Long-range magnetic order of the Er ions in $Er_2Ba_4Cu_7O_{14.92}$

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Neutron-diffraction investigations at temperatures down to 25 mK were performed for the high-temperature superconductor $Er_2Ba_4Cu_7O_{14.92}$ (Er-247), $T_c=89$ K on a polycrystalline sample prepared by a sol-gel technique. Below $T_N=0.54$ K, a three-dimensional antiferromagnetic ordering of the Er ions corresponding to a propagation vector $\mathbf{k}=(1/2,0,0)$ and a finite correlation length of about 130 Å along the c axis have been found. The magnetic Er moments are oriented parallel to the b axis. This work proves that Er-247 shows a long-range magnetic ordering. [S0163-1829(97)52518-5]

I. INTRODUCTION

The compounds of the $R_2Ba_4Cu_{6+n}O_{14+n-\delta}$ ($R=$most rare earth elements and Y; $n=0$, 1, 2; $0\leq\delta\leq 1$) family have closely related structures, containing superconducting CuO_2 planes and single and/or double Cu-O chains. The structure of the R-247 ($n=1$) can be described as composed of alternating blocks containing single and double chains forming a -123-124-123-124-stacking sequence. The single chains can be oxygen depleted as in the R-123 ($n=0$) phase, but the R-247 compound remains superconducting due to the presence of stoichiometric R-124 ($n=2$) blocks. The maximum $T_c=95$ K can be only reached in fully oxidized samples without stacking faults.[1]

The coexistence of high-temperature superconductivity and magnetism in the $R_2Ba_4Cu_{6+n}O_{14+n-\delta}$ family makes these compounds of special interest. The magnetic structures of the R-123 and R-124 phases have been studied extensively, e.g., Refs. 2 and 3, while little is known about the magnetic structures of R-247 compounds. Although the large separation between the R ions along the c axis in R-124 should favor two-dimensional antiferromagnetic ordering, also three-dimensional ordering has been found in, e.g., Er-124, Ho-124, and in R-123 compounds. To our knowledge only the magnetic structure of Dy-247 ($\delta\approx 0$) with $T_c=60$ K has been studied and a magnetic bilayer coupling has been found (antiferromagnetic in the (a,b) plane, ferromagnetic along the c direction).[4] We conclude that this Dy-247 sample ($\delta\approx 0$) contains a large amount of stacking faults since it has a $T_c\approx 60$ K which is 35 K below the highest possible value.

It is known that the superconducting transition temperature T_c in fully oxidized 247 samples is correlated to the number of stacking faults (Ref. 1). Er-247 was expected to be the only candidate among the $R_2Ba_4Cu_7O_{15-\delta}$ compounds for a three-dimensional Er ordering at low temperature.[4]

In this work we present neutron diffraction results on antiferromagnetic Er ordering on a $Er_2Ba_4Cu_7O_{14.92}$ sample with nearly maximum $T_c=89$ K.

II. EXPERIMENTAL DETAILS

A polycrystalline sample with a nominal composition $Er_2Ba_4Cu_7O_{15-\delta}$ ($\delta=0.08$) was prepared by polymerized complex synthesis.[5] The phase purity of our sample was examined by x-ray and neutron powder diffraction, where no impurities could be detected. The neutron diffraction experiment was performed on the powder diffractometer D1A ($\lambda=2.2776$ Å) at the high flux reactor of the Institute Laue-Langevin (ILL) in Grenoble, France. A bottom loading $^4He/^3He$ dilution refrigerator has been used, capable to reach 25 mK. In order to achieve a good thermal contact we used a special sample container made of copper with an Ag heat exchanger and condensed liquid helium into the sample voids. The neutron powder diffraction patterns were recorded at 25 mK, where we observed magnetic scattering, and at 3 K in the paramagnetic state. The oxygen content was determined both, by a very sensitive volumetric method[6] as well as by neutron diffraction.

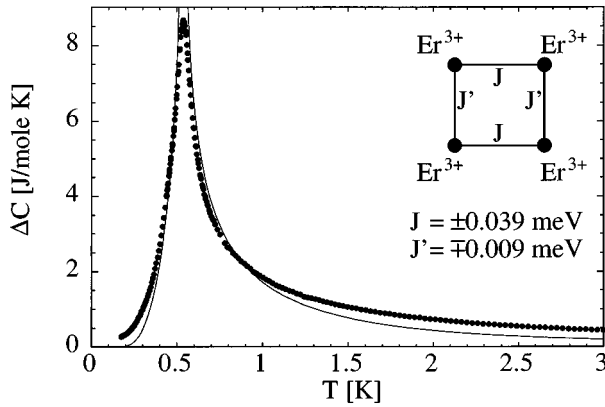For the characterization of superconducting properties, the dc magnetic susceptibility was measured by a Quantum

FIG. 1. Magnetic specific heat for $Er_2Ba_4Cu_7O_{14.92}$. The line is a fit with the anisotropic 2D-Ising model to the data using Onsager's solution (Ref. 10). While the absolute values of J and J' can be extracted from this model, their signs are indistinguishable. The opposite signs of the resulting coupling parameters J and J' in the (a,b) plane originate from the magnetic structure determined by neutron diffraction.

