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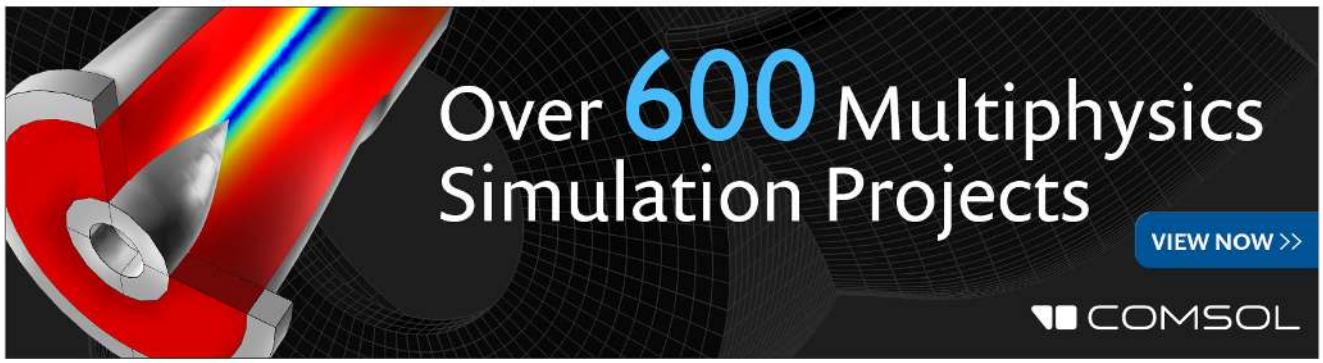

Origin of peak effect in the magnetization hysteresis loop of melt-processed $REBa_2Cu_3O_y$ superconductors

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Melt-processed Sm-123 samples with an excess Sm_2BaCuO_y (Sm-211) phase is studied by transmission electron microscope (TEM) and superconducting quantum interference device magnetometer, to explain the origin of the peak effect exhibited in the magnetization hysteresis loops of light RE-123 samples in intermediate fields at 77 K. TEM images of the sample showing peak effect reveal dilute concentrations of areas with ortho-II structure distributed on a nanometer scale within the twin regions of the sample. The ortho-II structure is an oxygen deficient 123 with a lower T_c than the bulk of the material and will be a source of flux pinning by turning normal in high fields, thus contributing to the peak effect. With an increase in Sm-211 concentration, the peak effect disappears from the hysteresis loop, as does the ortho-II structure in the TEM images. © *1997 American Institute of Physics.* [S0003-6951(97)03852-7]

Melt processing is an effective method for increasing the critical current density (J_c) of $REBa_2Cu_3O_y$ (RE-123) systems.[1] The enhancement of J_c in melt-processed samples is in part due to an increase in the flux pinning by the defects that occur at the Y-123/Y-211 interface.[2,3] A peak effect in the dc hysteresis loops ($M-H$ loops) showing an increase in J_c of single-crystal and melt-processed RE-123 samples have been observed at 77 K in high fields. It has been found that in Y-123 single crystals, the microstructure consists of a dilute mixture of ortho-I ($T_c=90$ K) and ortho-II ($T_c \sim 60-90$ K) structures generated by spinodal decomposition.[4] Because of the different superconducting properties of these structures, the oxygen-deficient regions have been proposed as flux pinning centers.[4–6] It has been reported[7,8] that melt-processed light rare-earth (Nd, Sm) 123 samples also show a large peak effect at 77 K. This peak effect was attributed to field-induced pinning from low-T_c solid-solution regions, which turn normal and act as flux pinning centers. Recently, Nakamura *et al.*[9] have reported that the low-T_c regions have a tweed morphology with a modulated structure containing compositional variations. This occurs because the Ba site can be substituted by a light rare-earth atom, leading to a different structure and a lower T_c. They also correlated the microstructural features with that resulting from spinodal decomposition in an aged alloy.[10] In the Nd-123 single crystal, tweed features were present within the twins only when the sample was oxygenated at 500 °C. If the annealing around 500 °C was avoided, those features were absent, and so also the peak effect. In the literature, to the best of our knowledge, there are no reports of observations of such regions in the melt-processed Sm-123 system. Here, we report the correlation between transmission electron microscope (TEM) images and $M-H$ loops of melt-processed Sm-123 samples having a starting composition with an excess (10 and 20 mol %) Sm_2BaCuO_y (Sm-211) phase. An explanation is offered for the origin of the peak effect based on the observations.

