Physical properties of structured high Tc thin films by laser ablation on SrTiO3, ZrO_2 and Si substrates

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Introduction

Laser ablation is a convenient and adequate technique for the deposition of high quality high Tc superconducting films on a large variety of substrates. These substrates are often monocrystalline but in the case of silicon a polycrystalline buffer layer is generally used. The granularity of the buffer layer may strongly influence the superconducting properties of the high Tc layer.

We are studying the effect of the substrate granularity on the properties of laser deposited YBaCuO films. Using an excimer laser (308 nm, 2 Hz, 80 mJ), we deposited layers under identical process parameters (1) on the single crystals $SrTiO_3$(100) and ZrO_2(100)on one hand, and on Si(111) with a polycrystalline ZrO_2 buffer layer on the other. The buffer layer was prepared according to the following procedure (2). An amorphous layer of NiZr (23-77) was deposited on Si(111) by electron gun evaporation and annealed during 2 hours at 500 °C in an oxygen atmosphere. The oxygen reacts with Zr and forms the polycrystalline ZrO_2 of about 100 nm thickness with a typical grain size of 1 µm. The Ni reacts with Si leading to an epitaxial layer of 40 nm $NiSi_2$. A study about interface reactions is presented by Aarnink et al. (3).

The YBaCuO films were investigated using three different measuring equipments. Temperature dependent resistivity measurements were performed with the substrate temperature during deposition as a parameter. Field cooled, zero-field cooled and remanent magnetization were measured on some high quality films and finally the field dependence of the resistivity was studied. This paper presents first results.

Results

Temperature dependent resistivity measurements

The effect of the substrate temperature during deposition of YBaCuO on $SrTiO_3$, ZrO_2, and Si($NiSi_2$)ZrO_2 on the resistivity is given in figure 1. Three temperature intervals can be distinguished in the T_{onset} and T_{zero} versus $T_{substr.}$ curves.

At low deposition temperatures the results for the different substrate types correspond very well. The onset temperature decreases with decreasing $T_{substr.}$ and the transition becomes broader leading to a low T_{zero}. From additional X-ray diffraction measurements it is found that the films are amorphous for a large part with grains of the correct phase, mostly c-axis oriented but some randomly. The oxygen stoichiometry, calculated on basis of the X-ray 2θ values of the (001) reflections, varies from 6.6 to 6.8 in the temperature range from 600 to 700 °C and is substrate independent. This effect causes the decrease of T_{onset}.

At the high temperature side the results with the single crystalline substrates are the best. The onset temperature is high (and constant over the substrate temperature range) and the transition is within 1 K, leading to zero resistivity at 90 K. X-ray diffraction analysis only shows c-axis orientation and from the peak position we calculated an oxygen stoichiometry of almost 7. The films deposited at Si(NiSi$_2$)ZrO$_2$ in this temperature range show a broadening of the transition, whereas the onset temperature does not change. The X-ray diffraction analysis again shows perfect c-axis orientation with an oxygen stoichiometry of 7. The temperature, at which zero resistivity is derived, decreases due to the diffusion of the silicon along the grain boundaries, which are clearly present now in the YBaCuO layer. The bulk properties of the material are still correct but the transport properties are determined by these grain boundaries.

