ISSN 1027-5495. Functional Materials, 22, No.3 (2015), p. 332-337.
doi:http://dx.doi.org/10.15407/fm22.03.332

Phase *H-T*-diagram for MgB_2 granular *BCS*-superconductor in weak magnetic fields

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Received March 24, 2015

For the purpose of plotting the *H-T* diagram for MgB_2 granular superconductors, the behavior of electrical resistivity-versus-temperature dependences were studied at ~35 to ~45 K in external magnetic fields with magnetic field strengths $\mathbf{H}_{ext}$ up to ~2 kOe. The effect of increasing the superconductivity transition width ΔT_c at increased H_{ext} has been revealed. A model has been developed for superconductivity transitions in a two-level system. The data obtained allowed establishing the existence of a weak links system in MgB_2 granular superconductors.
Keywords: MgB_2, HTSC, two-level system, H-T- phase diagram, critical fields, resistivity.

С целью построения фазовой *H-T*-диаграммы гранулярных сверхпроводников MgB_2 изучен характер температурных зависимостей электросопротивления при ~35–45 K во внешних магнитных полях $\mathbf{H}_{ext}$ напряженностью до ~2 kOe. Обнаружен эффект повышения ширины сверхпроводящего перехода ΔT_c при росте H_{ext}. Разработана модель сверхпроводящих переходов в двухуровневой системе. На основании обработки полученных данных установлено наличие системы weak links в гранулярных сверхпроводниках на основе MgB_2.

Фазова *H-T*-діаграма гранулярного *BCS*-надпровідника MgB_2 у слабких магнітних полях. *В.В.Дерев'янко, Т.В.Сухарева, В.О.Фінкель, Ю.М.Шахов.*
З метою побудови фазової *H-T*-діаграми гранулярних надпровідників MgB_2 вивчено характер температурних залежностей електроопору при ~35–45 K у зовнішніх магнітних полях $\mathbf{H}_{ext}$ напруженістю до ~2 kOe. Виявлено ефект підвищення ширини надпровідного переходу ΔT_c при рості H_{ext}. Розроблено модель надпровідних переходів у дворівневій системі. На підставі обробки отриманих даних установлено наявність системи "слабких зв'язків" у гранулярних надпровідниках на основі MgB_2.

1. Introduction

Superconductivity of magnesium diboride MgB_2 with critical temperature T_c ~ 39–40 K was discovered in 2001 [1]. From this time forward, physical properties of this superconductor are studied intensively in many laboratories of the world. The quantity of publications on this problem achieves tens thousands. Now it is established surely that the MgB_2 superconductivity is caused by the known from 1957 mechanism of electron-phonon interaction by Bardeen-Cooper-Schrieffer (further the *BCS*-theory) [2].

As it is known, in the framework of the *BCS*-model the superconductor critical temperature depends on three fundamental parameters: the Debye frequency ω_D ($\hbar\overline{\omega}_D = k\Theta_D$, where Θ_D is the Debye temperature, $\hbar$ is the Planck's constant, and k is the Boltzmann constant), electron-phonon interaction parameter V, and electron state density at the Fermi level $N(E_F)$:

$$kT_c = 1.13\hbar\overline{\omega}_D \cdot \exp\left[-\frac{1}{VN(E_F)}\right]. \quad (1)$$

Indeed, the high enough frequency ω_D of phonon oscillations provides attainment of the critical temperature T_c ~ 40 K.

In polycrystalline state, similarly to oxide high temperature superconductors (HTSC), formation of granular microstructure is characteristic for MgB_2. The behavior of the granular superconductors being a combination of two sub-systems: granules (grains) with strong superconductivity and intergranular boundaries — the Josephson's weak links with low superconductivity in magnetic field — can be described in the framework of a two-level model [3]. The correlations between critical parameters of the two-level system of the granular HTSC are given below.

For the superconducting granules (grains) [subscripts "g" (grain) or "A" (Abrikosov)] and the intergranular boundaries — Josephson weak links (subscript "J") we have:

$$T_c = T_{cg}(T_{cA}) = T_{cJ}, \quad (2)$$
$$H_{c1g}(T) > H_{c1J}(T),\ H_{c2g}(T) > H_{c2J}(T),$$
$$I_g(T,H) > I_J(T,H).$$

Here T_{cg} (T_{cA}) and T_{cJ} are critical temperatures; $H_{c1g}(H_{c1A})$ and H_{c1J} are critical fields for beginning the magnetic field penetration into subsystems of the granular superconductor; $H_{c2g}(H_{c2A})$ and H_{c2J} are critical fields for magnetic field total penetration into subsystems of the granular superconductor; I_g (I_A), I_J are critical currents.

