

# NON-BCS BEHAVIOUR OF THE SUPERCONDUCTING ORDER PARAMETER IN YBa<sub>2</sub>Cu<sub>3</sub>O<sub>7</sub> STUDIED WITH INFRARED SPECTROSCOPY

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We report on detailed measurements of the temperature dependence of the infrared reflectivity spectra of YBa<sub>2</sub>Cu<sub>3</sub>O<sub>7</sub>. It is shown that the prominent edge<sup>1</sup> at  $8k_BT_c$  shows practically no energy shift as a function of temperature, whereas the oscillator strength of this feature depends strongly on temperature with a marked critical behavior at the superconducting transition temperature. This behavior coincides with the temperature dependence of the superfluid fraction, which was determined from the spectra using two different methods. A precursor at the same energy position persists at temperatures above  $T_c$ . If the edge can be interpreted as the characteristic energy required to break up a pair, we arrive at the conclusion that the gap itself can not be considered as an order parameter in this case. The superfluid density on the other hand changes approximately as  $1 - (T/T_c)^4$ . We point out that a remarkable analogy exists to the situation in itinerant magnetism, where short range order persists above the Curie temperature resulting in an exchange splitting that does not vanish<sup>2</sup> at T<sub>c</sub>.

## 1. EXPERIMENTAL RESULTS

C-axis oriented thin films with thicknesses between 2000 Å and 4000 Å were prepared using the pulsed laser deposition technique set up for *in situ* Y-Ba-Cu-O thin film growth. Experimental details have been published elsewhere.<sup>3</sup> Characterization of the films using scanning electron microscopy, optical microscopy, and X-ray diffraction revealed, that the films are single phase and c-axis oriented with a smooth surface. The substrates are single crystals of SrTiO<sub>3</sub> and twinned crystals of LaAlO<sub>3</sub> ([100] surface) with a wedged backside. Transmission electron microscopic measurements demonstrated a well defined substrate/film interface on an atomic scale. T<sub>c</sub> of the samples was 89 K.

Reflectivities were measured in the range of 100 to 7000 cm<sup>-1</sup>. The data and details on data handling have been published elsewhere<sup>4</sup>. The spectra were taken at 5 K intervals between 20 and 150 K, allowing a detailed study of the temperature dependence of the reflectivity as well as the optical conductivity. The spectra are dominated by an absorption edge which rises steeply between 400 and  $800cm^{-1}$ . The nature of this edge has been extensively discussed<sup>5</sup>, and to date no concensus on the interpretation has been reached. From the spectra three important quantities related to the order parameter were determined:

- The superfluid fraction, using two different methods (see Ref.4 for details). Both methods give the same result, which is displayed in Fig. 1a. The temperature dependency closely follows a 1 (T/T<sub>c</sub>)<sup>4</sup> behaviour.
- The energy position of the above mentioned edge. Unlike the behaviour of the gap in a BCS superconductor the position of this edge is almost temperature independent. The result is displayed in Fig. 1a.
- The height (or oscillator strenght) of this edge, which very closely follows the temperature dependency of the superfluid fraction including the critical behaviour at  $T_c$ . This demonstrates an intimate relationship of the edge structure to the order parameter, although the relation is clearly very different from the gap-closing behaviour in a BCS superconductor. We have to add here, that a small precursor of the same edge structure exists already above  $T_c$ .

## 2. DISCUSSION

In weak and strong coupling BCS theory the absorption threshold corresponds to twice the superconducting gap parameter  $\Delta$  in the former case <sup>6</sup>,

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### FIGURE 1

Normalized amplitudes of the order parameter and energy splittings of a superconductor and an itinerant ferromagnet.

(a) Crosses: Superfluid fraction of YBa<sub>2</sub>Cu<sub>3</sub>O<sub>7</sub>. Open circles: Height of the threshold at 400 cm<sup>-1</sup>. The height was normalized to the conductivity at 95 K. The negative value at  $T > T_c$  reflects a weak edge structure above the superconducting transition. Bars: Energy of the edge. The length of the bars reflects the estimated accuracy. (b) Closed circles: The experimentally determined exchange splitting of the Ni 3d bands, using photo-electron spectroscopy. The results were taken from Ref. 2. The solid curve is a guide to the eye. Lower solid curve: Saturation magnetization of Ni. or to a Holstein process tied to  $2\Delta$  in the latter case<sup>7,8</sup>. In either case one expects the edge to shift considerably when  $T_c$  is approached, and close to the transition temperature  $\Delta$  is proportional to  $\sqrt{(n_s)}$ . So in this respect there is no one-to-one correspondence between the  $8k_BT_c$  edge structure and a BCS gap.

