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Neutron spectroscopic study of the pseudogap formation in $La_{1.81}Sr_{0.15}Ho_{0.04}CuO_4$ at ambient and elevated pressure

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Abstract

In order to understand the mechanism of high-temperature superconductivity in cuprates it is important to elucidate the nature of the pseudogap formation. We have investigated the temperature dependence of the relaxation rate of crystal-field excitations in the optimally doped superconductor $La_{1.81}Sr_{0.15}Ho_{0.04}CuO_4$ at hydrostatic pressures $p = 0$ and 0.8 GPa. The observed large pressure effect on the onset of the pseudogap formation temperature T^*, together with the huge isotope effects on T^* observed earlier for oxygen isotope substitution (^{16}O versus ^{18}O), qualitatively supports models based on phonon-mediated mechanisms of the pseudogap state.

(Some figures in this article are in colour only in the electronic version)

1. Introduction

Since the discovery of high-temperature superconductivity (HTSC) in cuprates [1], a wide variety of experimental techniques have demonstrated marked changes in their physical properties at temperatures $T_C < T \lesssim T^*$, suggesting a pseudogap-type electronic structure [2]. The existence of the pseudogap, a partial suppression of the accessible electronic states at the Fermi level in the normal state, is one of the most challenging problems in efforts to understand the microscopic pairing mechanism. Theoretical models of the origin of the pseudogap state are as diverse as the proposed pairing mechanisms in these materials [3]. Consequently, in order to identify the mechanism of the HTSC in doped cuprates, experiments which produce changes of the pseudogap temperature T^* are currently of particular importance.

Recently, a large oxygen isotope effect on T^* has been observed in $La_{1.94}Sr_{0.06}CuO_4$ by means of XANES [4] as well as in $La_{1.81}Sr_{0.15}Ho_{0.04}CuO_4$ and $HoBa_2Cu_4O_8$ by neutron crystalline-electric-field (CEF) spectroscopy [5, 6]. This oxygen isotope shift ΔT^* was

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quantitatively accounted for using models based on phonon-mediated mechanisms of the pseudogap formation, i.e. dynamical charge ordering [7], phonon-induced stripe formation [8] and bipolaron formation [9]. An essential ingredient in all these models is a characteristic phonon mode with energy $\hbar\omega_p$ that interacts with the charge carriers. Another experimental technique which affects the lattice degree of freedom is the application of hydrostatic pressure. To our knowledge, the results of very few pressure dependence experiments are controversial at the present time. The pressure dependence of the pseudogap temperature dT^*/dp obtained from NQR measurements for $YBa_2Cu_4O_8$ [10], as well as from the resistivity of quenched and Ca-doped $YBa_2Cu_3O_7$ compounds [11], is negative. In contrast, resistivity measurements for optimally doped $Hg_{0.82}Re_{0.12}Ba_2Ca_2Cu_3O_{8+\delta}$ show a linear increase of T^* with pressure [12].

The CEF transitions measured by the inelastic neutron scattering (INS) technique exhibit line broadening due to the interaction with the charge carriers, thus being a direct probe of the electronic density of states at the Fermi energy. The principle of neutron spectroscopic investigations in the application to rare-earth based HTSC is beyond the scope of this report and can be found in recent review articles [13–15]. In this paper we report on INS investigations of the pressure effect on the relaxation rate of CEF excitations in optimally doped $La_{1.81}Sr_{0.15}Ho_{0.04}CuO_4$.

2. Experimental details

For the present experiment the HTSC compounds $La_{1.85-x}Sr_{0.15}Ho_xCuO_4$, $x = 0$, 0.04, were synthesized by means of a conventional solid-state reaction, starting with high-purity oxides La_2O_3, Ho_2O_3, CuO and strontium carbonate $SrCO_3$. The powders were mixed, ground thoroughly, pressed into pellets and fired in air at 850 °C (30 h), 900 °C (40 h) and 950 °C (50 h) with intermediate grindings. The phase purity of the synthesis product was controlled by means of x-ray diffraction. The superconducting properties of the samples obtained were verified using ac magnetic susceptibility. The measurements were carried out on a Quantum Design PPMS system using a clamp cell made of non-magnetic Cu/Be alloy (pressures up to 1 GPa). The superconducting transition of the lead was used to measure the pressure [16].

