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1995 J. Phys.: Condens. Matter 7 L365

(http://iopscience.iop.org/0953-8984/7/27/003)

J. Phys.: Condens. Matter 7 (1995) L365–L369. Printed in the UK

LETTER TO THE EDITOR

Elastic light scattering in the YBCO system

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Received 13 March 1995, in final form 15 May 1995

Abstract. We present the measurements of the elastically scattered 'plasma' lines intensities as a function of temperature. We found two anomalies at the temperatures $T = T_c$ and $T = 150$ K. This behaviour indicates either the existence of charge fluctuation in the temperature region from T_c to $T = 150$ K, or unusual electronic transitions in the YBCO system.

Since the discovery of the high-T_c materials a huge number of papers have been devoted to light scattering experiments on different kinds of excitation in all high-T_c compounds. The investigation of the electronic structure of these materials, both experimentally and theoretically, attracts special attention to understand the unique electronic properties. Having in mind the nature of the interaction of light with electrons in the system, it is reasonable to expect that this kind of experiment gives useful information about the superconducting mechanism. In order to study the absorption properties we measured the elastic light scattering of YBCO materials. For this purpose we used argon ion laser 'plasma' lines and a Raman spectrometer. We investigated the intensities of the elastically scattered 'plasma' lines as a function of temperature. Such experiments allow the investigation of the dielectric function properties. Similar information is provided with ellipsometric measurements and these experiments have already been performed on the YBCO system [1–3]. From the different theoretical calculations of the electronic structure [4–7], the properties of the dielectric tensor function are obtained with a good agreement with experimental results. However, from these experiments there is no evidence of anomalous behaviour of the dielectric tensor as a function of the temperature. Furthermore, the electronic Raman scattering continuum is studied around the critical temperature [8]. The integrated intensities exhibit a sharp increase at the critical temperature that is explained through the formation of Cooper pairs and the resulting correlation. Similarly, powder absorption measurements in the near-IC region showed an anomaly of mead infrared peak at T_c [9]. In this letter we present the measurements of the elastically scattered 'plasma' line intensities as a function of the temperature. We found two anomalies at the temperatures $T = T_c$ and $T = 150$ K. These behaviours indicate either the existence of charge fluctuation in the temperature region from T_c to $T = 150$ K or unusual electronic transitions in the YBCO system.

The samples were prepared by the usual solid state reaction technique, already described in [10]. The samples were monophase with a sharp superconducting transition, within 2 K, which is confirmed by diffraction and d.c. conductivity measurements. The powder samples were pressed into pellets (Φ 13 mm, thickness 1–2 mm) using a pressure of 8 T cm^{-2}. Using the electronic microscope we obtained the grain size of 30 μm. The $YBa_2Cu_3O_6$ samples

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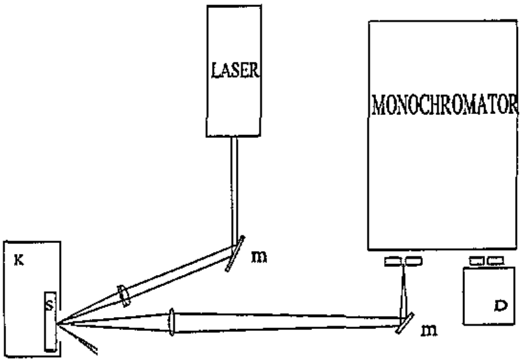

Figure 1. Experimental set-up: m—mirror, s—sample, k—cryostat, d—detector.

were obtained from ceramic $YBa_2Cu_3O_7$ samples after annealing in argon for 15 h and quenching in liquid nitrogen at 900 °C.

The spectral lines of an argon ion laser were used as excitation source. The experimental geometry was near backscattering as presented in figure 1. The light beam is focused on the sample surface using the usual cylindrical lens, with the spot (line) size around 10 mm. The laser power was controlled before and after each scan (the duration of the scans was 2 min) and we obtained the power stability within 2%. The scattered light is collected with an objective, aperture 1:1.4. The monochromator used was a Jobin–Yvon model U1000 equipped with a Peltier-effect-cooled RCA 313034 A photomultiplier with conventional photon counting system. The temperature is controlled with a closed-cycle helium cryostat and the measurements are performed in the heating regime. From the Raman spectra we selected the elastically scattered 'plasma' lines and measured their intensity as a function of temperature. The frequencies of the measured lines were chosen far from the phonon excitations.