It was assumed that the correlation like these were applicable to any granular HTSC. The validity of such assumption is not obvious.

The problem of temperature and external magnetic field influence on the evolution of vortex structure is not solved definitely. In spite of a lot of works devoted to attempts of plotting the *H-T*-phase diagrams for MgB_2 granular *BCS*-superconductors at low temperatures, yet the positions of H_{c1g} and H_{c2J} lines have not been defined at the *H-T*-phase diagram. Moreover, doubtful is the existence of weak links in the case of the MgB_2 granular *BCS*-superconductor.

In this connection, the aim of the present work is plotting the *H-T*-diagram for MgB_2 granular superconductors. The most effective way to attain the aim we consider precision measurements of electrical resistivity temperature dependences near T_c in relatively low magnetic fields. Obviously, appearance of the phase transition lines in the weak links subsystem is possible only in the range of weak external magnetic fields $\mathbf{H}_{ext}$. Note, that most of the works (see, for example, [4–12] were carried out at the high enough H_{ext} values.

2. Techniques

The subjects concerning synthesis of the MgB_2 *BCS*-superconductor and the technique of resistivity measurements at cryogenic temperatures under weak external magnetic fields were discussed earlier [13–17].

Experiments were reduced to measurements of electrical resistivity of the MgB_2 *BCS*-superconductor granular samples in the narrow enough range of low temperatures (from ~35 to~45 K) under external fields with strength to ~ 2 kOe. However, it should be emphasized that it is due to narrowness of the temperature range that the temperature measurements were to be carried out with precision not low than ~ 0.01–0.02 K. Only the curves $R(T)$ obtained with such precision allow an adequate treatment of the experimental data. Using the instruments applied in the work we could registered information with "step" ~0.01 K.

To organize the precise electro-physical measurements in the narrow temperature range the conditions of measurement process were strictly standardized. The following scheme of measurements was developed:

1. Measurements of electrical resistivity temperature dependence $\rho(T)$ for the MgB_2 granular *BCS*-superconductor sample under zero magnetic field in the temperature range from ~35 to ~45 K (at increasing temperature).
2. Cooling the sample in *FC*-regime to T ~ 35 K.
3. Measuring the electrical resistivity temperature dependence $\rho(T)_{Hext=const}$ in the temperature range from ~35 to ~45 K under given value of perpendicular magnetic field ($\mathbf{H}_{ext}\perp\mathbf{I}$, H_{ext} = const).
4. Dumping the magnetic field $\mathbf{H}_{ext}$.
5. Carrying out the control measurements of the electrical resistivity temperature dependence $\rho(T)$ at the zero magnetic field in the temperature range from ~35 to ~45 K in order to reveal possible residual effects in dependence $\rho(T)$ at H_{ext} = 0. The aim of these measurements is exclusion of

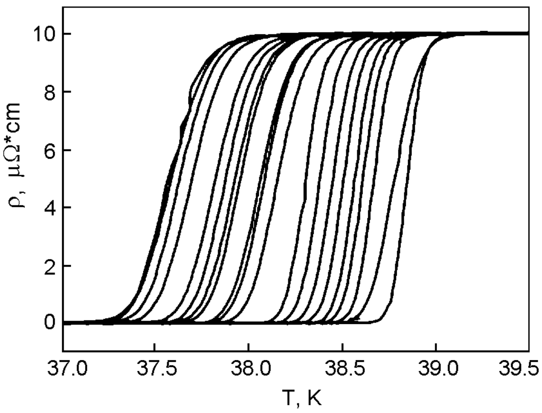

Fig. 1. Electrical resistivity temperature dependences $\rho(T)_{Hext=\ const}$ for MgB_2 granular samples in external magnetic fields H_{ext}. External field values are from right to left: 0, 100, 200, 400, 500, 600, 700, 800, 900, 1 000, 1100, 1200, 1300, 1400, 1500, 1600, 1700, 1800, 1900, and 1980 Oe.

possible errors caused by the instrumental failures.

6. Transfer to point 3 at the following value H_{ext} = const.

All the curves $\rho(T)$ have characteristic sigma-like shape (Fig. 1).

According to the scheme described, several series of $\rho(T)$ measurements were carried out for the MgB_2 granular *BCS*-superconductor samples. As a rule, each series included about 20 values of temperature in the range of H_{ext} "0 –1980 Oe".

