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ANOMALOUS MICROWAVE ABSORPTION IN Bi-2212 HIGH TEMPERATURE SUPERCONDUCTORS WITH THE PARAMAGNETIC MEISSNER EFFECT

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The results are reported on the study of an anomaly in low magnetic field microwave absorption in polycrystalline Bi-high temperature superconductors which show the so called Paramagnetic Meissner Effect. The data obtained witness for common origin of both effects which may occur due to the presence in the samples of weak links with inverse Josephson coupling (π-contacts).

1. In our recent experiments [1] we observed that in certain polycrystalline samples of Bi-2212 and Bi-2223 metal oxides the magnetic field cooled (FC) susceptibility in the superconducting (SC) state becomes paramagnetic in magnetic fields less than 1 *Oe* (the Paramagnetic Meissner Effect or the Wohlleben Effect (WE)[1]). No significant difference between samples with and without the WE was found in their stoichiometry, structure, microstructure, ac susceptibility, resistivity and specific heat. At the same time we found out that the samples showing the WE often exhibit a pronounced anomaly in their differential low magnetic field microwave absorption (MWA) [1]. The correlation which we noticed between the presence of the WE and appearance of the MWA anomaly gives us ground to believe that they both have common reason, and therefore the study of characteristics of the microwave absorption in such samples can shed more light on the nature of both phenomena.

In this paper we present the results of the study of the differential low magnetic field microwave absorption in those samples of Bi-2212 high temperature superconductor (HTS) which show the Wohlleben Effect. On the basis of the data obtained we discuss the properties of unusual Josephson junctions whose presence in the samples may cause the observed effects.

2. We studied the polycrystalline samples of $Bi_2Sr_2CaCu_2O_y$ compound prepared by a solid state reaction with subsequent annealing close to the melting point [3,4] (samples KN99F and KN100F) and by a melt cast process [4,5] (sample KAMU96). The measurements of *dc*-susceptibility at the field range from several Oe down to 10 mOe were carried out with a specially designed SQUID magnetometer previously described in [1]. The differential microwave absorption was measured

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[1] Since actually the observed phenomenon is not strictly related to the Meissner effect we would like to follow the suggestion by Sigrist and Rice [2] to call this from now as the Wohlleben Effect.

using a conventional X-band Bruker EPR spectrometer (for details of the technique see, e.g. [6]). The incident microwave power had a level 10-200mW, the amplitude of the ac modulating field (100kHz) was varied form 0.04 to 2Oe, and the dc magnetic field was swept from -20Oe to +20Oe.

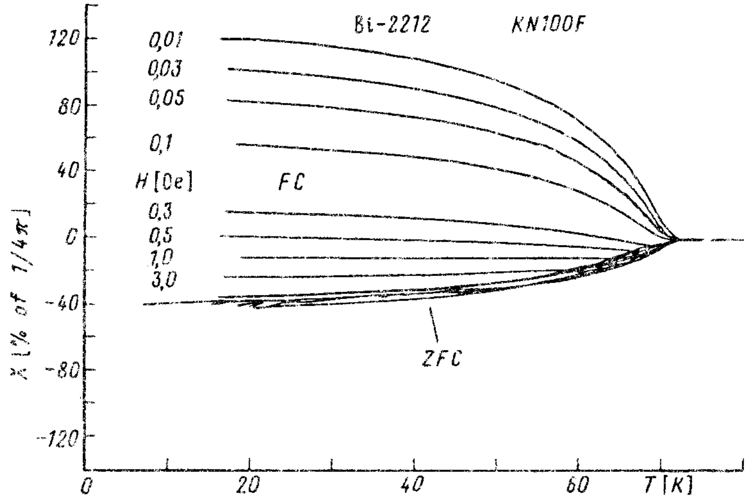

Fig.1. Temperature dependence of the dc susceptibility in the SC state for the sample KN100F.

Fig.1 shows the results of the measurements of the dc susceptibility χ for the sample KN100F with the onset of the resistivity SC transition $T_c^{onset} = 76$K. The zero field cooled (ZFC) susceptibility is diamagnetic and is almost field independent. At the same time the FC susceptibility decreases with decreasing the field H and becomes positive at $H < 0.5$Oe. The field dependence of χ can be fitted by the expression $\chi(H) = 0.187G/(4\pi(H + 0.12\text{Oe})) - 0.3/4\pi$. The temperature and field dependencies of the FC susceptibility for two other samples studied in the present work have the same qualitative features which are, in general, characteristic for the samples showing the WE.