Design superconducting quantum interference device (SQUID) magnetometer for an applied external field of 0.01 T. ZFC (zero-field cooled) and FC (field cooled) susceptibility data were collected on fine powdered samples, which were fixed with paraffin to prevent reorientation.

In order to observe a magnetic phase transition specific heat experiments between 90 mK and 3 K were carried out by a semiadiabatic heat pulse method using a dilution refrigerator.

III. RESULTS AND DISCUSSION

Low-temperature specific-heat versus temperature data of $Er_2Ba_4Cu_7O_{14.92}$, which are shown in Fig. 1, indicate a magnetic ordering at the Néel temperature $T_N=0.54$ K. Our data can be interpreted consistently with an anisotropic two-dimensional (2D) Ising model (line in Fig. 1). Thus, we regard the phase transition at T_N as two-dimensional in character. The entropy gain associated with the magnetic ordering effect is displayed in Fig. 2. The magnetic entropy saturates at the expected value of R ln2 corresponding to the
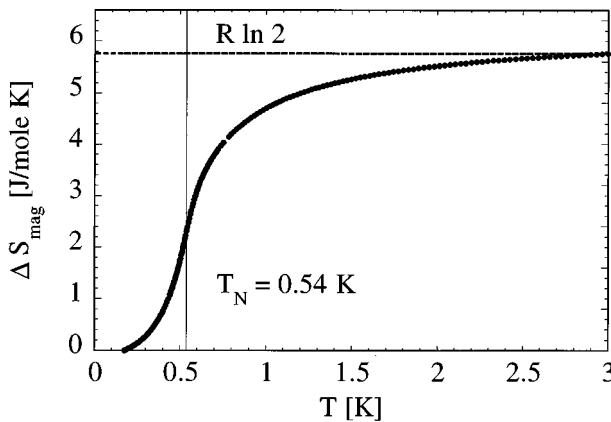

FIG. 2. Temperature dependence of the magnetic entropy for $Er_2Ba_4Cu_7O_{14.92}$.
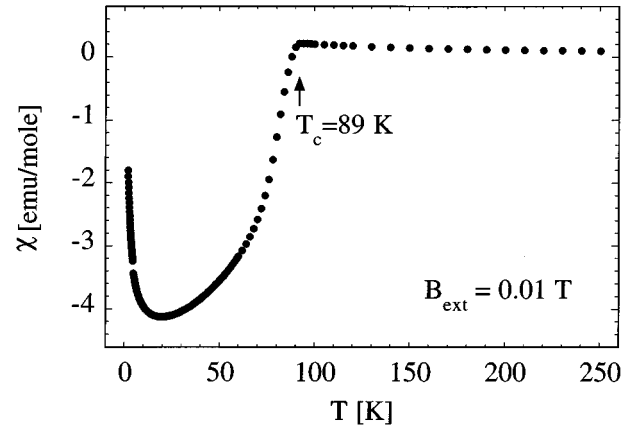

FIG. 3. Magnetic susceptibility data versus temperature for $Er_2Ba_4Cu_7O_{14.92}$ measured in the zero-field cooling mode in an applied external field of 0.01 T. The onset of superconductivity is at $T_c=89$ K (indicated by an arrow).

molar entropy of the doublet ground state of Er.

The superconducting transition temperature has been determined in a magnetic susceptibility measurement (Fig. 3). Our $Er_2Ba_4Cu_7O_{15-\delta}$ ($\delta=0.08$) sample shows a $T_c=89$ K which is close to 95 K, the highest value possible for a fully oxidized 247 sample ($\delta=0$). Recently, it has been found that the superconducting transition temperature T_c in fully oxidized 247 samples is correlated with the number of stacking faults.[1] Therefore, we conclude that our sample contains a very small number of stacking faults in the 123-124-123-124-sequence that is building up the structure. These stacking faults—if they are present at all—will be reflected in the magnetic ordering of the Er ions.

The chemical structure of $Er_2Ba_4Cu_7O_{14.92}$ has been refined in the space group $Ammm$. The refined lattice constants are $a=3.8263(2)$ Å, $b=3.8663(2)$ Å, and $c=50.3683(59)$ Å. The Thompson-Cox-Hastings pseudo-Voigt function was used to describe the profile and the full width at half maximum of the Bragg reflections. The best fit was achieved by introducing an anisotropic size model $Y \times \cos\phi$, where Y is the size parameter and ϕ is the angle between the scattering vector and the normal to the platelets which was assumed to be [001]. This model allows a good description of line broadening effects. The refinement yielded the size parameters $Y=0.407(22)$ for the chemical structure and $Y=1.366(27)$ for the magnetic structure.

