Absence of a magnetic-field effect on static magnetic order in the electron-doped superconductor $Nd_{1.86}Ce_{0.14}CuO_4$

M. Matsuda and S. Katano
Advanced Science Research Center, Japan Atomic Energy Research Institute, Tokai, Ibaraki 319-1195, Japan

T. Uefuji, M. Fujita, and K. Yamada
Institute for Chemical Research, Kyoto University, Gokasho, Uji 610-0011, Japan
(Received 2 July 2002; published 27 November 2002)

Neutron-scattering experiments were performed to study the magnetic-field effect on the electron-doped cuprate superconductor $Nd_{1.86}Ce_{0.14}CuO_4$, which shows the coexistence of magnetic order and superconductivity. The $(\frac{1}{2}\frac{3}{2}0)$ magnetic Bragg intensity, which originates from the order of both the Cu and Nd moments at low temperatures, shows no magnetic-field dependence when the field is applied perpendicular to the CuO_2 plane up to 10 T above the upper critical field. This result is significantly different from that reported for the hole-doped cuprate superconductors, in which the quasistatic magnetic order is noticeably enhanced under a magnetic field.

DOI: 10.1103/PhysRevB.66.172509 PACS number(s): 74.72.Jt, 75.25.+z, 75.50.Ee

Extensive neutron-scattering studies have been performed on high-T_c superconductors in order to clarify the interplay between the superconductivity and magnetism. In particular, in the hole-doped cuprate superconductor $La_{2-x}Sr_xCuO_4$ and related systems, static and dynamic properties of spin correlations have been studied in considerable detail.[1–5] A remarkable feature in the superconducting phase is that static and low-energy spin correlations are incommensurate, and the magnetic peaks are found at $(\frac{1}{2},\frac{1}{2}\pm\delta)$ and $(\frac{1}{2}\pm\delta,\frac{1}{2})$.[2,3] In the optimally doped region, there exists an excitation gap, and low-energy excitations are suppressed.[6] On the other hand, in the region where the hole concentration is $\sim\frac{1}{8}$, elastic incommensurate peaks, originating from both the spin-density wave and the charge density wave, are observed distinctly, suggesting the stripe model.[4] In this underdoped region, the coexistence of magnetic order and superconductivity is implied.[7,8]

In the electron-doped cuprate superconductor, however, the number of neutron-scattering studies is rather limited, probably because a large single crystal is difficult to grow. Yamada *et al.* reported that the superconducting $Nd_{1.85}Ce_{0.15}CuO_4$ (superconducting transition temperature $T_c\sim18$ K) shows a broad magnetic excitation peak at the commensurate position $(\frac{1}{2},\frac{1}{2})$.[9] It is also found that an excitation gap exists around 4.5 meV. Thus both hole- and electron-doped cuprate superconductors show a gap behavior in magnetic excitations, although the magnetic correlations are incommensurate and commensurate in hole- and electron-doped cuprate superconductors, respectively. The coexistence of magnetic order and superconductivity was also suggested in an electron-doped system.[10–12]

Neutron scattering under a magnetic field is one of the important techniques that can be used to study the interplay between magnetism and superconductivity. The magnetic-field effect has been studied in superconducting $La_{2-x}Sr_xCuO_4$ ($x=0.10$ and 0.12) and La_2CuO_{4+y}. These investigations showed that the static parallel stripe order is enhanced under a magnetic field perpendicular to the CuO_2 planes.[13–15] The enhancement of the elastic magnetic intensity is ascribed to vortices which stabilize the static magnetic order in a larger region than the vortex cores.[13–15] Theoretical studies were also performed intensively on the static magnetic ordering induced near the vortex cores, which is consistent with experiments.[16–24]

In the case of the electron-doped cuprate superconductor $Nd_{2-x}Ce_xCuO_4$, magnetic-field studies were performed only for undoped Nd_2CuO_4,[25] to the best of our knowledge. The main purpose of that study was to determine whether the magnetic structure is collinear or noncollinear. The magnetic field was applied in the CuO_2 plane and the magnetic structure was found to be noncollinear. In the present study, we examined the magnetic-field effect of the static magnetic correlations in $Nd_{1.86}Ce_{0.14}CuO_4$ ($T_c\sim25$ K). Since the coherence length is ~100 Å in the electron-doped system,[26] which is several times larger than that in $La_{2-x}Sr_xCuO_4$, a large magnetic-field effect can be expected. Furthermore, the upper critical field H_{c2} is less than 10 T in the electron-doped system, so that normal-state properties can easily be studied. It is found that the elastic magnetic peak is magnetic field independent up to 10 T above H_{c2}, suggesting that the interplay between magnetic order and superconductivity in this system is considerably different from that in the hole-doped system.