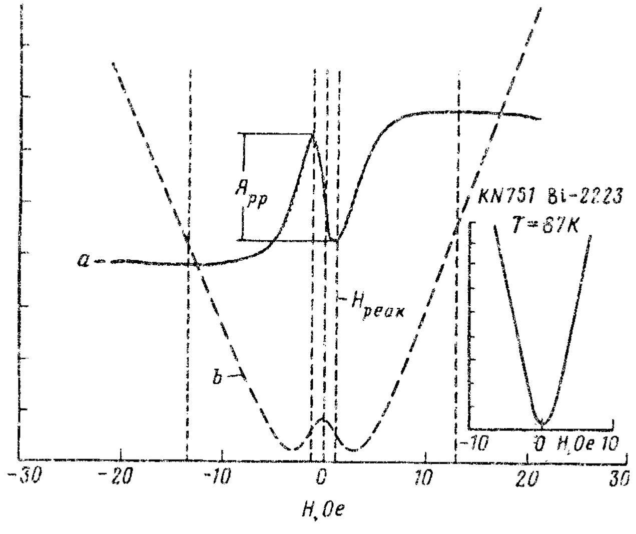

Fig.2. Curve a-dependence of the differential microwave absorption dP/dH on magnetic field H for the sample KAMU96 at $T = 62$K; curve b – $P(H)$ dependence obtained by numerical integration of the curve a. In insert is shown a typical $P(H)$ dependence for the sample whithout the WE.

The experimentally detected field derivative of absorbed power dP/dH for the sample KAMU96 is plotted as a function of H in Fig.2 (curve a). Curve b in the same figure is the result of numerical integration of the curve a, i.e. the field dependence of absorbed power $P(H)$. The field dependence of dP/dH appeared only in the SC state and was absent above T_c. The magnitude of the microwave response was a linear function of both the incident power and of the modulation field. It is evident from Fig.2 that the dP/dH curve consists of a broad and of a narrow line superimposed on each other and centered around $H = 0$. These two signals have opposite phases. The signs of the phases were compared with that of a derivative of the EPR absorption signal of a standard reference sample (like, e.g. organic radical DPPH) recorded during the same experiment. It turned out that the phase of the broad signal corresponds to minimum of absorption at $H = 0$, while the phase of the narrow signal corresponds to maximum of absorption at zero field. Let us call the first signal as a "normal" signal, and the second one as an "anomalous" signal. Thus, in general, the field dependence of the low magnetic field microwave absorption of the samples with the WE illustrated by the curve b is nonmonotonical. The characteristic feature is the presence of local maximum of absorption at zero field. Similar signals were observed for two other samples - KN99F and KN100F. We also stress that this peculiarity was not found in the samples without the WE. For such samples we always detected minimum of absorption at zero field. This is illustrated by insert of Fig.2 where $P(H)$ curve for one of the samples without the WE is shown (see also [1,6]).

In Fig.3a the temperature dependencies of the peak-to-peak amplitudes A_{pp} (defined in Fig.2) of "normal" and "anomalous" MWA signals for the samples studied in present work are presented. In Fig.3b we show the temperature dependence of the field at the peak of the "anomalous" signal H_{peak} i.e. the halfwidth of the local maximum around $H = 0$ (definition of H_{peak} is shown in Fig.2). As it can be seen from Fig.3a, upon transition to the SC state first the "normal" MWA signal appears and then the "anomalous" signal develops a few degrees below. On further cooling both signals reach maxima and then their amplitudes decrease. As to the value of H_{peak}, it increases rather rapidly near T_c and saturates at low temperatures. Simultaneously with saturation, the "anomalous" peak of dP/dH transforms into plateau which causes significant error in determination of the value of H_{peak} at about 20K.

3. It is known that the field dependence of microwave losses in HTS at $H < H_{c1}$ is caused by absorption of the microwave power P by a network of inter-and intragranular Josephson junctions [7]. Normally the shape of the dP/dH curve resembles that of the derivative of an EPR signal centered around $H = 0$. However it is a nonresonant effect and, moreover in contrast to the EPR, the centre of this line corresponds to minimum of absorbed microwave power. As it can be seen from Fig.2, the qualitative difference in behaviour of the HTS samples with the WE is that at $H = 0$ the magnitude of the microwave losses has local maximum.

