RESEARCH ARTICLE

Radiation Enhancement of Superconducting Critical Temperature of Fe-based HTSCs

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ABSTRACT: High Temperature Superconductivity (HTSC) with Superconducting Critical Temperature (T_c) up to 56 K in Fe Pnictides/Chalcogenide compounds baffled the classical idea that magnetic ions like Fe destroy superconductivity. These Fe-superconductors appear to be better than Cu-oxide HTSCs for making superconducting wires, considering the easier fabricability for Fe-HTSCs, for magnet and other applications, making Fe HTSCs an important group of emerging materials. Fusion and accelerator magnets work in intense radiation environment. So, as a first step, radiation damage of Ba $(Fe_{0.943}Co_{0.057})_2As_2$single crystal HTSC due to 1.5 MeV Ar^{6+}- irradiation has been investigated. A peak in the imaginary part (χ'') of magnetic susceptibility indicates T_c. The un-radiated (UR) sample shows T_c= 15.95 K from its χ'' peak. Irradiated T_c from χ' and electrical resistivity measurements are in excellent agreement with magnetic χ'' result of radiation enhanced T_c of 24.01 K, which is a new observation. Further studies under high magnetic fields will be discussed. Earlier report for Fe-HTSC films on enhancement of T_c from 18.0K to 18.5K due to 1×10^{15} p/cm^{2}irradiation of 190keV protons, support present observation. Physics needs to be investigated further.

KEYWORDS: Fe Pnictide superconductor, ion radiation damage, HTSC, upper critical field, $Ba(Fe_{0.943}Co_{0.057})_2As_2$ single crystals

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1.INTRODUCTION

Zero electrical resistivity and, more importantly, exclusion of magnetic flux on cooling a sample below itsSuperconducting Critical Temperature (T_c) confirm its entry into the superconducting state[1,2]. Superconductivity was discovered in 1911. Itis presenting new surprises like Cu-O based High Temperature Superconductors (HTSCs) in recent decades [3]. Latest surprise is superconductivity including high temperature superconductivity(wth$T_c$up to56 K) in Fe pnictide(Pn) /chalcogenide (Cn)compounds, with Fe-As or Fe-Te/Se layers as the seat of superconductivity[3-5]. Here, magnetic ions like Fe (or often Ni)is a major component. This shatters the textbook idea that magnetic impurities always destroy electron pairing and superconductivity. Search for pairing mechanism in Fe-HTSCs, most unlikely to be BCS, can be somewhat unearthed by radiation damage experiments. Stronger impetus for radiation modification investigations in Fe-HTSCs come from potential fabrication of Fe-HTSC cables and hence superconducting magnets and devices[6,7]. These may be used in radiation environment of accelarators and fusion reactors. So, an estimate of damage by fast neutrons and ions, and hence the working life of the Fe-HTSC magnets or devices must be made.

Expulsion of magnetic flux from the inside of asuperconductor leads toa peak at$T_c$in the imaginary part of magnetic susceptibility (χ'') and a step at$T_c$in the real part of magnetic susceptibility (χ'). Here, radiation damage by afastheavy ion beamto $Ba(Fe_{(i-x)}Co_x)_2As_2$, x = 0 and 0.057, have been investigated by XRD, Magnetic Susceptibility and Electrical Resistivity Measurements.

2. EXPERIMENTAL OUTLINE

2.1 Single Crystal Sample Preparation, XRD Characterization and Ion Irradiation

The single crystals (SXLs) of$Ba(Fe_{(i-x)}Co_x)_2As_2$were grown [5]by self-flux method, using a glassy carbon crucible. After heating Ba:(Fe,Co)As =1:5 mixture to1160°C in the crucible, it was cooled down very slowly at rates (0.22–0.30 °C/h).On completion of the growth the crucible was tilted to decant away the remaining flux. SXLs were collected from the crucible. XRDof UR samples in FIG.1 shows only 00l reflections from the large face ofour 1.7 – 3mm (length), 1 – 2 mm (width) and 0.2 – 1mm (thickness) samples. This proves the single crystal nature of the samples.Moreover, the peaks show gradual decrease of the lattice parameter c due to increasing Co-substitution: c =13.0314 Å for x=0.000, c = 12.999 Å for x=0.057 & 12.988 Å for x=0.102.[8]

Here, ~200 micron thick SXLs have beenirradiated at IUAC, New Delhi, India, by 1.5 MeV Ar^{6+}-beam. The

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crystals have been characterized first in UR (Un-Radiated) and then in AR (After Radiated) condition.

