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ALPHA IRRADIATION DAMAGE TO SPECIFIC HEAT IN THALLIUM 2212 AND 2223 OXIDE SUPERCONDUCTORS

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Abstract: Irradiation by $2x10^{16}$ He^{++}/cm^2 at 40 MeV reduces superconducting volume fraction, as probed by the specific heat jump, significantly in Tl-2223 and drastically to an unmeasurably (down to 20K) low value in Tl-2212. A.C. susceptibility study of the onset of superconductivity is also reported. The mechanism for higher damage in Tl-2212 is discussed.

Keywords: Radiation damage, Tl-superconductors

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1. Introduction

The present work is a preliminary report of radiation damage to specific heat, C, and a.c. susceptibility, acs, of the n = 2 and 3 members of $Tl_2Ba_2Ca_{(n-1)}Cu_nO_{(2n+4)}$ superconducting pellets [1-3] due to 40 MeV alpha-irradiation, there being no earlier data. The jump, ΔC, in the specific heat across the superconducting critical temperature, T_c, is [2] proportional to the superconducting volume fraction in the sample, which should be investigated in radiation damaged samples.

By making the sample thinner than the range of the irradiating alpha-beam, we ensured that it passes out of the ample, causing pure radiation damage and no implantation. The 40MeV alphas loose their energy through electronic excitations, important at such high energies, and elastic collisions with the lattice atoms. While looking for the mechanism of such radiation damage in oxide superconductors, it is often overlooked [4] that the process of electronic excitations can [5], unlike in the metallic superconductors [6], affect the covalent-like bonds in these oxides and thus damage their superconductivity. The specific heat , $C = C(e) + C(l) + C_o$, of solids arises from the electronic (e) and lattice-vibration (l) parts and other contributions collectively represented by C_o. Since the electronic as well as the lattice parts are directly affected by a high energy alpha irradiation in Tl-superconductors, and experimentally one measures the total specific heat , C, measurable changes in C can be anticipated in such irradiation experiments.

The present paper also addresses to the well-known problem of estimating $\Delta C/T_c$ on account of the large phonon background, C_l, at temperatures of the order of 100 K for T_c and adopts a logical semi-emperical solution .

2. Experimental Outline

Tl-2212 and Tl-2223 pellets with about 50% of the theoretical density were prepared [5] at the CRISMAT Laboratory, ISMRA, Caen from oxides, with X-ray showing no impurity phases. These were shaped into 500 and 300 micron thick Tl-2212 and Tl-2223 bars weighing about 55 and 66 mg, respectively, to avoid implantation as discussed earlier.

Specific heat has been measured by a continuous heating adiabetic technique [7,8] at the Nuclear Research Centre, Karlsruhe. Essentially the rate of temperature-rise of the sample, dT/dt, for its electrical heating, under nearly adiabetic condition, by a known heating power, P, is measured to determine the specific heat, C(T)= P/(dT/dt). Using 1 Oe a.c. magnetic field, susceptibility has been measured (fig. 1) to estimate the on-set T_c values.

For room temperature alpha-irradiations in the cyclotron at the VEC Centre, Calcutta, an aluminium target holder with secondary electron recollection arrangement [9] was used. Earthing the insulated target-holder through a current integrator, beam-current as well as the fluence, $2x10^{16}$ He^{++}/cm^2, were accurately measured.

3. Results and Observation

A.C. susceptibility data (fig. 1) shows that after irradiation the on-set T_c is practically unchanged to within about 1 K at about 110 K for for Tl-2212 and at about 119 K for Tl-2223.

Fig. 2 shows the measured (C/T) vs. T plots for Tl-2212. Compared to the clear kink at about 102 K for the unirradiated sample (graph a), later identified as the superconducting jump, there is no such structure in the irradiated sample (graph b). So there is no superconducting jump in the irradiated Tl-2212.

To make a jump in C/T vs. T data prominent, various authors subtract, from the measured specific heat, a phonon part, often estimated by fitting an un-explained [10] combination [11] of Debye and Einstein functions. Such fitting functions have more fitting parameters but not yet [12] a comprehensive physical model. Hence the present work estimates $(C/T)_{BG}$, the "background" appropriate for a non-superconducting