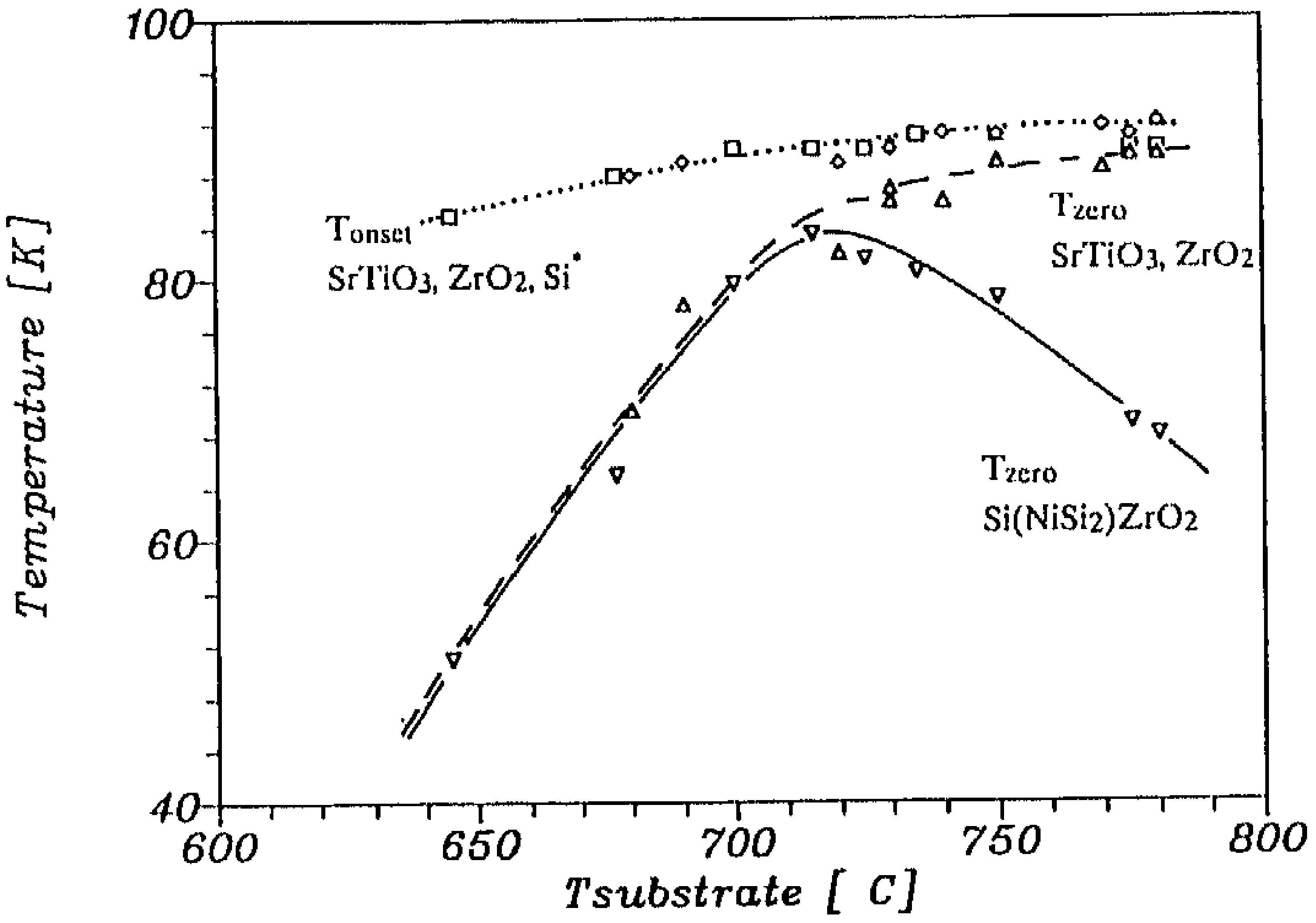

Fig. 1: Tonset and Tzero of YBaCuO films deposited on monocrystalline SrTiO$_3$ and ZrO$_2$ (diamant and uptriangle) and on polycrystalline Si(NiSi$_2$)ZrO$_2$ (Si*), represented by block and downtriangle.

The intermediate temperature range gives the optimum preparation temperature for the Si-substrate with respect to resistivity. It is the balance between amelioration due to reaching correct oxygen stoichiometry and deterioration due to Si diffusion along the grain boundaries.

Field cooled, zero-field cooled and remanent magnetization

Magnetization measurements at low fields have been performed on YBaCuO films on single crystalline ZrO$_2$, whereas the results on the polycrystalline ZrO$_2$ are in progress. The sample is placed in a SQUID magnetometer with the applied field parallel to the c-axis (perpendicular film). Zero-field (ZFC), field cooled (FC) and remanent (REM) magnetization were measured in background fields up to 2 mT. Fig. 2 shows the ZFC data obtained from a $5x5x2 \cdot 10^{-4}$ mm^3 YBaCuO film with Tons = 91 K and Tzero = 86 K. The magnetization values are divided by the applied field (1 a.u. is $3.5 \cdot 10^{-9}$ m^3). The ZFC and REM values are equal but opposite in sign and the FC values are equal to zero within the experimental accuracy. Thus there is no Meissner effect and the ZFC curves are determined by shielding currents. This explains that the magnetization is zero when Tzero is reached.

