almost unchanged to the lowest measured temperature $T=2$ K. In particular, it does not appear to weaken as superconductivity sets in at $T_c=16$ K. Additional evidence for the presence of ferromagnetic order is presented in Fig. 4, which shows that the isothermal magnetization loops at $T=5$ K and 50 K exhibit hysteretic magnetic behavior with a remanent magnetization $M_{\text{rem}} \approx 400$ Oe at 5 K and 200 Oe at 50 K.

Having established the existence of ferromagnetic order, the question arises of whether it involves the Ru-moments or the Gd-moments. In the following we present high-temperature susceptibility data which indicate that the ferromagnetic order involves only the Ru–moments, whereas the Gd-moments remain in the paramagnetic state below $T_{\text{Curie}}$. Figure 5 shows the inverse molar susceptibility, $1/\chi_m \approx (M_m/H^{\text{ext}})^{-1}$ obtained for different external fields in the range $5.5 \leq H^{\text{ext}} \leq 1000$ Oe (solid lines) in the temperature region 200 K$<T<400$ K. Shown by the plus signs (+) is the best fit to the experimental data using a two-component ‘Curie-Weiss + Curie-function’, $\chi = C_1/(T-\Theta) + C_2/T$, with $\Theta = T_{\text{Curie}}=133$ K kept fixed. This function describes the experimental data rather well and it gives us very reasonable values for the magnetic moments, with $\mu_1=1.05(5) \mu_B$ for the moments that order at $T_{\text{Curie}}$ and $\mu_2=7.4(1) \mu_B$ for the moments that remain paramagnetic below $T_{\text{Curie}}$. The magnetic moment of the paramagnetic component agrees reasonably well with the expected magnetic moment of Gd$^{3+}$ which for a free Gd$^{3+}$ ion is $\mu(\text{Gd}^{3+})=7.94 \mu_B$ and $\mu(\text{Gd}^{3+})=7.4 \mu_B$ for the structurally-similar GdB$_2$O$_{7-\delta}$ compound. On the other hand, the value of the Ru-moments with $\mu(\text{Ru})=1.05(5) \mu_B$ also appears to be reasonable. For Ru$^{5+}$ the number of 4d electrons is 3 and the free ion value of the magnetic moment is $3 \mu_B$ for the high spin state and $1 \mu_B$ for the low spin state. The experimentally observed value of $\mu(\text{Ru})=1.05(5) \mu_B$ therefore seems to imply that Ru$^{5+}$ is in the low spin state. Shown in the inset of Fig. 5 is the field-dependent magnetization for the temperatures $T=2, 30, 50, 100$ and 300 K. The low-temperature magnetization can be seen to saturate at a value of $\mu_{\text{sat}} \approx 8 \mu_B$, as may be expected for a system that contains one
Gd-moment per formula unit with $\mu(Gd) = 7 \mu_B$ plus one Ru-moment with $\mu(Ru) = 1 \mu_B$.

The idea that the Gd-moments do not participate in the ferromagnetic transition at $T_{Curie} = 133$ K is supported by the result of DC-magnetization measurements on the 10% Y ↔ Gd co substituted RuSr$_2$Gd$_{0.9}$Y$_{0.1}$Cu$_2$O$_8$. Figures 6(a) and 6(b) display the zero-field-cooled- and the field-cooled susceptibilities (dashed lines) and compare them with the corresponding data on the pure RuSr$_2$GdCu$_2$O$_8$ sample (solid line). It is evident that the ferromagnetic transition is not significantly affected by the partial substitution of non-magnetic Y$^{3+}$ for magnetic Gd$^{3+}$. Also shown in Fig. 6(a) and 6(b) by the dotted lines are the results for the Zn-substituted RuSr$_2$Cu$_{1.94}$Zn$_{0.06}$O$_8$ sample. The circumstance that the ferromagnetic order is not affected by the Zn-substitution supports our view that the majority of the Zn-impurities has been introduced into the CuO$_2$ layers while hardly any of them reside within the RuO$_2$ layers. Moreover, we infer from the rapid $T_c$-suppression upon Zn-substitution that only the CuO$_2$ layers host the superconducting charge carrier in RuSr$_2$GdCu$_2$O$_8$.

**B. Zero-field muon-spin-rotation (ZF-$\mu$SR)**

Next we discuss the result of the zero-field (ZF) $\mu$SR experiments. Figures 7(a) and 7(b) show representative ZF-$\mu$SR spectra for the evolution of the normalized time-resolved muon spin polarization $P(t)/P(0)$ at temperatures of $T = 5$ and 48 K.