A single crystal of $Nd_{1.86}Ce_{0.14}CuO_4$ was grown by the traveling solvent floating-zone method. The crystal was annealed in an Ar atmosphere at 920 °C for 12 h. T_c is ~25 K, as determined from a susceptibility measurement, and is shown in Fig. 1. From the data, the superconducting property is considered to be that of bulk in nature. The crystal used in this study is the one that was used in the previous study.[11] The Ce concentration dependence of magnetic and superconducting properties shows a systematic change in the transition temperatures, indicating that the doped electrons are homogeneously distributed.[10,11] For $x=0.14$, which was used in this study, similar volume of a magnetically ordered phase and superconducting phase coexist.[10] It is likely that a

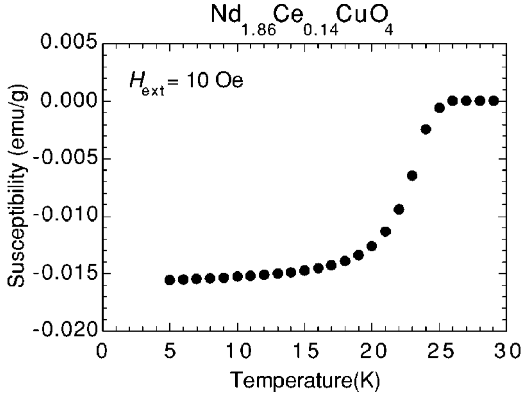

FIG. 1. Temperature dependence of the magnetic susceptibility under a zero-field-cooled condition with the magnetic field of 10 Oe for $Nd_{1.86}Ce_{0.14}CuO_4$.

slightly inhomogeneous distribution of electrons causes two phases that are spatially separated. This phase separation behavior is basically similar to that in $La_{2-x}Sr_xCuO_4$.

The neutron-scattering experiments were carried out on the three-axis spectrometer TAS2 installed in the guide hall of JRR-3M at the Japan Atomic Energy Research Institute. The typical horizontal collimator sequence was guide-20′-S-20′-80′ with a fixed incident neutron energy of $E_{\mathrm{i}} = 13.7$ meV. Contamination from higher-order beams was effectively eliminated using pyrolytic graphite filters. The single crystal was oriented in the $(HK0)$ scattering plane. Neutron-scattering experiments under magnetic fields were performed up to 10 T using a type of split-pair superconducting magnet cooled by cryocoolers. The field was applied vertically to the scattering plane.

In the $(HK0)$ scattering zone, magnetic Bragg peaks are observed at $(\frac{1}{2}+m,\frac{1}{2}+n,0)$, where m and n are integers, except at $(\pm\frac{1}{2}\pm m,\pm\frac{1}{2}\pm m,0)$ and $(\pm\frac{1}{2}\pm m,\mp\frac{1}{2}\mp m,0)$. It is

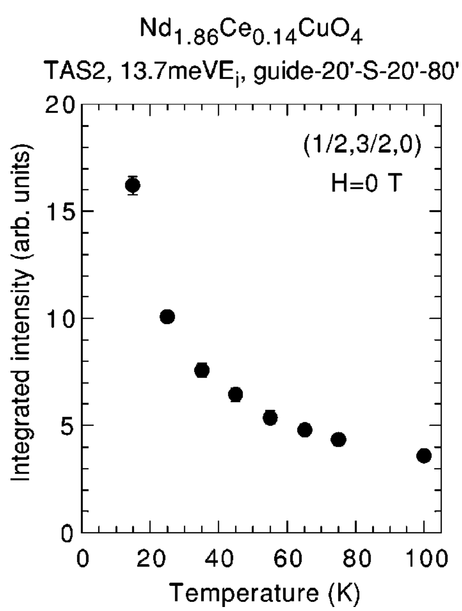

FIG. 2. Temperature dependence of the $(\frac{1}{2}\frac{3}{2}0)$ Bragg peak under zero magnetic field in $Nd_{1.86}Ce_{0.14}CuO_4$.

