

Phase transitions in the weak links system of granular $YBa_2Cu_3O_{7-\delta}$ HTSC

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Received July 2, 2009

The change dynamics of current-voltage characteristics (IVCs) in granular samples of $YBa_2Cu_3O_{7-\delta}$ ($\delta \approx 0.05$) high-temperature superconductor in a perpendicular ($\mathbf{I}\perp\mathbf{H}_{ext}$) magnetic field within the $0 \geq H_{ext} > H_{c2J}$ range at $T = 77.4$ K has been studied. The field dependences of the exponential equation for granular HTSC IVC, $E = A(j - j_c)^n$, where j_c is the superconducting current critical density, show a set of features caused by phase transitions occurring in the Josephson weak links system.

Изучена динамика изменения вольтамперных характеристик (ВАХ) гранулярных образцов высокотемпературного сверхпроводника $YBa_2Cu_3O_{7-\delta}$ ($\delta \approx 0.05$) в перпендикулярном ($\mathbf{I}\perp\mathbf{H}_{ext}$) магнитном поле в диапазоне $0 \geq H_{ext} > H_{c2J}$ при $T = 77.4$ К. Установлено, что полевые зависимости параметров степенного уравнения для ВАХ гранулярных ВТСП $E = A(j - j_c)^n$, где j_c — критическая плотность сверхпроводящего тока, обнаруживают ряд особенностей, обусловленных протеканием фазовых переходов в системе джозефсоновских слабых связей.

It is well known that the electromagnetic properties of granular (ceramic) oxide high-temperature superconductors (HTSC) can be described qualitatively within the frame of two-level model for the critical state [1, 2] where the granular superconductor is considered as a system consisting of two II kind superconductors, namely, grains with a high superconductivity and the natural grain boundaries, being the Josephson weak links. In contract to the "classical" critical state models for a macroscopically homogeneous II kind superconductor [3, 4] where the critical current density j_c is controlled by the density gradient of Abrikosov magnetic vortices, two critical current densities are considered for the granular superconductors: a high current density in the superconducting grains ($j_{cA} \approx 10^5 - 10^7$ A/cm^2) that depends on the density gradient of Abrikosov vortices and a low current density ($j_{cJ} \leq 10^3$ A/cm^2) depending on the density gradient of the Josephson vortices in the weal links, i.e. in the grain boundaries [5]. The transport properties of granular HTSC, first of all, the critical current density, are controlled completely by the behavior of weak links, *S-N-S* Josephson contacts (*S* being the superconductor, *N*, the material in non-superconducting (normal) state [6–8]).

In the two-level model of the granular HTSC critical state, the transition of each *S-N-S* links to the resistive state is controlled by three parameters [6]: temperature T, the local density j_L of the current running through the *S-N-S* contact and the local magnetic field H_L. The j_L and H_L values seem to depend on the weak links orientation with respect to the electric field $\mathbf{E}$ and the external magnetic field $\mathbf{H}_{ext}$. However, the studies of magnetic resistance orientational dependence in granular $YBa_2Cu_3O_{7-\delta}$ HTSC at $T = 77.4$ K in weak magnetic fields [9] evidence the absense of any appreciable dependence of the weak links critical field H_{c2J} on the mutual orientation of **E** and $\mathbf{H}_{ext}$ vectors: at a constant transport current density j = const, taking into account the demagnetizing factor, the effective critical field value H_{c2J}^{eff} = const. This means that, in spite of variety of weak links types in grained $YBa_2Cu_3O_{7-\delta}$ HTSC postulated in some works (see, e.g., [10–12]), only one links type forms extended Josephson contacts that influence determinatively the electric current flow in a granular superconducting medium.

