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Nature of the quantum spin correlations through the superconducting–normal phase transition in electron-doped superconducting $Pr_{0.88}LaCe_{0.12}CuO_4$

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ABSTRACT

We use neutron scattering and specific heat measurements to relate the response of the spin fluctuations and static antiferromagnetic (AF) order to the superconductivity in the electron-doped high-transition-temperature superconductor, $Pr_{.88}LaCe_{.12}CuO_{4-\delta}$ (PLCCO) ($T_c = 24\,K$), as the system is tuned via a magnetic field applied beyond the upper critical field (H_{c2}) and driven into the normal state. The strength of the collective magnetic excitation commonly termed "resonance" decreases smoothly with increasing field and vanishes in the normal state, paralleling the behavior of the superconducting condensation energy. The suppression of superconductivity is accompanied by a smooth reduction in the very low energy spin fluctuations, and the concomitant emergence of static AF order. Our results suggest an intimate connection between the resonance and the superconducting condensation energy.

The parent compounds of high-transition-temperature (high T_c) copper oxide superconductors are Mott insulators characterized by strong AF exchange in the CuO_2 planes and static long-range AF order. Doping holes or electrons into the CuO_2 planes suppresses the static AF order and induces a superconducting phase, with energetic short-range AF spin fluctuations that peak around the AF wave vector $\boldsymbol{Q} = (1/2, 1/2)$ in the reciprocal space subset of the two-dimensional CuO_2 planes (Fig. 1a) [1]. Although the precise mechanism responsible for the formation of the superconducting condensate in the high-T_c copper oxides remains elusive, vast experimental efforts over the past two decades have shown conclusively that AF spin fluctuations play an important role within the physics of high-T_c superconductivity. One key challenge remaining, however, is to experimentally identify the nature and extent through which magnetism in these high-T_c cuprates couples to the electronic charge carriers. Specifically, the fundamental mode of coupling between either static or fluctuating magnetic order and the quasiparticles responsible for Cooper pair formation remains a pivotal focus of research in the high-T_c community.

In this paper, we briefly review our recent work in studying the evolution of spin excitations in electron-doped $Pr_{.88}LaCe_{.12}CuO_{4-\delta}$ as the system is tuned from a superconductor without static AF order into a normal state semiconductor with static/quasi-static AF order. Recently, we have systematically studied the evolution of spin excitations of PLCCO as a function of oxygen annealing process [2–6]. One of our key discoveries is the observation of a spin resonance mode in the nearly optimally doped PLCCO with $T_c = 24\,K$ equivalent to those observed in hole-doped high-T_c cuprate classes [4]. Here, we explore the relationship between the magnetic resonance mode and the corresponding condensation energy of the superconducting phase in PLCCO [6]. Through capitalizing on the relatively low fields necessary to suppress superconductivity in the electron-doped cuprates, we measured the detailed changes in static and dynamic spin behavior in PLCCO as it is driven into its field-suppressed, non-superconducting ground state.

In searching for magnetic properties that demonstrate a clear coupling to the superconducting phase in the high-T_c cuprates, one particular feature—the magnetic resonance mode—is promising. The resonance is a collective spin excitation within the CuO_2 planes of the cuprates and is observed along the commensurate AF ordering wave vector at $\boldsymbol{Q} = (0.5, 0.5)$ in the $(ab)^*$-plane. Direct coupling between the superconducting phase and the resonance excitation has been observed through its complete disappearance above T_c in optimally doped cuprate systems and through the characteristic frequency of the resonance mode's direct proportionality to T_c [7,8]. This intimate link between the energy scale of the superconducting phase and that of the