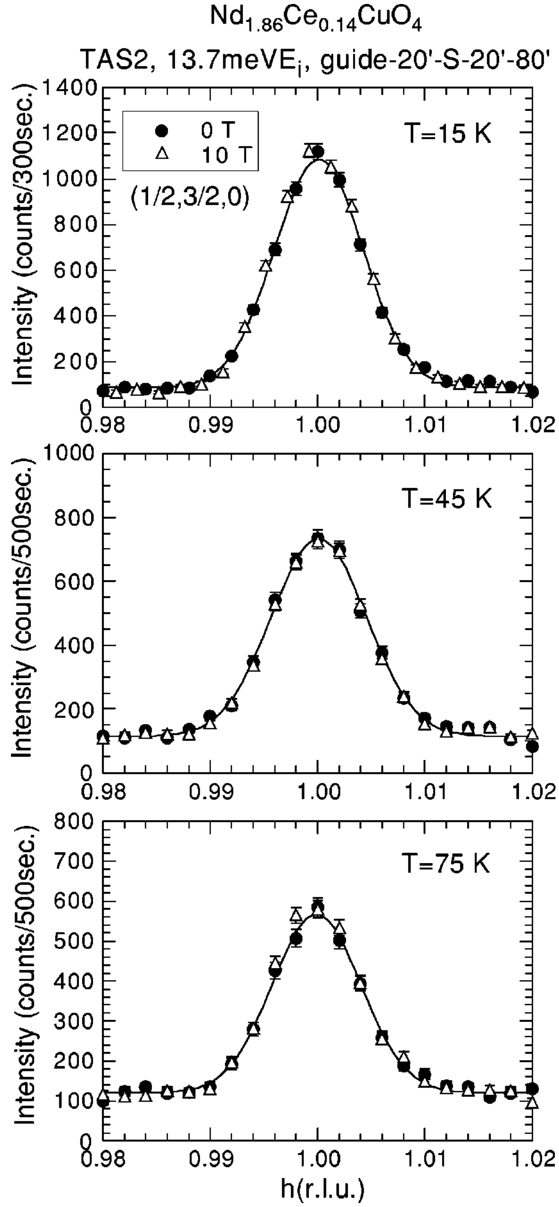

FIG. 3. Neutron elastic intensity around the commensurate position $(\frac{1}{2},\frac{3}{2},0)$ in $Nd_{1.86}Ce_{0.14}CuO_4$ under magnetic fields $H=0$ and 10 T and at $T=15$, 45, and 75 K. The magnetic field is applied perpendicular to the CuO_2 plane. The solid lines are the results of fits to a Gaussian function for the zero-field data.

also reported that superlattice peaks, which originate from a superstructure caused by the heat treatment and are almost temperature independent, are superimposed on these magnetic peak positions.[27] Figure 2 shows the temperature dependence of the $(\frac{1}{2}\frac{3}{2}0)$ Bragg intensity under zero magnetic field. The intensity at $(\frac{1}{2}+m,\frac{1}{2}+n,0)$ is described as

$$I=C\{M_{\mathrm{Cu}}(T)f_{\mathrm{Cu}}+2M_{\mathrm{Nd}}(T)f_{\mathrm{Nd}}\}^2+I_{\mathrm{lattice}}, \qquad (1)$$

where C is a constant, I_{lattice} is the superlattice scattering intensity, and $M(T)$ and f are ordered staggered moments and form factors for the Cu^{2+} and Nd^{3+} ions, respectively. Above 100 K, this reflection almost originates from the superstructure. With decreasing temperature, the Cu and Nd moments order gradually, and a contribution from the order of the Cu moments becomes comparable to that from the superstructure around 50 K. Below ~20 K the order of the Nd moments develops rapidly so that most of the scattering intensity originates from the magnetic order and the contribution of the Nd moments becomes comparable to that of the Cu moments. It was reported that $M_{\mathrm{Cu}}(10\ \mathrm{K}) \sim 0.1\mu_B$ and $M_{\mathrm{Nd}}(10\ \mathrm{K}) \sim 0.05\mu_B$ in $Nd_{1.86}Ce_{0.14}CuO_4$ if the moments are assumed to be homogeneously distributed.[11]

