## THERMAL AND MAGNETIC FIELD DEPENDENCE OF THE CRITICAL CURRENT OF 3D DISORDERED JOSEPHSON JUNCTIONS ARRAYS

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Tridimensional disordered Josephson junction arrays (3D-DJJA) have been produced in a controled manner by our group. Arrays were fabricated from granular superconductors, using powder of either low or high-temperature superconductors. All relevant signatures of JJAs are exhibited by the arrays, including the typical Fraunhofer dependence of the critical current with the applied magnetic field, a magnetic remanence and the Wohlleben effect. In this contribution we study the magnetic properties of 3D-DJJAs, regarded as multilevel granular systems for which a plaquette-structure response gives place to critical-state-likebehavior as the excitation increases above the threshold represented by the critical current of the Josephson coupling among grains.

Keywords: granularity, Josephson junction arrays, Wohlleben effect, critical current

## **1. Introduction**

The magnetic response of Josephson junction arrays (JJAs) to DC- and low-frequency AC-fields, have been studied recently by different techniques<sup>[1-5]</sup>. Some of the typical magnetic features of arrays include a Fraunhofer-like dependence of the critical current density with the applied magnetic field, J(H), a remanent magnetization within a limited temperature interval, M<sub>t</sub> (T), and the Wohlleben effect (WE, also known as Paramagnetic Meissner Effect). These aspects have been systematically studied by our group in recent years, using especially prepared samples of granular high- (HTS) and low-temperature (LTS) superconductors. These samples constitute tridimensional disordered Josephson junction arrays (3D-DJJAs) whose magnetic response exhibit all relevant features of arrays. The experimental approach adopted, including the strict control of sample granularity, allowed us to verify that all these notable the magnetic properties of JJAs are, in fact, manifestations of granularity.

The magnetized state of a JJA was first reported by Araujo-Moreira and co-workers <sup>[1,2]</sup>, based on simulations of the magnetic response of a single plaquette consisting of four resistively and capacitively shunted JJs. This state occurs in a window of temperatures, whose extent depends on the critical current density of the junctions. Also, there is a threshold value for the magnetic field in order to drive the JJA to the state where flux is retained after suppression of the field<sup>[1,2]</sup>. Studying our 3D-DJAs we have shown that the magnetized state actually occurs in a limited temperature interval, as predicted. Besides, the profile of the remanent magnetization,  $M_r$ , is sensitive to the critical current dispersion, so that this magnetic effect can be used to determine the critical current distribution of the array,  $N(J_c)^{[6]}$ .

Alternatively, 3D-DJJAs can be envisaged as especially assembled specimens of granular superconductors, so that their transport properties can be studied by the commonly employed approaches based on critical state models<sup>(7-9)</sup>. We have conducted a systematic study of the isothermal susceptibility response to the excitation field amplitude,  $\chi_{AC}(h)$ , of 3D-DJJAs fabricated from granular Nb and YBCO. The set of experiments included situations with a DC magnetic field, H, applied parallel to the excitation field. Critical state models<sup>(10,11)</sup> were employed to determine the average J<sub>c</sub> and its typical distribution width as a function of

temperature. To properly consider the significance of this result, obtained using a contactless configuration, one should bear in mind that performing conventional current-voltage measurements in the disordered arrays studied would be infeasible.

## 2. Sample Preparation

Niobium arrays are prepared from powder separated according to grain size, using a set of special sieves, with mesh gauges ranging from 38 to 44 **mm**. The powder is then uniaxially pressed in a mold to form a cylindrical pellet of 2.5 mm radius by 2.0 mm height. This pellet is a tridimensional disordered JJA in which the junctions are weakly-coupled grains, i.e., weak-links formed by a sandwich between Nb grains and a Nb-oxide layer originally present on the grain surface. As a consequence of the uniaxial pressure, samples produced in this way are anisotropic.