3. Results

The main results $\rho(T)_{Hext=const}$ obtained for one of the measurement series are shown in Fig. 1.

First of all we note some qualitative effects:

— way of cooling the samples (*ZFC* or *FC*) does not influence on the behavior of the curves $\rho(T)_{H_{ext}\ =\ const}$ under increasing temperature in the range of measurement precision;

— for the MgB_2 granular *BCS*-superconductor samples no differences were observed in the $\rho(T)_{H_{ext}=const}$ curves before and after application of H_{ext} external field;

— no hysteresis effects were observed in the $\rho(T)_{H_{ext}\ =\ const}$ curves.

On the "background" of these qualitative results we consider all the quantitative results below.

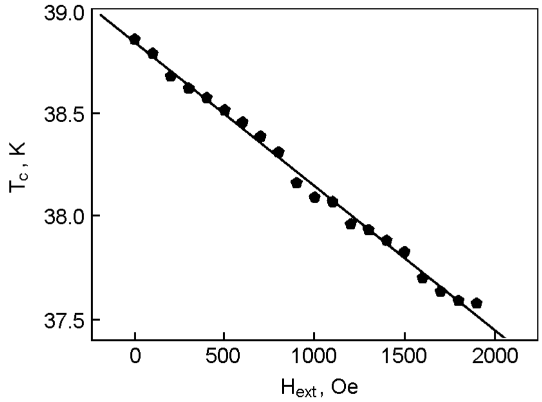

Fig. 2. Field dependence of the inflection point position in $\rho(T)_{H_{ext}=\ const}$ curves corresponding to critical temperature T_c for MgB_2 granular samples.

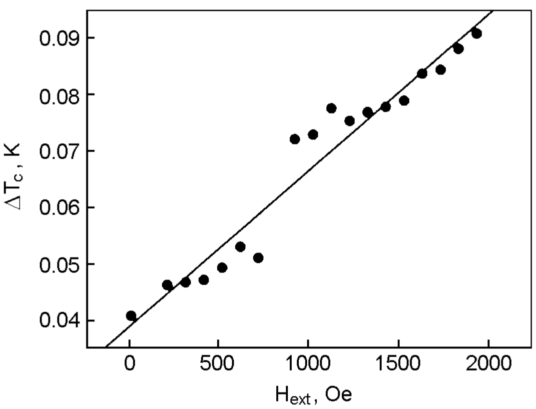

Fig. 3. Field dependence of the superconducting transition width ΔT_c for MgB_2 granular sample.

Obviously, the following parameters should be taken into consideration for analysis of the $\rho(T)_{H_{ext}\ =\ const}$ curves shown in Fig. 1:

— temperature T_c of superconducting transition;

— width of the transition ΔT_c;

— electrical resistivity jump $\Delta\rho$ at T_c.

As the external magnetic field strength $\mathbf{H}_{ext}$ varies, the following regularities are observed in the behavior of field dependences of abovementioned parameters:

1. At the $\rho(T)_{H'_{ext}\ =\ const}$ curves, the position of the inflexion point (practically coinciding with effective T_c) shifts linearly to the side of lower temperatures under increasing the field strength H_{ext} (Fig. 2).

2. The effective width of the transition ΔT_c increases with increasing the field H_{ext} (Fig. 3).

3. The electrical resistivity jump $\Delta\rho$ values at T_c do not depend on the applied external magnetic field H_{ext} (see Fig. 1).

As it is seen from Fig. 2, there is practically linear decrease of the critical temperature with increasing the magnetic field strength.

Principally important is the fact that in the range of weak magnetic fields, the effective width ΔT_c of superconducting transition increases practically linearly under increasing H_{ext}: $\Delta T_c = 38.84017 \pm 0.01259 + (-0.0007 \pm 0.00001)^* H_{ext}$; the correlation coefficient being $r^2 > 0.99$.

4. Discussion

It is worth to be noted that all the $\rho(T)_{H_{ext}\,=\,const}$ dependences near T_c have characteristic "sigma"-like shape. Such behavior of thermodynamic and kinetic values is described adequately by the known Boltzmann function which is applied widely in the theory of phase transitions followed by jumps of the corresponding physical values [18–21].

$$\rho(T)_{H_{ext}=const} = \frac{\Delta\rho_N}{1 + e^{T_c - \Delta T_c}}. \quad (3)$$

To the first approximation, the Boltzmann function can be used for describing the behavior of all dependences $\rho(T)_{H_{ext}=const}$ for the MgB_2 granular superconductors in the T_c vicinity (Fig. 1).

