Critical fields and spontaneous vortex state in the weak-ferromagnetic superconductor
RuSr$_2$GdCu$_2$O$_8$

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A spontaneous vortex state (SVS) between 30 K and 56 K was observed for the weak-ferromagnetic superconductor RuSr$_2$GdCu$_2$O$_8$ with ferromagnetic Curie temperature $T_C = 131$ K and superconducting transition temperature $T_c = 56$ K. The low field (±20 G) superconducting hysteresis loop indicates a narrow Meissner state region within average lower critical field $B_{c1}(T) = B_{c1}(0)[1 - (T/T_0)^2]$, with average $B_{c1}^0(0) = 12$ G and $T_0 = 30$ K. Full Meissner shielding signal in very low applied field indicates an ab-plane $B_{c1}^0(0) \sim 4$ G with an estimated anisotropic parameter $\gamma \sim 7$ for this layered system. The existence of a spontaneous vortex state between 30 K and 56 K is the result of weak-ferromagnetic order with a net spontaneous magnetic moment of $\sim 0.1 \mu_B$ per Ru, which generates a weak magnetic dipole field around 10 G in the CuO$_2$ bi-layers. The upper critical field $B_{c2}$ varies linearly as $(1 - T/T_c)$ up to 7-T field. The vortex melting line $B_m$ varies as $(1 - T/T_m)^{3.5}$ with melting transition temperature $T_m = 39$ K and a very broad vortex liquid region due to the coexistence and the interplay between superconductivity and weak-ferromagnetic order.

I. INTRODUCTION

Recently, high-$T_c$ superconductivity with anomalous magnetic properties was reported in the weak-ferromagnetic superconductor Ru$_8$GdCu$_2$O$_8$ (99.99 %), SrCO$_3$ (99.99 %), Gd$_2$O$_3$ (99.99 %), and CuO (99.99 %) preheated powders with the nominal composition ratio of Ru:Gd:Cu = 1: 1:2. The occurrence of high-$T_c$ superconductivity with Ru$^{5+}$ moment $\mu$ canted along the tetragonal basal plane resulting a small net spontaneous magnetic moment $\mu_s \ll \mu(Ru^{5+})$ too small to be detected in neutron diffraction. The Curie temperature $T_C \sim 130$ K observed from magnetization measurement in the prototype compound Ru$_8$GdCu$_2$O$_8$ is probably a canted G-type antiferromagnetic order with Ru$^{5+}$ moment $\mu$ canted at 45° from the tetragonal basal plane resulting a small net spontaneous magnetic moment $\mu_s \ll \mu(Ru^{5+})$. The occurrence of high-$T_c$ superconductivity with Ru$^{5+}$ moment $\mu$ canted along the tetragonal basal plane resulting a small net spontaneous magnetic moment $\mu_s \ll \mu(Ru^{5+})$ too small to be detected in neutron diffraction. 

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II. EXPERIMENTAL

The stoichiometric Ru$_8$GdCu$_2$O$_8$ samples were synthesized by the standard solid-state reaction method. High-purity RuO$_2$ (99.99 %), SrCO$_3$ (99.99 %), Gd$_2$O$_3$ (99.99 %), and CuO (99.99 %) preheated powders with the nominal composition ratio of Ru:Gd:Cu = 1: 1:2. The occurrence of high-$T_c$ superconductivity with Ru$^{5+}$ moment $\mu$ canted along the tetragonal basal plane resulting a small net spontaneous magnetic moment $\mu_s \ll \mu(Ru^{5+})$ too small to be detected in neutron diffraction. 

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III. RESULTS AND DISCUSSION

The powder x-ray diffraction pattern for the oxygen-annealed RuSr₂GdCu₂O₈ polycrystalline sample indicates close to single phase with the tetragonal lattice parameters of \( a = 0.5428(5) \) nm and \( c = 1.1589(9) \) nm. The space group \( P4/mbm \) is used for Rietveld refinement analysis, where neutron diffraction data indicate that a RuO₆ octahedra \( 14^\circ \) rotation around the \( c \)-axis is needed to accommodate physically reasonable Ru-O bond lengths.\(^{10}\) The refinement gives a good residual error \( R \) of 3.64 \%, weighted pattern error \( R_{wp} = 6.07 \% \), and Bragg error \( R_B = 5.95 \% \).