Tridimensional arrays of YBCO were prepared from granular material obtained using a modified method of polymeric precursors<sup>[12]</sup>. This route consists of mixing oxides and carbonates in stoichiometric amounts dissolved in HNO<sub>3</sub>, and then to an aqueous citric acid solution. A metallic citrate solution is then formed, to which ethylene glycol is added, resulting in a blue solution which was neutralized to pH~7 with ethylenediamine. This solution was turned into a gel and subsequently decomposed to a solid by heating at 400 °C. The sample was heat-treated at 850 °C for 12 h in air with several intermediary grindings, in order to prevent undesirable phase formations. Then, it was pressed into a pellet using controlled uniaxial (5,000 kgf/cm<sup>2</sup>) pressure and sintered at 950 °C for 6 h in O<sub>2</sub>. These pellets are 3D-DJJA, in which the junctions are weakly coupled grains, i.e., weak-links (WLs) formed by a sandwich of superconducting grains and intergrain material. As a consequence of the uniaxial pressure, samples produced in this way are anisotropic, a feature that can be either enhanced, by using higher pressures, or reduced, by applying isostatic pressures. Also, thermal treatment plays a fundamental role on creation and control of WLs and anisotropy<sup>[13]</sup>.

### **3. Experimental Results**

All measurements were performed using a Quantum Design MPMS-5T SQUID magnetometer. Both LTS and HTS samples exhibit the characteristic features of a genuine 3D-JJA as can be seen in Figure 1 and Figure 2.

The main picture of Figure 1 shows a low-field measurement of the reentrant magnetization (WE) for the Nb array. A typical Fraunhofer pattern for the critical current is presented in the inset, determined from the real part of  $\chi_{AC}^{[1]}$ . Similar behavior can be observed for the HTS granular sample, as is shown in the inset of Figure 2. It is also noticeable from the main curve in Fig.2 that the temperature at which the array ceases to respond is smaller than the critical temperature of the grains, T\* < T<sub>c</sub>.

To study the remanent state of the arrays, we performed an experimental procedure that consists of the following steps:

*i. the sample is submitted to an AC field h, consisting of a train of sinusoidal pulses, after what h is kept null;* 

ii. with h = 0, the magnetic moment of the sample, m(T), is measured;

iii. the temperature T is set to the next value and the steps are cyclically repeated.

Figure 3 shows m(T) measurements for a 3D-JJA of Nb, taken with h = 0, 0.01 Oe and 1 Oe. For h = 0, the typical response of a regular superconductor is seen. As h is increased, the array becomes magnetically active, and one observes a remanence which, as predicted, occurs only if h is above a threshold value  $h_b$ . The fact that the magnetic activity vanishes for temperatures above T\* is clearly seen: separate curves at lower temperatures collapse above T\*. From the three curves in Figure 3 we also recognize that the magnetic response of the 3D-JJA becomes more intense as h is increased. A similar behavior is exhibited by the YBCO array.

Figure 4 presents the remanent magnetization of a 3D-JJA of YBCO, normalized to its peak value,  $M_r/M_r^{max}$ . The temperature window within which the remanence occurs is clearly distinguished. A cognate curve is obtained for the Nb arrays. However, a residual

magnetic activity is also present at lower temperatures, which we ascribe to the fact that disordered arrays have a reasonably broad distribution of critical currents. To quantify the average value  $\langle J_c \rangle$  of the critical current density distribution, N(J<sub>c</sub>), and its typical dispersion, p, we measured the isothermal susceptibility  $\chi_{AC}(h)$ , of our 3D-DJJAs. Experiments including a DC magnetic field will be presented elsewhere <sup>[4]</sup>. We focus here on the results for H = 0, for which the exponential critical state model <sup>[7-9]</sup> can be employed to determine the average  $\langle J_c \rangle$  and p as a function of T.