The fact that all curves $\rho(T)_{H_{ext}\,=\,const}$ for MgB_2 are described adequately by equation (3) (correlation coefficient $r^2 > 0.99$) does not give enough reasons to assume the MgB_2 granular *BCS*-superconductor being an single-level system that is principally differs from two-level systems characteristic for the granular oxide HTSC. The question on belonging the MgB_2 granular *BCS*-superconductor to two-level systems remains open.

To obtain an argued enough answer to this question, it is necessary to pay attention at existence of two abovementioned peculiarities in the field dependences of $\rho(T)_{H_{ext}\,=\,const}$ curve parameters:

1. Width of the superconducting transition ΔT_c shows the evidently expressed field dependence (ΔT_c increases as the field strength rises).

2. Position of the upper boundary of the transition curves does not depend on the external $\mathbf{H}_{ext}$ magnetic field strength and, as a consequence, the value of electrical resistivity jump $\Delta\rho$ at T_c does not depend on H_{ext}.

4.1. Model of the superconducting transition (or transitions) in the two-level system of MgB2 granular BCS-superconductor

There are some grounds to assume that if the value of electrical resistivity jumps may be caused by existence of weak dependence of electrical resistivity on H_{ext} in normal state, the effect of substantial increasing the transition width with relatively small H_{ext} increase requires special consideration.

Indeed, in the phase *H-T*-diagram of the two-level granular superconductor there are four lines of the phase equilibrium: $H_{c1A}(T)$, $H_{c2A}(T)$, $H_{c1J}(T)$, $H_{c2J}(T)$ that coincide in the same point under the zero magnetic field according to the existing notions [3].

Obviously, that penetration of the Abrikosov's vortexes into the superconducting granules as well as the Josephson's vortexes into the weak links system begins under fields H_{c1}. Under the fields H_{c2}, destruction of superconductivity for both subsystems of the two-level superconductor takes place. Critical fields of the superconducting granules are substantially higher than the weak links critical fields [3].

It can be assumed that optimal way to search an answer to the question about possibility of existing the several lines of phase transitions in the *H-T*-diagram of the MgB_2 granular *BCS*-superconductor and also about the critical field values of both the superconducting granules and weak links is fitting the experimental data in the assumption that each $\rho(T)_{H_{ext}\,=const}$ curve being a superposition of two sigma-like curves described by the Boltzmann function:

$$\rho(T)_{H_{ext}=const} = \quad (4)$$
$$= \frac{\Delta\rho_{NA}}{1 + e^{T_{cA} - \Delta T_{cA}}} + \frac{\Delta\rho_{NJ}}{1 + e^{T_{cJ} - \Delta T_{cJ}}}.$$

Obviously, for description of the superconducting transition "smeared" over magnetic field, the model described by equation (4) can be applied. In the framework of this model, the MgB_2 granular *BCS*-superconductor is considered as a "classical" two-level system.

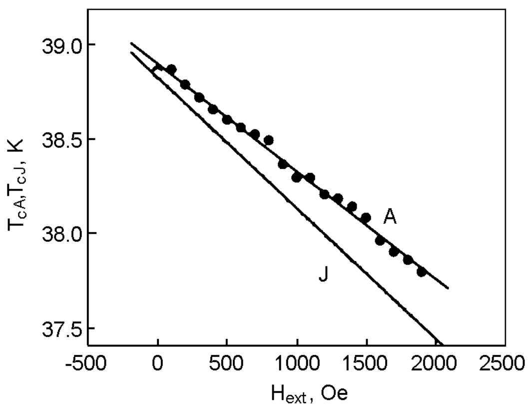

Fig. 4. Phase *H-T*-diagrams for MgB_2 granular *BCS*-superconductor. Symbols "A" and "J" correspond to superconducting granules and weak links, respectively.