The magnetic structure was determined at 25 mK. Figure 4 shows the magnetic diffraction pattern for $Er_2Ba_4Cu_7O_{14.92}$, obtained by subtracting the scattering observed above T_N, measured at 3 K, from the data observed at $T=25$ mK. A three-dimensional configuration for the magnetic Er moments inferred from the refinement of the magnetic diffraction pattern is shown schematically as an inset in Fig. 4. The refinement was performed using the program FULLPROF,[7] which allows the simultaneous treatment of several chemical and magnetic phases. The magnetic cell can be described by the propagation vector $\mathbf{k}=(1/2,0,0)$, i.e., the magnetic unit cell is doubled along the a direction compared to the chemical unit cell. The magnetic structure consists of chains of parallel (ferromagnetic) Er moments along the b direction with adjacent chains in the (a,b) plane coupled
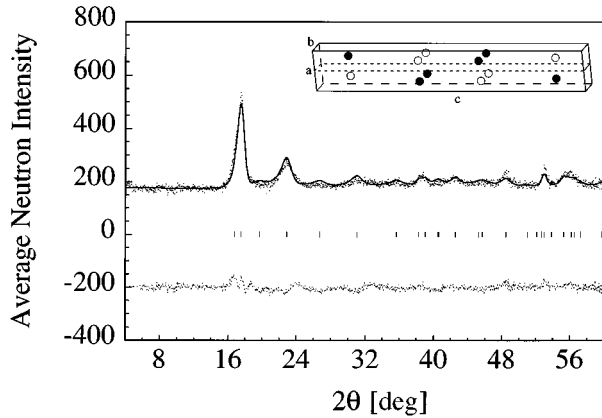
FIG. 4. Observed (dots) and calculated (solid line) difference neutron diffraction pattern (25 mK–3 K) of $Er_2Ba_4Cu_7O_{14.92}$. The calculated peak positions are shown as vertical lines and the difference of observed and calculated pattern is shown as a dashed line. The inset shows the magnetic unit cell of $Er_2Ba_4Cu_7O_{14.92}$. The magnetic moments are oriented parallel to the b axis. White and black colors correspond to the antiparallel configuration of the magnetic moments.
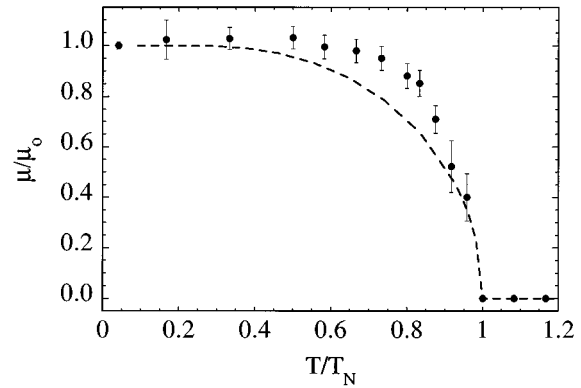

FIG. 5. Temperature dependence of the sublattice magnetization of Er^{3+} in $Er_2Ba_4Cu_7O_{14.92}$. The dashed line denotes a mean-field calculation on the basis of the crystal field parameters.

antiferromagnetically. The magnetic moments are antiparallel in the next layer up, which is translated by $1/2b + 1/2c$. The refinement of the magnetic diffraction pattern yields a correlation length $\lambda_c = 133(4)$ Å along the c axis which is due to magnetic stacking faults along this direction as expected from magnetic susceptibility measurements.

In addition to the basic configuration of magnetic moments, we determined their magnitude. In a mean-field calculation based on the crystal field level scheme of Er^{3+} we obtained a magnetic Er moment $\mu_{cal} = 3.92\mu_B$. Our experimental value of $\mu_{Er} = 3.68(10)\mu_B$ at saturation determined by neutron diffraction is in good agreement with μ_{cal} (see Fig. 5). The same type of ordering has been observed in the related $ErBa_2Cu_4O_8$ (Er-124) compound,[8] with an Er moment $\mu_{Er} = 3.86(15)\mu_B$ at saturation, whereas $4.2(1)\mu_B$ was reported for $ErBa_2Cu_3O_7$.[9]

IV. CONCLUSION

In conclusion, we have shown that the magnetic ordering of the Er^{3+} ions in well crystallized $Er_2Ba_4Cu_7O_{14.92}$ with $T_c = 89$ K taking place below $T_N = 0.54$ K is very similar to the ordering known for Er-124. We found a three-dimensional antiferromagnetic order of the Er ions with a finite correlation length of $\xi_c = 133(4)$ Å. The correlation length is more than twice as large as the lattice constant c, so that at least 10 magnetic ions are coupled along the c axis. The magnetic Er moments are aligned parallel to the b axis. The ordered magnetic moment of the Er ions at saturation is determined to be $\mu_{Er} = 3.68(10)\mu_B$.

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