The intensity versus temperature, for three characteristic elastically scattered lines in the $YBa_2Cu_3O_7$ sample, with energies within the range 2 to 2.7 eV, are shown in figure 2. The results are normalized with $T = 10$ K intensity value for each energy. As can be seen from figure 2, the intensity exhibits a decrease from 200 K to 150 K where its reaches a minimum value. This value remains approximately constant down to the temperature of the superconducting phase transition. Then, the intensity sharply increases until 80 K, where it has a constant value again, or a slightly increasing one. The arrow in figure 2 denotes a superconducting transition temperature obtained from resistivity measurements (see inset in figure 3(a)). The elastically scattered light intensities versus *temperature for* $YBa_2Cu_3O_6$ *and* $YBa_2(Cu_{0.97}Zn_{0.03})_3O_7$ *samples are presented in figure* 3. Evidently, the increase of the intensity can be associated with the superconducting transition since the $YBa_2(Cu_{0.97}Zn_{0.03})_3O_7$ sample exhibits the same behaviour at T_c (50 K for $YBa_2(Cu_{0.97}Zn_{0.03})_3O_7$ according to d.c. conductivity measurements, see inset figure 3(b)) as the $YBa_2Cu_3O_7$ one. While the increase of intensity for the Zn-doped sample is shifted, the decrease of intensity remains at the same temperature as in $YBa_2Cu_3O_7$, denoted by the dashed arrow in figure 3. Furthermore, in the case of the $YBa_2Cu_3O_6$ sample we obtained no anomalies of intensity in the whole temperature region we considered (10 K to 300 K). All measurements were repeated three or four times with excellent reproducibility.

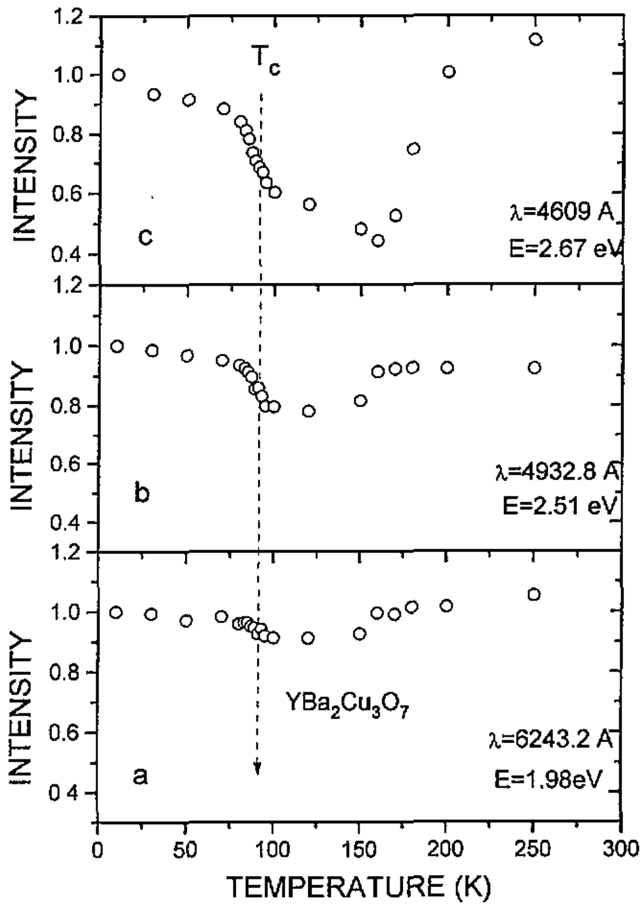

Figure 2. Intensity of the scattered light versus temperature for (a) $E = 1.98$ eV, (b) $E = 2.51$ eV and (c) $E = 2.67$ eV energy of incident light.

Finally, in figure 4, we show the intensity of the effect, the abrupt jump of the intensity in the vicinity of T_c, as a function of the incident light energy in the $YBa_2Cu_3O_7$ sample. As can be seen, the intensity of the effect increases as function of incident energy of light up to the highest value we measured, 2.67 eV.

There is no doubt that the behaviour of the elastic light scattering intensity is strongly related to the superconductivity phase transition. It remains to be discussed whether the decrease of the intensity ($T = 150$ K) is connected with the abrupt increase of the intensity at $T = T_c$ or needs to be considered separately, like two independent phenomena. In the first case we should consider the temperature region from T_c to $T_c + 50$ K as a charge fluctuation domain. Still, it is not clear why the absorption of light appears in such a wide temperature region (in the case of the Zn-doped sample the scattering region is about 100 K) if it is caused by a charge fluctuation effect. On the other hand this behaviour might be explained taking into account some electronic transitions within the material. Considering the superconductivity effect, the increase of the intensity at T_c can be attributed to the creation of the ordered superconductivity state that is less absorption efficient than the normal state as the manifestation of the electronic correlation of the Cooper pairs. Then, the