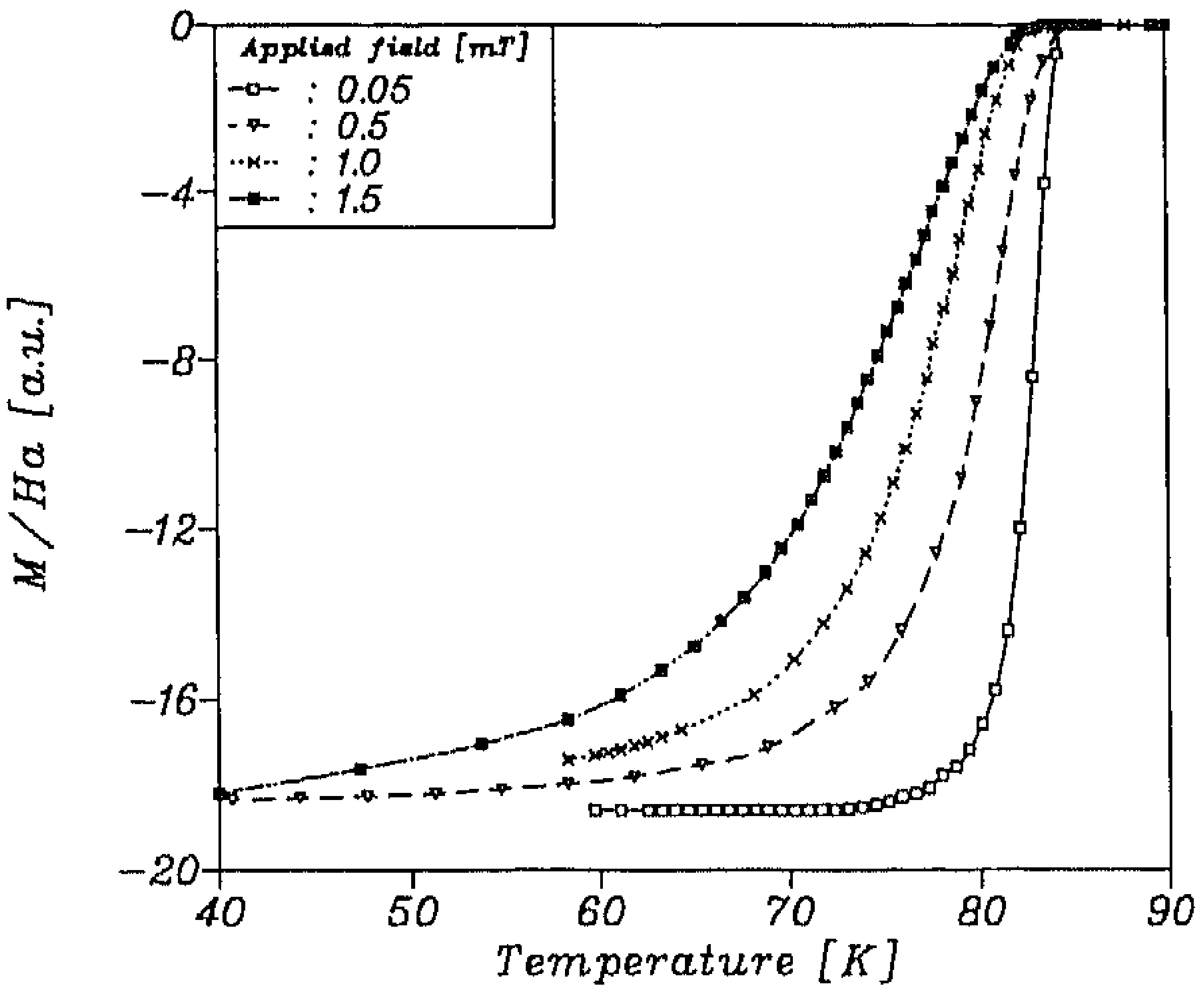

Fig. 2: ZFC values obtained from a $5x5x2 \cdot 10^{-4}$ mm^2 YBaCuO layer on ZrO_2 with a field step of 0.05, 0.5, 1.0, and 1.5 mTesla. The magnetization values are divided by the applied field (1 a.u.≈ $3.5 \cdot 10^{-9}$ m^3).

The relation between magnetization and the applied field in the superconducting transition region is given in fig. 3. The magnetization is expressed in arbitrary units (1 a.u. is about $2.6x10^{-8}$ Am^2). Calculations based on the Bean's model are currently performed.

