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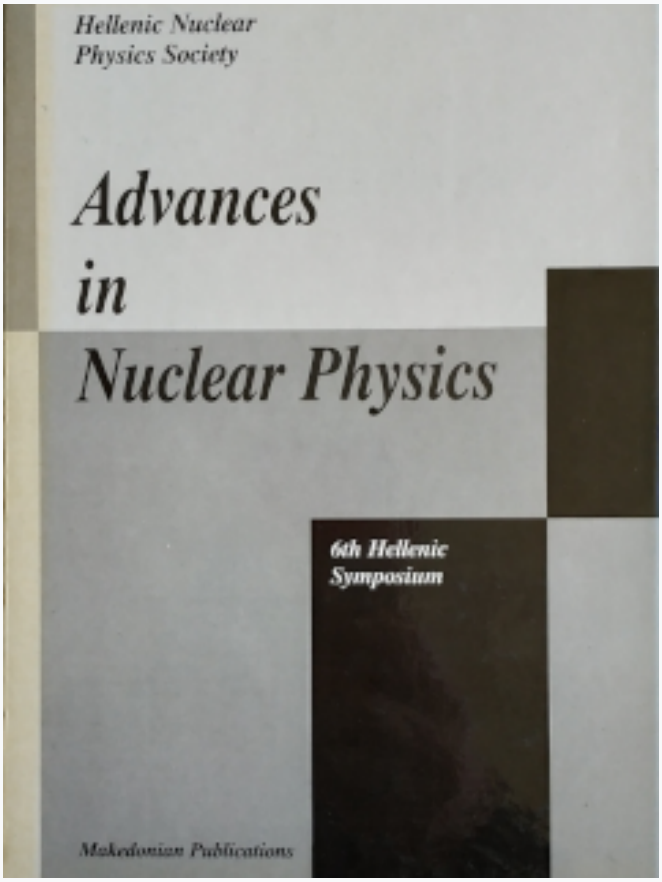

Radiation damage of thin films of YBa2Cu3O7 superconductors

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Radiation damage of thin films of $YBa_2Cu_3O_7$ superconductors

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Abstract

The effect of ion-beam irradiation of $YBa_2Cu_3O_7$ superconductors has been studied by Raman spectroscopy. The ion beams ^4He, ^{16}O and ^{127}I have been used at energies 4, 25 and 200 MeV, respectively, in an attempt to investigate the radiation damage with respect to the mass of the bombarding ions. Further, different doses of irradiation have been tried for each ion beam in order to investigate at which critical dose the phase transition from crystalline to amorphous and the loss of superconductivity occur.

1 Introduction

The effects of radiation on high temperature superconductors (HTSs) are of particular interest from both basic and practical points of view. The role of phonons in the superconductivity, the phase transformations as well as the correlation between structural defects and properties like transition temperature and resistivity, have attracted much experimental and theoretical effort in connection with basic problems in understanding the superconductivity in HTSs [1,5]. From the practical point of view, many applications of superconductors such as superconducting magnets in fusion reactors and high energy accelerators require investigation of the HTS property changes in a radiated enviroment. In addition, the controlled introduction of defects with ion-beams, have been used for patterning circuits onto $YBa_2Cu_3O_7$ films which may lead also to device applications such as SQUIDs [6,7].

In this article we report on the study of structural changes of $YBa_2Cu_3O_7$, thin films induced by energetic particle irradiation. The films were bombarded with ^4He, ^{16}O and ^{127}I ions at various doses and examined by means of Raman spectroscopy in order to get comprehensive understanding of the irradiation induced defects and damaging process in this material.

2 Experimental

Films of nominal composition $YBa_2Cu_3O_7$ of about 200-300 nm in thickness, grown on MgO substrate were irradiated at room temperature with ion beams at various doses. The 200 MeV ^{127}I and 25 MeV ^{16}O beams were provided by the Munich 14MV tandem accelerator, while the 4 MeV ^4He beam was supplied by the tandem T11/25 accelerator of the NRCPS "Democritos".

