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17 keV-photon induced damage of Bi-2212 whiskers by synchrotron μ-beam exposure

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Abstract

Experimental data show that the normal state resistivity of superconducting $Bi_2Sr_2CaCu_2O_{8+\delta}$ (Bi-2212) whiskers increases after a six-hours irradiation by a synchrotron μ-beam with 17 keV photons. We analyze this result on the basis of previously reported effects in Bi-2212 whiskers due to ageing or heating processes. A finite element model of the experimental setup clarifies that the heat load induced by the microbeam has to be excluded as a possible cause for the material changes. The knock-on the interstitial, loosely bound, O species by secondary electrons is discussed as the most likely mechanism responsible for this effect.

1.Introduction

The $Bi_2Sr_2CaCu_2O_{8+\delta}$ (Bi-2212) high temperature superconductor (HTSC) exhibits a layered structure along the crystal *c*-axis consisting of alternating superconducting and insulating planes, which induces the Intrinsic Josephson Effect (IJE) in this material [1]. This fact opens the way to a novel class of solid state devices consisting of large arrays of Josephson junctions, densely integrated on an atomic scale level, e.g. THz radiation emitters and voltage standards [2, 3].Therefore, the ability of Bi-2212 to tolerate harsh working conditions is an important issue for the efficiency of such devices.

The effect of heavy ion irradiation on this material has been widely studied, showing that the critical temperature T_c starts degrading at fluences higher than $\approx 1\text{-}2\times 10^{11}$ ions/cm^2 [4]. Concerning protons and α particles, it has been proved that they can knock O atoms out of the material at doses $\approx 10^{16}$ particles/cm^2, affecting both T_c and the normal state resistivity [5]. Also electron irradiation can induce polycrystallinity and amorphization in Bi-2212 at fluences of $\approx 10^{22}$ e$^-$/cm^2 [6].

On the other hand, much less is known about the effects induced by energetic photons. To our knowledge, there has been only one experiment about the effect of γ rays on Bi-2212, which showed that photons of about 1.3 MeV are able to induce the desorption of extra oxygen atoms from the BiO layers and to reduce the size of the crystallites [7]; however, other investigations confirmed that γ-rays with similar energy affect the oxygen concentration or carrier distribution in different HTSC compounds, as well [8, 9].

Less energetic photons are commonly considered ineffective from the point of view of radiation damage, nevertheless this assumption can be questioned according to the work by Piñera *et al.*, who showed, by means of simulations, that photon energies as low as 122 keV can induce non-negligible effects on loosely bound O atoms, like in the case of the Cu-O chains of $YBa_2Cu_3O_{7-\varepsilon}$ (Y-123) [10]. From the experimental point of view, recent developments of micro- and nano-beams at third generation synchrotrons make it possible to test such stability at low energy (≈20 keV) and high power density (≈3×10^5 W/m^2) conditions [11]. Recently, we have used such set up to monitor aging or thermally induced variations in the stoichiometry of Bi-2212 whiskers [12], without investigating possible damages by the μ-beam. In the present paper, we provide evidence of modifications in the electrical behaviour of Bi-2212 whiskers before and after the μ-beam exposure, indicating the existence of low energy X-ray damage in this material. The role played in this effect by the heat load induced by the beam is also investigated by means of a finite element model.

2.Experimental method

Bi-2212 whiskers were grown, positioned on sapphire substrates and electrically contacted with Ag/Au stripes [13]. Electrical characterizations were performed along the *a*-axis direction by standard four-probe technique with I=1μA in the range 77.4K≤ T≤ 295 K. Resistivity values were obtained via accurate geometrical characterization by AFM and SEM.

The experiment was performed at the ID22 beam-line of European Synchrotron Radiation Facility (ESRF).The experimental setup is described elsewhere [12, 14]. The microprobe spatial resolution was 1.7 μm (vertical) X 5.3 μm (horizontal), with a photon flux at the focal spot of about 10^9 photons·s^{-1} at 17 keV for a ring current of 200 mA and a depth of focus of the order of 300 μm. The sample was scanned by the beam for a total time of 6 hours and the exposed area was practically all the region between the voltage contacts. The sample was kept in air atmosphere during the whole X-ray irradiation experiment.