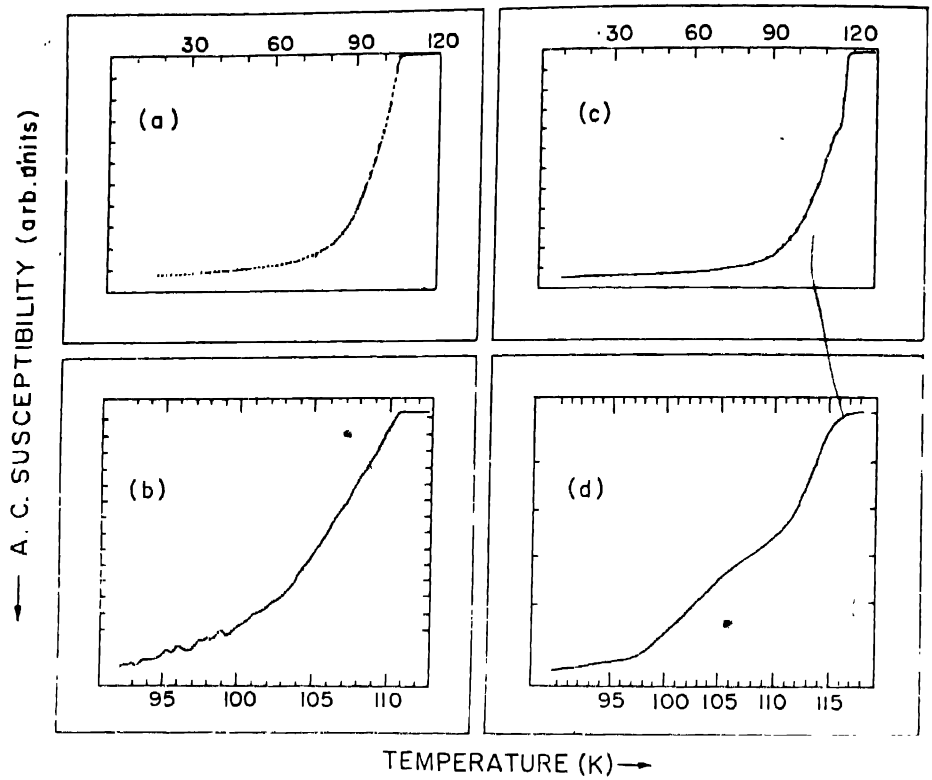
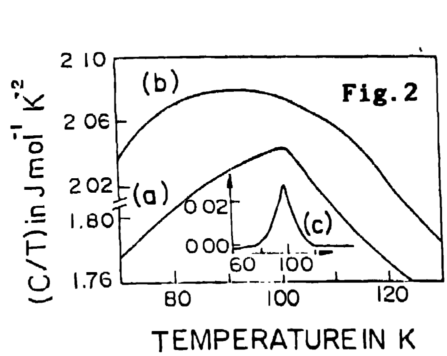


Fig. 1. Superconductivity onset in a.c. susceptibility for (a) unirradiated Tl-2212, (b) irradiated Tl-2212, (c) unirradiated Tl-2223 & (d) irradiated Tl-2223 samples.

but otherwise equivalent sample, by fitting [13] a simple polynomial: $C/T = A_o + B/T^2 + A_i T^i$, i= 1 to 5, to our data on both sides of the superconducting transition region. For example data below 80K and above 109K of the unirradiated Tl-2212 have been fitted to give $(C/T)_{BG}$ vs. T, which is subtracted from the experimental data (graph a) to get graph c in fig.2 and hence conclude a jump: ($\Delta C/T_c$) = (33+3)mJ $mol^{-1}K^{-2}$at T_c=102 K. This compares well with the values 35+10 and 25 to 29 mJ mol^{-1} K^{-2} reported [7,14] earlier.

Similarly, "background"-corrected specific heat data for



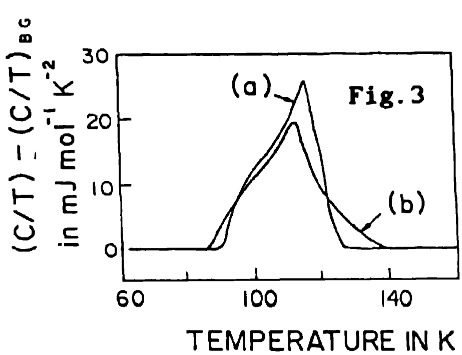


Fig.2. Specific heat, C, vs. temperature T, data in C/T vs. T form for (a) unirradiated and (b) irradiated Tl-2212 pellets. Graph c in the inset is the unirradiated data minus its non-superconducting background (explained in the text) and represents C/T vs. T.

Fig.3. Measured specific heat, C, minus its non-superconducting background, $(C)_{BG}$, for (a) unirradiated and (b) irradiated Tl-2223 sample in $[(C/T)-(C/T)_{BG}]$ vs. T form.

Tl-2223 is presented in fig.3. It shows a jump of (28±3) mJ $mol^{-1}K^{-2}$ at 116K for the unirradiated and (18±2) mJ $mol^{-1}K^{-2}$ at 113K for the irradiated sample. The unirradiated value is again comparable to the reports [7,12] of (20±10) and 29 mJ $mol^{-1}K^{-2}$.

Discussion & Conclusion

The main result is that the superconducting fraction, as probed by our specific heat measurement, is reduced by the alpha irradiation, significantly in Tl-2223 and drastically to a unmeasurably low value in Tl-2212. However, the onset T_c, as measured by the a.c. susceptibility is practically unchanged by this irradiation. This tends to indicate that some traces of the original superconducting phases are left undamaged in both the samples to provide shielding currents and thus show almost unchanged onset T_c in the acs measurement.

Radiation damage in Tl-superconductors via electronic energy loss of Xe and other heavy ion irradiations at a few GeV energy is well-known [5] and already studied by electron microscopy. Such electronic contribution for our 40 MeV alpha-irradiation appears to be responsible for more damage to Tl-2212, as discussed in the next paragraph. In addition, there is evidence of lattice damage by the 40 MeV irradiation in our observed change of high temperature (above 120K)specific heat due to irradiation.

More damage to Tl-2212 than to Tl-2223, can be explained from the difference that Tl-2212 has relatively more blocking/non-conducting layers, in which electronic exitations by the alpha-particles can cause [5] effective damage. Metal-like CuO_2-layers, relatively more in Tl-2223, are less affected by the electronic energy loss of the alpha-beam, as we have already indicated. This explains our observation and indirectly establishes the significant role of damage [5] via electronic energy loss in case of oxide superconductors.

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