Figure 3 shows the magnetic field dependence of the neutron elastic intensity at $(\frac{1}{2},\frac{3}{2},0)$ in $Nd_{1.86}Ce_{0.14}CuO_4$. The magnetic field is applied perpendicular to the CuO_2 plane. The magnetic peak width is slightly broader than the instrumental resolution[11] although the superlattice peak is almost resolution limited in the (*HK*0) plane.[27] At 75 K, where most of the scattering intensity comes from the structural distortion, there is no magnetic-field effect up to 10 T, which is reasonable. At 45 K above T_c, where about one third of the intensity comes from the static magnetic order, mostly of the Cu moments,[28] the magnetic-field effect is still missing. Finally, at 15 K below T_c, where about 80% of the intensity is magnetic in origin and the Cu and Nd contributions are comparable,[28] there is almost no magnetic-field dependence even at 10 T, which is above H_{c2}. This result is significantly different from that reported for the hole-doped system, in which the quasistatic magnetic order is enhanced under a magnetic field.[13–15]

As mentioned at the beginning for hole-doped $La_{2-x}Sr_xCuO_4$, it is theoretically predicted that the static magnetic order is stabilized and enhanced around the vortex cores with an application of magnetic field,[16–24] indicating that dynamic spin fluctuations in the superconducting phase can be easily pinned by the vortices. If such a strong pinning effect also exists in the electron-doped system, the magnetic field should enhance the static magnetic order. Since almost the same volume of magnetic and superconducting phases coexist in $Nd_{1.86}Ce_{0.14}CuO_4$,[10] the enhancement of the elastic magnetic intensity is expected to be clearly observable.

A puzzling question is how the magnetically ordered phase and superconducting phase coexist in the electron-doped system. We mentioned that the two-phase behavior probably originates from the phase separation of the doped carriers, which is also probable in $La_{2-x}Sr_xCuO_4$. Although the correlation length of the magnetically ordered phase in the CuO_2 plane is similar in the both systems (~100 Å), the difference between the two systems is the correlation length perpendicular to the CuO_2 plane. In a nearly optimum-doped region of $La_{2-x}Sr_xCuO_4$, magnetic correlations are almost two dimensional. On the other hand, the correlation length in $Nd_{1.86}Ce_{0.14}CuO_4$ is estimated to be about 100 Å,[11] which is fairly large. Therefore, the magnetically ordered state is expected to be robust in $Nd_{1.86}Ce_{0.14}CuO_4$ against magnetic fields, which is consistent with the experimental results. Even in this case, however, a static magnetic order might appear in the superconducting region when the magnetic field exceeds H_{c2} and the superconducting region turns to be in the normal state. Therefore, the absence of a magnetic-field effect is surprising.

A probable scenario for the absence of the magnetic-field effect would be as follows. The superconducting phase has an excitation gap as reported in $Nd_{1.85}Ce_{0.15}CuO_4$,[9] and the gap does not close even above H_{c2}, so that a quasielastic component still does not appear. This behavior is similar to that observed in optimally doped $La_{2-x}Sr_xCuO_4$,[29] in which an in-gap state develops but the gap still remains under a magnetic field, although the applied field is much smaller than H_{c2}. It is also possible that the superconducting phase in the $Nd_{2-x}Ce_xCuO_4$ system lies in the overdoped phase, in which the magnetic fluctuations are shorter ranged,[3] and thus a long-range magnetic order does not appear easily. In order to clarify this in detail, the magnetic-field dependence of the excitation spectra should be measured. We plan to perform neutron inelastic scattering experiments under a magnetic field in future.

In summary, our neutron-scattering experiments under a magnetic field in the electron-doped cuprate superconductor $Nd_{1.86}Ce_{0.14}CuO_4$ demonstrate that the static magnetic order of both the Cu and Nd moments shows no magnetic-field dependence up to 10 T which is above H_{c2}. This is in sharp contrast to the static magnetic order in the hole-doped system.

We would like to thank S.-H. Lee for stimulating discussions and Y. Shimojo for technical assistance. This study was supported in part by a Grant-in-Aid for Scientific Research from the Japanese Ministry of Education, Culture, Sports, Science, and Technology.