The value of the initial muon spin polarization, $P(0)$, has been determined by a transverse field (TF) $\mu$SR experiment performed on the same sample at a temperature above $T_{Curie}$. In the ferromagnetic state performed at $T_{Curie} = 133$ K we find that the spectra are well described...
by the relaxation function:

\[ P(t)/P(t=0) = A_1 \cdot \exp(-\lambda t) \cdot \cos(2\pi \nu_\mu t) + A_2 \cdot \exp(-\lambda t), \]  

(1)

where \( \langle \nu_\mu \rangle \) is the average muon spin precession frequency which corresponds to the average value of the spontaneous internal magnetic field at the muons sites, \( \langle \nu_\mu \rangle = \gamma_\mu / 2\pi \cdot \langle B_\mu \rangle \), with \( \gamma_\mu = 83.4 \, \text{MHz/T} \) the gyromagnetic ratio. The damping rate of the non-oscillating (longitudinal) component, \( \lambda \), is proportional to the dynamic spin-lattice relaxation rate, \( \lambda \sim 1/T_1 \), whereas the relaxation rate of the oscillating (transverse) component, \( \lambda \), is dominated by the static distribution of the local magnetic field, i.e., \( \lambda \approx \gamma_\mu \cdot \langle B_\mu \rangle \). Figure 8 shows the temperature dependence of (a) the precession frequency, \( \langle \nu_\mu \rangle(T) \), (b) the transverse relaxation rate, \( \lambda(T) \), and (c) the longitudinal relaxation rate, \( \Lambda(T) \).

Before we discuss the ZF-\( \mu \)SR data in more detail, we first emphasize the most important implications, which are evident from Figs. 7 and 8. Firstly, the presence of an oscillating component in the ZF-\( \mu \)SR spectra for \( T < T_{\text{Curie}} = 133 \, \text{K} \) gives unambiguous evidence for an ordered magnetic state which is homogeneous on a microscopic length scale (of typically 20 \( \AA \)). Secondly, from the amplitude of the oscillating component (\( A_1 \approx 2/3 \)) we can deduce that the magnetically-ordered state accounts for more or less the entire volume of the sample. And thirdly, from the temperature dependence of the \( \mu \)SR signal it becomes clear that the magnetic order persists almost unchanged in the superconducting state.

1. The volume fraction of the magnetic phase

In the following we outline how the volume fraction of the magnetically-ordered phase is obtained from the amplitude of the oscillating component of the ZF-\( \mu \)SR spectra. For a polycrystalline sample with randomly-oriented grains in zero external field the local magnetic field, on average, is parallel (perpendicular) to the direction of the muon spin direction with probability 1/3 (2/3). For a homogeneous magnetically-ordered sample one therefore expects that 2/3 of the amplitude of the ZF-\( \mu \)SR signal (the transverse component) exhibit an oscillatory behavior, while 1/3 of the signal (the longitudinal component) is non-oscillating and only slowly damped due to spin-flip excitations. On the other hand, for a sample with inhomogeneous magnetic order, for example containing non-magnetic regions, the amplitude of the oscillating signal will be accordingly reduced and a second non-oscillating transverse component will appear. If the non-magnetic regions are microscopically small, this non-oscillating component is likely to have a rather large damping rate of the order of \( \lambda \sim \gamma_\mu \cdot \langle B_\mu \rangle \) due to stray fields which are imposed by the neighboring magnetic domains. From Fig. 7(a) and 7(b) it can be seen that the ZF-\( \mu \)SR data on RuSr\(_2\)GdCu\(_2\)O\(_8\) give no indication for such an inhomogeneous magnetic state. As was mentioned above, the amplitude of the initial muon spin polarization \( P(t = 0) \) has been determined from a transverse-field (TF)-\( \mu \)SR measurement. From the size of the amplitude of the oscillatory component we deduce that more than 80 % of the sample is magnetically ordered below \( T_{\text{Curie}} = 133 \, \text{K} \). Based on this analysis we estimate that the volume fraction of any disordered magnetic- or non-magnetic phase must be well below 20%. Note that some of the muons (typically 10-20 %) do not stop inside the sample but somewhere in the cryostat walls. In the ZF-\( \mu \)SR experiment these muons give rise to a missing fraction since their spin-polarization is much more slowly damped than for the rest of the signal. In the TF-\( \mu \)SR experiment, however, this very slowly damped component can be detected via its precession in the external field and it contributes to the to-
2. Local magnetic field at the muon site

