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On the possibility of antiferromagnetic vortex cores in superconducting $YBa_2Cu_3O_{7-\delta}$

David Vaknin *, Jerel L. Zarestky, Lance L. Miller

Ames Laboratory, Department of Physics and Astronomy, Iowa State University, Ames, IA 50011, USA

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Abstract

Neutron scattering studies of optimally doped single crystal $YBa_2Cu_3O_{7-\delta}$ under applied magnetic field were used to assess the existence of antiferromagnetic (AF) correlations in vortices. In the superconducting (SC) state, a very weak signal at the $(\frac{1}{2}\,\frac{1}{2}\,0)$ reciprocal lattice point was found. It is argued that this signal can be associated with AF correlations in vortex cores. An upper-limit average magnetic moment per vortex in each layer is estimated. Above T_c, weak paramagnetic scattering was observed, which we hypothesize is due to scattering from nearest-neighbor copper spins that are coupled ferromagnetically by a hole on an oxygen site between them. © 2000 Elsevier Science B.V. All rights reserved.

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1. Introduction

From the outset, the intimate relationship between superconductivity and magnetism in the copper-oxide family of superconductors has intrigued researchers [1–4]. Evidence for this link came from neutron scattering experiments on the parent compound of the single-layer cuprates, La_2CuO_4, that showed long-range antiferromagnetic (AF) ordering of Cu^{2+} spins ($S=1/2$) in the CuO_2 layers [5]. Subsequently, it was established that all members of the superconducting (SC) cuprates are essentially derivatives of insulating copper-oxide antiferromagnets [6–8]. In particular, it was found that the end member $YBa_2Cu_3O_6$ is an antiferromagnet, and that doping with oxygen destroys the long range AF order and establishes superconductivity with $T_c = 90$ K for $\delta \approx 0$. Previous neutron scattering studies of optimally doped and underdoped samples identified magnetic excitations associated with the spin system [10–12]. Polarized neutron scattering experiments on a single crystal $YBa_2Cu_3O_{6.92}$, identified a resonance centered at 41 meV at the (π,π) reciprocal lattice position, that was both energy- and temperature-dependent [11]. These results gave impetus to a newly developed SO(5) model that unifies superconductivity and antiferromagnetism in the high temperature superconductors [9]. One of the consequences of this theory is that AF correlations exist in the cores of underdoped cuprates. Although our initial motivation for this work stems from that proposal, we decided to conduct the search for AF cores in a nearly

* Corresponding author. Tel.: +1-515-294-6023; fax: +1-515-294-0689.
E-mail address: vaknin@ameslab.gov (D. Vaknin)

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optimally doped system, to avoid ambiguities due to sample inhomogeneity. The present magnetic neutron scattering study deals with the question about the nature of the spin system associated with the CuO_2 sheets in the optimally doped regime $YBa_2Cu_3O_{7-\delta}$ ($\delta \approx 0$).

2. Experimental

The twinned single crystal (approximate dimensions $4 \times 4 \times 0.5$ mm^3, weight 50 mg) used in the present neutron scattering investigation was grown in air from a mixture $8YBa_2Cu_3O_7 + BaCuO_2 + CuO$ in yttria-stabilized zirconia crucibles. The furnace used for the growth was modified for enhanced temperature stability and a small vertical temperature gradient. The mixture was preheated at 1035°C for 12 h and cooled from 960°C to 920°C over 5 days. This was followed by cooling to room temperature over a day and a half. The essentially free standing crystals (up to $6 \times 6 \times 0.6$ mm^3) were cut apart and heated to 475°C under flowing oxygen for 4 months with heating and cooling rates of 45°C/h. Crystal quality and homogeneity were examined by DC magnetic susceptibility ($\chi = M/H$) vs. temperature and standard neutron diffraction measurements. In Fig. 1, the magnetic susceptibility $\chi(T)$ under magnetic field $H = 10^{-4}$ Tesla (T) shows a sharp SC transition (Meissner effect) at $T_c \approx 90$ K with a temperature width on the order of 1 K, confirming the high homogeneity of the crystal. In addition, the lattice parameters, extracted from our neutron diffraction $a = 3.8130$ Å, $b = 3.8851$ Å and $c = 11.67$ Å, indicate that the crystal is optimally doped, with $\delta \approx 0.0$ [13].

