Magnetic-field effect on static antiferromagnetic order above the upper critical field in $Nd_{1.85}Ce_{0.15}CuO_4$

M. Matsuura,[1] Pengcheng Dai,[2,1] H. J. Kang,[2] J. W. Lynn,[3] D. N. Argyriou,[4] Y. Onose,[5] and Y. Tokura[5,6,7]
[1]*Condensed Matter Sciences Division, Oak Ridge National Laboratory, Oak Ridge, Tennessee 37831-6393, USA*
[2]*Department of Physics and Astronomy, The University of Tennessee, Knoxville, Tennessee 37996-1200, USA*
[3]*NIST Center for Neutron Research, National Institute of Standards and Technology, Gaithersburg, Maryland 20899, USA*
[4]*Hahn-Meitner Institute, Glienicker Str. 100, Berlin D-14109, Germany*
[5]*Spin Superstructure Project, ERATO, Japan Science and Technology, Tsukuba 305-8562, Japan*
[6]*Correlated Electron Research Center, Tsukuba 305-8562, Japan*
[7]*Department of Applied Physics, University of Tokyo, Tokyo 113-8656, Japan*
(Received 1 October 2003; published 19 March 2004)

We use neutron scattering to study the effect of a *c*-axis-aligned magnetic field on superconducting $Nd_{1.85}Ce_{0.15}CuO_4$ for fields above its upper critical field. We also determine the effect of such a field on the cubic impurity phase $(Nd,Ce)_2O_3$. By comparing these data with previous field-induced results on other electron-doped materials, we conclude that while the impurity phase is responsible for scattering at (1/2,0,0), application of a magnetic field does induce a quantum phase transition from the superconducting to an antiferromagnetic state in electron-doped high-T_c superconductors.

DOI: 10.1103/PhysRevB.69.104510 PACS number(s): 74.72.Jt, 75.25.+z, 75.50.Ee, 61.12.Ld

One of the most important unresolved problems in high-transition-temperature (high-T_c) copper oxides concerns the nature of the correlated electron state at low temperature when superconductivity is suppressed.[1] While such a state in conventional Bardeen-Cooper-Schrieffer superconductors is a homogeneous diamagnetic metal, there are reasons to believe that many other quantum states compete with superconductivity in high-T_c copper oxides.[2–9] Determining the closest competing ground state and establishing the evolution of such a state when superconductivity is destroyed by a magnetic field will strongly constrain possible physical explanations for superconductivity. For *p*-type underdoped $La_{2-x}Sr_xCuO_4$ (LSCO), transport measurements have revealed that the field-induced normal state is a charge insulator.[10] Although subsequent neutron scattering experiments showed that application of a magnetic field enhances incommensurate spin-density-wave (SDW) order,[11,12] the transition from the superconducting state to the SDW state was never observed because the upper critical field B_{c2} for LSCO is beyond the capability of the current neutron measurements. Similarly, neutron scattering experiments for fields up to 7 T failed to confirm static SDW as a competing ground state to superconductivity in underdoped superconducting $YBa_2Cu_3O_{6.6}$.[13,14]

Fortunately, *n*-type doped high-T_c copper oxides generally have much lower B_{c2}.[15–17] While initial measurements found no observable field-induced effect for $Nd_{1.86}Ce_{0.14}CuO_4$,[18] we showed that B_{c2} (for a *c*-axis-aligned field) is only 6.2 T for $Nd_{1.85}Ce_{0.15}CuO_4$ (NCCO) and suppression of superconductivity by such a field induces a commensurate SDW order.[19,20] For two-dimensional antiferromagnetic (AF) ordered copper oxides, structure factor calculations show that magnetic scattering should center around (1/2,1/2,0) and/or its equivalent positions.[21] While the observation of a field-induced AF peak at (1/2,1/2,0) suggests that antiferromagnetism competes with superconductivity, the appearance of (1/2,0,0) and (0,1/2,0) magnetic reflections is puzzling.[19] Recently, Mang *et al.*[22] found that the annealing process necessary to make superconducting NCCO also induces epitaxial cubic $(Nd,Ce)_2O_3$ as an impurity phase. While this impurity phase is lattice matched with the *a*-*b* plane of NCCO, its lattice parameter along the *c*-axis is about 10% smaller than that of NCCO. Mang *et al.*[22] also claim that the epitaxial cubic $(Nd,Ce)_2O_3$ has long-range order parallel to the CuO_2 planes of NCCO but extending only $\sim 5a_c$ along the *c* axis, thus giving rise to structural peaks that occur at one subclass of half-integer NCCO Bragg peaks with $L=0$ (zero momentum transfer along the *c* axis) such as (1/2,1/2,0), where AF peaks occur. Although we confirmed the presence of such an impurity phase, $(Nd,Ce)_2O_3$ in our samples has a *c*-axis coherence length of about 220±20 Å full width at half maximum (FWHM) [Fig. 1(c)] and therefore essentially forms three dimensional long-range structural order.[20]