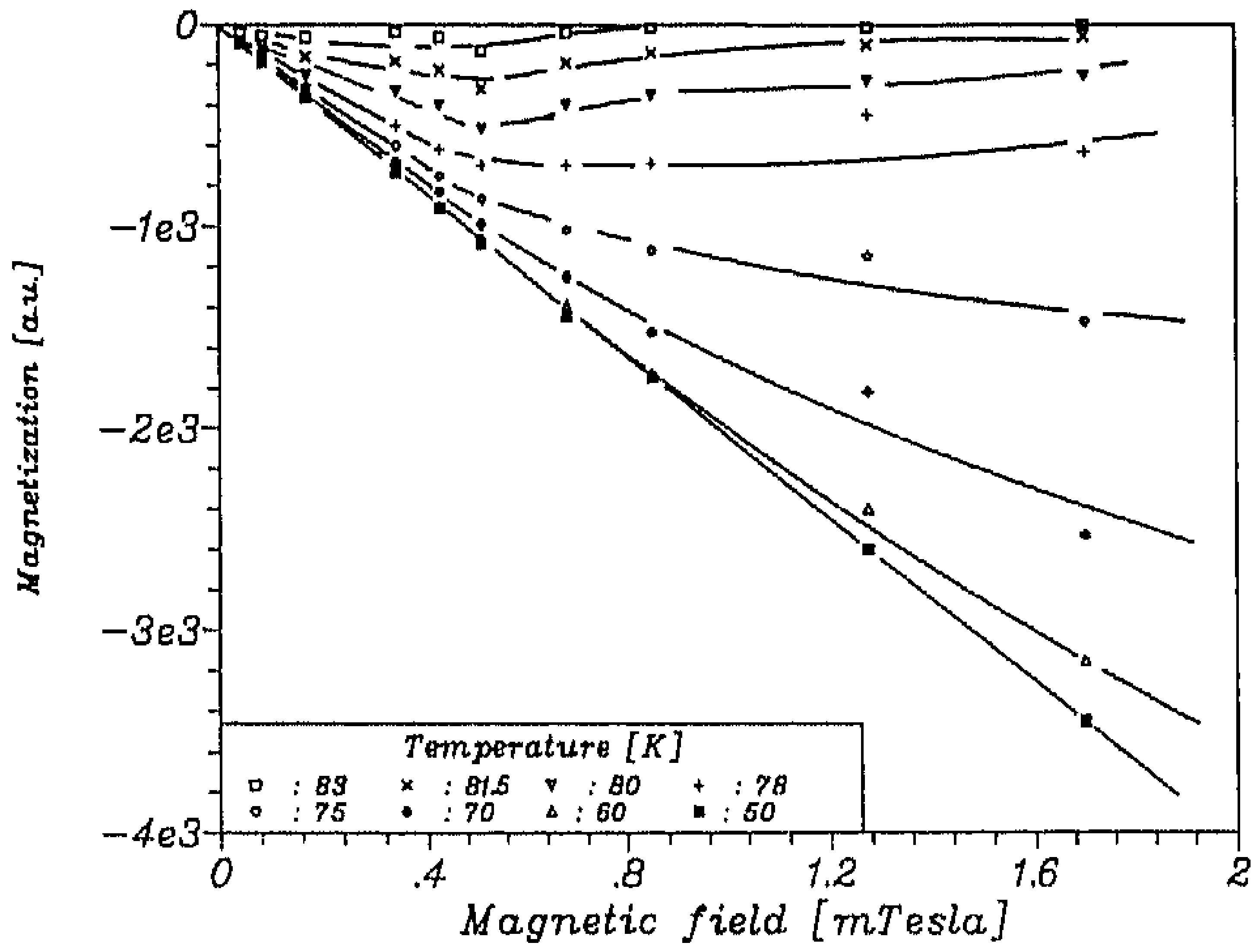

Fig. 3: Magnetization versus applied magnetic field for different temperatures (-1e3 a.u.=$-2.6 \cdot 10^{-5}$ A/m^2)

Field-dependent resistivity measurements

Resistivity measurements at different magnetic fields have been performed on an YBaCuO film on single crystalline ZrO_2. The film had a bridge with a width of 10 μm and a length of 100 μm, structured by the excimer laser. The magnetic field was varied between 0 and 4 Tesla.