Although a microscopic understanding of the behavior of the gap is lacking, and no satisfactory agreement was found at all temperatures with either strong coupling theory or weak coupling theory, a fairly good fit could be made with the following empirical expression for the dielectric function<sup>4</sup>

$$\epsilon(\nu) = f_s(T)\epsilon_s + (1 - f_s(T))\epsilon_n$$

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where  $f_s(T)$  is the temperature dependent superconducting component, which we take proportional to  $1 - (T/T_c)^4$ ,  $\epsilon_s$  is the dielectric function of the superconducting component, and  $\epsilon_n$  is the dielectric function of the normal component. In Ref. 4 it is shown that these dielectric functions of the two components can be expressed using simple assumptions of weak coupling theory with a large and temperature independent gap for the superconducting component and Drude theory for the normal component. The phenomenology underlying the properties of the superconducting condensate as displayed in Fig. 1a, calls for an analogy with other strongly correlated systems near the phase transition. In that context we like to discuss an interesting analogy of the present system to itinerant ferromagnetism of materials with a strong on-site exchange interaction, where again an interplay between short range interactions and long range coherence takes place.

One of the simplest mean field treatments of a ferromagnet is the Stoner model. In this model an overall magnetization is created via an exchange splitting, resulting in a shift in energy of the spin-up bands relative to the spin-down bands<sup>9</sup>. The magnetization then follows from a different filling of the spin-split bands. Clearly in such a model the magnetization and the exchange splitting are proportional to each other. This is not unlike the situation in an ordinary BCS superconductor, where the superconducting gap parameter  $\Delta$  is proportional to  $\sqrt{(n_s)}$ . Interestingly the transition metals on the right side of the 3d series do not behave like this, as has been beatifully demonstrated by Eastman et al. who used photo-emission to determine the exchange splitting of the Ni 3d bands as a function of temperature<sup>2</sup>. What is observed in those cases is, that the exchange splitting persists above the Curie temperature. This has been understood as a consequence of the fact, that there is

still short range magnetic order above  $T_c$ , although the long range order is lost. The results of Eastman et al. were replotted in Fig. 1b to facilitate comparison to our data. The close similarity of both plots seems to indicate, that the cuprate high  $T_c$  superconductors are characterized by a phase transition where some short range superconducting order persists above T<sub>c</sub>, although the long range order, as reflected in the superfluid density, disappears more or less in the usual way at T<sub>c</sub>. So far no bulk superconductors are known that exhibit a phase transition between states with long range order and short range order. On the other hand, this is a well studied phenomenon in 2-dimensional arrays of Josephson junctions<sup>10</sup>. In these systems a Kosterlitz-Thouless transition takes place below the transition temperature of the superconducting islands.

There are at least two factors which distinguish the high  $T_c$  cuprates from ordinary superconductors, and which may favor the rather unusual phenomenology outlined above. The first is the scale of the coherence length, which is of comparable magnitude as the average distance between charge carriers. The second is the layered two-dimensional nature of these materials. Due to the small coherence length Cooper-pairs have much smaller overlap than in classical superconductors, where the coherence length is of the order of thousands of lattice spacings. In a number of theoretical papers<sup>11,12,13</sup> such a situation has been adressed, although superconducvity in this regime of parameter space has not been fully understood.

## 3. CONCLUSIONS

The temperature depedency of the superfluid component and the position of the gap as observed in YBa<sub>2</sub>Cu<sub>3</sub>O<sub>7</sub> as observed with infrared spectroscopy is demonstrated to show non-BCS like behavior. The gap position does not shift with temperature, while the superfluid component is well approximated with a  $1 - (T/T_c)^4$  behavior. A small signature at the gap position below  $T_c$  persists above  $T_c$ . It is discussed that this behavior may indicate that we have a situation analogous to what is observed in certain itinirant ferromagnets, where the long range order vanishes at  $T_c$ , whereas the short range order persists above the transistion temperature resulting in a non-vanishing exchange splitting.

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