It is evident from Fig. 8(a) that the muon spin precession frequency (the local field at the muon site) does not exhibit any strong anomaly at the superconducting transition temperature $T_c$. Instead, as shown by the dashed line, the temperature of the muon spin precession frequency ($\nu_\mu(T)$) and thus of the magnetic order parameter) is well described by the function $\nu_\mu(T) = \nu_0 (1 - T/T_c)^\beta$, with $\nu_0 = 9.7(1)$ MHz (corresponding to $\langle B_\mu/2 \rangle (T \to 0) \approx 720(10)$ G), $T_c = 133(1)$ K, and $\beta = 0.333(5)$). This functional form is strictly valid only in the critical regime close to $T_{Curie}$ but it can be seen to provide a reasonable description of the magnetic order parameter over a fairly wide temperature range of $T_{Curie} \geq T \geq 5$ K. The anomaly at very low temperature arises most likely from the magnetic ordering transition of the Gd-moments at $T_N \approx 2.6$ K. Note that for the structurally related compound GdBa$_2$Cu$_3$O$_{7-\delta}$ (Gd-123) the antiferromagnetic ordering transition of the Gd-moment occurs at a very similar temperature of $T_N = 2.3$ K [16,17]. The value of the critical exponent $\beta = 0.333$ is close to the theoretical value 0.345 in the 3D XY model. We cannot determine with certainty the number of components in the spin system with these data and, in particular, distinguish between the 2-component XY ($\beta = 0.345$) and the 3-component Heisenberg ($\beta = 0.365$) models. The contribution of ferromagnetic fluctuations above $T_{Curie}$ to the susceptibility provides better discrimination as will be discussed later.

The oscillating transverse component exhibits a damping rate of the order of $\lambda \approx 10-15 \mu$s$^{-1}$ corresponding to a spread in the local magnetic field of $\langle \Delta B_\mu \rangle / \langle B_\mu \rangle \approx 0.2$. This $20\%$ spread of the local magnetic field does not seem to agree with a scenario where the ferromagnetic order is assumed to exhibit a spiral modulation (with a wavelength shorter than the superconducting coherence length of typically 20 Å in the cuprates) and/or to be spatially inhomogeneous as in ErRh$_2$B$_4$, HoMo$_6$S$_8$, and Y$_3$Co$_4$. Instead, we emphasize that the observed spread in the local magnetic field can be accounted for by the grain boundary effects and by the differences in the demagnetization factors of the individual grains which naturally arise for a polycrystalline sample that has a very small average grain size of about $1 \mu$m. Also, we point out that recent transmission-electron-microscopy (TEM) studies have revealed that our present Ru-1212 sample contains [100] rotation twins and also exhibits some cationic disorder due to the intermixing of Sr ↔ Gd and to a lesser extent of Ru ↔ Cu. These kinds of structural imperfections certainly tend to further increase the transverse relaxation rate $\lambda$ of the ZF-$\mu$SR spectra. Meanwhile, we have prepared Ru-1212 samples which are structurally more perfect (by sintering at slightly higher temperature and for longer periods). Recent DC-magnetization measurements have shown that these crystallographic defects do not affect the fundamental magnetic and superconducting behavior. In fact, both the superconducting- and the ferromagnetic transitions become somewhat sharper and $T_c$ and $T_{Curie}$ are slightly increased for these structurally more perfect samples. Additional $\mu$SR measurements on these samples are presently under way.