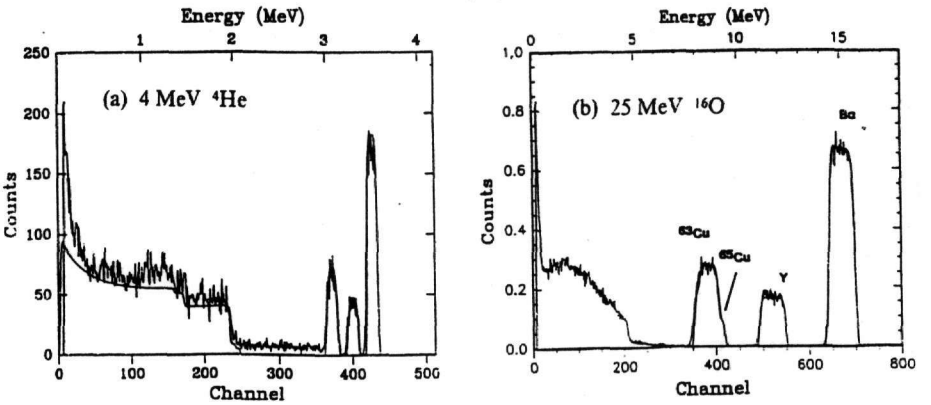

Fig. 1 . RBS spectra of $YBa_2Cu_3O_7$ films on MgO, taken with 4 MeV ^4He (a) and 25 MeV ^{16}O (b). The solid lines represent simulation by RUMP code.

The composition and thickness of the samples were measured by the Rutherford Back Scattering (RBS) spectrometry and their quality concerning both oxygenation and homogeneity was tested by means of Raman spectroscopy prior to irradiation. For the RBS measurements and irradiation with the ^4He beam, the newlly installed chamber and goniometer at "Democritos" , manufactured by Charles Evans and Associates, was used [8]. The vacuum of the chamber was 10^{-6} Torr during the measurements.The surface barrier RBS detector was positioned at 160° scattering angle and at a distance of 14cm

from the target. For the irradiation with ^{16}O, the RBS spectrum was taken again during the bombardment at the higher dose, by using the RBS facility of the Munich MP tandem, described in ref. [9]. For the irradiation with ^{127}I, carried out in Munich, the samples were tested with RBS at "Democritos" by using the 4 MeV alpha beam. All the RBS spectra were analysed by utilizing the computer code RUMP [10]. As an example, RBS spectra taken with ^{16}O and ^{4}He beams are shown in Fig. 1. The solid line represents the RUMP simulation.

After irradiation the samples were studied by Raman spectroscopy by comparing the spectra taken fom the irradiated and non-irradiated regions of the samples. For these measurements, carried out at room temperature, the apparatus of the Departement of Physics of the NTU was used, which consists of a Jobin-Yvon T64000 triple spectrometer, equipped with a microscope and a liquid N_2 cooled CCD detector, as well as the 514.5 nm line of an Ar^+ laser at 1-3 mW.