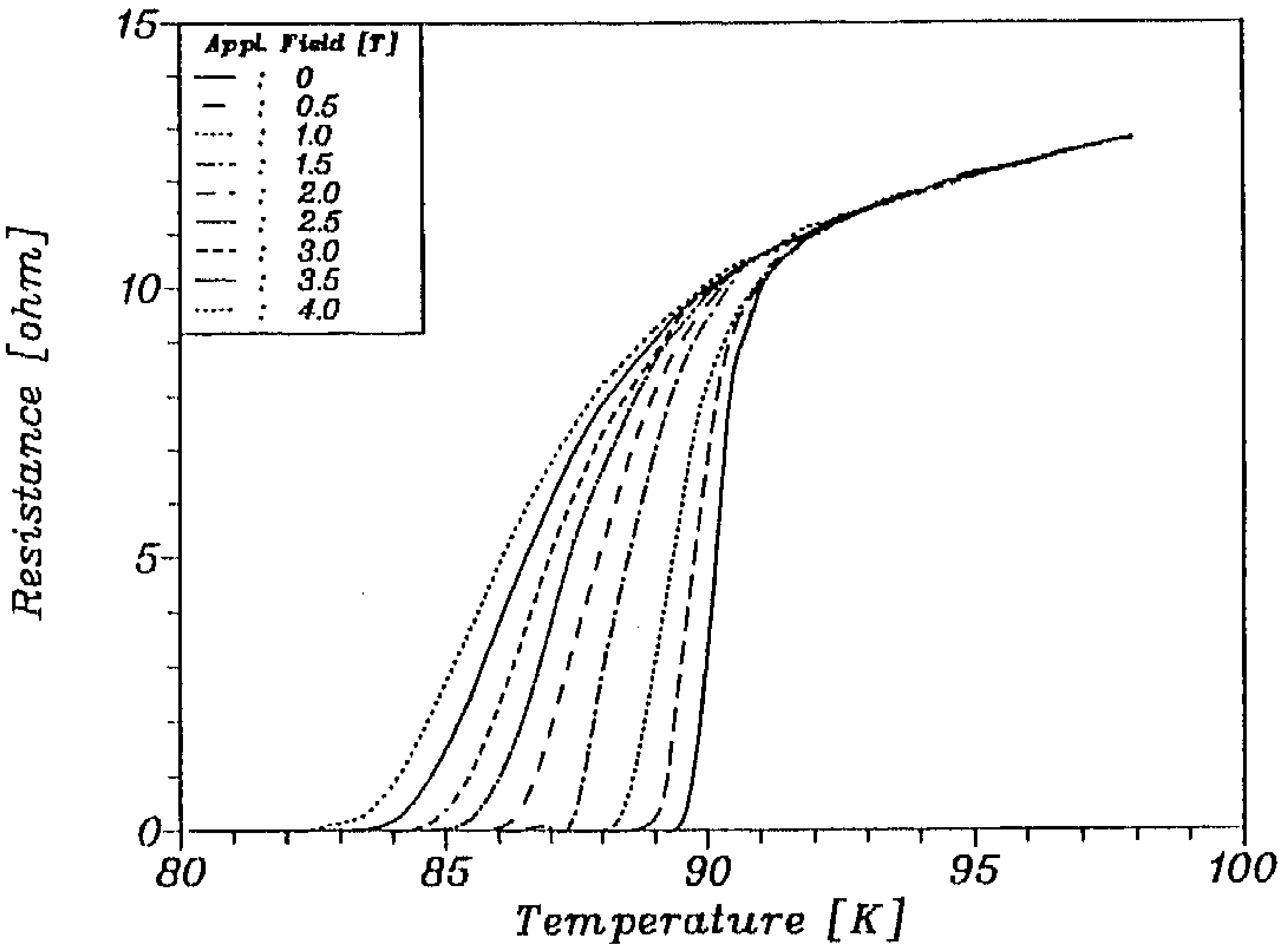

Fig. 4: Field dependent resistivity for a 10 x 100 μm bridge of a 200 nm YBaCuO layer on ZrO_2. Applied field (0, 0.5, 1.0, 1.5, 2.0, 2.5, 3.0, 3.5, and 4 T) parallel to the c-axis.