It is natural that, according to the existing concepts of the II kind superconductors [13], the H-T phase diagrams of both "subsystems" in a two-level system should include the existence regions of the Meissner phase, mixed state (Shubnikov phase), and normal phase. However, the lower and upper critical magnetic fields for superconducting grains [$H_{c1A}(T)$ and $H_{c2A}(T)$] and weak links [$H_{c1J}(T)$ and $H_{c2J}(T)$] that separate the mentioned regions differ significantly: in most HTSCs, the fields corresponding to the starting penetration of Abrikosov vortices into the superconducting grains, H_{c1A}, attains several hundreds Oe, the H_{c2A} fields amounting several hundreds kOe, while the fields providing the starting penetration of Josephson vortices into the weak links, H_{c1J}, are estimated to be in the range of 10^{-3} to 10^2 Oe (see, e.g., [5–7]), and the fields of the complete Josephson vortice penetration, H_{c2J}, amount several tens Oe (see, e.g., [14–21]).

Although the charge transfer processes in granular HTSC are controlled completely by the behavior of intergrain Josephson contacts, the study of the specific kinetic properties, in particular, of electric conductivity, at phase transitions in the weak links system meets some difficulties. In fact, when the H_{c2J} field values can be determined easily using the resistivity onset, $\rho \neq 0$, or the critical current zeroing, $j_c = 0$ (see, e.g., [7, 14, 16–19]), there are no similar simple criteria determining the H_{c1J} field values: the Josephson vortices start to penetrate into weak links of a granular superconductor at $H_{ext} = H_{c1J}$ occurs at $\rho = 0$, $j_c \neq 0$.

This fact means that at $H_{c1J} > H_{ext} > H_{c2J}$, the HTSC current-voltage characteristic (IVCs) must not differ qualitatively in shape from that measured at $H_{ext} < H_{c1J}$. However, quantitative differences is possible between the IVCs obtained in the $H_{ext} < H_{c1J}$ and $H_{c1J} > H_{ext} > H_{c2J}$ ranges. In fact, in relatively weak magnetic fields, the IVCs of a granular HTSC with randomly distributed critical currents is described satisfactorily by a power function (see, e.g., [11, 22–25])

$$E = A(j - j_c)^n. \quad (1)$$

Within the range of mixed weak links state, $H_{c1J} \le H_{ext} \le H_{c2J}$, the A value in the frame of percolational model of granular HTSC (see, e.g., [26–29]) is in proportion to the number of weak links where the phase coherence is destroyed, i.e. those passed to the resistive state [11, 30]), and characterizes the electrical resistance of intergrain boundaries. At $H_{ext} > H_{c2J}$, the parameter A should obviously be correlated directly with the averaged resistance of those boundaries.

The magnetic field dependences of A, j_c and n (the IVC nonlinearity extent) are to be supposed to reveal singularities at phase transitions in the weak links system of a grained HTSC. The character of those singularities was not studied systematically before, as far as we know. A behavior study of $A(H_{ext})$, $j_c(H_{ext})$ and $n(H_{ext})$ dependences in the vicinity of critical fields H_{c1J} and H_{c2J} seems to make it possible to judge of the nature of phase transitions connected with the Josephson vortice penetration onset into the weak links at $H_{ext} = H_{c1J}$ and with the superconductance deterioration in the weak links subsystem resulting from the complete Josephson vortice penetration at $H_{ext} = H_{c2J}$.

In this connection, the purpose of this work is to study the variation dynamics of current-voltage characteristics for granular $YBa_2Cu_3O_{7-\delta}$ HTSC within a wide range of the external magnetic field strength $\mathbf{H}_{ext}$: from $H_{ext} = 0$ to $H_{ext} > H_{c2J}$ at the liquid nitrogen boiling point (77.4 K).

Studied were the $YBa_2Cu_3O_{6.95}$ HTSC samples synthesized using the "standard" ceramic technique (see, e.g., [31]). The sample size was about 3×2×20 mm^3. The current and potential contacts were applied using a silver-based conductive adhesive. To attest the samples, XRD, critical temperature T_c and critical current I_c measurements. All the samples understudy were essentially single-phase ones. The superconducting transition middle temperature $T_c^{1/2}$ was 92.5 ± 0.1 K, the transition width $\Delta T_c \approx 0.4$ K.