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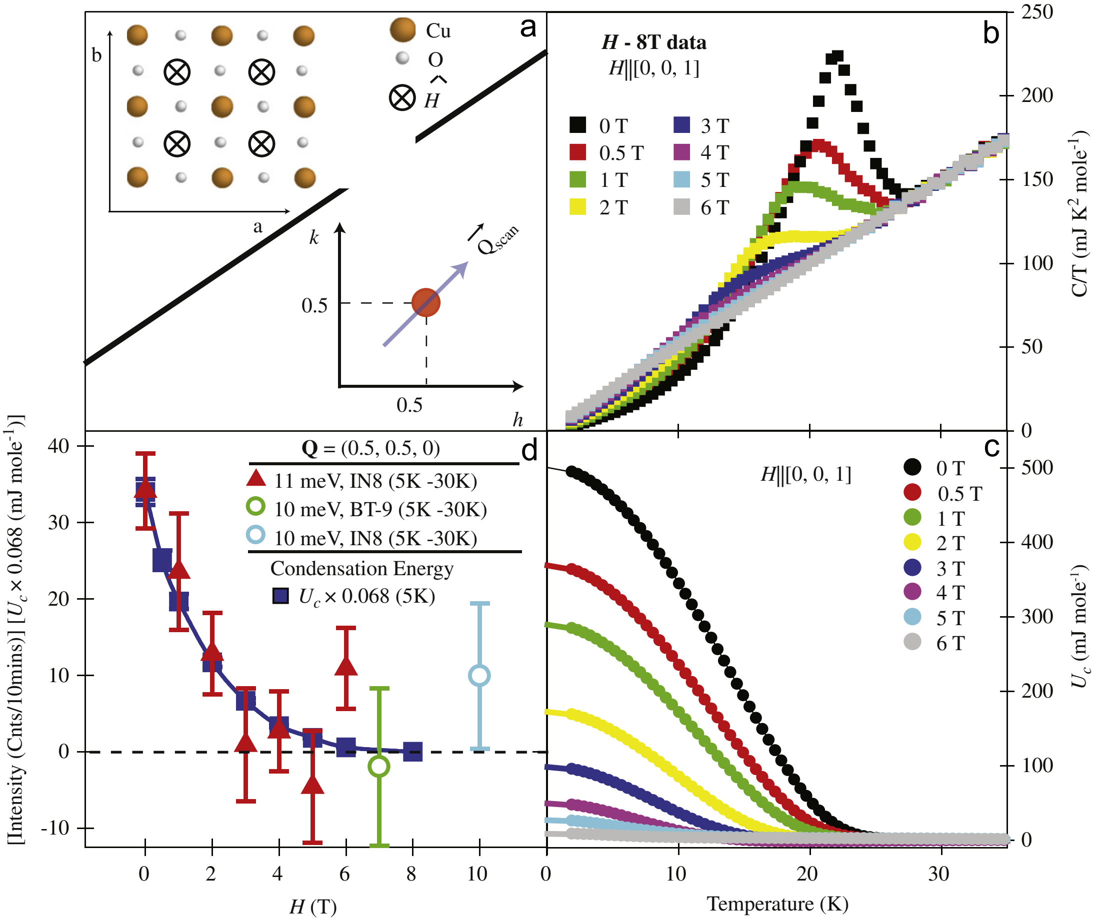

Fig. 1. (a) Upper panel: The two-dimensional CuO_2 plane. Lower panel: Schematic of typical constant-energy scans through reciprocal space. Spin excitations are centered at $\mathbf{Q} = (1/2, 1/2)$. (b) Field dependence of the total electronic specific heat versus temperature. Data taken at 8 T were established to be above H_{c2} and were used to isolate and subtract background contributions from the normal state phonon/electronic heat capacity. To obtain the normal state electronic specific heat γT, 8 T data are fitted by $C = \gamma T + \beta T^3$, where βT^3 is the phonon contribution. The resulting linear electronic contribution γT ($\gamma = 5$) was added back to the field-subtracted data to obtain the total electronic specific heat. (c) Condensation energy, U_c, determined from $U_c(T) = \int_T^{T_c+15K}[S_N(T') - S_{SC}(T')]\,dT'$ as a function of temperature at different fields. (d) Magnetic field dependence of the condensation energy (solid blue square symbols) connected by a solid line. Intensity of the resonance mode plotted as a function of applied field. Red triangles denote peak intensity measurements at $\mathbf{Q} = (1/2, 1/2, 0)$, $\hbar\omega = 11$ meV at $T = 4$ K with the normal state background at $T = 30$ K subtracted.

resonance excitation ($E_R = 5.8k_BT_c$) along with the resonance mode's universal presence in various classes of optimally doped high-T_c cuprates has led to speculation that the resonance is intimately related to electron pairing and superconductivity [9].

To demonstrate that the resonance mode is indeed related to electron–electron pairing process in the high-T_c cuprates, an experimental signature must be established that shows a definitive coupling between the resonance mode and the electronic quasiparticle behavior within the superconducting phase. At moderate field strengths, a c-axis aligned magnetic field introduces vortices into the CuO_2 planes of the cuprates and effectively weakens the superconducting state. Once the field strength reaches above the upper critical field (H_{c2}) of the system, the non-superconducting vortex regions prevent the establishment of a coherent superconducting phase. Thus, through parallel magnetic field measurements of quasiparticle excitations observed through heat capacity measurements and inelastic neutron measurements of the resonance mode, possible links between quasiparticle behavior and the resonance can be studied as a function of the suppression of superconductivity. Previous measurements on the hole-doped $YBa_2Cu_3O_{6.6}$ ($YBCO_{6.6}$) system have attempted such a study through demonstrating that the resonance mode is partially suppressed under the application of a modest c-axis aligned magnetic field; however, the mode remains completely unaffected in the presence of ab-plane aligned field [10]. While these results suggest a link between the resonance mode and the suppression of superconductivity, the experiments were unable to completely suppress T_c and the corresponding condensation energy due to the experimentally intractable H_{c2} of the YBCO system ($H_{c2} > 40$ T). Until now, the extremely high H_{c2}'s manifested by all the hole-doped cuprates have prevented any comprehensive measurement of the relationship between the superconducting condensation energy and the magnetic resonance mode.