Since application of a magnetic field will induce a net ferromagnetic (FM) moment in the impurity phase at positions (1/2,1/2,0) and (1/2,0,0),[22] it is important to determine how much of the scattering at (1/2,1/2,0) and (1/2,0,0) in the $\mathbf{B}\|c$-axis geometry[19] originates from $(Nd,Ce)_2O_3$. While the magnetic field dependence of (1/2,0,0) appears to behave similarly with that of (1/2,1/2,0) below 7 T,[19] we show here that these two families of reflections have quite different field-dependent behavior at higher fields. By comparing the field dependence of the scattering with impurity scattering at these positions, we conclude that suppression of superconductivity by a *c*-axis-aligned field enhances scattering at (1/2,1/2,0) and (1/2,3/2,0). However, the field-induced intensity at (1/2,0,0) is likely due to the impurity phase.

The single crystals of superconducting NCCO used in present work were described in detail before.[19,20] They have a T_c of 25 K and a B_{c2} of 6.2 T at 5 K as determined from the resistivity measurements. The high field measurements

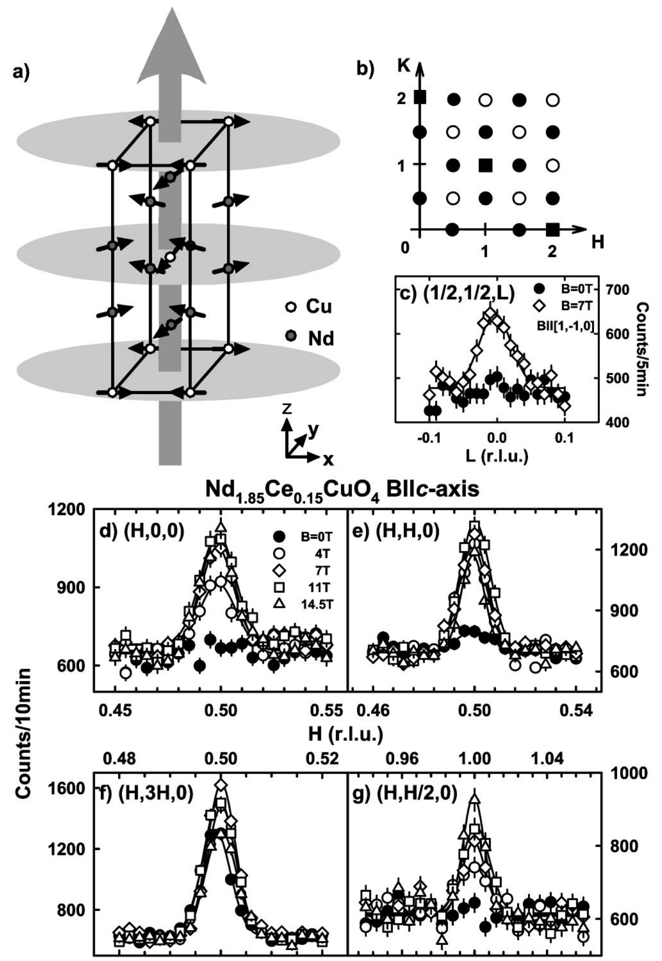

FIG. 1. Spin structure models, summary of reciprocal space probed in the neutron experiments, and effect of a magnetic field on the AF and impurity peaks at 5 K. (a), The non-collinear type-I/III spin structure of the AF order in NCCO. (b), Classification of field-induced magnetic peaks at 5 K($<T_c$). Moderate field induces peaks at $((2m+1)/2,\ (2n+1)/2,0)$, $(m,(2n+1)/2,0)$, and $((2m+1)/2,n,0)$, where $m,n=0,1$. The field-induced peaks can be classified into two types with different magnetic field dependence above B_{c2}. The closed circles represent peaks with increasing intensity above B_{c2}, while the open circles show peaks with decreasing intensity above B_{c2}. The closed squares indicate structural Bragg peaks where the intensity scales with increasing field and displays only a small anomaly across B_{c2}. (c) L-scans around the AF peak (1/2,1/2,0) at 0 T and 7 T magnetic field along the $[1,-1,0]$ direction. Since the applied magnetic field is parallel to the CuO_2 planes, the observed field-induced enhancement can be attributed almost entirely to the polarization of Nd moments in the impurity phase of $(Nd,Ce)_2O_3$. The FWHW of the field-induced scattering along the c-axis (ΔL) is 0.056 ± 0.006 (r.l.u.), indicating a minimum coherence length ξ of about $18a_c$ or 220 ± 20 Å along the c-axis of $(Nd,Ce)_2O_3$ (using $\xi\approx a_c/\Delta L$). Radial scans along (d) $(H,0,0)$, (e) $(H,H,0)$, (f) $(H,3H,0)$, and (g) $(H,H/2,0)$ directions at various fields. The data were collected on the E4 two-axis spectrometer at HMI.