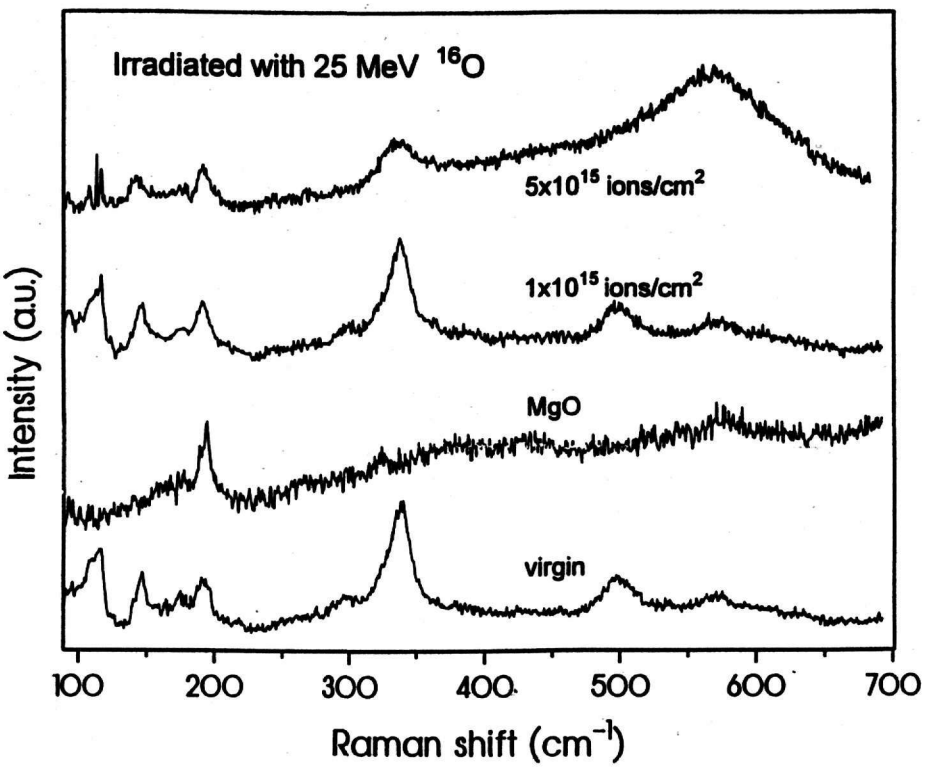

Fig. 2. Raman spectra of $YBa_2Cu_3O_7$ films on MgO as deposited (virgin) and irradiated at two different doses with 25 MeV ^{16}O . The Raman spectrum of the MgO substrate is also shown for comparison.

The Raman spectra of $YBa_2Cu_3O_7$ films exhibit two vibration modes at about 115 and 148 cm^{-1}, corresponding to vibrations of Ba and plane Cu atoms, respectively and another two at 338 cm^{-1} associated with the out of phase motion of the O(3)- O(2) atoms of the Cu planes and at 500 cm^{-1} associated with the displacement of the O(4) apical oxygen along the c-axis. The vibrations of the O(1) atoms of the Cu chains are not Raman active, due to their mode of symmetry [11]. Such Raman spectra are shown in Figs 2, 3 and 4 and are characterizd by the label "virgin", meaning that they are taken from the non-irradiated part of the sample. In these Figures the Raman spectra taken from $YBa_2Cu_3O_7$ samples irradiated with ^{16}O, ^{127}I and ^{4}He at different doses are also presented and will be discussed in the next section.

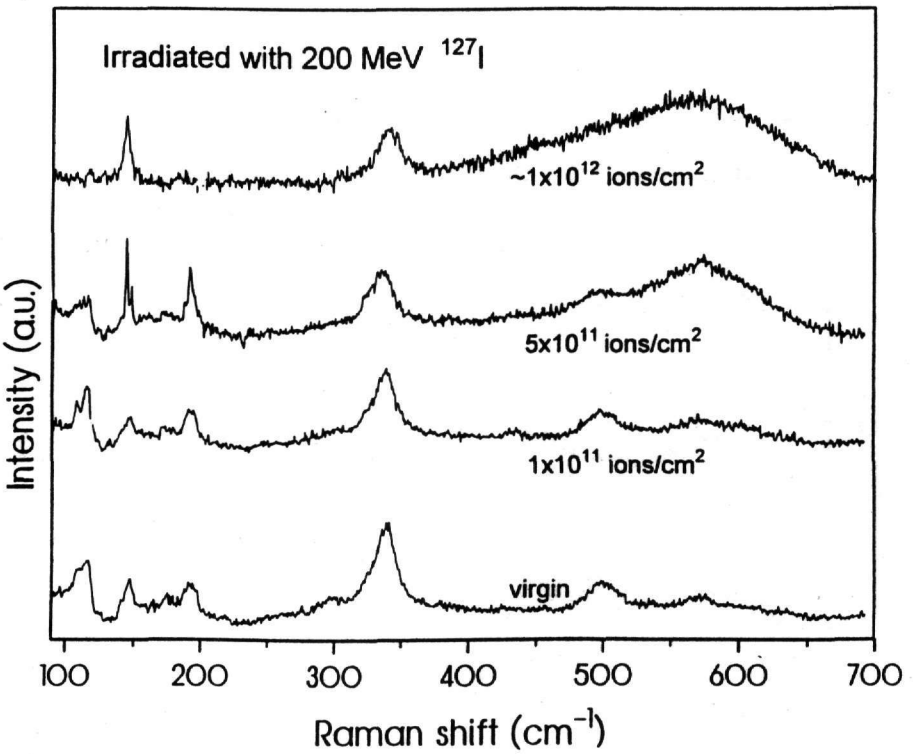

Fig. 3. Raman spectra of $YBa_2Cu_3O_7$ films on MgO as deposited (virgin) and irradiated at three different doses with 200 MeV ^{127}I. The peak at 190 cm^{-1} corresponding to the MgO substrate is not present in the upper spectrum (irradiation dose ~ $1x10^{12}$ ions/cm^2) since the sample was thick enough so that the substrate could not be seen by the laser beam.