Fortunately, we have recently demonstrated that the resonance mode is also present in electron-doped materials [4,9]. This discovery, along with the advantageous feature of an experimentally accessible H_{c2} ($H_{c2} \sim 8$–10 T) now enables us to study the detailed evolution of the resonance mode as superconductivity is driven into its field-suppressed ground state. For our experiments, we used inelastic neutron-scattering experiments to map out the evolution of the magnetic scattering function, $S(\mathbf{Q}, \omega)$, over a range of energies ($0 \leqslant \hbar\omega \leqslant 18$ meV) in electron-doped PLCCO ($a = b = 3.98$ Å, $c = 12.27$ Å; space group: $I4/mmm$) as the system is driven into its superconductivity-suppressed ground state by a magnetic field. Neutron scattering experiments were carried out around $\mathbf{Q} = (1/2, 1/2, 0)$ on the IN-8 and IN-22 thermal triple-axis spectrometers at Institute Laue-Langevin, Grenoble, France; BT-9

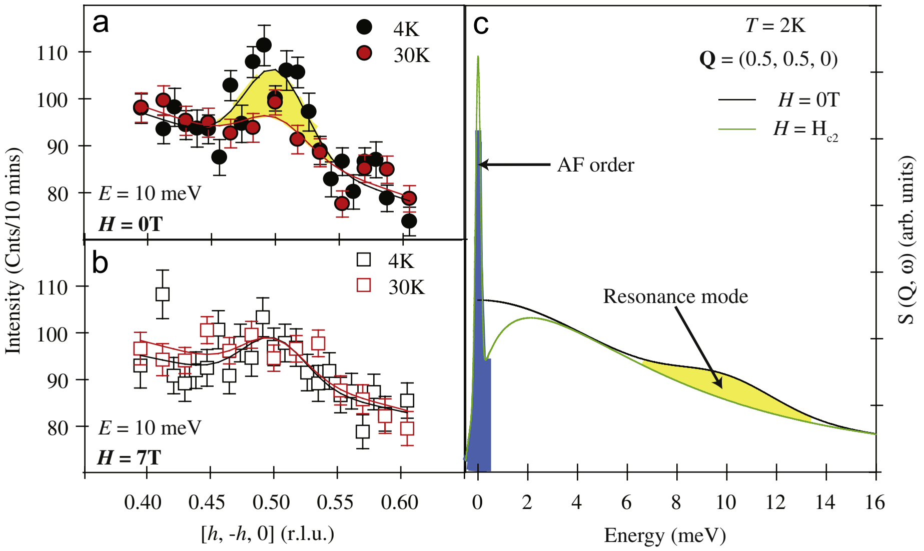

Fig. 2. (a) BT-9 momentum scans through the $\mathbf{Q} = (0.5, 0.5)$ in-plane AF ordering wave vector at the resonance energy in PLCCO, $\hbar\omega = 10$ meV. The enhancement observed upon cooling into the superconducting phase (yellow shaded region) at (0.5, 0.5) is indicative of the resonance mode. (b) The 7 T momentum scans through $\mathbf{Q} = (0.5, 0.5)$ at the resonance energy. The resonance mode has completely vanished under this field strength. (c) $S(\mathbf{Q}, \omega)$ versus energy at $T = 2$ K at the AF ordering wave vector for PLCCO under both 0 and 7 T. Upon the application of a superconductivity-suppressing field, quasi-2D AF order is stabilized, and the resonance mode is completely suppressed.

at NIST center for neutron research, Gaithersburg, Maryland; and on the V2 cold triple-axis spectrometer at Hahn-Meitner-Institut, Berlin, Germany. To compare neutron-scattering results with measurements sensitive to superconducting quasiparticles, we cut a small piece from one of the crystals [4] and carried out the superconducting heat capacity anomaly measurements at Institute of Physics, Beijing, China.