2.2 Electrical Resistivity Measurement

Using a standard DC 4-probe technique, electrical resistivity of our un-radiated and radiated SXL samples have been measured from room temperature down to 2K in a liquid helium cryostat under different magnetic fields (0 to 6 Tesla), applied along c axis direction of the samples. For resistivity measurements, rectangular shaped samples have been cut out with a wire-saw. Electrical contacts for conduction along the large face that turned out from XRD to be ab-plane have been made by attaching thin copper wires with silver epoxy.

2.3Magnetic Susceptibility Measurement

A Vibrating Sample Magnetometer from Quantum Design has been used for measurement down to 2 K, of real and imaginary parts of Magnetic Susceptibility.

3. RESULTS AND DISCUSSION

FIG. 1(a) depicts full XRD pattern, with all the observed peaks indexed for $Ba(Fe_{(1-x)}Co_x)_2As_2$, as a function of irradiation dose (zero to 10×10^{15}Ar-ions/cm^2). Gradual broadening of the peaks on irradiation can be observed, more clearly in FIG. 1(b), an enlarged view for004 peak only.

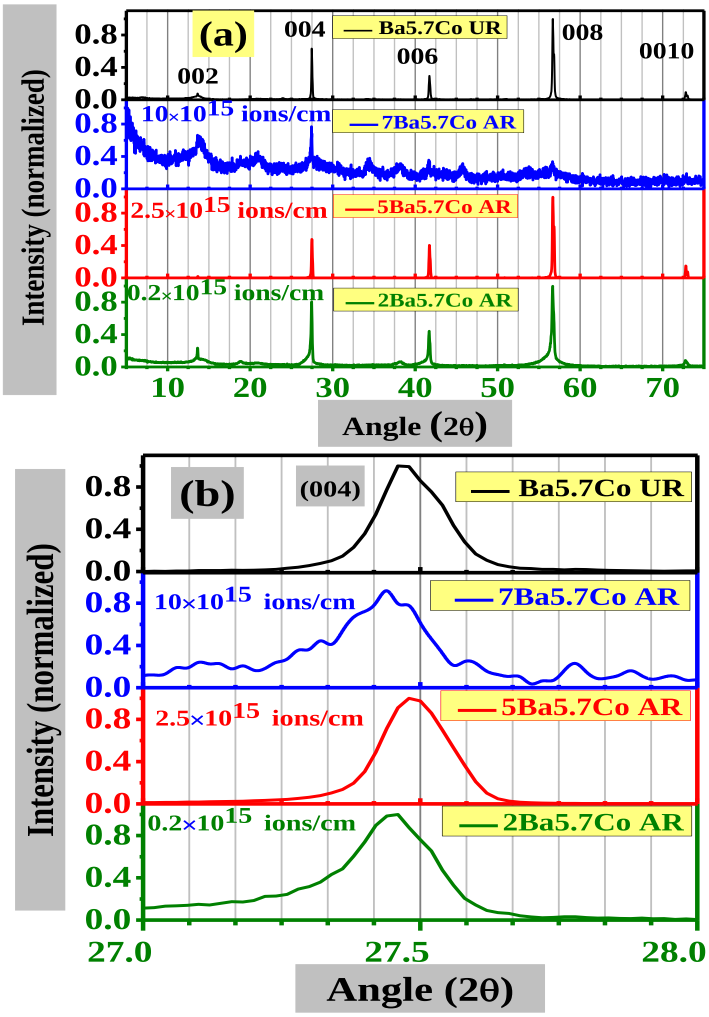

FIG. 1: (a) XRD (x-ray diffraction) pattern of plate-like Single Crystal $Ba(Fe_{1-x}Co_x)_2As_2$, x = 0.057 or 5.7% samples showing only 00l reflections – in UR (Un-radiation) & AR (After radiations, by 1.5 MeV Ar-beam) conditions. Sharp peaks in un-irradiated Ba122 samples (called "Ba5.7Co UR") become progressively broader after irradiation fluences 0.2×10^{15} (sample "2Ba5.7Co AR"), 2.5×10^{15} (sample "5Ba5.7Co AR") and10×10^{15} ions/cm^2. (sample "7Ba5.7Co AR"). (b) XRD in 2θ range 27° to 28° showing the 004 peak in details.

