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Physica C 223 (1994) 106–110

PHYSICA C

Compton-profile measurements on $(Bi_{1.6}Pb_{0.4})Sr_2Ca_2Cu_3O_{10+x}$ superconductor

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Received 4 January 1994; revised manuscript received 1 February 1994

Abstract

In this paper the measured Compton profiles (CP's) of the ceramic superconductor $(Bi_{1.6}Pb_{0.4})Sr_2Ca_2Cu_3O_{10+x}$ above and below T_c are presented. The observed difference in the CP's between the normal and superconducting state suggests a redistribution of the electron momentum during the superconducting transition.

1. Introduction

Since the discovery of HT_c superconductivity by Bednorz and Muller [1] and enormous amount of activity has been devoted to the investigation of the electronic structure and related physical parameters of systems showing a similar behavior using various experimental techniques [2]. In this work we present the first measurements on $(Bi_{1.6}Pb_{0.4})Sr_2Ca_2Cu_3O_{10+x}$, a ceramic high-$T_c$ superconductor, using Compton-scattering spectroscopy.

In a Compton-scattering experiment, the intensity of the inelastically scattered photons is measured for a fixed scattering angle, as a function of their energy. This energy spectrum can be converted to a momentum scale (Compton profile) and is directly related to the momentum distribution of the electrons of the scattering material.

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The Compton profile $J(p_z)$ is the projection of the scatterers electron momentum distribution $n(\boldsymbol{p})$ along the scattering vector $\boldsymbol{k}$ (generally chosen as the z-axis):

$$J(p_z)=\int_{p_x}\int_{p_y} n(p_x,p_y,p_z)\,\mathrm{d}p_x\,\mathrm{d}p_y\,. \qquad (1)$$

In the case of an isotropic system Eq. (1) is usually rewritten in terms of the radial momentum distribution $I(p)=4\pi p^2 n(p)$ and a scalar momentum variable $q=\boldsymbol{kp}/|\boldsymbol{k}|\,|\boldsymbol{p}|$. Since $n(\boldsymbol{p})$ and $I(p)$ are probability distributions the Compton profile is subject to the normalization rules [3]

$$\int_{-\infty}^{+\infty} J(p_z)\,\mathrm{d}p_z=\int_{-\infty}^{+\infty} J(q)\,\mathrm{d}q=Z\,, \qquad (2)$$

where Z is the total number of scattering electrons.

In this paper we report measurements of the Compton profile (CP) for the ceramic superconduc-

SSDI 0921-4534(94)00038-H

tor $(Bi_{1.6}Pb_{0.4})Sr_2Ca_2Cu_3O_{10+x}$ in the normal and superconducting state. A difference in the CP's between these two states is observed. In the discussion following the experimental results, the significance of this difference is pointed out. Possible causes to which this difference might be due are also presented together with an attempt to interpret it.

2. Experimental procedure

The experimental set-up comprises a Compton spectrometer with a HPGe detector and a liquid-nitrogen cryostat. The ^{241}Am Compton spectrometer used in this experiment has been described elsewhere [4]. The collimation system has a mean scattering angle of 160° and an FWHM of 3°. The total resolution of the detecting system is 0.54 a.u. of momentum. The channel width was fixed at 61.2 eV while the drift of the detecting system was found to be less than 0.1 channels at 59.54 keV.

The $(Bi_{1.6}Pb_{0.4})Sr_2Ca_2Cu_3O_{10+x}$ sample used in this study was prepared by the well known solid-state sintering technique in the Laboratoire de Physique de Solides de Bellevue. The necessary amount of raw materials, Bi_2O_3, PbO, $SrCO_3$, $CaCO_3$ and CuO were thoroughly mixed in an agate mortar and calcinated in an alumina boat at 850°C for about 15 h. This was followed by another grinding and pressing into pellets. The pellets were sintered at 850°C in air for about 120 h and cooled at a rate of 80°C/h. The resulting product had the following lattice parameters: $a=b=5.4$ and $c=37$ Å. From the X-ray diffraction pattern, the sample was estimated to contain at least 75% of the (2223) phase. Both the SQUID and the resistivity data showed a T_c of 107 K [5].