were taken at the E4 two-axis diffractometer using the VM-1, 14.5-T vertical field magnet at the Berlin Neutron Scattering Center, Hahn-Meitner-Institute (HMI). The collimations were 40′-40′-Sample-40′ with one pyrolytic graphite (PG) filter and the incident beam energy (E_i) was fixed at 13.6 meV. For fields below 7 T, we used BT-9 and BT-2 triple-axis spectrometers at the NIST Center for Neutron Research with PG monochromator, $E_i=14.7$ meV, two PG filters, and collimations of 40′-46′-40′-80′. For the experiments, we label wave vectors $Q=(q_x,q_y,q_z)$ in Å^{-1} as $(H,K,L)=(q_xa/2\pi,q_ya/2\pi,q_zc/2\pi)$ in the reciprocal lattice units (rlu) appropriate for the tetragonal unit cell of NCCO (space group $I4/mmm$, $a=3.92$ and $c=12.07$ Å). The impurity phase $(Nd,Ce)_2O_3$ has space group $Ia3$ and lattice parameter $a_{NO}=11.702$ Å.[20]

TABLE I. Miller indices of $Nd_{1.85}Ce_{0.15}CuO_4$ and equivalent cubic $(Nd,Ce)_2O_3$ reflections assuming $a_{NO}=2\sqrt{2}a_{NCCO}=11.072$ and $c_{NCCO}=12.07$ Å.

NCCO	NO	$\lvert F_{NO}(H,K,L)\rvert$	NO	NCCO
(0,1/2,0)	(1,1,0)	0	(1,0,1)	(1/4,1/4,1.1)
(1/2,1/2,0)	(2,0,0)	11.14	(0,0,2)	(0,0,2.2)
(1/2,3/2,0)	(4,2,0)	16.85	(4,0,2)	(1,1,2.2)

Figures 1(a) and 1(b) plot the schematic diagram of canted AF NCCO and reciprocal space probed in the experiment, respectively. At zero field, weak superlattice peaks were observed at (1/2,0,0), (1/2,1/2,0), (1/2,3/2,0), and (1,1/2,0).[20] Application of a magnetic field induces magnetic scattering at (1/2,1/2,0) and (1/2,0,0) type positions as well as intensity at FM Bragg peak positions such as (1,1,0).[19,20] As a function of applied field, the field-induced scattering at (1/2,1/2,0) was found to initially increase linearly with field, saturate around B_{c2}, and finally decreases for larger fields. On the other hand, the field-induced FM scattering at (1,1,0) increases continuously from 0 T to 14.5 T and shows no major anomaly around B_{c2}. These results suggest the presence of a phase transition from the superconducting state to an antiferromagnetically ordered state at B_{c2}.[19] While the new measurements at the AF position (1/2,3/2,0) [Fig. 1(f)] confirm the earlier results at (1/2,1/2,0) [Fig. 1(e)],[19] scans around (1/2,0,0) [Fig. 1(d)] and (1,1/2,0) [Fig. 1(g)] positions behave quite differently. Instead of decreasing for fields above B_{c2}, the field-induced scattering appear to saturate above 8 T ($>B_{c2}$).