A Dewar flask with liquid nitrogen containing the sample holder was placed into a copper wire solenoid coil. All the IVCs of the

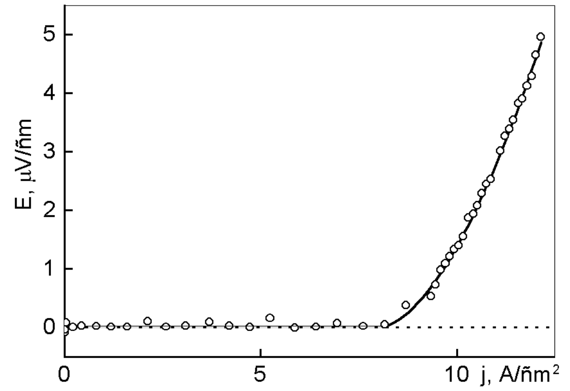

Fig. 1. A representative E–j characteristic of ceramic $YBa_2Cu_3O_{6.95}$ HTSC sample at T = 77.4 K, H_{ext} = 7.96 Oe. The parameters of Eq.(1): $A = 0.42029 \pm 0.01292$ a.u., j_c = 8.0 ± 0.1 A/cm^2, $n = 1.72294 \pm 0.02464$. The correlation factor $r^2 = 0.99746$.

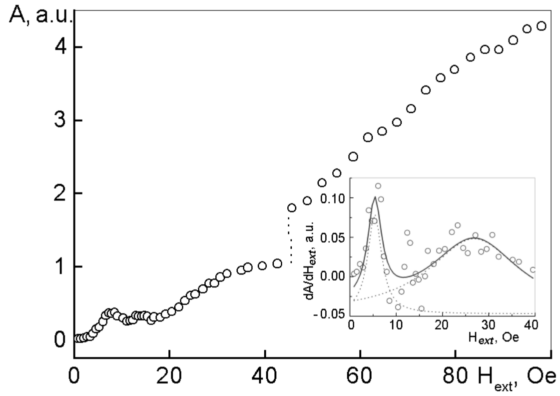

Fig. 2. Field dependence of the parameter A in Eq.(1) for a ceramic $YBa_2Cu_3O_{6.95}$ HTSC sample at T = 77.4 K. Inset: the field dependence of $dA/dH_{ext}(H_{ext})$ derivative.

$YBa_2Cu_3O_{6.95}$ HTSC samples at T = 77.4 K were obtained in the perpendicular ($\mathbf{I} \perp \mathbf{H}_{ext}$) magnetic field at $0 \le H_{ext} \le \approx 100$ Oe.

To obtain the IVCs at $H_{ext} = const$, a special setup [32] was used consisting of a control unit for the magnetic field source (solenoid) current I_{sol} and that for the measuring (transport) current I_{meas} flowing through the HTSC sample. All the measurements were done in automated mode: at a constant value of the solenoid current I_{sol} (corresponding to the pre-specified H_{ext} value), the transport current $I = I_{meas}$ was increased smoothly till the voltage $U \approx 5$ μV on the HTSC sample was attained. Then the data set $E(j)$ at $H_{ext} = const$ was recorded in a PC memory, the measuring current I_{meas} was zeroed, the next I_{sol} value was pre-specified and the measuring cycle was repeated.

The j–E characteristics were processed as follows. Using the experimental IVC for $E = 0$, approximate critical current density j_c values were found, the A and n parameter values for were calculated using the power law (1), then the A, j_c and n values were improved using the functional minimization technique till the correlation coefficient $r^2 \ge 0.99$ was attained. As an example, Fig. 1 presents the IVC for a sample obtained at T = 77.4 K, H_{ext} = 7.96 Oe, and the figure caption gives its processing results.