The high-energy-transfer INS spectra at ambient pressure were measured on the MARI chopper spectrometer (incident beam energy of 100 meV) at the spallation neutron source ISIS, Rutherford Appleton Laboratory, UK. Inelastic neutron spectra obtained from $La_{1.85}Sr_{0.15}CuO_4$ were used to establish the phonon density of states. The powder samples were mounted inside an aluminium can on the cold head of a helium closed-cycle refrigerator, and all measurements were performed at temperatures of 14 and 100 K.

The pressure dependence of the high-resolution INS spectra associated with the transition from the ground state to the lowest excited level was measured on the time-of-flight spectrometer FOCUS installed at the spallation neutron source SINQ at Paul Scherrer-Institut, Villigen, Switzerland. The data were collected in the temperature interval of 10–100 K (10–70 K for $p = 0.8$ GPa) using an incoming neutron wavelength $\lambda = 5.75$ Å, giving an energy resolution at the elastic position (full width at half-maximum (FWHM)) of 50 μeV. The axially symmetric pressure cell made of hardened aluminium with an inner diameter of 7.2 mm allowed a total sample volume of about 1600 mm^3 [17]. Fluorinert FC-77 was used as a pressure medium. The pressure was determined from the shift in the lattice parameters of NaCl added to the sample [18]. The raw data were corrected for absorption, detector efficiency and detailed balance effects according to standard procedures.

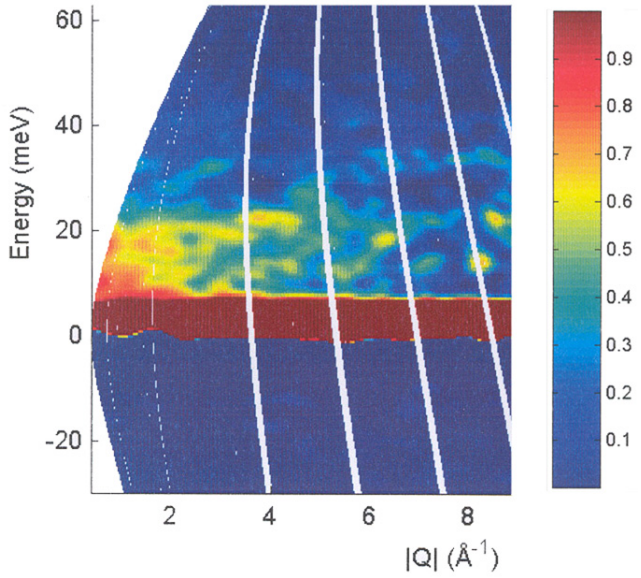

Figure 1. A contour plot of the magnetic excitations obtained at 14 K in $La_{1.81}Sr_{0.15}Ho_{0.04}CuO_4$. The non-magnetic scattering measured for $La_{1.85}Sr_{0.15}CuO_4$ has been subtracted.

3. Results and discussion

The ac susceptibility measurements revealed that the dependence of T_C on the pressure ($T_C = 32.5 \pm 0.3$ and 35.2 ± 0.3 K for $p = 0$ and 1.0 GPa, respectively) is in an agreement with previously published results [19, 20].