Neutron-diffraction measurements were carried out on the HB1A fixed-initial-energy triple-axis spectrometer at the High Flux Isotope Reactor (HFIR) at the Oak Ridge National Laboratory (ORNL). This instrument uses a double monochromator system of highly oriented pyrolytic graphite (HOPG) crystals, (002) planes, to produce monochromatic incident neutrons of wavelength $\lambda = 2.358$ Å. The $\lambda/2$ component present in the beam was removed with two sets of pyrolytic graphite filters located before and after the monochromator to better than 1 part in 10^6 of prominent nuclear Bragg reflections. The beam scattered from the sample was energy analyzed with an HOPG (002) crystal analyzer. A collimation of 40′ (effective), monochromator-40′ (effective), sample, 40′-analyzer, 136′-detector yielded energy resolution of about 1 meV and a momentum-transfer (Q) resolution of about 0.01 Å^{-1}. All scattered beam counting was performed vs. monitor detector counts to account for variations in reactor flux. The sample was wrapped in aluminum foil and affixed to a

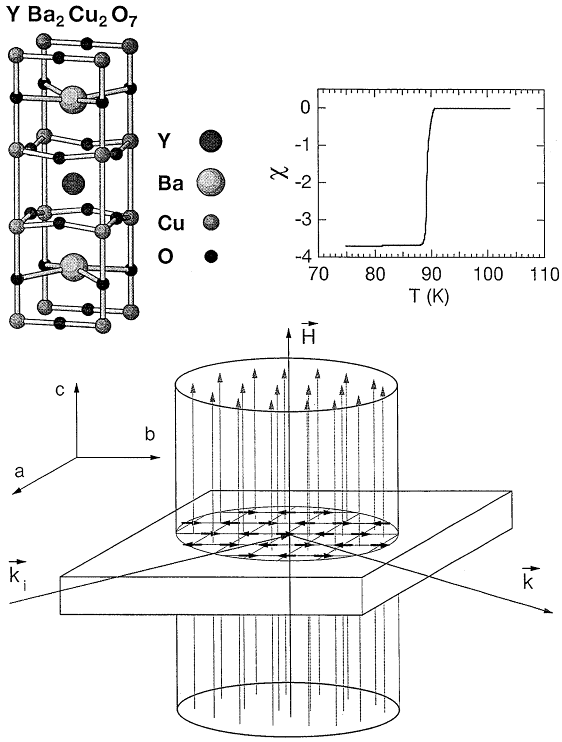

Fig. 1. The crystal structure of $YBa_2Cu_3O_7$, showing the two adjacent CuO_2 layers, and the CuO_x chains. The superconducting transition temperature T_c of the single crystal used in this study was measured by magnetic susceptibility ($\chi = M/H$) vs. temperature with an applied magnetic field of 10^{-4} T. The sharp transition, 1 K in width, is a measure of the high homogeneity of oxygen distribution in the crystal. The scattering configuration depicts the crystal orientation with respect to the scattering plane (defined by the incoming, $\boldsymbol{k}_i$, and the outgoing, $\boldsymbol{k}_f$, wave-vectors). The magnetic field was applied along the crystallographic c-axis (normal to the scattering plane). AF ordering within a vortex core is shown schematically.

0.5-mm aluminum plate mounted such that the c-axis of the sample was perpendicular to the scattering plane, (defined by $\boldsymbol{k}_i$ and $\boldsymbol{k}_f$, as shown in Fig. 1). The area where the foil was affixed to the aluminum plate was painted with gadolinium-oxide paint to reduce parasitic scattering. The sample was then mounted in an Oxford Instruments 7 T Cryomagnet to obtain the required temperature and magnetic field (vertical to the scattering plane) for this experiment. The temperature was controlled to within a few tenths of a Kelvin using a carbon–glass sensor in the 4–20 K range, and a rhodium–iron sensor at higher temperatures. The (002), (200), and (110) nuclear Bragg reflections were used to monitor and correct for crystal misorientation at each magnetic field and temperature.