3. Longitudinal relaxation rate, $\Lambda \sim 1/T_1$

The temperature dependence of the relaxation rate of the non-oscillating component of the ZF-$\mu$SR signal, $A(T) \sim 1/T_1$, is shown in Fig. 8(c). As a function of decreasing temperature $A(T)$ can be seen to exhibit a cusp-like feature at the ferromagnetic transition of the Ru-moments at $T_{Curie} = 133$ K and a step-like increase at very low temperature which most likely is related to the ordering of the Gd moments. The cusp feature at $T_{Curie} = 133$ K characterizes the slowing down of the spin dynamics of the Ru-moments as the ferromagnetic transition is approached. The cusp maximum occurs when the spin fluctuation rate, $\tau_c$, equals the typical $\mu$SR time scale for $\tau_c \approx 10^{-15}$ s. Note, that in the ferromagnetically-ordered state the longitudinal relaxation rate remains unusually large with values of $A(T < T_{Curie}) \approx 0.3-0.4 \mu$s$^{-1}$ that are at least an order of magnitude larger than expected for a classical ferromagnet (where two-magnon excitations provide the major contribution to spin dynamics). We have confirmed by a $\mu$SR measurements in a longitudinal field of $H^{LF} = 6$ kOe that this large relaxation rate is indeed characteristic for the longitudinal component of the $\mu$SR signal. At present we cannot provide a definite explanation of the origin of the unusually large value of $\Lambda$. However, we emphasize that the Ru$_2$(Gd,Co)$_2$Cu$_3$O$_8$ system can be expected to exhibit a rather complex magnetic behavior since, besides the ferromagnetically-ordered Ru moments, it also contains the larger Gd moments with $\mu(Gd^{3+}) \approx 7.4 \mu_B$ which remain paramagnetic below $T_{Curie}$. The magnetic ordering transition of the Gd moments at $T\approx 2.6$ K is evident in the ZF-$\mu$SR data in Fig. 8(a)-(c) from the sudden increase in the local magnetic field (or the $\mu$SR precession frequency, $\langle \nu_\mu \rangle$) and a corresponding increase in both relaxation rates, $\lambda$ and $\Lambda$. In addition, we note...
that recently it has been shown by $\mu$SR measurements that in strongly underdoped high-$T_c$ cuprate superconductors (like the present RuSr$_2$GdCu$_2$O$_8$ compound) also the Cu moments exhibit a spin-glass type freezing transition at low temperature. Finally, it appears that the longitudinal relaxation rate $\Lambda$ exhibits an additional weak anomaly at a temperature of $T\approx 20$ K, i.e. in the vicinity of the superconducting transition at $T_c=16$ K. At present we are not sure whether this effect is related to the onset of superconductivity. From Fig. 8(b) it appears that the transverse relaxation rate also exhibits a steplike increase in the same temperature range. The local magnetic field at the muon site, however, (see Fig. 8a) does not seem to exhibit any anomaly in the vicinity of $T_c$. We expect that further $\mu$SR measurements on rare earth (RE) substituted RuSr$_2$Gd$_{1-x}$RE$_x$Cu$_2$O$_8$ samples, as well as on less strongly underdoped samples with higher critical temperatures of $T_c$ up to 40 K, should shed more light on the complex magnetic behavior and its interplay with superconductivity in the Ru-1212 system.

C. Dipolar Field Calculation

While the ZF-$\mu$SR data give clear evidence for the presence of a homogeneous magnetically ordered state, they do not provide any direct information about the origin of the magnetic moments, the type of the magnetic order and its direction. Based on dipolar-field calculations of the local magnetic field at the muon site, however, one can test the consistency with an assumed magnetic structure. The result of these calculations depends on the location of the interstitial muon site and also on the orientation of the Ru-moments. Unfortunately, for the Ru-1212 system neither of these is accurately known at present. Nevertheless, it seems plausible that the muon site is similar to that in YBa$_2$Cu$_3$O$_{7-\delta}$ (and other related cuprate compounds) where the positive muon forms a hydroxyl bond with the apex oxygen and is located at the so-called ‘apical-site’ near the point $(0.12a,0.22b,0.14c)$. Indeed, as is summarized in table 1, we obtain rather good agreement with the experimental value of $\langle B_\mu \rangle (T=0) = 720$ G if we take a similar apical-site near the point $(0.13a,0.22b,0.16-0.17c)$ and assume that the ferromagnetically-ordered Ru-moments ($\mu$(Ru)=1 $\mu_B$) are oriented along the RuO$_2$ plane either along the Ru-O bond, [100], or along the diagonal [110] (see table 1). For the [110] orientation, however, there exist two magnetically inequivalent muon sites which should give rise to two distinct precession frequencies in the $\mu$SR spectra (which are not observed experimentally). For the Ru-moments oriented perpendicular to the RuO$_2$ layer along [001] the resulting local magnetic field at the apex-site is significantly larger than the experimental value. In order to obtain reasonable agreement with experiment for the [001]-orientation one has to assume that the muon site is located much closer to the CuO$_2$ planes. Such a muon site, however, is not very realistic (simply speaking the positive muon is repelled by the positively charged CuO$_2$ planes) and has not been observed in any of the related cuprate compounds. We thus tentatively conclude that the moments align in-plane consistent with the 2-component XY scenario. While this result is rather convenient in terms of the coexistence of the ferromagnetic order of the Ru moments and the superconductivity which resides within the CuO$_2$ layers as discussed below, one has to keep in mind that the underlying assumptions are rather crude. For more detailed and decisive information on the structure and the orientation of the Ru spin order we must await the result of neutron scattering experiments.