To compare the field-induced scattering from the impurity phase with that at (1/2,0,0), (1/2,1/2,0), and (1/2,3/2,0) for $\mathbf{B}||c$ axis, we realigned the crystal in the (H,H,L) zone where the applied vertical field is along the $[1,-1,0]$, the cubic edge direction of $(Nd,Ce)_2O_3$.[20] By measuring the field dependence of the scattering at (1/4,1/4,1.1), (1,1,2.2) and (0,0,2.2) positions, we are probing the magnetic field effect of the impurity phase on (1/2,0,0), (1/2,3/2,0), and (1/2,1/2,0) for the $\mathbf{B}||c$-axis geometry,[20,22] respectively, as summarized in Table I. Figure 2 shows the outcome of the experiment for fields up to 14.5 T. Since (1/4,1/4,1.1) is not an allowed structural Bragg position for $(Nd,Ce)_2O_3$, the scattering there is featureless at zero field [Fig. 2(a)]. On increasing the magnetic field from zero to 14.5 T at 5 K, the field-induced intensity increases with increasing field and essentially saturates for fields above 7 T (Fig. 2(a)). For

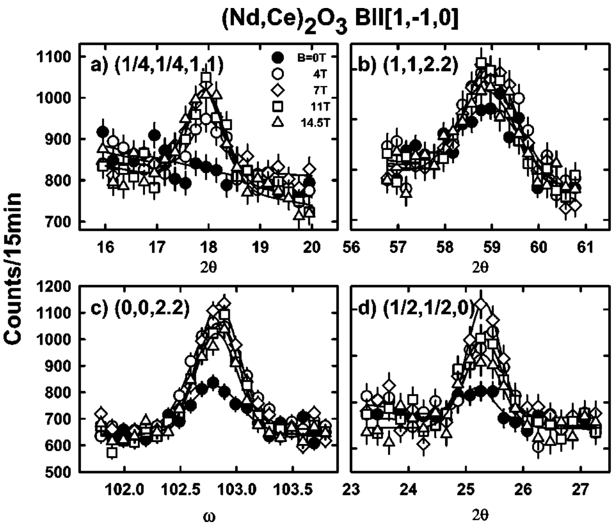

FIG. 2. Effect of a $\mathbf{B}||[1,-1,0]$ field on the impurity $(Nd,Ce)_2O_3$ peaks at 5 K. At zero field, $(Nd,Ce)_2O_3$ peaks were observed at (b) (1,1,2.2), (c) (0,0,2.2), and (d) (1/2,1/2,0). Moderate field induces peaks at (a) (1/4,1/4,1.1), and enhances (b) (1,1,2.2), (c) (0,0,2.2) and (d) (1/2,1/2,0). The spectrum shows radial scans for (a), (b), and (d), while a rocking curve is presented in (c) because of a large background from a strong Bragg reflection that is found in a radial scan (Ref. 20). The data were collected on the E4 two-axis spectrometer at HMI.