The samples were prepared by taking a mixture of Sm-211 plus liquid phases ($BaCuO_2$ and CuO). Melt processing was done by melting the samples at 1115 °C for 10 min and then subsequently slow cooling from 1060 to 950 °C at a rate of 1 °C/h, in commercial argon atmosphere (with an oxygen content of <4 ppm). The melt-processed samples were oxygenated in flowing oxygen atmosphere, by cooling from 600 to 250 °C as per the schedule:

$$\begin{array}{cccc} 6\text{ h} & 2\text{ h} & 24\text{ h} & 48\text{ h} \\ 600\ ^\circ\text{C} \rightarrow & 500\ ^\circ\text{C} \rightarrow & 450\ ^\circ\text{C} \rightarrow & 350\ ^\circ\text{C} \rightarrow 250\ ^\circ\text{C}. \end{array}$$

The x-ray diffraction patterns of the samples showed the formation of Sm-123 with some amount of the Sm-211 phase. The samples were characterized by TEM and superconducting quantum interference device (SQUID) magnetization measurements. The superconducting transition temperature (T_c) of the samples (defined as the onset of diamagnetism in the low-field temperature variation of dc magnetization) as measured by the SQUID magnetometer was 90 and 91 K, respectively, for the samples with 10 and 20 mol % Sm-211.

Figure 1 shows the dc $M-H$ loop of the 10 and 20 mol % samples at 77 K with the field applied parallel to the c axis of a rectangular sample with dimensions 2.1 mm×1.6 mm×1 mm for the 10 mol % sample, and 2.75 mm×1.8 mm×1 mm for the 20 mol % sample, respectively. The c axis is along the larger dimension of the samples. The 10 mol % sample exhibits a peak around the field region of the 1–2 T region. The TEM image [Fig. 2(a)] of this sample at high magnification shows microtwins, defects like stacking faults, and dislocations. Though a tweed contrast could not be observed, the features within the twins

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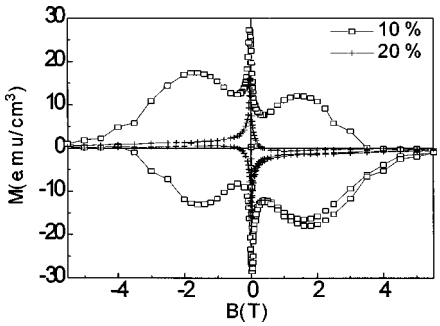

FIG. 1. $M-H$ loop of the Sm-123 samples containing 10 and 20 mol % Sm-211 at 77 K.

at high magnification resemble those reported in Ref. 9. There are no modulated structures present, but the material remains as a single phase with clusters of a different structure. The exact composition could not be obtained due to the limitations in the TEM setup. The small-area electron diffraction (SAD) pattern within the twin region [Fig. 2(b)], did not contain any satellite spots, confirming the absence of modulated structures. This pattern could be indexed to an orthorhombic structure of Sm-123, with no spots left unindexed. Figure 2(c) shows the electron diffraction pattern from another region in the sample, which exhibits streaks around the central bright spot. This is a feature indicating the presence of the ortho-II structure in the region, which is an

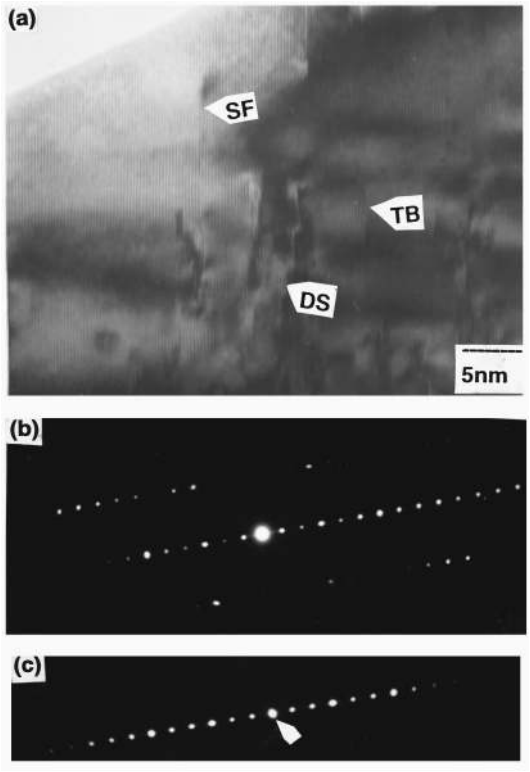

FIG. 2. (a) TEM image of the 10 mol % sample, showing the second phase within the sample. Also marked are TB=twin boundary, SF=stacking faults, and DS=dislocation. (b) SAD pattern of the 10 mol % sample within the twin boundary. The zone axis is [010]. a and c-axes are lying in the plane of the image. (c) SAD pattern of the 10 mol % sample in another region of the sample. Due to the fine scale distribution of the second phase, streaks around the central bright spot are faintly visible. This is an indication of the presence of ortho-II structures.