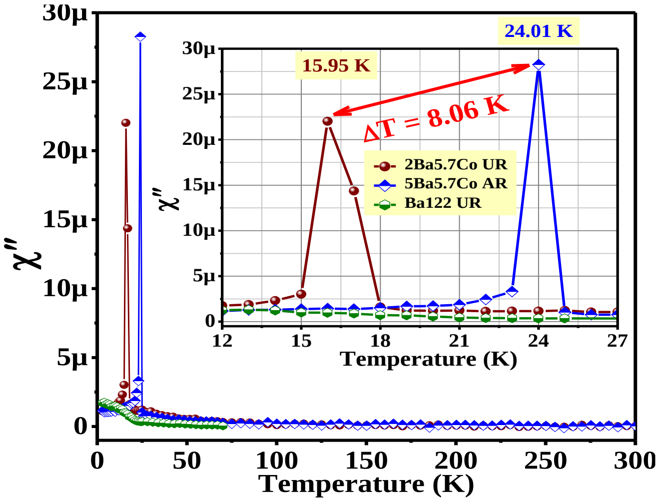

FIG. 2. Imaginary part of magnetic susceptibility (χ"in emu unit) (i) for the un-radiated pure $BaFe_2As_2$ (called Ba122 UR sample and indicated by hexagonal: half-filled symbol with olive colour) sample;(ii) un-radiated 5.7% Co doped $BaFe_2As_2$ sample (called 2Ba5.7Co UR sample shown by wine-coloured solid spheres; and(iii)5.7% Co doped sample after radiation to 2.5 × 10^{15} Ar-ions.cm^{-2} (called "5Ba5.7Co AR" sample indicated by blue-coloured half-filled diamond symbols.

Graphs for 2 to 300 K have been supplemented by enlarged view of the transitions in the inset. Irradiation, increases (FIG. 4, H=0 graphs)T_c from ~15.6 K to 23.8 K i.e. by 8.2 K in excellent agreement with the magnetic result. Wefind that the step in χ′ data show same T_c as χ″ peak (unpublished).

X-ray peaks in the un-irradiated samples are sharper. They broden progressively on irradiation to fluences 0.2×10^{15}, 2.5×10^{15} and 10×10^{15} Ar-ions/cm^2, indicating defect generation.[8]

Neither the imaginary part (χ″) nor the real part (χ′) of magnetic susceptibility show superconductivityfor the undoped sample, expected to be non-superconducting. We call it"Ba122UR" with olive colored graph in FIG. 2.

Results of magnetic (Fig.2) and electrical (Figs. 3 & 4) measurements will now be discussed. Fig. 2 for the x = 5.7% crystal showsT_c= 15.95 K from its χ″ peak in the un-radiated ("2Ba5.7Co UR" in figure in wine color) sample. After our Ar-irradiation of 2.5 × 10^{15} ions/cm^2, this sample "5Ba5.7Co AR", shows a surprising and significant increase of T_c toT_c= 24.01 K. Independent measurement of T_c from electrical resistivityalso records that 2.5 × 10^{15} Ar-ions/cm^2

In the present resistive transition, completion of the superconducting transition is being quoted as T_c. Onset superconducting transition temperature, real beginning of superconductivity, utilized in many publications, can be seen in our graphs to be much higher. Here, T_c from completion of superconducting transition has been preferred, as it usually matches theχ″ peak.