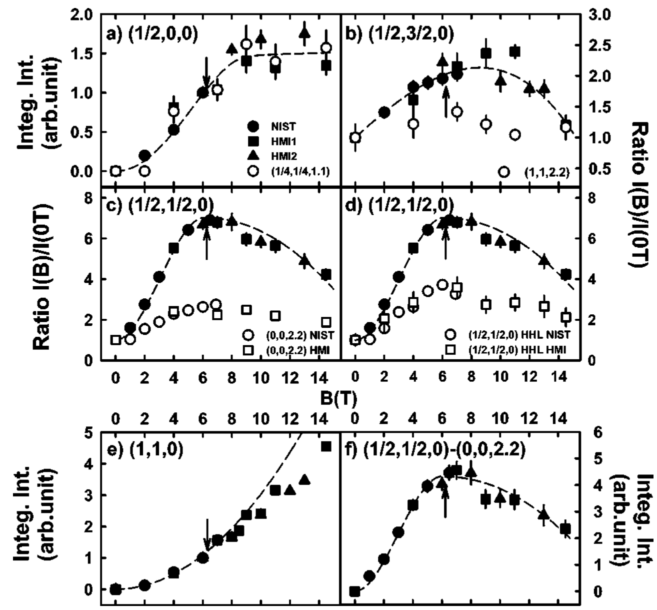

FIG. 3. Field dependence of the integrated intensities of AF, FM and impurity peaks as a function of field at 5 K. The magnetic field was applied along the c axis for AF and FM peaks (closed markers) and the $[1,-1,0]$ direction for impurity peaks (open markers). The circles are data taken at NIST while triangles and squares are two separate experiments at HMI. The integrated intensity at (b), (1/2,3/2,0)-residual AF and (1,1,2.2); [(c) and (d)], (1/2,1/2,0) and (0,0,2.2); are normalized to 1 at 0 T, while (a), (1/2,0,0) and (1/4,1/4,1.1) data are normalized to 1 at 6 T since there is no $(Nd,Ce)_2O_3$ peak at 0 T for (1/4,1/4,1.1). (e) shows the field dependence of induced FM peak (1,1,0). The dotted line in (e) shows a quadratic fit to the data below and upper critical field ($B_{c2}=6.2$ T). (f) represents the difference between (1/2,1/2,0) and (0,0,2.2) shown in (c). The dashed lines in (a)–(d) and (f) are guides to the eye and arrows indicate the upper critical field as determined from transport data (Ref. 19).

the (1,1,2.2) peak, which is an allowed reflection of $(Nd,Ce)_2O_3$ (Table I), the applied magnetic field has a weak effect on its intensity [Fig. 2(b)]. As a function of increasing magnetic field, the field-induced intensity increases slowly and essentially saturates for fields above 7 T [Fig. 2(b)]. In comparison, the field-induced scattering at (1/2,3/2,0) in the $\mathbf{B}||c$-axis geometry initially increases with increasing field but then decreases with increasing field for B larger than 7 T [Fig. 1(f)]. The field-dependence of the scattering at the (0,0,2.2) and (1/2,1/2,0) maximizes around B_{c2} and decrease slightly above B_{c2}.

For AF ordered copper oxides, the magnetic scattering should only appear at (1/2,1/2,0) and/or its equivalent positions. Assuming that the field-induced intensity is entirely magnetic, we normalized the structural integrated intensity measured at NIST and HMI to 1 at 0 T [except at (1/2,0,0) and (1/4,1/4,1.1), where the superlattice peak (1/2,0,0) arises from lattice distortion of the CuO_2 plane and scattering across (1/4,1/4,1.1) is featureless as required by the $Ia3$ space group of $(Nd,Ce)_2O_3$]. The field-induced magnetic intensities of (1/2,0,0) and (1/4,1/4,1.1) are normalized to 1 at 6 T. Since a 7 T c-axis-aligned field has no effect on (1/2,3/2,0) in the parent compound Nd_2CuO_4 and nonsuperconducting as-grown $Nd_{1.85}Ce_{0.15}CuO_4$,[20] we assume that such a field also has no effect on the residual AF order in superconducting $Nd_{1.85}Ce_{0.15}CuO_4$. At (1/2,1/2,0) [Figs. 3(c) and 3(d)] and (1/2,3/2,0) [Fig. 3(b)], the field-induced scattering peaks around B_{c2} and decreases for larger fields. At (1/2,0,0) [Fig. 3(a)] and (1,1/2,0) (not shown), the field-induced intensity behaves similarly to that of (1/2,1/2,0) and (1/2,3/2,0) for fields less than 6.2 T but clearly does not decrease for fields up to 14.5 T. Finally at the Bragg peak position (1,1,0) [Fig. 2(e)], the field-induced scattering has a quadratic field dependence for fields below B_{c2}, and then increases further with increasing field with a reduced slope.

To estimate the contribution of the impurity scattering at different lattice positions, we normalize the nuclear Bragg peaks from the impurity phase to the superlattice reflections. Assuming the latter has only the contributions from the impurity phase (which is the upper bound), we can determine the field dependence of $(Nd,Ce)_2O_3$ and compare that with the field-induced effect at (1/2,1/2,0) and (1/2,3/2,0) quantitatively. The open circles in Fig. 3 show the field-dependence of the impurity scattering based on intensity gains from impurity nuclear Bragg peaks. Consistent with earlier results,[20] we find that impurity scattering cannot account for the observed field-induced intensity at AF peaks. However, we confirm that the scattering at (1/4,1/4,1.1) and (1/2,0,0) positions are indistinguishable to within the error of the measurements.[22] In an independent experiment on superconducting $Pr_{0.89}LaCe_{0.11}CuO_4$,[23] a similar electron doped material where the cubic impurity phase $(Pr,La,Ce)_2O_3$ has a nonmagnetic ground state and no field dependence below 7