3.Results and discussion

Figure 1(a) shows three resistivity curves obtained on the analyzed Bi-2212 whisker. The electrical measurements were performed on the as-grown sample, after annealing for 6h in air at T=363K, and after synchrotron light irradiation. As observed in previous experiments [12, 15], the annealing process at T=363 K changed the resistivity and the T_c of the samples: the as-grown crystal exhibits a slightly overdoped behaviour with a critical temperature of about T_c=77 K, while the 6-hours annealed sample shows a higher resistivity accompanied by a clear downwards curvature and a higher T_c=77.7 K. We have already proved that these changes are associated with an elongation of the *c*-axis and therefore are due to an oxygen depletion process [12]. The curve measured after the 6-hours μ-beam exposure has a similar curvature as the previous measurement, but also shows a resistivity increase that is beyond uncertainties, while the critical temperature is almost constant (T_c=77.5K). To our knowledge, this is the first observation of a change in the electrical behaviour induced by so low energy photons (17 keV).

On the basis of the oxygen decrease occurring during the annealing part of the experiment, it seems reasonable to interpret such modifications as changes of the oxygen content or distribution due to the X-ray exposure. Actually, a similar increase of resistivity was observed in a Y-123 microbridge under irradiation by 20 keV electrons and was explained as the knocking on O atoms, which induced disorder in their distribution, but no out-diffusion [16]. This experiment also provided the value of 2.8 eV as the upper limit for displacement energy of the oxygen atoms in the Cu-O chains. By comparing the activation energy for the diffusion of the interstitial O atoms in Bi-2212 (≈0.93 eV) [17] with the activation energy of O in Y-123 (≈1.3 eV) [18], a displacement threshold energy well below 2.8 eV can be deduced for the O atoms in Bi-2212. Since it has been shown that γ-rays produce an energy distribution of secondary electrons that is peaked very close to the energy of the incident photons [10], it is very likely that the secondary electron flux induced by 17 keV photons can displace interstitial O atoms in a macroscopically detectable amount, especially if we consider that in our experiment the absorbed dose ($\approx 2\times10^9$ Gy) well exceeds the typical dose of damage experiment using ^{60}Co radiation sources ($2\text{-}4\times10^6$ Gy).