FIG.3 shows (a)a superconductivity-like sharp fall of electrical resistivity measured (at different magnetic fields:0 to6Tesla) directed along the c-axis of the single crystals of un-radiated $BaFe_2As_2$ ("Ba122 UR" sample, x = 0) on cooling from 21 K towards 10 K to a small non-zero value, a minimum; (b) detailed view of a slow semiconducting-type rise in resistivity of the same sample on cooling in the temperature range 10 to 2K; and (c) 2.5×10^{15}Ar-ion/cm^2irradiatedx = 0 crystal (called 4Ba122 AR sample) shows sharp fall of resistivity to zero valuewith enhanced T_c, and no semiconductor-like rise of resistivity on further cooling to T < T_c.

Since Fig. 2(i) does not show χ″ peak, bulk or sufficiently bulk superconductivity can be concluded to be absent in $Ba(Fe_{(1-x)}Co_x)_2As_2$, x = 0 sample. The sharp resistive transition of Fig. 3(a) in this crystal is what is called spurious superconductivity or trace superconductivity. It has to be due to a somehow completed or almost completed superconducting path formed from possible superconducting impurity phase/s,of such low volume fraction that detection by magnetic susceptibility failed (Fig. 2). Present pathcanbe assumed to bea case of "almost completed superconducting path" consisting of fewlong superconducting parts linked by semiconducting grains, to explain the semiconducting rise at lower temperatures.AboveT_c, combined normal state resistace of the longer length superconducting paths must be high compared to that of the few semiconducting links so that the metallic character dominates. Butbelow T_c, resistance of the semiconducting links is the only resistance so as to dominate and show the semiconducting rise of resistance on cooling in FIG. 3 (a) & (b) in the low temperature region(10 to 2 K).

A possible sources of above-mentioned superconducting (SC)impurity phases (providing SC paths) at crystal surface or domain boundaries in x = 0 sample can be growth of these SC phase/s due to inadvertant exposure to moisture, as in$FeTe_{0.8}Se_{0.2}$ powder[9,10]. The physics of the water exposure causing superconductivity is not yet clarified. But, the surface of $SrFe_2As_2$ film,as discussed also byKatase et al. in 2009,showsgrowth of Fe_2As, a SC phase, after an exposure to water[9,11]. Even this trace superconductivity appears to improve its $T_c$in

FIG. 3(c) due to our 1.5 MeV Ar irradiation. Actually, our observed decrease of T_c due magnetic field in Figs. 3 (a) & (c) should be taken as proof of real superconductivity, although it is non-bulk trace superconductivity.

Now, comes the detailed discussion (Fig. 4) of the bulk superconding sample, $Ba(Fe_{0.943}Co_{0.057})_2As_2$ single crystal HTSC,which responded positively, in FIG. 2, to magnetic measurements. FIG. 4shows superconducting transitions under 0, 1 and 6 T magnetic fields for $Ba(Fe_{0.943}Co_{0.057})_2As_2$ crystals before (Fig. 4(a)) and after (Fig. 4(b)) 1.5 MeV Ar^{6+}-irradiation.As expected, superconducting transition clearly shifts to lower temperatures on applying increasing magnetic fields, in un-radiated as well as irradiated samples.

Surprising increase of the completion of transition$T_c$of the 5.7% Co doped crystal due toAr-irradiation has already been discussed.This resistively observed 8.2 K increase of H = 0 T superconducting transition temperature compare well with 8.06 K increase observed from magnetic susceptibility measurements. It is a significant discovery in view of widely reportedradiation damage or decrease of $T_c$in Fe-HTSCs [12] and other superconductors due to energetic electron, ion and neutron irradiation.Here, (i) Radiation-induced vanishing, in Fig. 3(c), of semiconductor-like rise of resistivity on cooling the x = 0 sample in 10 to 2 K range, (ii) no semiconductor-like rise of resistivity in ~16 to 2 K range, on cooling the un-radiated x = 5.7% sample (Fig. 4(a)),and (iii)return in the irradiated sample (i.e. in FIG. 4(b)) of much smaller but non-zero resistivity below $T_c$and semiconductor-like rise of resistivity on further cooling, demand new explanation if these are internal phenomena in the sample.This issue will presently be left unsettled, as it does not affect the main investigation i.e. howT_c is modified by the irradiation used.