[1] M.A. Kastner, R.J. Birgeneau, G. Shirane, and Y. Endoh, Rev. Mod. Phys. **70**, 897 (1998).
[2] S.-W. Cheong, G. Aeppli, T.E. Mason, H. Mook, S.M. Hayden, P.C. Canfield, Z. Fisk, K.N. Clausen, and, J.L. Martinez, Phys. Rev. Lett. **67**, 1791 (1991).
[3] K. Yamada, C.H. Lee, K. Kurahashi, J. Wada, S. Wakimoto, S. Ueki, H. Kimura, Y. Endoh, S. Hosoya, G. Shirane, R.J. Birgeneau, M. Greven, M.A. Kastner, and Y.J. Kim, Phys. Rev. B **57**, 6165 (1998).
[4] J.M. Tranquada, B.J. Sternlieb, J.D. Axe, Y. Nakamura, and S. Uchida, Nature (London) **375**, 561 (1995).
[5] M. Fujita, H. Goka, K. Yamada, and M. Matsuda, Phys. Rev. Lett. **88**, 167008 (2002).
[6] K. Yamada, S. Wakimoto, G. Shirane, C.H. Lee, M.A. Kastner, S. Hosoya, M. Greven, Y. Endoh, and R.J. Birgeneau, Phys. Rev. Lett. **75**, 1626 (1995).
[7] J.M. Tranquada, J.D. Axe, N. Ichikawa, A.R. Moodenbaugh, Y. Nakamura, and S. Uchida, Phys. Rev. Lett. **78**, 338 (1997).
[8] Ch. Niedermayer, C. Bernhard, T. Blasius, A. Golnik, A. Moodenbaugh, and J.I. Budnick, Phys. Rev. Lett. **80**, 3843 (1998).
[9] K. Yamada, K. Kurahashi, Y. Endoh, R.J. Birgeneau, and G. Shirane, J. Phys. Chem. Solids **60**, 1025 (1999).

[10] T. Uefuji, T. Kubo, K. Yamada, M. Fujita, K. Kurahashi, I. Watanabe, and K. Nagamine, Physica C **357-360**, 208 (2001).
[11] T. Uefuji, K. Kurahashi, M. Fujita, M. Matsuda, and K. Yamada, Physica C **378-381**, 273 (2002).
[12] M. Fujita, T. Kubo, S. Kuroshima, T. Uefuji, K. Kawashima, K. Yamada, I. Watanabe, and K. Nagamine, cond-mat/0203320, Phys. Rev. B (to be published).
[13] S. Katano, M. Sato, K. Yamada, T. Suzuki, and T. Fukase, Phys. Rev. B **62**, R14677 (2000).
[14] 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).
[15] B. Khaykovich, Y.S. Lee, R. Erwin, S.-H. Lee, S. Wakimoto, K.J. Thomas, M.A. Kastner, and R.J. Birgeneau, Phys. Rev. B **66**, 014528 (2002).
[16] D.P. Arovas, A.J. Berlinsky, C. Kallin, and S.-C. Zhang, Phys. Rev. Lett. **79**, 2871 (1997).
[17] M. Ogata, Int. J. Mod. Phys. B **13**, 3560 (1999).
[18] M. Ichioka, M. Takigawa, and K. Machida, J. Phys. Soc. Jpn. **70**, 33 (2001).
[19] J.-X. Zhu and C.S. Ting, Phys. Rev. Lett. **87**, 147002 (2001).
[20] J.-P. Hu and S.-C. Zhang, cond-mat/0108273 (unpublished).
[21] D.-H. Lee, Phys. Rev. Lett. **88**, 227003 (2002).
[22] M. Franz, D.E. Sheehy, and Z. Tešanović, Phys. Rev. Lett. **88**, 257005 (2002).
[23] Y. Zhang, E. Demler, and S. Sachdev, Phys. Rev. B **66**, 094501 (2002).
[24] Y. Chen, H.Y. Chen, and C.S. Ting, Phys. Rev. B **66**, 104501 (2002).
[25] S. Skanthakumar, J.W. Lynn, J.L. Peng, and Z.Y. Li, Phys. Rev. B **47**, 6173 (1993).
[26] Y. Hidaka and M. Suzuki, Nature (London) **338**, 635 (1989).
[27] K. Kurahashi, H. Matsushita, M. Fujita, and K. Yamada, J. Phys. Soc. Jpn. **71**, 910 (2002).
[28] The relative contribution of the Cu and Nd moments was calculated in Ref. 11 using magnetic Bragg reflections in the (*HOL*) scattering plane. See, e.g., M. Matsuda, Y. Endoh, K. Yamada, H. Kojima, I. Tanaka, R.J. Birgeneau, M.A. Kastner, and G. Shirane, Phys. Rev. B **45**, 12548 (1992).
[29] B. Lake, G. Aeppli, K.N. Clausen, D.F. McMorrow, K. Lefmann, N.E. Hussey, N. Mangkorntong, M. Nohara, H. Takagi, T.E. Mason, and A. Schröder, Science **291**, 1759 (2001).