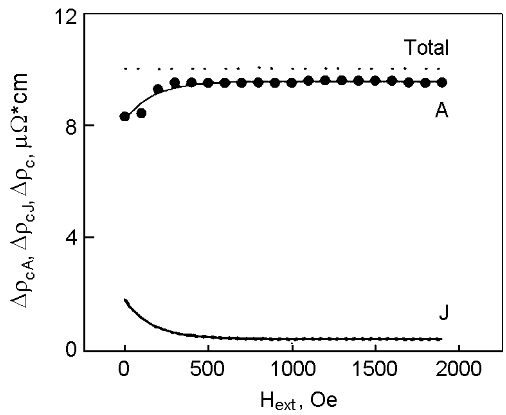

Fig. 5. Field dependences of electrical resistivity jumps at superconducting transition for MgB_2 granular superconductor. Symbols "A" and "J" correspond to superconducting granules and weak links, respectively. *Total* — the total value of the electrical resistivity jumps.

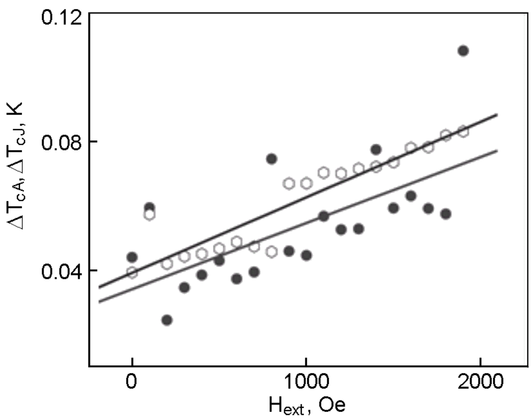

Fig. 6. Field dependences of width of superconducting transition in MgB_2 granular superconductor. Symbols "A" and "J" correspond to superconducting granules and weak links, respectively.

4.2. Treatment of experimental results

The results of mathematical treatment of the data obtained, namely, the curves $\rho(T)_{H_{ext} = const}$, (see Fig. 1–3) in the framework of the two-level model using the method of functional minimization are shown in Fig. 4–6.

The following regularities are observed in the behavior of Eq.(2) parameters depending on fields:

1. Superconducting transition critical temperatures for the granules and weak links are practically similar for the zero magnetic fields; as the H_{ext} increases, the difference between T_{cA} and T_{cJ} noticeably increases.

2. Electrical resistivity jumps for the superconducting granules at T_{cA} exceed significantly the jumps at T_{cJ}.

3. Differences between the widths of superconducting transitions ΔT_{cA} and ΔT_{cJ} do not exceed the limits of possible measurement errors.

Let us comment the results shown in Figs. 4–6.

1. Treatment of the results on the base of notions about description of the transition curves as a convolution of two Boltzman functions for A- and J-subsystems of the two-level system undoubtedly justifies the assumption on existing the two phase transitions with close values of the critical temperatures and fields.

The following dependences of linear equation parameters for the field dependences of critical temperatures:

$$T_{cA} = 38.90184 \pm 0.01183 -$$
$$- (0.00057 + 0.00001)^* H_{ext}(r^2 = 0.99377);$$
$$T_{cJ} = 38.8292 \pm 0.01402 -$$
$$- (0.00069 \pm 0.00001)^* H_{ext}(r^2 = 0.99408)$$

— indicate undoubtedly the presence of real signs of existing the lines $T_{cA}(H_{ext})$ and $T_{cJ}(H_{ext})$ in the *H-T*-diagram for the MgB_2 granular *BCS*-superconductor (see Fig. 4).

It should be admitted that resistivity measurements of the MgB_2 granular *BCS*-superconductor samples under the weak fields resulted in plotting a simplified variant of the H-T-diagram, we could not establish the positions of $H_{c1}(T)$ lines. However, this does not exclude existence of the weak links in the MgB_2 granular BCS-superconductors.

2. The behavior of field dependences of the Abrikosov's and Josephson's electrical resistivity jumps (see Fig. 5) should obviously consider reasonable because of the observed tendency to closing their values with approaching to T_c.

3. Comparison of the experimental (Fig. 3) and calculated (Fig. 6) field dependences of the superconducting transition widths indicates similarity of both their character (increase with field increasing) and absolute values. Surely, the behavior of the electrical resistivity jumps and superconducting transition widths requires further investigations under the stronger magnetic fields.

5. Conclusions

Fundamentally important result of the present work is establishment of the possibility to apply the critical state two-level model developed for describing the electromagnetic properties of oxide HTSC to MgB_2 granular BCS-superconductors.

Obtaining the main result of the work was provided by the following:

— precise measuring the electrical resistivity near T_c in the weak magnetic fields;

— revealing the effect of broadening the superconducting transition under increasing the strength of magnetic field;

— developing the model of superconducting transitions in the two-level system;

— plotting the phase H-T-diagram for the MgB_2 granular BCS-superconductor.

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