IV. A POSSIBLE SCENARIO FOR COEXISTENCE OF FERROMAGNETIC- AND SUPERCONDUCTING ORDER

Having established that the ferromagnetic and the superconducting order parameter coexist on a microscopic scale, we arrive at the important question as to how this system manages to avoid strong pair-breaking effects. We suspect that the answer is closely related to the layered structure of the hybrid ruthenate-cuprate compound and, in particular, to the purely two-dimensional coherent charge transport in the strongly underdoped CuO$_2$ planes. We envisage a scenario where the ferromagnetically ordered Ru spins are aligned in the RuO$_2$ planes. We thus tentatively conclude that the moments align in-plane consistent with the 2-component XY scenario. While this result is rather convenient in terms of the coexistence of the ferromagnetic order of the Ru moments and the superconductivity which resides within the CuO$_2$ layers as discussed below, one has to keep in mind that the underlying assumptions are rather crude. For more detailed and decisive information on the structure and the orientation of the Ru spin order we must await the result of neutron scattering experiments.
for $T > T_{Curie}$. The slope of $-2.30(3)$ indicates a critical exponent of $\gamma = 1.30(3)$ consistent with 3D XY fluctuations for which $\gamma = 1.32^{18}$ and again consistent with orientation of the Ru-moments within the a-b plane.

Finally, we note that an alternative (and highly speculative) explanation for the coexistence of high-$T_c$ superconductivity and ferromagnetic order in the present Ru-1212 superconductor could be that the superconducting order parameter has a non-zero angular momentum which itself breaks time-reversal symmetry. Such a highly unconventional order parameter symmetry has been discussed also in the context of the Sr$_2$RuO$_4$ superconductor. We point out, however, that at present we have no evidence in favor of such a scenario.

V. CONCLUSIONS

In summary, we have performed DC-magnetization and zero-field muon spin rotation (ZF-$\mu$SR) measurements which characterize the superconducting- and the magnetic properties of the hybrid cuprate-ruthenate compound RuSr$_2$GdCu$_2$O$_8$. The DC-magnetization data establish that this material exhibits ferromagnetic order (or at least magnetic order with a sizeable ferromagnetic component) below $T_{Curie} = 133$ K and becomes superconducting at a much lower temperature of $T_c = 16$ K. We obtain evidence that superconducting charge carriers originate from the CuO$_2$ planes, while the ferromagnetic order is associated with the Ru moments with $\mu$(Ru) $\approx 1$ $\mu_B$. The larger Gd moments with $\mu$(Gd) $\approx 7.4$ $\mu_B$ do not appear to participate in the ferromagnetic transition but remain paramagnetic to very low temperature and undergo most likely an antiferromagnetic transition at $T_N$=2.6 K. The ZF-$\mu$SR experiments provide evidence that the ferromagnetic phase is homogeneous on a microscopic scale and accounts for most of the sample volume. Furthermore, they indicate that the magnetically ordered state is not significantly modified by the onset of superconductivity. This rather surprising result raises the question as to how ferromagnetic and superconducting order can coexist on a microscopic scale while avoiding strong pairbreaking effects that tend to destroy superconductivity. We have outlined a possible scenario which relies on the two-dimensional charge dynamics of the CuO$_2$ planes and the assumption that the ferromagnetic order parameter of the Ru-moments is confined to the RuO$_2$ layers.

VI. ACKNOWLEDGMENTS

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* Permanent address: Institute of Experimental Physics, Warsaw University, Hoża 69, 00-681 Warsaw, Poland; # Present address: 1318 Tenth St., Saskatoon, Canada S7H 0J3

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Table 1: The local magnetic field at the muon site, \(\langle B_\mu \rangle\), obtained from the dipolar-field calculation. The results are shown for two different muon sites and for three different orientations with the Ru moments (\(\mu(\text{Ru}^{5+})=1 \mu_B\)) ferromagnetically ordered along the Ru-O bond [100], along the diagonal [110], or perpendicular to the RuO\(_2\) planes [001]. Note that for the [110] orientation there exist two magnetically inequivalent muon sites.

| muon-site | [001] |
|-----------|-------|
| (0.13, 0.225, 0.16) | 1060 G, 1068/805 G, 1471 G |
| (0.128, 0.222, 0.175) | 740 G, 824/665 G, 1231 G |