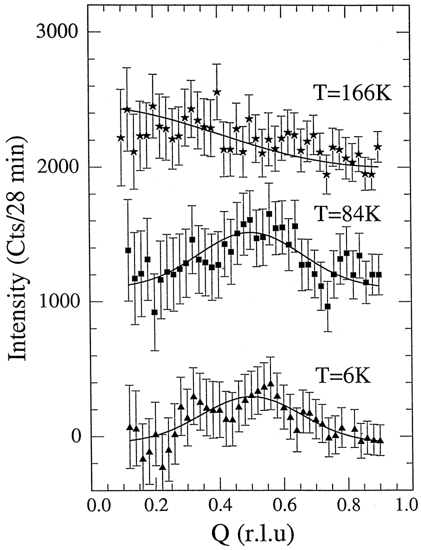

Fig. 3. Intensity vs. momentum transfer along the (110) direction in reciprocal space, calculated by subtracting a zero field scan from a scan measured under a magnetic field, (as shown in Fig. 2) at various temperatures. The applied magnetic field at $T=6$ K and at $T=166$ K was $H=5$ T; and at $T=84$ K, the field was $H=7$ T (scans are displaced vertically by 1000 cts units for clarity). It is proposed that the peaks below T_c can be associated with the inplane short-range AF ordering in vortex cores. At $T=166$ K, the scattering is indicative of paramagnetic scattering. It is argued that the paramagnetic moments consist of two copper spins that are coupled ferromagnetically by a hole on an oxygen site between them.

3. Results and discussion

Fig. 2 shows relatively wide scans along the $(Q\,Q\,0)$ direction in reciprocal space for $H=5$ T and $H=0$ T, at $T=6$ K. The gradual fall off of the count rate with the increase in Q is mainly due to the fact that the detector is moving away from the direct beam. The scan in Fig. 2 shows no evidence of the (110) Bragg peak due to the $\lambda/2$ component in the beam, at the nominal $(\frac{1}{2}\,\frac{1}{2}\,0)$ position. It shows, however, some other reproducible features that are basically temperature and field independent. [1] Fig. 2 shows that the scattered intensity under magnetic field is slightly stronger than that at $H=0$ T. Fig. 3 shows the difference between these two scans at $T=6$ K, and similarly at $T=84$ K ($H=7$ T), and well above the SC transition at $T=166$ K. Note that the magnetic field applied in this study at $T=84$ K is below H_{c2} (≈ 12 T according to Ref. [14]).

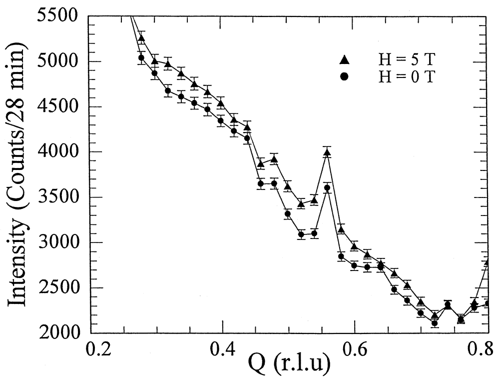

Fig. 2. In-plane scans vs. momentum transfer, Q (r.l.u. stands for reciprocal lattice units) along the (110) direction at $T=6$ K. The scans were measured under external magnetic field $H=5$ T (triangles) and at $H=0$ (circles). The $H=0$ scan was measured on a zero-field cooled sample after warming it well above T_c.

The most prominent result in this study is the observation of Q-dependent scattering below T_c, at $T=6$ K and $T=84$ K, with a peak that is roughly centered around $(\frac{1}{2}\,\frac{1}{2}\,0)$, indicating the formation of an enlarged inplane unit cell that is $\sqrt{2}\times\sqrt{2}$ the CuO_2 square lattice crystallographic unit cell. We

[1] The origin of the weak peaks observed at $Q=0.57$ and 0.8, is not clear to us yet, they might be due to multiple scattering effects.

propose that this reflection is due to short-range two-dimensional (2D) AF ordering similar to the inplane ordering observed in the parent compounds [5]. The observation of scattering at the $(\frac{1}{2}\,\frac{1}{2}\,0)$ suggests poor bilayer correlations. By contrast, in the undoped 3D AF case of $YBa_2Cu_3O_6$, the two adjacent layers are coupled in such a way that the lowest order reflection is the $(\frac{1}{2}\,\frac{1}{2}\,1)$ (see Ref. [6])[2] We hypothesize that the AF correlations $\xi_{AF}(6\text{ K}) = 8 \pm 3$ Å reflect the size of a vortex core. The vortex core diameter in a superconductor is believed to be closely related to the SC coherence length [15], ξ_{SC}, which in terms of the critical field, H_{c2}, and a fluxoid $(\Phi_0 = 2\pi\hbar/2e)$ is estimated by:

$$\xi_{SC} \approx \sqrt{\frac{\Phi_0}{\pi H_{c2}}}\,. \tag{1}$$

Supporting evidence that the scattering is due to vortex cores is that, at $T = 6$ K, the magnetic scattering at $(\frac{1}{2}\,\frac{1}{2}\,0)$ persists after lowering the magnetic field to zero, with an average count rate that is higher than that measured for a zero field cooled sample. At this temperature, the system is in the irreversible regime with a long time core-creep rate. Therefore, the zero field scan subtracted from the scan at $T = 6$ K under $H = 5$ T, was measured on a zero-field-cooled sample. To examine the feasibility of observing magnetic scattering from cores, with the present crystal, we estimate the average ordered Cu^{2+} magnetic moment $(S = 1/2)$ per flux-line, and the peak intensity (I^{diff}_{peak}). Based on the correlation length, we assume that within each core there are on average N_{AF} (≈ 10) ordered spins per CuO_2 layer, and that the total scattering is an incoherent sum over all the cores in all layers in the crystal such that:

$$I^{diff}_{peak} \approx 2 I_0 [r_0 f(Q) N_{AF}]^2 \rho_{core} V \Delta_{rod}/c. \tag{2}$$

I_0 is the neutron flux $(\approx 10^7$ neutrons/s/cm$^2)$, the factor 2 is due to the fact that there are two CuO_2 layers in each unit cell, $r_0 = 0.54 \times 10^{-12}$ cm, and the form factor $f(Q) \approx 0.9$. The 2D core density is given by $\rho_{core} = H/\Phi_0 \approx 2.5 \times 10^{11}$ cores/cm^2 at $H = 5$ T, V is the volume of the crystal, $c = 11.67$ Å, and Δ_{rod} is the fraction of the 2D rod-like signal over which the scattered beam is integrated by the detector, $(\approx 1\%)$. Eq. 2 yields an estimated value $I^{peak}_{diff} \approx 0.08$ counts/s, which compares reasonably well with the experimental value $I^{peak}_{diff} \approx 0.16$ counts/s at $T = 6$ K. The estimated upper limit of the average magnetic moment is then given by (N_{AF}/N_{fl} where N_{fl} is the total number of spins in one flux line per layer) $\mu \leq 0.004\mu_B$ per flux line in a layer.

Another intriguing result in this study is the observation that magnetic field induced featureless scattering above T_c, at $T = 166$ K, as shown in Fig. 3. The intensity falls off monotonically as a function of Q approaching zero counts at about $Q_{min} \approx 2.0$ Å^{-1}. This kind of scattering is typical of a paramagnetic system, basically yielding the form factor of an average magnetic moment of radius, R. From Q_{min}, the radius of the magnetic moment can be estimated, $R \approx \pi/Q_{min} \approx 1.6$ Å. This is about half the distance between two nearest neighbor Cu spins in the plane $(a/2 \approx 1.9$ Å). We hypothesize that this magnetic moment is formed when a hole resides between two copper spins in an oxygen site with an effective ferromagnetic coupling between such nearest neighbor copper spins, as proposed by Aharony et al. [16]. In this scenario, the magnetic moment would have an elongated shape with its principal axis either along (100) or (010), thus the scan along the (110) in Fig. 3 is probing a 45°-rotated object. The solid line through the data is calculated by a simplified form factor $(\sin(QR)/QR)^2$, where $R = (a+b)\sqrt{2} = 1.36$ Å is the projection on the diagonal of an average inplane unit cell. For such scattering to occur, the hole must reside on the same site for at least 10^{-12} s, as estimated from the energy resolution of the experimental setup. It is interesting to note that this paramagnetic component of the scattering completely disappears in the SC state, at $T = 6$ K. More studies of this magnetic field induced scattering along other directions in reciprocal space, its temperature dependence, and the hole life time on one site are required to clarify the exact nature of these magnetic entities.

In conclusion, the results of the present study suggest that the vortex cores in optimally doped

[2] In the experimental setup, the vertical resolution after the sample was relaxed to increase integration over 2D rod-like Bragg reflections.

$YBa_2Cu_3O_{7-\delta}$ might be AF, although more experimental evidence is crucial to corroborate this suggestion. In a conventional superconductor, the core vortex is believed to be a normal metal. By contrast, if the vortex core in the cuprates is AF, as our results suggest, the core has to be an insulator, since it is well established that a minute amount of carriers in the lightly doped cuprates destroy the AF order. In this picture, carriers have to be expelled out of the cores by the external magnetic field. Both the underlying AF interactions and the redistribution of carriers could shed light on the mechanism for high temperature superconductivity and a variety of peculiar experimental results, such as the anomalous Hall effect [17,18] and tunneling spectroscopy experiments [19].

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