Fig. 1b shows the magnetic field dependence of the superconducting heat capacity anomaly. In the case of hole-doped superconductors, the large normal state fluctuation effect means that estimation of the superconducting condensation energy is difficult [11]. One measure of the importance of fluctuation effects in measurements of the specific heat in superconductors is to check the entropy balance condition for the quantity $(C/T)_{\text{Superconducting State}}-(C/T)_{\text{Normal State}}$ integrated up to T_c. Previous measurements on optimally electron-doped PCCO samples have shown that contrary to the case of hole-doped superconductors, such a balance is met at T_c with no need to invoke the presence of a pseudogap contribution to the "normal state" electronic density of states [12]. An analysis of the specific measurements presented in Fig. 1b confirms that a similar balance condition is also met for the PLCCO ($T_c = 24$ K) system. We can then calculate the temperature and magnetic field dependence of the superconducting condensation energy for the PLCCO (Fig. 1c and d). From these data, we find that the upper critical field (H_{c2}) necessary for the complete suppression of the superconductivity is $H_{c2}(T = 0) \sim 7$ T (Fig. 1d).

Fig. 2 shows neutron-scattering measurements probing the effect of a c-axis aligned magnetic field to the neutron-spin resonance at 10 meV in PLCCO [4]. Our results reveal that, for fields greater than H_{c2}, the resonance excitation completely vanishes. Now, over plotting the magnetic field dependence of U_c and the resonance mode in Fig. 2d surprisingly reveals that the resonance mode's rapid suppression as a function of field closely follows the suppression of U_c in PLCCO. The relative spectral weight of the resonance is therefore sensitive to the quasiparticles participating in the superconducting condensate.

Our magnetic field experiments on this PLCCO ($T_c = 24$ K) system have also revealed the emergence of a competing AF phase with the suppression of superconductivity [6]. Previous experiments on underdoped PLCCO concentrations ($T_c < 23$ K) have already demonstrated that a quasi two-dimensional AF state coexists with superconductivity [2]; whereas for nearly optimally doped PLCCO ($T_c > 23$ K) concentrations, no zero field static magnetic order exists [4]. A direct competition between this quasi-2D AF phase in underdoped samples and superconductivity was shown through an enhancement in the coexisting AF phase upon the application of a c-axis aligned magnetic field [3]. This competition, however, was only observed in underdoped systems in which the AF phase already coexisted with superconductivity under zero field. Instead, studies on optimally doped PLCCO concentrations (perhaps due to small signal or insufficient sample mass) were unable to resolve the appearance of any field-induced, competing AF phase in samples that initially possessed no AF order. Our recent neutron studies of the PLCCO ($T_c = 24$ K) system have now shown the clear appearance of a field-induced AF phase following the application of a c-axis aligned magnetic field [6]. The induced AF magnetic signal exhibits an anisotropic response with respect to the applied field direction with no observable order induced under the application of a magnetic field parallel to CuO_2 planes. This is a clear indication that the induced AF signal arises through a direct competition with the superconducting phase (which is strongly suppressed when the field is oriented parallel to c yet only slightly effected when the field is parallel to the ab-plane). This behavior is strikingly similar to the competition between the superconducting phase and the incommensurate spin density wave states in the $La_{2-x}Sr_xCuO_4$ (LSCO) hole-doped system [13], thereby suggesting that for both these monolayer hole- and electron-doped cuprates, the presence of a competing magnetic phase coexisting with superconductivity is a common feature.

Through our high-field neutron studies of the static order, low energy excitations, and resonance mode in the PLCCO ($T_c = 24$ K) cuprate, we have mapped out a comprehensive picture of the modification of spin behavior in an electron-doped cuprate superconductor as it is driven into its field-suppressed, non-superconducting ground state (Fig. 2c). Our results have shown that the spectral weight of the resonance in PLCCO is directly connected to the system's condensation energy and that the resonance mode vanishes entirely with the field-suppression of the superconducting phase. Field-induced changes in the magnetic spectra previously observed in two separate hole-doped systems, such as the suppression of the resonance in $YBa_2Cu_3O_{6.6}$ [10] and the competing static magnetic order in LSCO [14], have both been observed in this single electron-doped PLCCO system. Even more intriguing is the possible observation of the transfer of spectral weight from the suppressed resonance mode into a field-induced AF phase; however, future experiments are required to quantitatively test this. The picture through which the spin excitations and static magnetic order in the cuprates evolve as these systems are driven to be non-superconducting, provides invaluable insight into the detailed nature of the ground state from which high T_c manifests itself.

In summary, Fig. 2c plots the temperature dependence of the $S(\mathbf{Q},\omega)$ at $\mathbf{Q}=(1/2,1/2,0)$ under zero field which shows the appearance of the resonance at $\hbar\omega=10\,\text{meV}$ below T_c. After application of an $H=H_{c2}$ magnetic field, the resonance disappears and spectral weight losses at the resonance and quasi-elastic energies are compensated in part by the intensity gain at elastic AF position.

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