Review of literature [13-15] showed that essentially the same irradiation dosage 1.8 C/cm^{2}of 2.5 MeV electron beam produced (a) a suppression of 5% of T_c in $BaFe_2(As_{0.67}P_{0.33})_2$, (b) in contrast to an unusal T_c increases by about the same amount in FeSe. Different mechanisms and effectsin these two systems have, therefore, been believed to be at the root of the opposite effects in T_c. Infact, in above-mentioned FeSe (single crystals) the superconducting transition temperature T_c increases by 0.4 K from $T_{c0} \approx 8.8$ K, while the structural transition temperature T_s decreases by 0.9 K from $T_{s0} \approx 91.2$ K after the electron irradiation.After discussing several explanations for the $T_c$enhancement,a local strengthening of the pair interaction by irradiation-induced Frenkel defects has been proposed as the most likely cause of T_c increase. T_c can be affected also by irradiation induced strain in the sample.

More literature survey for the rare cases of $T_c$increase on fast ion and electronirradiation, follows, to illustrate conditions leading to such increase:

Sarkar et al [16] reported the enhancement and degradation of T_c in Bi-HTSC due to Li^{3+} irradiations"showedan increase of T_c only in such$Ba_2Sr_2CuO_{8+x}$(Bi-2212) samples that missed best$T_c$due to our deliberately different chemical preparation. These samples had excess oxygen than is optimal. Removal of this excess oxygen from these samples by 50 MeV Li irradiation caused the increase of T_c – as $T_c$vs. O-content is an inverted parabola. Li irradiation induced decrease ofT_c has been observed in the same work for samples having optimal O-content or less.

We reported earlier, an increase in critical temperature of over-doped $Bi_2Sr_2CaCu_2O_x$ superconductors due to alpha particle irradiation [17]. Our 13.6 MeV alpha irradiation of Nb_3Sn on Hastealloy (in a tape) in radiation damage studies of superconducting magnet materials as reported by De et al. in 1984 [18] showed small increase in T_c only at low dose, with expected decrease at higher doses. We conjecture that the initial increase of T_c must have been due to removal of mechanical strain in the Nb_3Sn layer, CVD-deposited on the metallic substrate.Mizukami et al. [19], reported in 2017 thatfor $BaFe_2(As_{1-x}P_x)_2$with x = 0.16 & 0.24, a final reduction of the temperature of the onset of superconductivity (here called T_c) upon the introduction of disorder by an electron beam with an incident energy of 2.5MeV.Authors found T_c to gradually increase (by up to 2K) with increasing radiation dose up to ≈ 3C/cm^2. Although the resitivity did not reach zero for x = 0.16, the initial increase of $T_c$is clear for both x = 0.16 and 0.24. Then, T_c decreased at higher doses. That means, although the superconducting transition temperature T_c is depressed at $x \gtrsim 0.28$ concentrations, it showed an initial increase at lower x as discussed. This implies that the superconducting dome tracked the shift (due to irradiation) of the antiferromagnetic phase. Ozaki et al. [20], used low-energy proton irradiation and created cascade defects in $FeSe_{0.5}Te_{0.5}$ (=FST) films. The FST films were irradiated with 1x10^{15}p/cm^2 dose of 190keV protons. This increased T_c,(R=0) from 18.0 to 18.5K due to the nanoscale compressive strain and proximity effect. Finally, it is to be appriciated that modification of superconductivity by various irradiations is an interplay of many factors, leading to T_c increase only in few cases.