The sample, in the form of rectangular bar (2×2 cm, 0.2 cm thick) was attached to a liquid-nitrogen cryostat. The temperature of the sample was continuously monitored and always stabilized at 83 K and at room temperature (295 K) for the superconducting and normal-state measurements, respectively.

Compton-profile measurements have been repeated for each state several times. A resistivity measurements of the sample against temperature at the end of each cycle was used to ensure that the sample was retaining its superconducting properties. No change of T_c was observed in any set of measurements. In order to improve the statistical accuracy, the independent measurements sets for each state have been added. More than 4×10^6 counts for the total of six sets of measurements taken were accumulated under each Compton profile.

Data processing includes background subtraction from the raw data of the measured spectra as well as corrections for the energy-dependent Compton cross-section and absorption in the sample. The signal-to-noise ratio was 600:1. The above mentioned corrections practically did not change the difference of the profiles. In order to avoid accumulation of errors the spectra were not corrected for the finite resolution of the spectrometer.

The two Compton profiles were normalized to the total number of electrons between −7 a.u. and +7 a.u. The total number of electrons is 424 and corresponds to all electrons, valence and core, of the elements Bi, Pb, Sr, Ca, Cu, and O contributing to the Compton profile of the compound, except the K and L electrons of Bi and Pb, the K of Sr, and the K electrons of the Cu from 3.2 to 7.0 a.u. of momentum which are not excited since their binding energies are greater than the transferred energy.

3. Results and discussion

In order to detect any change, if existing, in the Compton profile of the ceramic superconductor $(Bi_{1.6}Pb_{0.4})Sr_2Ca_2Cu_3O_{10+x}$ between the two states, the difference $\Delta J(q)$ is taken by subtracting point by point the normalized CP of the superconducting state from the normal one. This difference expressed as a percentage of the Compton peak $J(0)$ at 295 K, versus q (projection of the electron momentum on the scattering vector) is shown in Fig. 1. Fig. 1(a) shows the entire spectrum, which is symmetric to the Compton peak (0.0 a.u.) while Fig. 1(b) shows the same spectrum folded (addition of the two symmetric parts).

A change of 1% is revealed around $p=0.0$ a.u. as well as smaller differences between 1 and 4 a.u. of momentum. The error bars correspond to 2σ ($\pm\sigma$) in all figures. It is obvious, considering the normalization process, that the two areas, above and below the axis q, are equal.

Compton scattering is expected to be temperature

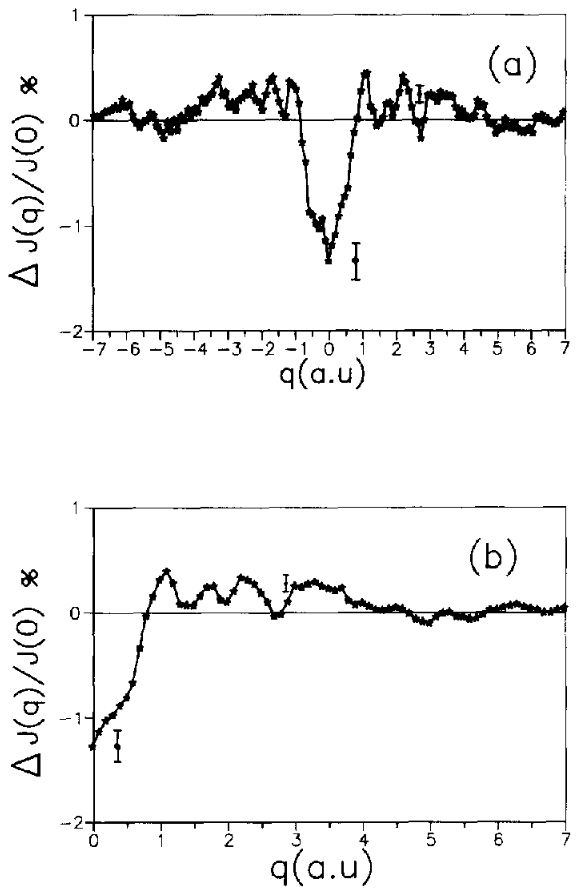

Fig. 1. Difference of Compton profiles for $(Bi_{1.6}Pb_{0.4})Sr_2Ca_2Cu_3O_{10+x}$ between normal (295 K) and superconducting state (83 K) as a percentage of the Compton peak $J(0)$. (a) The entire spectrum; (b) spectrum folded around $J(0)$. Lines are drawn as eye guides.