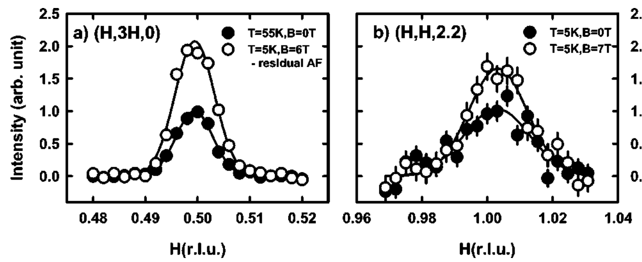

FIG. 4. Comparison of the field-induced effect at residual AF peak (1/2,3/2,0) in the $\mathbf{B}||c$-axis geometry with that at (1,1,2.2) in the $\mathbf{B}||ab$-plane geometry (along the $[1,-1,0]$ direction). At (1/2,3/2,0), the scattering intensity grows below $T_N \approx 38$ K. To compare the field-induced intensity at (1/2,3/2,0) with that from the polarization of $(Nd,Ce)_2O_3$ at (1,1,2.2), we normalize the integrated intensity of the structural superlattice reflection at (1/2,3/2,0) to that of (1,1,2.2) at 0 T and 5 K.

T,[24] Fujita *et al.* found enhanced AF order at (1/2,3/2,0) for fields up to 5 T. Above 5 T, this AF order decreases with increasing field, consistent with the field dependence of (1/2,1/2,0) in Ref. 19 and (1/2,3/2,0) of NCCO [Figs. 1(e) and 2(b)]. However, Fujita *et al.*[23] did not find any field-induced effect at the (1/2,0,0) and (1,1/2,0) positions in their experiments. In view of this fact and the similarities between the field-induced scattering at (1/2,0,0) and (1/4,1/4,1.1) [Fig. 3(a)], we conclude that the (1/2,0,0) scattering reported in Ref. 19 is due to the impurity $(Nd,Ce)_2O_3$ phase. Although this is in agreement with Mang *et al.*,[22] we emphasize that the central conclusion of Ref. 19, i.e., AF order is enhanced with the suppression of superconductivity, remains unchanged and is completely consistent with measurements on $Pr_{0.89}LaCe_{0.11}CuO_4$.[25]

In Fig. 4, we compare the field-induced scattering from $(Nd,Ce)_2O_3$ at (1,1,2.2) with that of (1/2,3/2,0). For NCCO with type-I/III magnetic structure, the long-range AF order of NCCO will contribute to the magnetic scattering at (1/2,3/2,0). Since superlattice peaks are temperature independent below 50 K,[19,20] we can use the intensity of (1/2,3/2,0) superlattice peak at zero field and 50 K for comparison with the (1,1,2.2) peak from $(Nd,Ce)_2O_3$. Figure 4 shows that the field-induced intensity at (1/2,3/2,0) has only a small contribution from $(Nd,Ce)_2O_3$.

In summary, we have performed neutron scattering experiments on the effect of a *c*-axis aligned magnetic field on superconducting $Nd_{1.85}Ce_{0.15}CuO_4$ for fields above its upper critical field. We determined the field-effect on the cubic impurity phase $(Nd,Ce)_2CuO_3$. By comparing these data with our previous field-induced results, we conclude that the impurity phase is responsible for the unexpected scattering at (1/2,0,0). This simplifies the interpretation of the results for the unusual AF NCCO reflections. However, an applied field does induce a quantum phase transition from the superconducting state to an AF state in electron-doped high-T_c superconductors.

We are grateful to Y. Ando, D.-H. Lee, H. A. Mook, Sudip Chakravarty, Steve Kievelson, K. Yamada, and S. C. Zhang for helpful discussions. We thank K. Prokes for technical assistance. This work was supported by the U.S. NSF DMR-0139882 and DOE under Contract No. DE-AC-00OR22725.