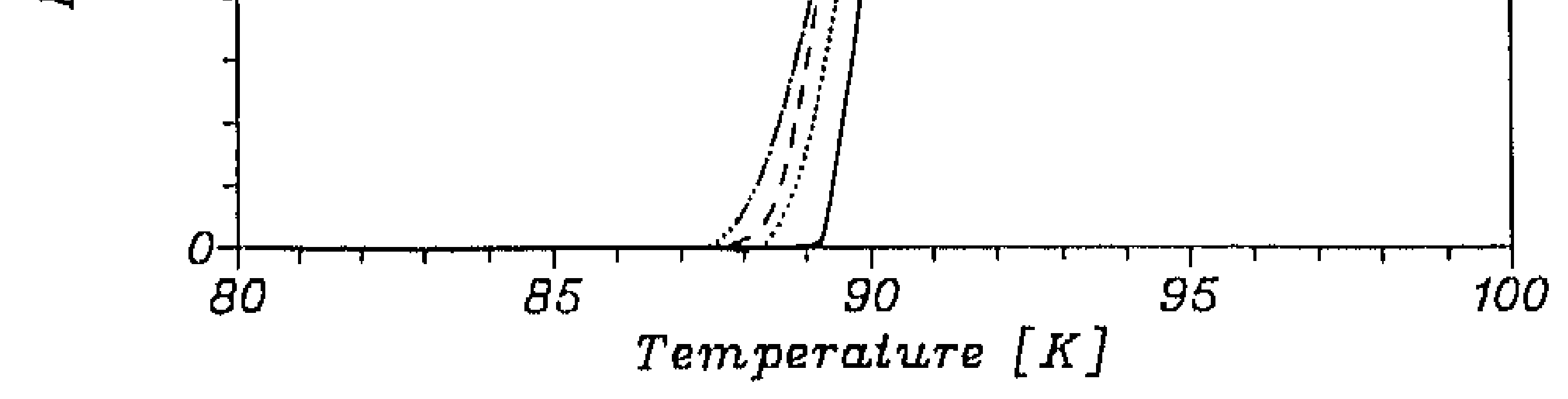

Fig. 5: Field-dependent resistivity for the sample of fig. 4. Applied field (1.0, 2.0, 3.0, 4.0 T) parallel to the a-b plane.

The direction of the magnetic field was parallel or perpendicular to the c-axis of the film and in both cases perpendicular to the current. The results are shown in fig. 4 and fig. 5. With the field parallel to the c-axis the onset temperature remains about the same at different fields, while the T_{zero} decreases. In the case of the magnetic field perpendicular to the c-axis (fig. 5), the decrease in the T_c is much smaller.

For the analysis we follow the theory of Tinkham (4) where the resistance at the transition R, relative to the normal state resistance Rn, is expressed by $R/R_n = I_o^{-2}(x)$, where Io is the modified Bessel function with argument $x = A \cdot (1-t)^{3/2} / 2B$. Here $t = T/T_c$, H the applied field and $A \approx 3.5\ \beta\ J_{co}(0)$ for YBaCuO ($\beta \approx 1$, $J_{co}(0) = J_c\ (B{\approx}0,\ T{=}0)$).

For simplicity we have taken the points $R/R_n = 0.5$, so that t can be easily obtained from the measurements. It is found for both directions that (1-t) is proportional to $B^{2/3}$ in the field range from 1.5-4 T, whereas at lower field a steeper dependence is found. Taken ß equal to 1, the critical current $J_{co}(0)$ is derived as 3×10^6 A/cm^2 for H ∥ c-axis and as 3×10^7 A/cm^2 for H ∥ a-b plane, leading to the anisotropy ratio of 10. This value is somewhat larger than the one presented by Tinkham for a single crystal.

Conclusions

We are studying the differences in electrical and magnetic properties of YBaCuO on monocrystalline and polycrystalline substrates. The superconducting transition temperature range is substrate independent at low deposition temperatures but at high temperatures Si diffusion through polycrystalline ZrO_2 on Si substrates causes a severe deterioration of the zero-resistance temperature. Low-field magnetization experiments for monocrystalline substrates (field parallel to c-axis) exhibit a non-measurable Meissner effect whereas zero-field cooled and remanent magnetization are equal but opposite in sign. Field-dependent resistivity measurements show a strong field effect on the superconducting transition range when the field is parallel to the c-axis and a much weaker influence with the field parallel to the a-b plane. The flux creep model of Tinkham has been applied leading to $J_{co}(0) = 3 \cdot 10^6$ A/cm^2 when H ∥ c-axis and $3 \cdot 10^7$ A/cm^2 with H ∥ a-b plane.

References

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