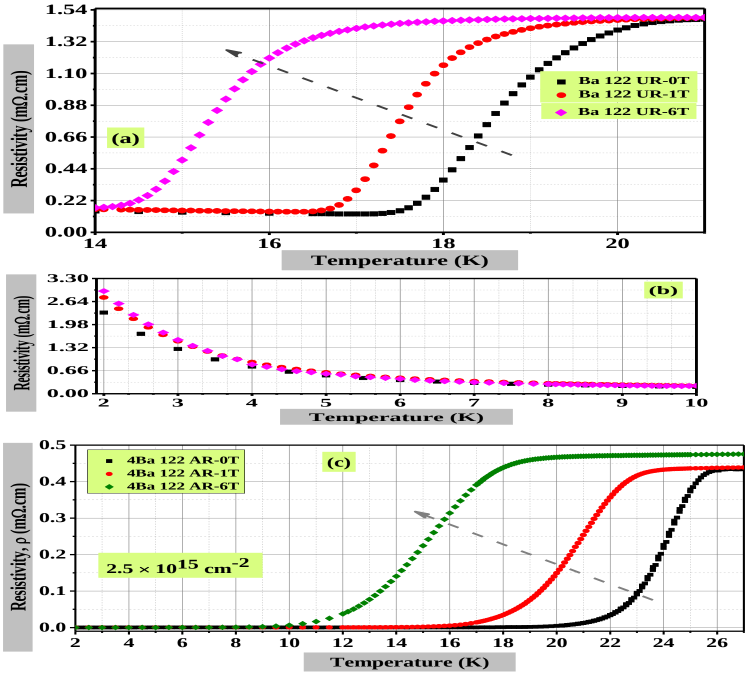

FIG. 3.Temperature dependence of electrical resistivity at different magnetic fields of0, 1, and 6 Tesla applied along the c-axis for single crystals of (a) un-radiated $BaFe_2As_2$ (called “Ba122 UR”) in the temperature range 21 to 10 K; (b) un-radiated $BaFe_2As_2$ (called “Ba122 UR” sample) in the temperature range 10 to 2 K, where resistance decreases with temperature showing a semiconducting nature; and (c) 2.5 × 10^{15} cm^{-2}Ar-radiated $BaFe_2As_2$ (called “4Ba122 AR” sample) in the temperature range 26 to 2K.

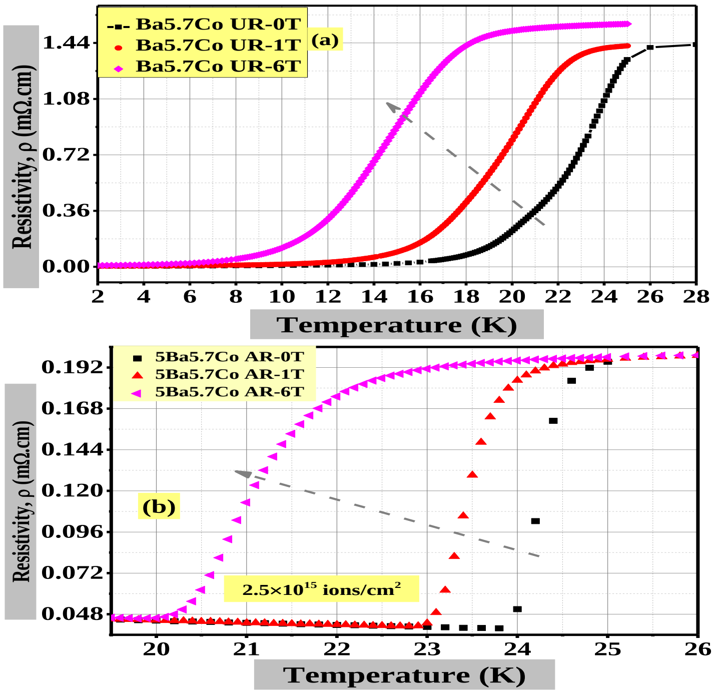

FIG. 4.Temperature dependence of electrical resistivity at different magnetic fields of 0, 1and 6 Tesla) applied along c axis for single crystals of (a) un-radiated 5.7% Co doped $BaFe_2As_2$ (“Ba5.7Co UR”) in temperature range25.5 to 2 K, showing superconducting transition, (b) 2.5 × 10^{15}Ar-ion/cm^2 irradiated 5.7% Co-doped $BaFe_2As_2$ (“5Ba5.7Co AR”) in the temperature range 25 to 15K.

4. CONCLUSION

All characterizations detect radiation induced changes in these $Ba(Fe_{(1-x)}Co_x)_2As_2$Single Crystal samples due to irradiation with 1.5 MeV (entering energy) Ar^{6+} beam. We observe T_c increase on 1.5 MeV Ar irradiation, but with broadened superconducting transition. Single Crystal samples with x = 5.7% Co show convincing T_c increse of ~ 8 K from electrical as well as magnetic measurements.

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