The CEF Hamiltonian of Ho^{3+} in $La_{2-x}Sr_xCuO_4$ has not been established experimentally so far, since most of the transition matrix elements have extremely small values. Instead, we derived the CEF level scheme by extrapolation from that of $HoBa_2Cu_4O_8$ [21] based on the procedure described in [14] and [15]. This extrapolation procedure predicts the first excited CEF level to lie at 0.2 meV, well separated from the next excited level by about 10 meV. The measurements on the MARI spectrometer were thus needed to establish the upper energy limit of the CEF excitations in the $La_{1.81}Sr_{0.15}Ho_{0.04}CuO_4$ and to prove the viability of the extrapolated level scheme. Figure 1 shows a typical contour plot summarizing the data taken for $La_{1.81}Sr_{0.15}Ho_{0.04}CuO_4$ at a temperature of 14 K. Magnetic scattering is most prominent at small values of the scattering vector Q; thus we conclude that ground state CEF excitations occur at energies below 25 meV. The energy spectrum in the low-energy window ($\Delta E < 10$ meV) exhibits just one inelastic peak at $\sim$0.2 meV, which corresponds to the lowest-lying CEF transition, in full agreement with the extrapolation mentioned above. Thus, the analysis of the linewidth of this CEF excitation could be done in the framework of a two-level approximation.

Figure 2 shows the evolution of the lowest-lying CEF transition with temperature at $p = 0$ and 0.8 GPa. The inelastic peak was described by a convolution of the temperature-independent Gaussian function, due to instrumental resolution and local structural distortions around the Ho^{3+} ions resulting from the doping with Sr^{2+} ions ($\sim$150 μeV), and a Lorentzian broadening function. The position, amplitude and intrinsic linewidth of the Lorentzian were obtained from a least-squares fitting procedure at each temperature.

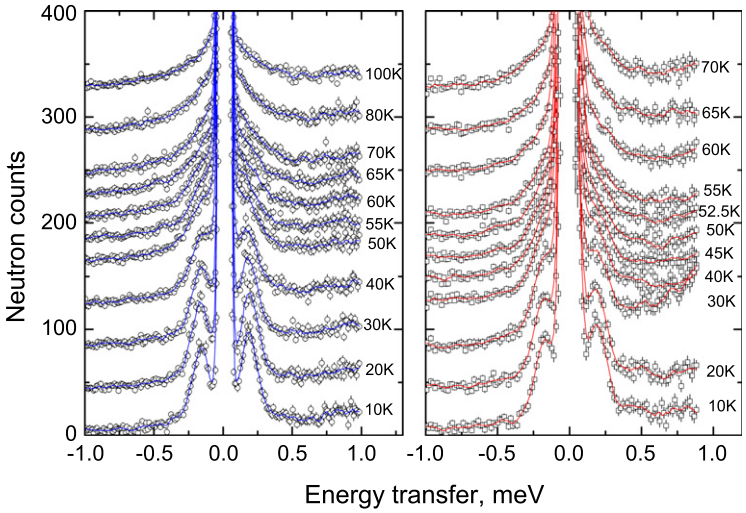

Figure 2. Evolution of the energy spectra of neutrons with temperature in the $La_{1.81}Sr_{0.15}Ho_{0.04}CuO_4$ at ambient pressure (left panel) and $p = 0.8$ GPa (right panel) as taken on the TOF spectrometer FOCUS at SINQ. The solid curves are the results of a least-squares fitting procedure as explained in the text.

In the two-level approximation the reduction in linewidth of the CEF transition caused by the isotropic superconducting (pseudo)gap is given by

$$\Gamma_s(T) = \Gamma_n(T) \exp\left(-\frac{\Delta}{k_B T}\right), \tag{1}$$

where Γ_n is the intrinsic CEF linewidth in the normal state and Δ is an isotropic energy gap [5]. Note that for a d-wave anisotropic gap function [22] certain relaxation channels exist even at the lowest temperature, and the calculation of Γ_s requires an integration in k-space as described in detail by Mesot *et al* [23].

For the normal state, on the other hand, the linewidth of the CEF transition with energy $\hbar\omega \ll k_B T$ increases linearly with temperature according to the Korringa law

$$\Gamma_n(T) = 4\pi M_{ij}^2 J_{ex} N^2(E_F) k_B T, \tag{2}$$

where M_{ij} is the matrix element of the $|i\rangle \rightarrow |j\rangle$ CEF transition, J_{ex} is the exchange integral for the 4f electrons of the Ho^{3+} ions and $N(E_F)$ is the density of states at the Fermi level [24, 25]. Therefore, the pseudogap formation temperature T^* can be directly obtained from the temperature dependence of the CEF linewidth, as the temperature where the curve starts to deviate from the linear behaviour.