It was assumed earlier [1] that the WE may be caused by appearance in a SC phase of the spontaneous orbital currents and connected with them orbital magnetic moments forming a kind of "orbital glass" [8]. The reason for appearance of such currents could be the existence in the samples of weak links with inverse Josephson coupling, or negative critical current (π-contacts). During the tunneling through this junction the Cooper pair acquires the phase shift π, e.g. due to

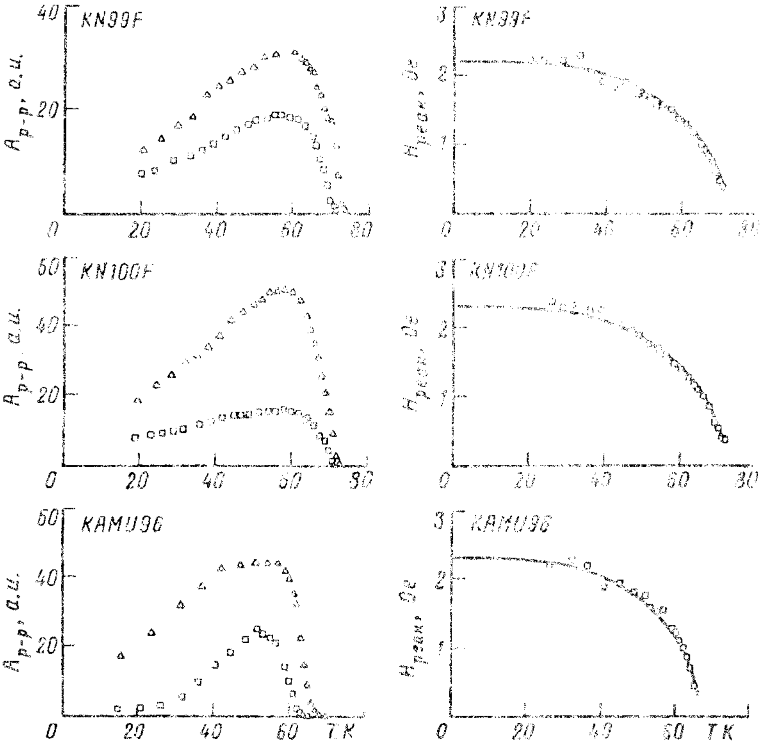

Fig.3. Temperature dependences: (*a*) of the amplitude of the "normal" (triangles) and "anomalous" (squares) MWA signals (left panel); (*b*) - field at the "anomalous" MWA peak H_{peak} (right panel) for the samples KN99F, KN100F and KAMU96.

spin-flip tunneling via magnetic impurities in the barrier. The properties of a loop with such a π-contact were theoretically investigated in^9 $^{2)}$. The authors showed that under the condition $(2\pi/c)LI_c > \Phi_0$ the ground state of the loop is the state with a current and with a flux (here L is the inductance of the loop, I_c is the Josephson critical current and Φ_0 is the quantum of flux). If this condition is satisfied the dependence of the current I_s in the SC loop of the area S on the external flux $\Phi_{ext} = HS$ has hysteresis [10]. The centre of hysteresis corresponds to $\Phi_{ext} = \Phi_0/2$ for the zero phase shift of the Cooper pair (0-contact) and to $\Phi_{ext} = 0$ for the phase shift π(π-contact). This hysteretic dependence of I_s on Φ_{ext} centered around $\Phi_{ext} = 0$ may lead both to appearance of spontaneous magnetization (i.e. to the WE) and to absorption of microwave power by a SC loop with a π-contact in the absence of dc external field [2]. When such a loop is threaded only by a microwave flux, due to hysteretic behaviour of $I_s(\Phi_{ext})$ the phase slips will occur leading to dissipation of the microwave energy. If the studied system consists of SC loops with both π- and 0-contacts, the absorption should first have maximum at $\Phi_{ext} = 0$ and then should decrease and reach next maximum when the external flux approaches the hysteresis region centered about $\Phi_{ext} = \Phi_0/2$ for the loop with

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$^{2)}$ The SC pairing of *d*-type as an alternative reason for appearance of the phase shift π was discussed recently in [2] the 0-contact [2]. In reality, due to random orientation of the loops relative to the direction of the external field and also due to scattering of the values of S and I_c, the smooth dependence of dP/dH on H with two peaks with the opposite phases may be expected. The values of the magnetic field at these peaks will correspond to some effective area and critical current of loops with π- and 0-contacts (for the system of SC loops with 0-contacts this situation was modeled in [11]). As it can be seen from Fig.2 just these peculiarities of the MWA are indeed observed for the samples with the WE.