The temperature dependence of the electrical resistivity \( \rho(T) \) and the volume magnetic susceptibility \( \chi_V(T) \) at 1-G field-cooled (FC) and zero-field-cooled (ZFC) modes for RuSr₂GdCu₂O₈ are shown collectively in Fig. 1. The high temperature resistivity decreases monotonically from room temperature value of 9.2 mΩ·cm (not shown) to 6.4 mΩ·cm at 200 K, and extrapolated to 2.8 mΩ·cm at 0 K with a good resistivity ratio \( \rho(300 K)/\rho(0 \ K) \) of 3.3 for the polycrystalline sample. High temperature resistivity shows a non-Fermi-liquid-like linear T-dependence down to Curie temperature \( T_C \) of 131 K, then changes to a \( T^2 \) behavior below \( T_C \) due to magnetic order.

The superconducting onset temperature of 56 K is determined from the deviation from the \( T^2 \) behavior, with a zero resistivity \( T_{\rho(0)} \) at 39 K. The broad transition width \( \Delta T \) at 17 K observed is the common feature for all reported Ru-1212 resistivity data, which indicates that the superconducting Josephson coupling along the tetragonal \( c \)-axis between Cu-O bi-layers may be partially blocked by the dipole field \( B_{dip} \) of ordered Ru moments in the Ru-O layer.\(^{13,4,5,29,38}\) The diamagnetic \( T_C \) at 39 K was observed in the 1-G ZFC susceptibility measurement. The full Meissner shielding signal \( 4\pi\chi_V \) in an applied field \( B_a \sim -1.5 \) (Gaussian units) was recorded at 5 K. This value is identical to the Meissner shielding signal expected for a superconducting sphere with a demagnetization factor \( N \) of -4\( \pi /3 \) and in an applied field \( B_a \) well below lower critical field \( B_{c1} \). The large diamagnetic signal in 1-G ZFC mode is the best data observed so far from various reported susceptibility measurement techniques.\(^{15,28,29,38}\) Since our measurements were performed with the standard moving-sample SQUID magnetometer, it is clear that sample quality is more crucial than measuring techniques. Both ZFC and FC data reveal a Curie temperature \( T_C \) of 131 K. However, in 1-G FC mode, no diamagnetic field-expulsion signal can be detected below 39 K due to strong flux pinning where superconductivity coexists with weak-ferromagnetic order.

The zero-field-cooled (ZFC) volume susceptibility \( \chi_V(T) \) at 1 G, 10 G, and 100 G applied fields are shown collectively in Fig. 2. All data show the same magnetic order \( T_C(Ru) \) of 131 K. Although the diamagnetic \( T_C \) of 39 K was still observed at 10-G ZFC measurement, the diamagnetic signal at 5 K is reduced to 60% of the full Meissner signal. Consider the polycrystalline nature of sample with varying microcrystallite size and orientation, the average superconducting lower critical field \( B_{c1} \) at 5

![Graph 1: Electrical resistivity \( \rho(T) \) and volume magnetic susceptibility \( \chi_V(T) \) at 1-G field-cooled (FC) and zero-field-cooled (ZFC) modes for oxygen-annealed RuSr₂GdCu₂O₈.](image1)

![Graph 2: ZFC volume susceptibility \( \chi_V(T) \) for RuSr₂GdCu₂O₈ at 1 G, 10 G, and 100 G. Note that the full Meissner shielding signal was observed only at low applied field and low temperature.](image2)
K is estimated to be close to 10 G. No net diamagnetic signal can be detected at 100-G ZFC mode where the sample is already in the vortex glass or lattice state and the small diamagnetic signal is overshadowed by a large weak-ferromagnetic background.