3 Results and discussion

3.1 Irradiation with ^4He,^{16}O and ^{127}I

The Raman spectra of $YBa_2\ Cu_3O_7$ thin films on MgO, irradiated with ^{16}O and ^{127}I are shown in Figs 2 and 3, respectively. The spectra called "virgin" are taken from the non-irradiated part of the samples and contain the four peaks associated with the $YBa_2Cu_3O_7$ modes of vibration mentioned in the previous section. Apart from these peaks, another well developed peak appears at about 190 cm^{-1} and a weak one at about 570 cm^{-1} , which correspond to the vibrations of the MgO substrate. A spectrum taken from the MgO substrate is also shown for comparison in Fig. 2. The spectra taken from the irradiated part of the samples are labelled by the corresponding doses of irradiation. In the two Figures the spectra corresponding to the doses $1x10^{15}$, $1x10^{11}$ ions/cm2 , respectively, look identical with the corresponding "virgin", meaning that the doses are too low to affect the crystal. At the higher doses ($5x10^{15}$, $1x10^{12}$ ions/cm^2 , respectively), however, a large broad peak emerges, in the region of 550 to 600 cm^{-1}, which covers the peak of O(4) apical oxygen at 500 cm^{-1}. In the case of ^{127}I irradiation, a spectrum taken at an intermediate dose $5x10^{11}$ ions/cm^2 is also shown in Fig. 3 , which illustrates the onset of the distortion induced by irradiation, while the O(4) peak still keeps its identity, with reduced intensity. In both Figures, the Ba peak at 115 cm^{-1} decreases significantly, or even vanishes, at the higher doses and a shift to lower frequencies of the Cu peak (from 147 to 144 cm^{-1}) appears, which indicate oxygen loss from the Cu chains [11]. The peak of the O(2)-O(3) of the Cu planes at 338 cm^{-1}, is broadened to some amount but otherwise does not seem to be seriously affected by the irradiation.

Irradiation of $YBa_2Cu_3O_7$ films with 4 MeV ^4He has aso been carried out at several doses up to $1x10^{16}$ ions/cm 2. The results are shown in Fig. 4, where the Raman spectra for the non-irradiadiated (virgin) and the irradiated parts of the samples at the higher doses, are presented. It is seen that the spectra look identical, indicating that the dose is too low to affect the crystallinity of the film.

4 Discussion

The physical properties of $YBa_2Cu_3O_{7-x}$ HTSs depend on the concentration and ordering of the oxygen atoms. For the high oxygen content $0 < x < 0.2$, the structure of the compound is orthorhombic and the critical temperature reaches the maximum value of Tc$\sim$ 92K. For higher values of x the critical

temperature decreases and for $x > 0.6$ the orthorhombic to tetragonal transition occurs and the superconductivity is lost. This phase transition is related to the loss of O(1) atoms in the Cu chains, while the O(4) apical atoms are also of importance since they transfer the charge between CuO chains and CuO_2 planes.

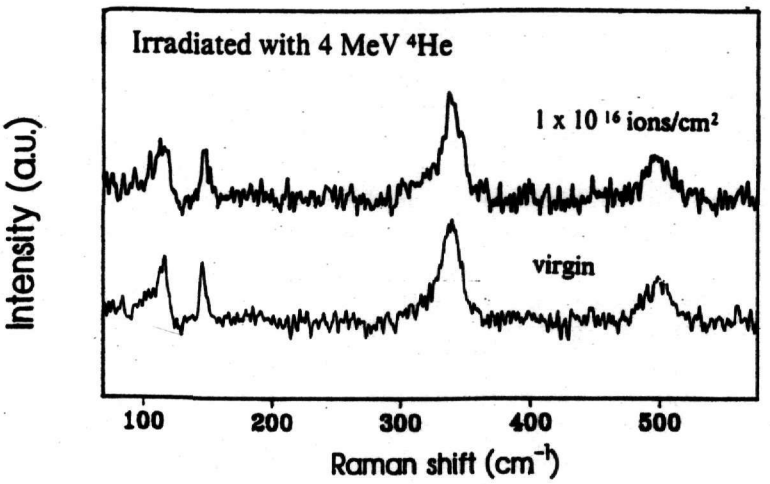

Fig. 4. Raman spectrum of $YBa_2Cu_3O_7$ films on MgO as deposited (virgin) and irradiated with 4 MeV ^4He.