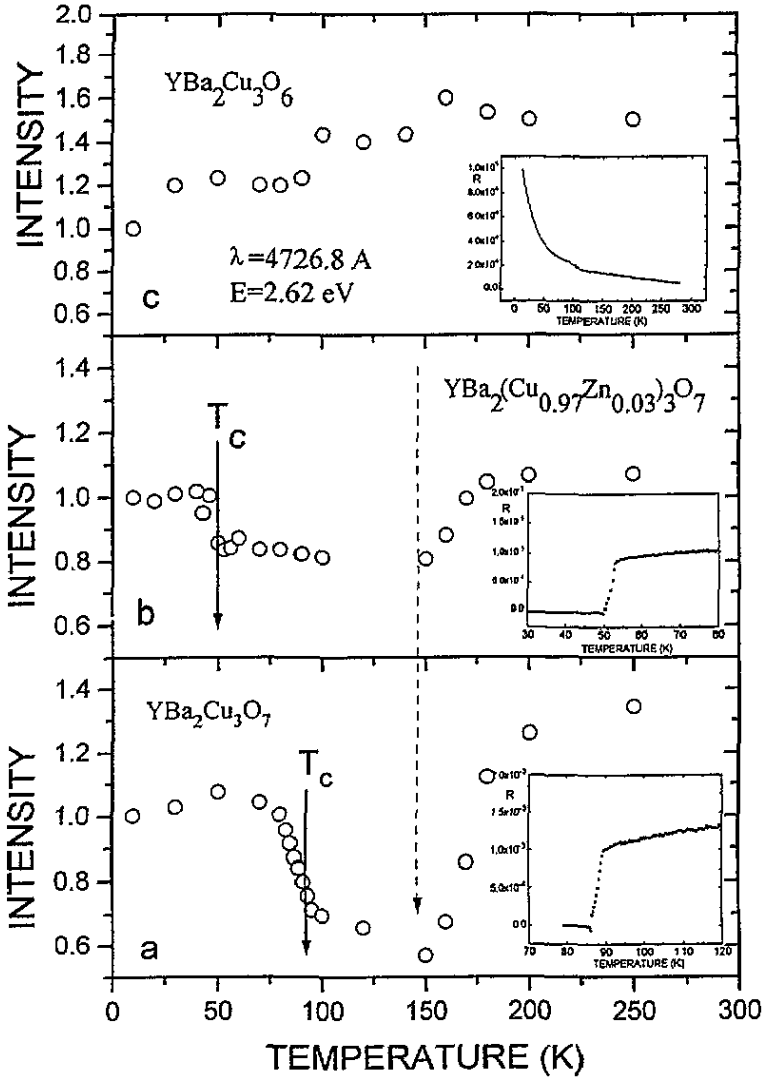

Figure 3. Intensity of the scattered light versus temperature for $E = 2.62$ eV incident light in (a) $YBa_2Cu_3O_7$, (b) $YBa_2(Cu_{0.97}Zn_{0.03})_3O_7$ and (c) $YBa_2Cu_3O_6$ samples. *The insets show the* resistivity curves for each sample.

decrease in the vicinity of T_c+50 K is related to the rearrangement of the electron levels that is caused by structural phase transition or some other mechanism. From the ellipsometric measurements, in the energy region of our experimental conditions, a structure around 2.7 eV is obtained [1]. This band is associated with CuO_2 planes since it is observed in all copper oxide compounds. Furthermore, according to the anisotropy dependence of the dielectric tensor [2] the structure between 2 and 3 eV is *c* polarized and originates from the transitions into the antibonding Cu(1)–O(1)–O(4) band. Concerning the behaviour presented in figure 4, the effect we obtained might be associated with these transitions since it is increasing up to 2.7 eV, the peak position of the 2.7 eV band. Also, some structural phase transition along

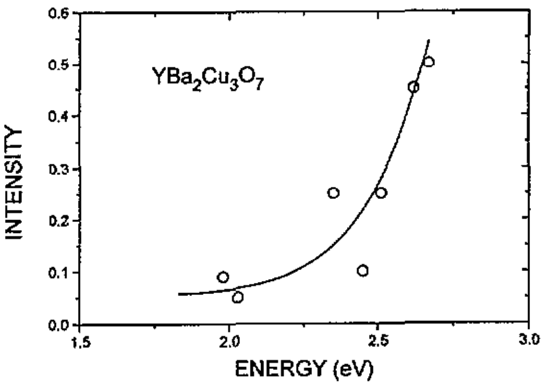

Figure 4. Intensity of the effect as a function of energy in the $YBa_2Cu_3O_7$ sample. The solid line is a guide for the eye.

the c axis is indeed observed around $T = 150$ K using neutron diffraction measurements [11]. Finally, from the present results we conclude that the structural phase transition at $T = 150$ K provokes the appearance of the strong transition into a Cu(1)–O(1)–O(4) band while at $T = T_c$, according to high electronic ordering, there is no absorption in the material. Still, for a better understanding of this effect further investigations are necessary, specially on single-crystal samples as well as on another high-T_c material. These investigations are in progress.

We would like to thank to Z V Popović and Z Radović for helpful discussions. This work was supported by the Serbian Ministry of Science and Technology under project 0104.

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