[1] P.W. Anderson, Science **235**, 1196 (1987).
[2] S.C. Zhang, Science **275**, 1089 (1997); D.P. Arovas, A.J. Berlinsky, C. Kallin, and S.C. Zhang, Phys. Rev. Lett. **79**, 2871 (1997); H.D. Chen, J.P. Hu, S. Capponi, E. Arrigoni, and S.C. Zhang, *ibid.* **89**, 137004 (2002).
[3] X.G. Wen and P.A. Lee, Phys. Rev. Lett. **76**, 503 (1996).
[4] S. Chakravarty, R.B. Laughlin, D.K. Morr, and C. Nayak, Phys. Rev. B **63**, 094503 (2001).
[5] Y. Zhang, E. Demler, and S. Sachdev, Phys. Rev. B **66**, 094501 (2002).
[6] D.-H. Lee, Phys. Rev. Lett. **88**, 227003 (2002).
[7] Y. Chen and C.S. Ting, Phys. Rev. B **65**, 180513 (2002).
[8] S.A. Kivelson, D.-H. Lee, E. Fradkin, and V. Oganesyan, Phys. Rev. B **66**, 144516 (2002).
[9] J. Zhu, I. Martin, and A. Bishop, Phys. Rev. Lett. **89**, 067003 (2002).
[10] Y. Ando, G.S. Boebinger, A. Passner, T. Kimura, and K. Kishio, Phys. Rev. Lett. **75**, 4662 (1995).
[11] S. Katano, M. Sato, K. Yamada, T. Suzuki, and T. Fukase, Phys. Rev. B **62**, R14677 (2000).
[12] B. Lake, H.M. Ronnow, N.B. Christensen, G. Aeppli, K. Lefmann, D.F. McMorrow, P. Vorderwisch, P. Smeibidl, N. Mangkorntong, T. Sasagawa, M. Nohara, H. Takagi, and T.E. Mason, Nature (London) **415**, 299 (2002).
[13] P. Dai, H.A. Mook, G. Aeppli, S.M. Hayden, and F. Doğan, Nature (London) **406**, 965 (2000).
[14] H.A. Mook, P. Dai, S.M. Hayden, A. Hiess, J.W. Lynn, S.-H. Lee, and F. Doğan, Phys. Rev. B **66**, 144513 (2002).
[15] Y. Hidaka and M. Suzuki, Nature (London) **338**, 635 (1989).
[16] P. Fournier, P. Mohanty, E. Maiser, S. Darzens, T. Venkatesan, C.J. Lobb, G. Czjzek, R.A. Webb, and R.L. Greene, Phys. Rev. Lett. **81**, 4720 (1998).
[17] R.W. Hill, C. Proust, L. Taillefer, P. Fournier, and R.L. Greene, Nature (London) **414**, 711 (2001).
[18] M. Matsuda, S. Katano, T. Uefuji, M. Fujita, and K. Yamada, Phys. Rev. B **66**, 172509 (2002).
[19] H.J. Kang, P. Dai, J.W. Lynn, M. Matsuura, J.R. Thompson, S.-C. Zhang, D.N. Argyriou, Y. Onose, and Y. Tokura, Nature (London) **423**, 522 (2003).
[20] M. Matsuura, P. Dai, H.J. Kang, J.W. Lynn, D.N. Argyriou, K. Prokes, Y. Onose, and Y. Tokura, Phys. Rev. B **68**, 144503 (2003).
[21] J. W. Lynn and S. Skanthakumar, in *Handbook on the Physics and Chemistry of Rare Earths*, edited by K. A. Gschneidner, Jr., L. Eyring, and M. B. Maple (Elsevier, Amsterdam, 2001), Vol. 31, p. 315.
[22] P.K. Mang, S. Larochelle, and M. Greven, Nature (London) **426**, 139 (2003).
[23] M. Fujita, M. Matsuda, S. Katano, and K. Yamada, cond-mat/0311269 (unpublished).
[24] In an independent experiment on overdoped superconducting

$Pr_{0.88}LaCe_{0.12}CuO_4$ [H. J. Kang *et al.* (unpublished)], we find that the impurity phase $(Pr,La,Ce)_2O_3$ in the sample has no observable field-induced effect at (0,0,2.2) below ~7 T. This is consistent with the results of Fujita *et al.* and confirms that Pr in $(Pr,La,Ce)_2O_3$ has a nonmagnetic singlet ground state.

[25] H.J. Kang, P. Dai, J.W. Lynn, M. Matsuura, J.R. Thompson, S.-C. Zhang, D.N. Argyriou, Y. Onose, and Y. Tokura, Nature (London) **426**, 140 (2003).