Isothermal measurements of  $\chi_{AC}(h)$  for an excitation frequency of 100 Hz were carried out using the AC-module of a Quantum Design SQUID magnetometer. Figure 5 shows the real ( $\chi'$ ) and imaginary ( $\chi''$ ) parts of  $\chi_{AC}(h)$  for a Nb 3D-DJJa ( $T_c= 8.9$  K,  $T^* = 8.0$  K) at some values of T close to T\*. The field at which  $\chi''$  peaks,  $h_{\rm b}$ , is an indirect measure of the average critical current density of the intergranular matrix  $^{[5,6]}$ , i.e.,  $\langle J_c \rangle$  of the array. For a sample of cylindrical shape of radius a,  $h_p = a < J_c >$ . The exponential critical state model (ECSM)<sup>[7-9]</sup> was used to simultaneously fit  $\chi'$  and  $\chi''$ , from which the temperature dependence of  $\langle J_c \rangle$ , its typical distribution width, p(T), and the granular fraction of the sample,  $f_g(T)$ , are determined. It is worth mentioning that, as expected, the ECSM fits well the experimental data above h, but fails to adjust the curve below it. It is reasonable to expect that, for such low fields, the total flux imposed to the array is so small that its value per plaquette is much below the flux quantum  $\phi_0$ . It is thus conceivable that, in this conduction, flux penetrating the array could not be described by a critical state model. Consistently, up to  $h = h_{p}$  the array gives a positive (WE) contribution to the real part of  $\chi_{AC}$ , so that  $\chi' > -1$  and the sample is not perfectly diamagnetic. On the other hand, the dispersive activity of the vortices differs from that of an ordinary granular sample, being either lower, when the flux lines are pinned, or higher, when they are temporarily free to relocate, depending on the value of h.

The main curve in Figure 6 shows the average  $J_c(T)$ , whereas the insets depict p(T) (lower left) and  $f_g(T)$  (upper right). The line connecting the experimental points for the average  $J_c(T)$  is a fit of the form  $J_c(T) = J_{co}(1-T/T_c)^{2.38}$ , as introduced by Wright and coworkers for a matrix formed by grains linked by Josephson couplings <sup>[15,16]</sup>. Here,  $T_c$  is the critical temperature of the array, which was obtained from the fitting as been 8.05 K, in excellent agreement with the value of T\* determined by independent means in Ref. [3] for the

same sample. Not surprisingly, the numbers obtained for  $\langle J_c \rangle$  of the 3D-DJJA are comparable to those reported previously for the intergranular critical current of a melt-textured YBa<sub>2</sub>Cu<sub>3</sub>O<sub>7-δ</sub> sample<sup>[9]</sup>, an ordered 2D-JJA of Nb-AlO<sub>x</sub>-Nb<sup>[1]</sup> and a 3D-DJJA of YBCO<sup>[6]</sup>, among others. As could be anticipated, the critical current distribution of the array broadens as T approaches T<sub>c</sub>, as can be inferred by the continuous decrease of its typical dispersion,  $p(T)^{[8]}$ . A corresponding decrease on the granular fraction, measured by the volume fraction of superconducting grains to the normal matrix, f<sub>g</sub>, occurs as the superconducting properties degrade with increasing T, weakening at the grain boundaries and, from there, towards the center of the grains.

### 4. Conclusions

In conclusion, we have measured the predicted magnetic remanence of JJAs in 3D-DJJAs fabricated from granular LTS and HTS. The remanence is intense within a limited interval of temperatures. The m(T) profile, which is sensitive to the critical current dispersion, reveals a reasonably broad  $N(J_c)$  for the arrays. The exponential critical state model fits quite well the non-JJA regular behavior of the samples above the intergranular penetration field  $h_p$  but, predictably, fails to match the experimental data below  $h_p$ , since the array is magnetically active and responds in its own manner, which is admittedly non-critical-state.

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## FIGURE CAPTIONS

- Figure 1 Field-cooled magnetization curve of a Nb 3D-DJJA showing WE (reentrance). The inset shows the Fraunhofer pattern of  $J_c$ , as measured by the real part ( $\chi'$ ) of the AC susceptibility.
- Figure 2 Positive magnetization (WE) of a 3D-DJJA of YBCO, measured on cooling with H = 30 mOe. Inset shows the Fraunhofer pattern of  $\chi'$  for the sample
- Figure 3 Magnetic remanence exhibited by a 3D-DJJA of Nb.
- Figure 4 Magnetic remanence exhibited by a 3D-DJJA of YBCO.
- Figure 5 Real ( $\chi'$ ) and imaginary ( $\chi''$ ) parts of  $\chi_{AC}(h)$  for three values of T. The field at which  $\chi''$  peaks is an indirect measure of the average critical current density of the array.
- Figure 6 Main curve: average critical current of the 3D-DJJA; lower left: dispersion of critical current distribution, p(T); upper right: granular fraction,  $f_g(T)$ . Line connecting  $J_c(T)$  points is a fit of the expression  $J_c(T) = J_{co} (1-T/T_c)^{2.38}$ . Lines on insets are only guides to the eye

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