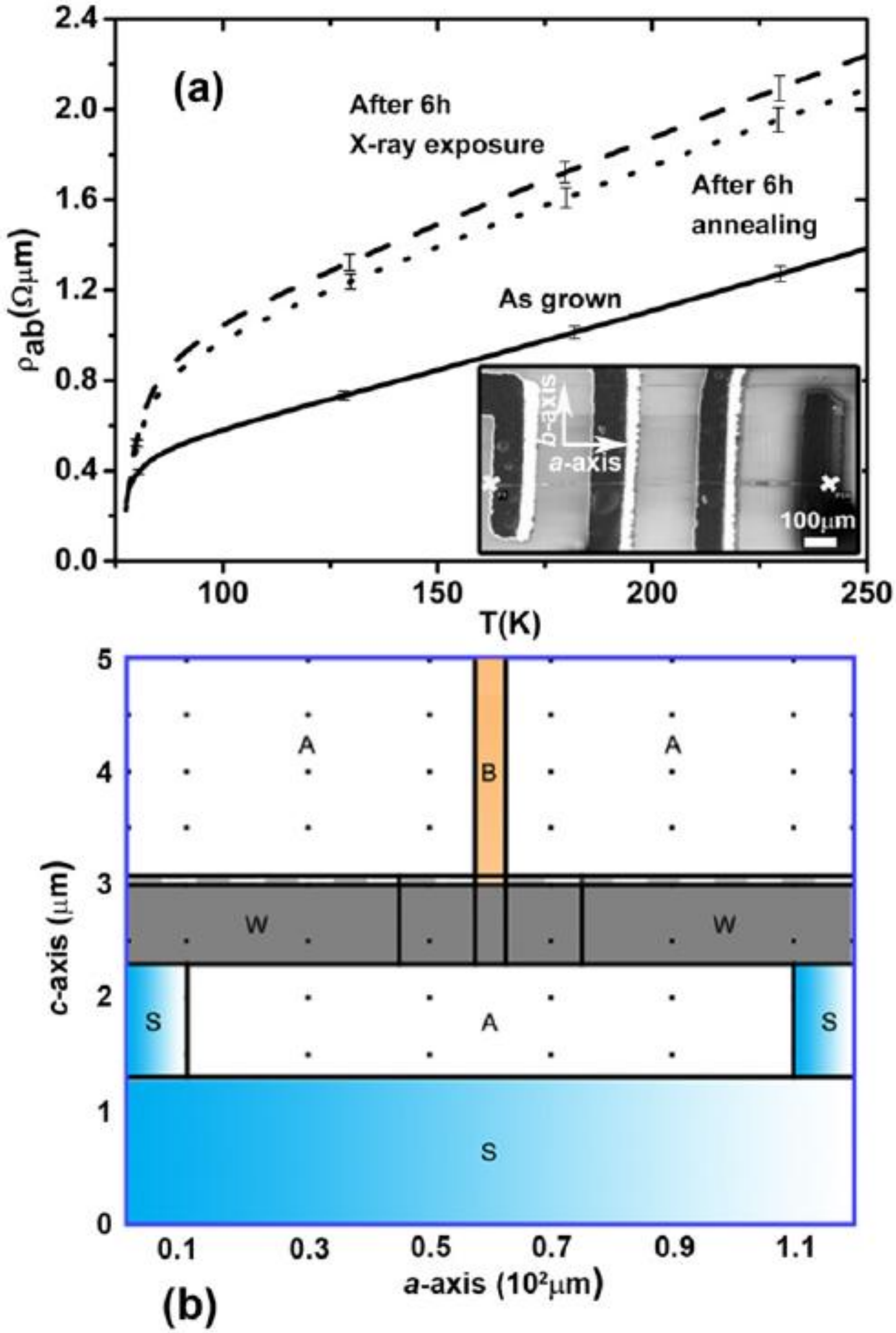

Figure 1. Panel (a): *ab*-plane resistivity vs. temperature curves obtained on the analyzed whisker. The solid curve refers to the as-grown sample, the dotted curve refers to the sample after 6 hours annealing at *T*=363K in air, the dashed curve refers to the sample after 6 hours μ-beam irradiation. Error bars on resistivity are reported. Inset: SEM picture of the electrically contacted whisker. The crystal is the horizontal thin line marked by two white crosses, the four contacts are the dark vertical stripes, *a*- and *b*-axis of the crystal are indicated. Panel (b): sketch of the central part of the geometry used in the simulation. The whisker is represented in gray and labelled as **W**, the sapphire substrate is represented in blue and labelled as **S**, the air regions (both below and over the whisker) are white and labelled as **A**. A Bi_2O_3 passivated layer is also present on the whisker upper surface and is represented with vertical stripes. The incoming beam is drawn in orange and labelled as **B** (Colours online).

Even if this picture seems very reasonable, this effect could in principle also be ascribed to a local temperature increase induced by the final phononic decay of the power delivered by the μ-beam. Such a locally increased temperature could promote the out-diffusion of the interstitial oxygen atoms, so that no real difference would exist in the mechanism responsible for the resistivity increase during the thermal annealing and during the X-ray exposure. We investigated this possibility by means of a finite element model aimed at simulating the temperature field in the whisker under irradiation.

The following partial differential equation was implemented, which represents the energy balance for the whisker considering the heat transfer both by conduction and convection:

$$\nabla \cdot \left(\mathbf{k} \nabla \mathbf{T} \right) + \rho \mathbf{C_p} \mathbf{u} \cdot \nabla \mathbf{T} = \mathbf{Q},$$

where k is the thermal conductivity, T the temperature, ρ is the crystal density, C_p the heat capacity, $\mathbf{u}$ is the air velocity and $\mathbf{Q}$ represents the heat source. By considering the material absorption coefficient α=610 cm^{-1}, we estimated an absorbed power density $\mu_a q_r \approx 3 \cdot 10^5$ W/m^2 (μ_a is the surface absorption coefficient, and q_r is surface density of the incoming heat flow).

The geometrical model reproduces the average whisker size, i.e. 700 x 5 x 0.7 μm^3. A sketch of the central part of this geometry is shown in figure 1(b). An air layer is inserted between the crystal and the substrate, as suggested by microscope observations, and its thickness was varied to investigate the effect on the thermal load. Two sapphire layers are also placed in perfect contact with the crystal at a distance of about 50 μm from the beam, simulating the thermal anchoring induced by the presence of the electrical contacts. A thin layer of Bi_2O_3 was added on the whisker upper surface (gray stripes area in Fig. 1(b)) in order to consider also the passivated layers usually observed in this material [19, 20]. Since it is not possible to estimate the thickness of such layer, we performed different simulations for thicknesses in the range 0÷200nm and compared the results. Materials physical parameters used in the simulation are summarized in Table 1.

Table 1. Material physical parameters used in the simulations.