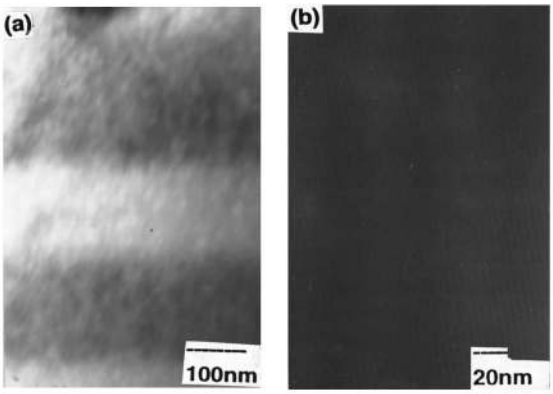

FIG. 3. (a) TEM image of the sample with 20 mol % of Sm-211, showing the twin boundary at low magnification. (b) High magnification TEM image of the 20 mol % sample. The sample does not contain any secondary structure within the twin regions.

oxygen-deficient RE-123 structure with a doubling of the unit cell. Khachaturyan and Semenovskaya[11] have shown that the presence of diffraction maxima at 1/2{100} generic points is a feature of the ortho-II structure. It has also been mentioned by them that if the 123 sample is cooled very slowly while being oxygenated, a secondary tweed structure will form within the twins by an oxygen ordering, which contains the ortho-I and ortho-II structures. The presence of the ortho-II structure has been reported in Y-123 by many groups,[4,12] and also in an oxygen depleted Y-123 sample,[13] in which the oxygen depletion was due to an irradiation effect. In the present study on the Sm-123 sample, the possibility of irradiation in the TEM was minimized by using an accelerating voltage of 160 kV, which is below the value that would have irradiated the sample.[14] Thus, we conclude that the two structures present are: ortho-I with a T_c above 90 K and an ortho-II structure with a $T_c \sim 60-90$ K. The ortho-II structure in the sample is distributed on a nanometer scale and will be a source of flux pinning at 77 K in high fields as it turns normal.

In support of the above it may be mentioned that, Khachaturyan[15] has reported that it is thermodynamically possible for the 123 phase to decompose ''spinodally'' around 500 °C during oxygenation. As it passes sequentially through a series of stoichiometric compositions of Magneli-type phases, a cell doubled ortho-II structure having oxygen deficiency will result. Thus, it is possible that ''spinodal decomposition'' into the ortho-I and ortho-II structures is also taking place in the LRE-123 systems at 500 °C during oxygenation. The idea of field-induced pinning from the low-T_c solid-solution regions may not be valid, because the present data did not indicate their presence. The solid-solution regions will have a lower c-lattice parameter because of the substitution of a Ba atom by a rare-earth atom.[16] But in the SAD patterns, the values of the c parameters were identical. Also, the solid-solution regions would appear in the SAD pattern as different reflection spots, but no such extra unindexed spots were present.

Similar studies were done on the sample having a starting composition of 20 mol % of Sm-211. A peak effect was not observed in the M–H loop (Fig. 1), unlike in the earlier sample. The TEM image [Fig. 3(a)] shows the presence of

twin boundaries, and the high magnification picture [Fig. 3(b)] within the twin boundaries does not have the features as observed in the earlier sample. The data clearly show that a spinodal decomposition is not taking place in the sample containing 20 mol % Sm-211.

It is known that[17] for an oxide alloy to decompose spinodally, the three important controlling parameters are composition of the alloy, the annealing temperature, and the elastically soft directions of the solid. Variations in any one of them may disallow the spinodal decomposition. In the literature, it is reported that stoichiometric melt-processed RE-123 samples and single crystals show the peak effect because of spinodal decomposition during oxygenation. We have shown that a sample with an addition of 10 mol % 211 also exhibits this phenomenon. The sample with 20 mol % 211 does not exhibit the peak effect because of the absence of ''spinodal decomposition.'' The exact reason for the absence of ''spinodal decomposition'' in this sample is difficult to isolate, because of the many factors involved as mentioned above.

In conclusion, the origin of the peak effect at 77 K in the M–H loop is explained for a melt-processed Sm-123 sample with excess Sm-211. The peak effect is due to the ''spinodal decomposition'' of the 123 during oxygenation. The sample remains as a single phase but has clusters with an ortho-II structure distributed on a nanometer scale within the twin regions. The ortho-II structure, having a lower T_c than the bulk due to oxygen deficiency, acts as flux pinning centers by turning normal at high fields. The sample having 10 mol % of 211 showed this effect. An increase in the 211 concentration (i.e., 20 mol %) for a second sample disallowed the spinodal decomposition, and hence, no peak effect was present.

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