Based on this information, the low-field (±20 G) isothermal superconducting hysteresis loops M-B are measured and collectively shown in Fig. 3(a) (5 K, 10 K, 15 K, and 20 K) and 3(b) (25 K, 30 K, and 35 K). The initial magnetization curve deviates from straight line in 4 G at 5 K, 3.5 G at 10 K, 3 G at 15 K, 2 G at 20 K, and 1 G at 25 K. This is the narrow region that full Meissner signals are detected and is roughly corresponding to the anisotropic lower critical field in the ab-plane B_{c1}^{ab}(T) with B_{c1}^{ab}(0) = 4 G. The average lower critical field B_{c1}^{ave} for polycrystalline sample is determined from the peaks of initial diamagnetic magnetization curves. B_{c1} decreases steadily from 12 G at 5 K, 11 G at 10 K, 9 G at 15 K, 6 G at 20 K, 3 G at 25 K and below 1 G at 30 K. A simple empirical parabolic fitting gives B_{c1}(T) = B_{c1}(0)[1 - (T/T_0)^2], with average B_{c1}^{ave}(0) = 12 G and T_0 = 30 K (see Fig. 4). Using the anisotropic Ginzburg-Landau formula B_{c1}^{ave} = [2B_{c1}^{ab} + B_{c1}^{c}]/3, c-axis B_{c1}^{c} ∼ 28 G and anisotropy parameter γ ∼ 7 is estimated. This value is close to reported anisotropic γ-value for YBa$_2$Cu$_3$O$_7$ where the 123-type structure can be written as Cu-1212 CuBa$_2$YCu$_2$O$_7$. An average penetration depth λ_{ave}(0) = [Φ_0/2πB_{c1}^{ave}(0)]^{1/2} of 520 nm was derived with estimated λ_{c}(0) = 340 nm and λ_{c}(0) = 2400 nm from B_{c1}^{ab} = Φ_0/2πλ_{c}^2 and B_{c1}^{ab} = Φ_0/2πλ_{ab}λ_{c}, where Φ_0 is the flux quantum.

Since T_0 = 30 K is well below T_c(onset) = 56 K and T_c(zero) = 39 K in zero applied field, a spontaneous vortex state (SVS) indeed exists between 30 K and 56 K. The low field phase diagram B_{c1}(T) for polycrystalline sample is shown in Fig. 4, with the average B_{c1}(T) separates the Meissner state from the vortex state and a smaller B_{c1}^{ab}(T) inside the Meissner region for reference. T_c(zero) = 39 K in the broad resistive transition is the onset of vortex pinning by driving current. This temperature is very close to the melting transition temperature T_m from the spontaneous vortex glass or lattice state to the spontaneous liquid state due to nonzero dipole field B_{dip} of weak-ferromagnetic order. The upper critical field B_{c2} defined from T_c(onset) and the vortex melting field B_{c2}(T) defined from T_c(zero) are temperature independent for small applied fields below 20 G. The internal dipole field generated by a weak-ferromagnetic order can be estimated using a simple extrapolation [B_{c2}(0) + B_{dip}]/B_{c2}(0) = T_c/T_0 = 56 K/30 K, which results with a dipole field B_{dip} = 10.4 G on the CuO$_2$ bi-layers. A small net spontaneous magnetic moment μ_s of ~0.11 μ_B per Ru is estimated using B_{dip} ∼ 2μ_s/d^3 with d = c/2 = 0.58 nm is the distance between midpoint of CuO$_2$ bi-layers and two nearest-neighbor Ru moments. If the weak-ferromagnetic structure is a canted G-type antiferromagnetic order with Ru moments μ = 1.5 μ_B for Ru$_3^{5+}$ in t$_{2g}$ states canted along the tetragonal basal plane, the small net spontaneous magnetic moment gives a canting angle of 4° from the tetragonal c-axis and is difficult to be detected in neutron diffraction with a resolution around 0.1 μ_B.

At 5 K, the shape of superconducting hysteresis loop with a large remanent molar magnetization M_r of 83 G.cm$^{-3}$/mol indicates strong pinning as well as a good indication of bulk nature of superconductivity for the oxygen-annealed sample. The remanent M_r decreases.
to 4 G.cm$^{-3}$/mol at 30 K and 1 G.cm$^{-3}$/mol at 35 K, where a weak-ferromagnetic background can be clearly seen. Fluctuation in the hysteresis loop is probably also related to the weak-ferromagnetic order.