The temperature dependence of the intrinsic linewidth together with the fitted values of the pseudogap formation temperature T^* (according to equations (1), (2) and [23]) are shown in figure 3. It is seen from equation (1) that the temperature dependence of the linewidth of a CEF transition caused by the pseudogap should have an exponential form. Therefore, the use of a logarithmic scale indicates this fact visually. At the same time, the linear behaviour of the linewidth above T^* is not bent much in the short temperature interval. In our analysis we use the fact that hydrostatic pressure application does not affect the slope of the Korringa line $d\Gamma_n/dT \sim N^2(E_F)$ (see equation (2)) [19]. Thus, we found the pressure-induced shift of the pseudogap temperature $dT^*/dp = (-7.1 \pm 1.6)$ K GPa^{-1}, in reasonable agreement with $dT^*/dp \sim -10$ K GPa^{-1} determined for $YBa_2Cu_4O_8$ [10]. The opposite effects of

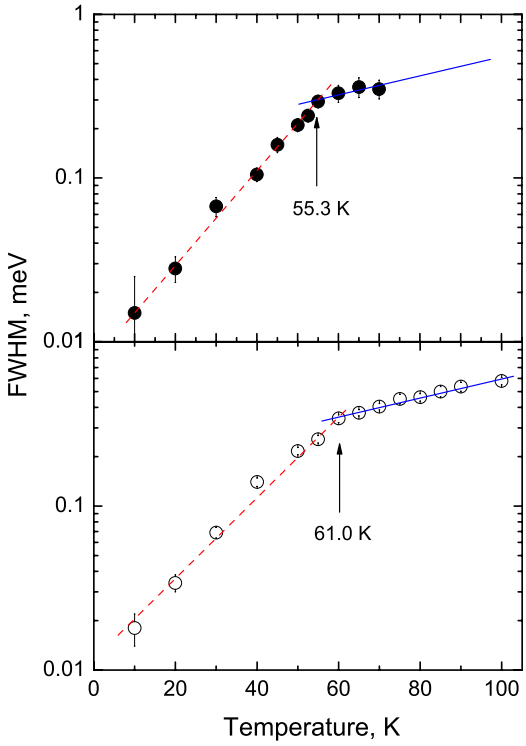

Figure 3. The temperature dependence of the intrinsic linewidth corresponding to the lowest ground state CEF transition in $La_{1.81}Sr_{0.15}Ho_{0.04}CuO_4$ at ambient pressure (lower panel) and at $p = 0.8$ GPa (upper panel).

pressure and isotope substitution can be correlated with the characteristic phonon energy $\hbar\omega_p$. Replacement of ^{16}O by ^{18}O reduces $\hbar\omega_p$ and results in an enhancement of T^* [7–9], while the application of hydrostatic pressure increases $\hbar\omega_p$ by a factor $\gamma\Delta V/V$, where γ is the Grüneisen parameter and $\Delta V/V$ is the pressure-induced volume change. It should be noted that the temperature dependence of the relaxation rate does not exhibit an anomalous behaviour around T_C, in contrast to the case for conventional superconductors. This is in agreement with the ARPES data obtained on $Bi_2Sr_2CaCu_2O_{8-\delta}$ [22] and supports the idea that the superconducting gap emerges smoothly from the normal state pseudogap [26].

4. Conclusion

In conclusion, high-resolution neutron spectroscopy was applied to study the relaxation rate of the lowest-lying CEF excitation of Ho^{3+} ions in the $La_{1.81}Sr_{0.15}Ho_{0.04}CuO_4$ HTSC compound at pressures $p = 0$ and 0.8 GPa. The observed large pressure effect, together with the corresponding oxygen isotope shift (^{16}O versus ^{18}O) of T^* found earlier [4, 5], suggest that a phonon-mediated pairing mechanism plays an important role in the pseudogap formation.

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