In Figs. 2 through 4, presented are the magnetic field dependences of the power equation parameters for the IVCs of the $YBa_2Cu_3O_{6.95}$ granular samples studied. All the $A(H_{ext})$, $j_c(H_{ext})$ and $n(H_{ext})$ dependences are clearly non-monotonous.

In the $A(H_{ext})$ dependence (Fig. 2), the following characteristic features are observed against the background of the trend to a considerable increase of the intergrain boundaries (parameter A in (1)) as the external magnetic field strength rises. First, a pronounced maximum appears at $H_{ext} \approx 8$ Oe; second, the $A(H_{ext})$ dependence changes its shape appreciably at $H_{ext} \approx 30$ Oe; moreover, the parameter A shows a "jump" at $H_{ext} \approx 48$ Oe, while at $H_{ext} > \approx 48$ Oe the $A(H_{ext})$ dependence becomes essentially linear.

The field dependence of $dA/dH_{ext}(H_{ext})$ derivative (inset in Fig. 2) is worth to attention: a sharp peak is observed at $H_{ext} \approx 5$ Oe and a rather spread-out maximum at $H_{ext} \approx 30$ Oe.

As to the $j_c(H_{ext})$ dependence (Fig. 3), a slight knee is observed at $H_{ext} \approx 5$ Oe besides of the general trend to decreasing critical current density as the magnetic field rises. The knee point in the $j_c(H_{ext})$ dependence is confirmed clearly by a rather sharp minimum ("negative peak") in the field dependence of the $dj_c/dH_{ext}(H_{ext})$ derivative (inset in Fig. 3). At $H_{ext} > \approx 30$ Oe, the j_c becomes zeroed within the measurement accuracy.

The $n(H_{ext})$ dependence (Fig. 4) is rather complex. The "nonlinearity parameter" n in the HTSC IVC equation (1) decreases from very high values ($n \approx 3.5$) at $H_{ext} \to 0$ down to $n \approx 1$ (being typical of ohmic resistance of intergrain contacts) at $H_{ext} \ge \approx 30$ Oe. Two pronounced deep minima are observed

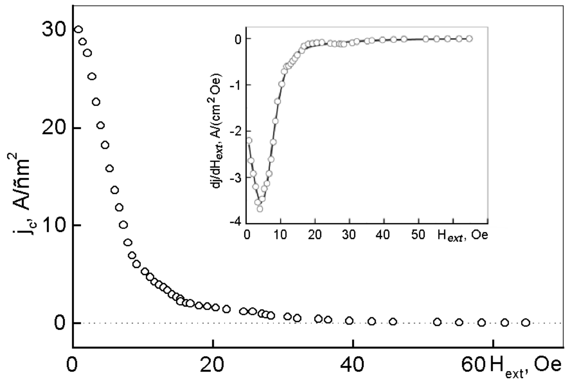

Fig. 3. Field dependence of the critical current density (parameter j_c in Eq.(1)) for a ceramic $YBa_2Cu_3O_{6.95}$ HTSC sample at T = 77.4 K. Inset: the field dependence of $dj_c/dH_{ext}(H_{ext})$ derivative.

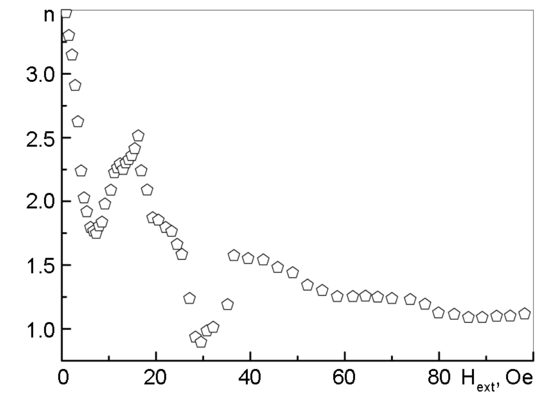

Fig. 4. Field dependence of the "nonlinearity parameter" n in Eq.(1) for a ceramic $YBa_2Cu_3O_{6.95}$ HTSC sample at T = 77.4 K.