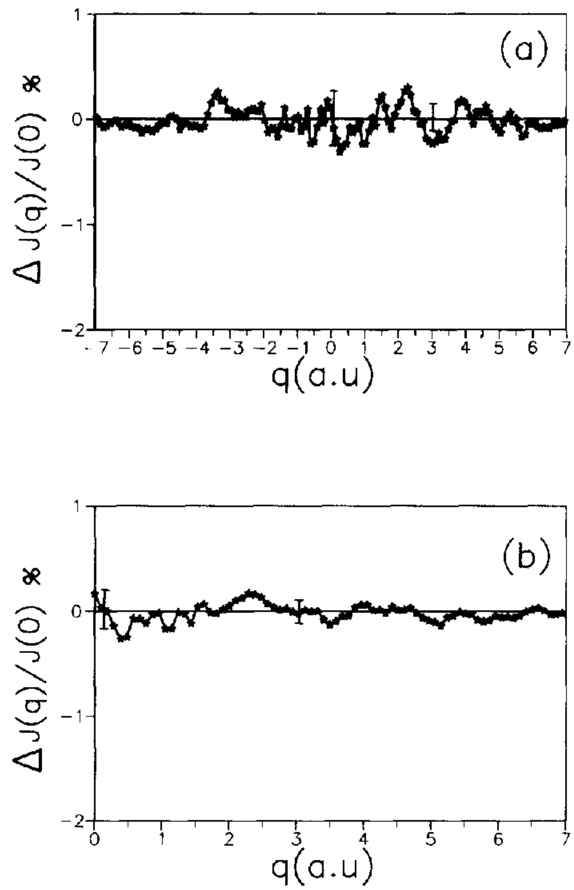

Fig. 2. Difference of Compton profiles for polycrystalline Copper between 295 K and 80 K as a percentage of the Compton peak $J(0)$. (a) The entire spectrum; (b) spectrum folded around $J(0)$. Lines are drawn as eye guides.

independent. The temperature dependence in Compton scattering is a second-order effect involving a photon–phonon–electron or photon–electron–phonon scattering and should increase as the temperature increases. If this contribution was affecting the observed CP of the copper oxide superconductor this should also affect the CP of another material having a characteristic Debye temperature near room temperature (i.e. copper, $\Theta_D = 315$ K) although these materials are quite different.

As a reliability test of the spectrometer, Compton profiles of polycrystalline Cu were obtained at 80 K and room temperature using the same experimental procedure and data processing. The subtraction result of these two Compton profiles is shown in Fig. 2. No changes were observed. Any fluctuations are within the limit of the experimental error.

As an additional confirmation that the observed difference is not due to temperature-dependent effects we have taken measurements on the high-T_c material at the intermediate temperature of 155 K. The difference $\Delta J(q)$ obtained by subtracting the CP taken at 155 K from the CP at 295 K expressed as a percentage of $J(0)$ at 295 is plotted in Fig. 3. No changes are observed. Again, any fluctuations fall within the limits of the experimental errors.

The results presented in Figs. 2 and 3 indicate that the observed changes shown in Fig. 1 cannot be attributed to parasitic or any temperature-dependent effect. These changes are attributed to the superconducting transition.