Component	Density (kg m^{-3})	Thermal conductivity (W m^{-1} K^{-1})	Heat capacity (J kg^{-1} K^{-1})
Bi-2212	6700 [21]	0.86 [22]	425 [23]
Sapphire	4000 [24]	40 [25]	780 [26]
Air	1.2 [27]	0.026 [28]	1012 [28]
Passivated layer (Bi_2O_3)	8929 [29]	0.9 [30]	236 [31]

For the simulations, the COMSOL Multiphysics commercial software was used. A regular 2D-mesh was implemented and the computation algorithm was adapted for 2D analyses of the temperature profile. Considering the large thickness of the sapphire (500 μm), a constant temperature of 300 K was set in the substrate.

Figure 2 shows two representative results for the two cases of air thickness of 1 μm (Fig. 2(a)) and 3 μm (Fig. 2(b)): the graphs show maximum temperature increases of 3.2K and 5.6K, respectively. In this case the Bi_2O_3 surface layer thickness was 150 nm. By fixing the air thickness layer in 1 μm and varying the Bi_2O_3 layer thickness to 75nm and 20nm, we found maximum temperature increases of 2.9K and 5.1K, respectively, while for the case of 0nm thickness a 4K increase was obtained.

On the basis of our previous experience on ageing and annealing processes in Bi-2212 whisker [12, 15], such temperature values are not high enough to induce electrical modifications on timescales of hours. This rules out the possibility that the resistivity increase induced by the X-ray exposure is simply due to thermally activated oxygen out-diffusion and supports the of O atom displacement by 17 keV secondary electrons, anyway, more specific measurements are needed to verify this hypothesis.

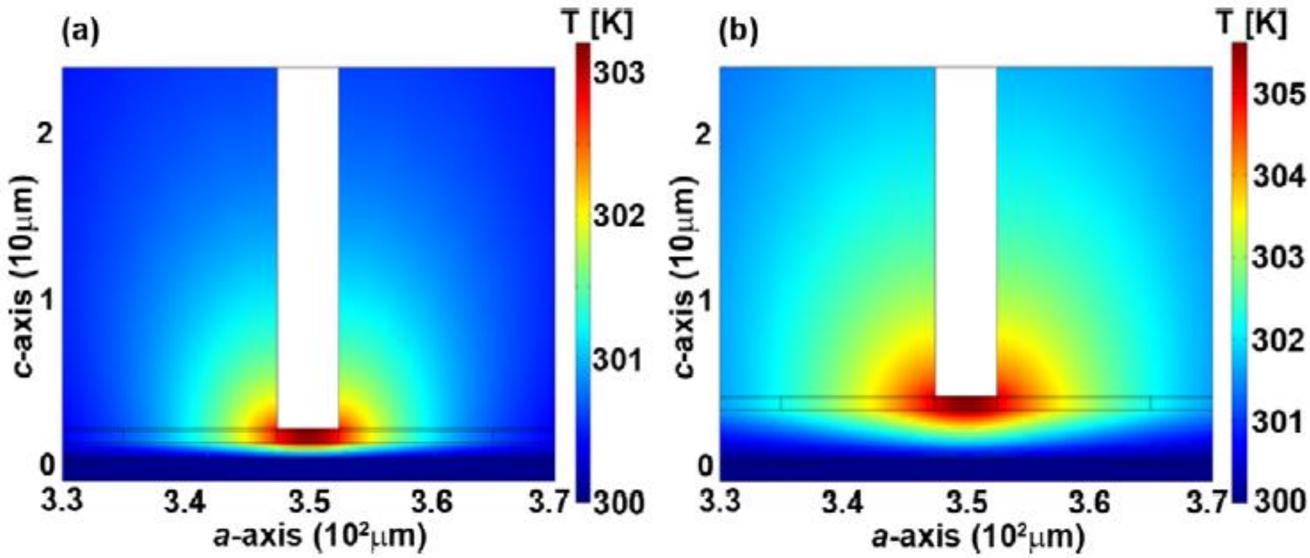

Figure 2. Typical results of the simulation of the temperature field induced by the synchrotron μ-beam (150 nm Bi_2O_3 oxide thickness). Panel (a) refers to 1 μm air layer below the whisker; panel (b) refers to the 3 μm air layer case (Colours online).

4.Conclusions

We have observed electrical changes in Bi-2212 whiskers under synchrotron μ-beam exposure. By means of a finite element simulation of the experimental setup we can exclude the radiation heating as the primary source of such electrical modifications, which must thus be due to pure photon-induced O atomic disorder. This effect should be carefully considered in synchrotron radiation experiments on Bi-2212, especially at μ-beams. Similar irradiations are of great interest to extend the knowledge on the effects of synchrotron μ/nano-beams on inorganic materials.

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