To study the high-field effect on superconductivity, the magnetoresistivity $\rho(T,B_a)$ for RuSr$_2$GdCu$_2$O$_8$ up to 7 T are collectively shown in Fig. 5. The broadening of resistive transition in magnetic fields is the common features for all high-$T_c$ cuprate superconductors. The normal state resistivity is field independent and follows a $T^2$-dependence below $T_c$, with superconducting $T_c$ (onset) of 56 K in zero field decreases slightly to 53 K in 7-T field. The temperature dependence of upper critical field $B_{c2}(T)$ can be fitted with a linear function $B_{c2}(0)[1 - T/T_c]$ with average $B_{c2}(0) = 133$ T. An average coherence length $\xi_c = \sqrt{\Phi_0/(2\pi B_{c2}^2)}$ of 0.5 nm with the Ginzburg-Landau parameter $\kappa$ of 1040 and the thermodynamic critical field $B_{c1}(0) = (B_{c1}B_{c2})^{1/2} = 0.32$ T. No anisotropic $\xi_\perp$ and $\xi_\parallel$ values can be estimated from present data. The $T_c$ (zero) decreases from 39 K in zero applied field to 32 K in 1-kG, 28 K in 5-kG, 25 K in 1-T, 22 K in 2-T, 19 K in 3-T, 17 K in 4-T, 16 K in 5-T, 15 K in 6-T, and 14 K in 7-T field. If the zero resistivity is taken as the lower bound of the vortex melting temperature $T_m$, then the temperature dependence of the vortex melting transition line $B_m(T)$ can be fitted roughly by the formula $B_m(T) = B_m(0)[1 - T/T_m]^{3.5}$ with $B_m(0) = 35$ T and large exponent 3.5. In the lower field region, $B_m(T)$ rises as $[1 - T/T_m]^2$ as predicted by the mean-field approximation for temperature near $T_m = 39$ K. The full phase diagram $B_a(T)$ of RuSr$_2$GdCu$_2$O$_8$ is shown in Fig. 6 to exhibit both high field and low field features. The very broad vortex liquid region with $\Delta T = 17$ K in zero field and $\Delta T = 42$ K in 7-T field is extraordinary and is most likely originated from the coexistence and the interplay between superconductivity and weak-ferromagnetic order. This magnetic order is so weak that superconductivity can coexist with the magnetic order, but the effect of a weak spontaneous magnetic moment

![FIG. 5: Temperature dependence of magnetoresistivity $\rho(T, B_a)$ for RuSr$_2$GdCu$_2$O$_8$ in applied field up to 7 T.](image1)

![FIG. 6: Full phase diagram $B_a(T)$ of RuSr$_2$GdCu$_2$O$_8$.](image2)

![FIG. 7: Field dependence of magnetoresistivity $\rho(B_a)$ for RuSr$_2$GdCu$_2$O$_8$ in the vortex state at 20 K, 30 K, and 40 K.](image3)
with 93 K for YBa$_2$Cu$_3$O$_7$ or 103 K for TiBa$_2$CaCu$_2$O$_7$. The depression of $T_c$ by small spontaneous magnetic moment can be partially recovered by substitution of nonmagnetic Cu ions at Ru site. For example, in the Ru$_{1-x}$Cu$_x$Sr$_2$GdCu$_2$O$_8$ system, $T_c$ on set up to 65 K for $x = 0.1$ and 72 K for $x = 0.4$ was reported.

IV. CONCLUSION

The lower critical field with $B_{c1}(0) = 12$ G and $T_c = 30$ K indicates the existence of a spontaneous vortex state (SVS) between 30 K and $T_c$ of 56 K. This SVS state is closely related with the weak-ferromagnetic order

with a net spontaneous magnetic moment of $\sim 0.1 \mu_B$ per Ru. The broad vortex liquid region observed above vortex melting line $B_m(T)$ is also due to the coexistence and the interplay between superconductivity and weak-ferromagnetic order.

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