Let us now discuss the temperature dependence of $H_{peak}(T)$ shown in Fig.3b. On the basis of the above discussion one may assume that H_{peak} is the measure of the width $\Delta\Phi$ of the hysteresis of the curve $I_s(\Phi_{ext})$ centered at $\Phi_{ext} = 0$. Since $\Delta\Phi \sim LI_c$, the variation of the peak position of the "anomalous" MWA signal with temperature should reflect the temperature dependence of the Josephson critical current of the π-contact $I_c(T)$. Using the Ambegaokar-Baratoff expression [12] for the Josephson critical current $I_c(T) \sim \Delta(T)\tanh[\Delta(T)/2k_BT]$ and solving numerically the well-known BCS integral equation for the SC gap $\Delta(T)$, one can simulate the temperature dependence of H_{peak} (solid lines in Fig.3b). The result is in good accordance with the experimental data. From the condition of hysteresis - $(2\pi/c)LI_c > \Phi_0$, one can roughly estimate the lower limit of the area S of a loop with π-contact. If one assumes that $LI_c/c = SH_{peak}(0)$, then for $H_{peak}(0) \approx 2.5\,\mathrm{Oe}$ (Fig.3b) we would get $S \gtrsim 1.3 \mu m^2$. The small value of the estimated loop's area may speak for intragranular localisation of π-contacts.

4. In summary, in this paper we present the results of experimental investigation of an anomaly in differential low magnetic field microwave absorption in the samples of Bi-2212 HTS which have what we call the Wohlleben Effect (paramagnetic FC susceptibility at low fields). The analysis of field and temperature dependencies of differential absorption dP/dH allows one to conclude that the anomaly in the MWA has common origin with the WE and is probably caused by presence in the samples of the Josephson weak links with π-contacts. It should be noted that there are different points of view as to the possible microscopic origin of such contacts e.g. existence of magnetic impurities in the junction or anisotropic (i.e. d-wave) pairing in HTS.

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1. W.Braunisch, N.Knauf, V.Kataev et all., Phys. Rev. Lett., **68**, 1908 (1992); W.Braunisch, N.Knauf, G.Bauer et all. Phys. Rev., **B48**, (1993); N.Knauf, W.Braunisch, G.Bauer et all., to appear in Physica C, Proc. from LT20 in Eugene, Oregon, Aug.4-11, 1993.
2. M.Sigrist and T.M.Rice, J.Phys.Soc.Jap. **61**, 4283 (1992); M.Sigrist and T.M.Rice, unpublished.
3. N.Knauf, J.Harnischmacher, R.Müller et all, Physica C, **173**, 414 (1991).
4. N.Knauf, PhD thesis, Universität zu Köln (in preparation).
5. J.Bock and E.Preisler, Solid State Commun., **72**, 453 (1989).
6. V.Kataev, N.Knauf, B.Büchner and D.Wohlleben, Physica C, **184**, 165 (1991).
7. K.W.Blazey, K.A.Müller, J.G.Bednorz, et all., Phys.Rev., **B36**, 7241 (1987); K.Khachaturyan, E.R.Weber, P.Tejedor, et all., Phys.Rev., **B36**, 8309 (1987).

8. F.V.Kusmartsev, Phys.Lett. **A160**, 108 (1992); Phys.Rev.Lett. **69**, 2268 (1992).
9. L.N.Bulaevskii, V.V.Kuzii and A.A.Sobyanin, Pis'ma v Zh.Eksp.Teor.Fiz., **25**, 314 (1977) [JETP Lett., **25**, 290 (1977)].
10. A.H.Silver and J.E.Zimmermann, Phys.Rev., **157**, 317 (1967).
11. F.J.Owens, Physica **C171**, 25 (1990).
12. V.Ambegaokar and A.Baratoff, Phys.Rev.Lett., **10**, 486 (1963); **11**, 104(E) (1963).