From the Raman spectra in Figs 2, 3 and 4, described in section 3.1, it is obvious that the main effect of the irradiation is the appearence of a broad structure in the region of 550-600 cm^{-1} . Such broad structures have been observed in oxygen defficient samples which have not been carefully annealed and can be attributed to an IR mode at 555 cm^{-1} which becomes Raman active due to the disorder [12]. This could also be the case in this work, with the disorder of the oxygen atoms being induced by the irradiation. Thus the irradiation could result in a fluctuation of the position of O(1) atoms and of the lenghts of their bonds to the neighbouring atoms, thus producing distortion of the chains and even amorphization. As a consequence the O(4) as well as the O(2)-O(3) vibrations are also affected to some extend as indicated by the broadening of their peaks, but the structural order of the CuO_2 planes does not seem to be disturbed by the irradiation. In addition, the disappearance of the Ba peak together with the shift of the Cu peak by three wave numbers, suggest oxygen deficiency of the order of $x \sim 0.6$ associated with O(1) loss, which leads to amorphization of the chains. These results are in agreement with the conclusions of ref [13] where $YBa_2Cu_3O_7$ films were irradiated with 173 MeV ^{129}Xe atoms. For such irradiation the electronic stopping power is dominant compared to the nuclear stopping power leading in an amorphization

rather than point-defect concentration.

In addition, the critical dose at which the amorphization of the crystal takes place has been determined to be of the order of $5x10^{15}$ and $1x10^{12}$ ions /cm^2 for the 25 MeV ^{16}O and 200 MeV ^{127}I ion beams, respectively. These values for ^{16}O and ^{127}I are in agreement with the ones extracted from measurements of the resistivity and critical temperature of $YBa_2Cu_3O_7$ films irradiated with 25 MeV ^{16}O and 173 MeV ^{129}Xe, reported in ref. [14]. In the same reference, the critical dose for irradiation of $YBa_2Cu_3O_7$ films with 6 MeV ^{4}He turns out to be $> 5 \times 10^{16}$ ions/cm^2. This explains why the Raman spectra of $YBa_2Cu_3O_7$ films irradiated with 4 MeV ^{4}He ions are seen in Fig. 4 to be unaffected by the irradiation.

5 Summary

This article reports on the investigation of structural changes of $YBa_2Cu_3O_7$ thin films, induced by energetic particle irradiation, by means of Raman spectroscopy. The films were bombarded at room temperature with beams of ^{4}He, ^{16}O and ^{127}I at 4, 25 and 200 MeV, respectively and to several doses in order to determine the critical dose at which amorphization occurs. At the higher doses of the heavy-ion irradiations, the Raman spectra show a broad structure in the region of 550-600 cm^{-1} . This could be associated to the O(1) vibration (IR mode) which has become Raman active due to the disorder of the oxygen atoms induced by the irradiation. The decrease and even disappearence of the Ba peak, together with the shift of the Cu peak to lower frequencies is an indication of oxygen deficiency due to the irradiation. It is thus concluded that the energetic particle irradiation of $YBa_2Cu_3O_7$ thin films results in a distortion and even amorphization of the chains. The B_{1g} mode of the O(2)-O(3) as well as the vibration of Cu are not seriously influenced, which means that the structural order of the CuO_2 planes is not disturbed by the irradiation. The critical doses at which amorphization occurs have been determined to be of the order of $5x10^{15}$ and $1x10^{12}$ ions /cm^2 for the 25 MeV ^{16}O and 200 MeV ^{127}I ion beams, respectively. For the light ion ^{4}He beam the critical dose seems to be much higher than the one tried in the present work and has to be determined in a future experiment.

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