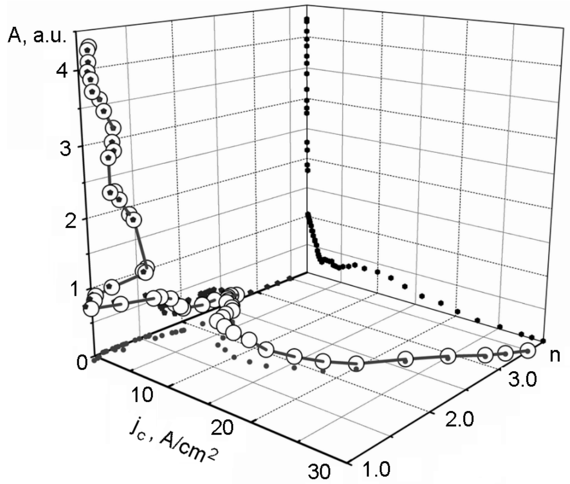

Fig. 5. IVC variation dynamics for $YBa_2Cu_3O_{6.95}$ in the "parameter space" of Eq.(1) A, j_c, and n, T = 77.4 K.

in the $n(H_{ext})$ curve (at $H_{ext} \approx 8$ and ≈ 30 Oe).

It follows from the above data that the field dependences of all three parameters (A, j_c, and n) of the power equation describing the IVC of grained $YBa_2Cu_3O_{6.95}$ HTSC show a series of singularities. Namely, (i) at $H_{ext} \approx 5$–8 Oe, maxima are observed in $A(H_{ext})$ and $dA/dH_{ext}(H_{ext})$ dependences while minima in $n(H_{ext})$ and dj_c/dH_{ext} ones; (ii) at $H_{ext} \approx 30$ Oe, j_c is zeroed, a maximum is observed in $dA/dH_{ext}(H_{ext})$ dependence and a minimum in $n(H_{ext})$ one; (iii) at $H_{ext} \approx 48$ Oe, the parameter A increases stepwise and at the further H_{ext} increase, the IVC takes the ohmic character ($n \to 1$).

The characteristic behavior features of the power equation (1) for the $YBa_2Cu_3O_{6.95}$ HTSC connected with strength variation of the external magnetic field $\mathbf{H}_{ext}$ and the interrelation thereof are reflected adequately in the IVC variation dynamics with the three-dimensional "space of parameters" A, j_c, and n (Fig. 5). The "maximum" at $H_{ext} \approx 8$ Oe, "minimum" ($H_{ext} \approx 30$ Oe) and "jump" ($H_{ext} \approx 50$ Oe) are clearly seen. The latter effect is most pronounced in the projection of the IVC parameter variation onto the j_c–A plane.

When discussing the results obtained, it is to note first of all that the field dependence of dA/dH_{ext} (inset in Fig. 2) shows two characteristic peaks (at $H_{ext} \approx 5$ Oe and $H_{ext} \approx 30$ Oe), similar in appearance to the λ-anomalies of heat capacity at the 2nd order phase transitions. Note that the parameter A describes de facto the electric resistance of intergrain boundaries. The behavior of the electric resistance with respect to temperature (and perhaps with respect to H_{ext}) is known to be similar (in the frame of fluctuation theory of phase transitions [33]) to the behavior of second derivatives of thermodynamic potentials, in particular, of heat capacity. Two peaks in the (H_{ext}) curve testifies considerably the the observed anomalies are connected with the 2nd order phase transitions with respect to magnetic field in the weak links system of the granular $YBa_2Cu_3O_{6.95}$ HTSC. This assumption is confirmed by the extremes in the $n(H_{ext})$ dependence (see Fig. 4) and the IVC "trajectory" change in the A–j_c–n space (see Fig. 5). The reasons for the critical current density zeroing at $H_{ext} \approx 30$ Oe (see Fig. 3) are obvious: as mentioned above, this means a

2^{nd} order $S-N$ phase transition in the weak links system at the external magnetic field strength H_{ext}, close to the critical field of complete Josephson vortice penetration H_{c2J}, at $j = 0$. The dependence of the field value H_{c2J} on the transport current density, $H_{c2J}(j)$, can be obtained directly from the $j_c(H_{ext})$ curve similar to that presented in Fig. 3 [19].