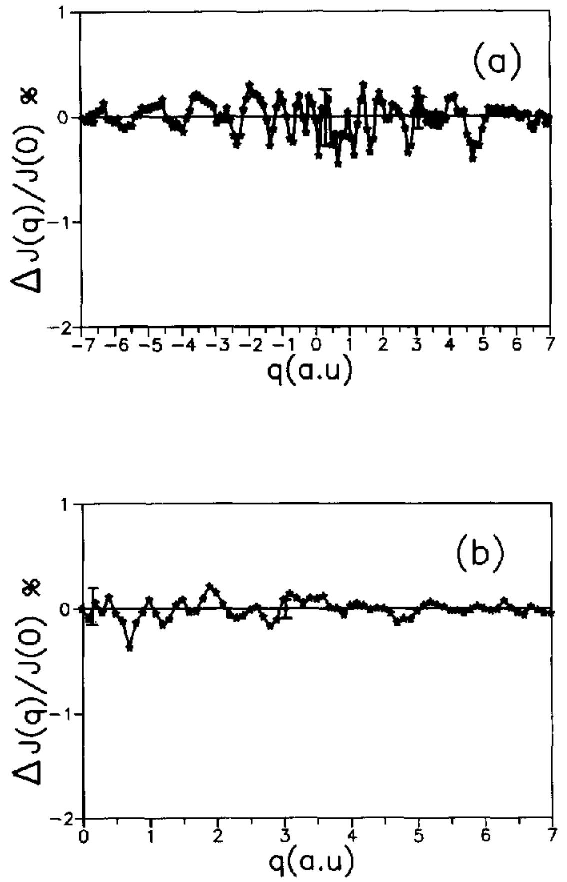

Fig. 3. Difference of Compton profiles for $(Bi_{1.6}Pb_{0.4})Sr_2Ca_2Cu_3O_{10+x}$ between 295 K and 155 K as a percentage of the Compton peak $J(0)$. (a) The entire spectrum; (b) spectrum folded around $J(0)$. Lines are drawn as eye guides.

It is clear that by taking the difference of the Compton profiles the contribution of the core electrons is cancelled and the only changes that remain are those due to electrons responsible for the superconductivity. Thus, as the Compton profile is a measure of the electron-momentum distribution (EMD) the observed difference of the CP's shown in Fig. 1 suggests a redistribution of the EMD of the electrons responsible for the superconductivity.

Tang et al. [6] reported a difference on Doppler-broadening annihilation radiation (DBAR) for the $Bi_{1.7}Pb_{0.3}Sr_2Ca_2Cu_3O_{8+\delta}$ between the normal (300 K) and the superconducting (77 K) state. They attributed this difference to a change of the EMD but their results are in disagreement with ours. This fact may be due to the inherent complexity of positron-annihilation spectroscopy [7,8] where the positron–electron momentum density is measured instead of the EMD as in Compton scattering. Another reason, related to the above, could be that they have taken measurements in only two quite distant temperatures and consequently one cannot rule out the influence of positron trapping by temperature-dependent defects.

The observed difference $\Delta J(q)$ in our results cannot be explained in the context of the BCS theory of superconductivity. According to BCS the EMD in the superconducting state crosses the Fermi surface with a finite slope [9] instead of having a sharp cut-off at p_F as is the case in the normal state. For a high-T_c superconductor ($T_c=100$ K, $\xi_0=10$ Å) the above mentioned change of the slope is of the order of $\delta p/p_F=10^{-2}$, a change that cannot be observed with current Compton-scattering experiments resolution. However, even if this change of the slope were detectable, its effect on the difference of the two CP's $\Delta J(q)$ (as defined in the present work), would have the opposite sign of the observed one.

The experimentally observed difference $\Delta J(q)$ between the Compton profiles which suggests a redistribution of the EMD of the electrons responsible for the superconductivity may be due to one or both of the following causes:

(1) Structural changes below T_c, or

(2) changes in the electronic density. In an attempt to give an interpretation of the experimental results, we continue with some speculation as follows.

It is well known that valence electrons, especially of s or p character, contribute to the Compton profile up to 1.0 a.u. of momentum. Valence electrons may contribute to higher momentum components say 2–4 a.u. of CP via an Umklapp process if they are in d character states [10]. The d electron states of the Cu–O bands fall in this case. In this context the results shown in Fig. 1(b) can be interpreted by assuming that there are more electrons in d-like states above T_c while below T_c we have more electrons in p-like states.

The above interpretation could be valid if local structural changes below T_c are accepted [11]. In that way electrons of d-like states in Cu–O bands which contribute to momentum values higher than 1.0 a.u. above T_c are diminished below T_c as they are in increased coupling with electrons of p-like states of Bi–O bands [12]. Thus for $T<T_c$ more electrons in p-

like states appear, contributing to Compton profile around $p=0.0$ a.u.

In conclusion we would like to point out that Compton-profile measurements can give unique information for the high-T_c superconductors behavior. However, for a more detailed interpretation of the results presented, more experimental work is needed on other superconducting compounds and especially monocrystals.

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