The rather strong spread-out of anomalies in $A(H_{ext})$, $dA/dH_{ext}(H_{ext})$ and $n(H_{ext})$ behavior with respect to the magnetic field seems to be connected with different orientations of the external magnetic field strength vactor $\mathbf{H}_{ext}$ with respect to intergrain boundaries (weak links). This means that the transition of all weak links into the resistive state occurs within a rather wide H_{ext} range and is defined by local density values j_L of the current flowing through the $S-N-S$ contact and local magnetic field values H_L, both parameters being dependent on the grain boundary orientation [6].

The fulfillment of $j_c = 0$ condition is to be assumed to point only on the $S-N$ transition in the system of weak links forming the continuous Josephson contacts, i.e. the breakdown of percolation ways for the superconducting transport current. While the "jump" in the $A(H_{ext})$ curve (see Fig. 2), i.e., in fact an electric conductance "jump" in the weak links system associated with the $S-N$ transition, as well as a pronounced trend to the ohmic IVC behavior ($n \to 1$, see Fig. 4) at $H_{ext} > H_{c2J}$ must evidence the complete transition of all the weak links into the resistive state. Thus, for the granular $YBa_2Cu_3O_{6.95}$ HTSC, the maximum upper critical field is $H_{c2j}^{max} \approx 48$ Oe.

To conclude, when studying the variation dynamics of IVCs for granular $YBa_2Cu_3O_{7-\delta}$ HTSC in magnetic fields of the $0 \geq H_{ext} > H_{c2J}$ range at $T = 77.4$ K, some novel magnetoresistive effects have been revealed caused by phase transitions in the Josephson weak links system. The IVCs of the studied samples are shown to be described adequately by the power function $E = A(j - j_c)^n$ within the whole magnetic field range (correlation factor $r^2 \geq 0.99$). Basing on this fact, the following regularities in the IVCs variation have been established that are connected with evolution of grain boundaries under action of magnetic field and transport current. At the penetration onset of the Josephson vortices into the weak links system in the field H_{c1J}, maxima appear in the $A(H_{ext})$ and $dA/dH_{ext}(H_{ext})$ dependences and minima in the $n(H_{ext})$ and $dj_c/dH_{ext}(H_{ext})$ ones. As the percolation paths of transport currents are broken down in the field H_{c2J}, j_c is zeroed, a maximum appears in the $dA/dH_{ext}(H_{ext})$ curve and a minimum in the $n(H_{ext})$ one. When all the weak links transit into resistive state in the H_{c2J}^{max} field, the parameter A increases stepwise, and the IVCs becomes ohmic at further H_{ext} increase.

Authors are thankful to V.V.Derevyanko who assisted in the experiments.

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Фазові переходи в системі слабких зв'язків гранулярних ВТНП $YBa_2Cu_3O_{7-\delta}$

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Вивчено динаміку зміни вольтамперних характеристик (ВАХ) гранулярних зразків високотемпературного надпровідника $YBa_2Cu_3O_{7-\delta}$ ($\delta \approx 0.05$) у перпендикулярному (**I** ⊥ **H**IVext) магнітному полі у широкому діапазоні значень напруженості зовнішнього магнітного поля: $0 \geq H_{ext} > H_{c2J}$ при $T = 77.4$ K. Встановлено, що польові залежності параметрів степеневого рівняння для ВАХ гранулярних ВТНП $E = A(j - j_c)^n$, де j_c — критична щільність надпровідного струму, виявляють ряд особливостей, обумовлених протіканням фазових переходів у системі джозефсонівських слабких зв'язків.