Peculiarities of nonlinear response of YBaCuO ceramic superconductors near the transition point

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The nonlinear properties of ceramic YBaCuO samples near the superconducting-transition point are experimentally investigated. The temperature and field dependences of the detection signal are obtained. It is shown, in particular, that in a strong field the nonlinear response is proportional to H^{-q}, with the exponent q dependent on the temperature. The self-consistent equations of the dynamics of a Josephson junction are used to derive for the detection signal expressions that agree qualitatively with the experimental results.

1. INTRODUCTION

Various studies of different aspects of the magnetic behavior of the new oxide semiconductors (see, e.g., Ref. 1) have shown that ceramic or cast polycrystalline samples are Josephson media (JM). Such media are characterized by the existence of small (compared with the sample size) regions (with characteristic dimension larger than or of the order of the field penetration depth λ) with relatively large value of the typical condensation energy ε_c (granules, crystals) and with weak Josephson bonds between these regions, with a characteristic energy ε_j ($\varepsilon_j \ll \varepsilon_c$). Note that certain classes of oxide superconductors (for example, of type YBaCuO) behave as JM even in the single-crystal state, owning to the presence of a branched network of twins or of other macroscopic defects that suppress the superconducting order parameter.

Research into JM began long before the advent of high-temperature superconductivity (HTSC) (see, e.g., Ref. 2), but only the study of HTSC has revealed a number of heretofore unknown aspects of the behavior of a JM. These include primarily many specific nonlinear properties. As a rule, the nonlinearities of these objects are determined mainly by a very low value of the field H_{c1} for weak bonds between the granules of the substance. In JM. the value of H_{c1} is estimated to be of the order of 10^{-2}–10 Oe. The spectrum of the harmonics excited at low frequencies ($\omega_0/2\pi \approx 1 - 10$ kHz) therefore contains harmonics of very high number, up to 23 inclusive.[3]

In addition to experiments on harmonic generation, measurements are usually made also of the detecting properties of superconductors.[4,5] In these experiments, however, samples with contacts are used, and this leads to additional difficulties in the interpretation of the results. These experimental shortcomings can be avoided by using contactless measurement methods. Such an experimental procedure was used in Refs. 6 and 7 to study detection in normal metals at low temperatures in the microwave and the rf bands. It was shown there that observation of nonlinear properties of normal metals requires samples with very large carrier mean free paths. Ceramic HTSC have a very small electron mean free path, and become therefore nonlinear only in the superconducting phase. We report here a study of detection in the HTSC ceramic $YBa_2Cu_3O_{7-\delta}$ by a procedure somewhat different from that of Ref. 7. We show in the next section that the study of detection yields interesting physical information on the purely Josephson properties of the medium.

2. BASIC THEORETICAL PREMISES OF THE EXPERIMENT

The foundations of the theoretical description of JM were developed in a 1974 study, by Rosenblatt,[8] of coherent and paracoherent states of such media. This theory was further developed in a large number of studies (see, e.g., Ref. 2). Dersch and Blatter[9] used one of the self-consistent averaged variants of this theory to construct a model of the critical state of ceramic HTSC. It should also be noted that a quasilinear variant of this theory, used by Sonin and Tagantsev[10] to describe the frequency dependence of the impedance of HTSC, is apparently in fair agreement with experiment.

We shall find useful here the basic relations describing the dynamics of a JM. To this end we consider first a granulated sample without any bonds between the granules. Then, introducing as usual ρ_n and ρ_s as the fractions of the normal and superconducting phases, we express the magnetic permeability of the ceramic, following for example Ref. 9, in the form

$$\mu_c = \rho_n + \rho_s \mu_g,$$

where μ_g is the effective magnetic permeability of the superconducting grains, defined as the ratio of the magnetic fluxes that penetrate into a grain in the superconducting (Φ_s) and normal (Φ_n) states, i.e., $\mu_g = \Phi_s/\Phi_n$. Since the characteristic dimension a of the granules used in this experiment is of the order of the penetration depth $\lambda(0)$ of the magnetic field at $T = 0$, it follows that μ_c can be substantially dependent on temperature in some interval in the vicinity of T_c, since μ_g takes, for example for spherical granules, the form

$$\mu_g = \frac{3}{\gamma(T)}\left[\operatorname{cth}\gamma(T) - \frac{1}{\gamma(T)}\right], \tag{1}$$

and for cylindrical granules[9]

$$\mu_g = \frac{4}{\gamma}\frac{I_1(\gamma(T)/2)}{I_0(\gamma(T)/2)}, \tag{2}$$

where $\gamma(T) = a/\gamma(T)$ and I_n is a Bessel function of imaginary argument.

It is seen from Eqs. (1) and (2) that, regardless of the granule shape, there exists at $a \sim \lambda(0)$ a rather wide temperature interval in which μ_g, and hence also μ_c changes perceptibly with temperature. When $\gamma(0) \gg 1$ the interval where the temperature dependence of μ_c is substantial shifts to a narrow temperature region $\Delta T = T_c - T$ near T_c: $\Delta T \approx T_c \gamma^{-2}(0)$. The value of μ_c of the samples em-

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ployed amounted in the vicinity of 77 K to several tenths.

Turning on the bonds between the granules in the case $\varepsilon_j \gg T$ establishes a coherent gauge-invariant phase difference Φ_{ik} between granules i and k, in the form

$$\Phi_{ik}=\chi_i-\chi_k-\frac{2\pi}{\Phi_0}\int_i^k d\mathbf{r}\,\mathbf{A}, \tag{3}$$

where χ_i is the phase of the order parameter of granule i, $\mathbf{A}$ is the vector potential, and Φ_0 is the magnetic-flux quantum ($\Phi_0 = ch/2e$). The current between the granules is known to be determined by the Josephson relation

$$J_{ik}=J_j \sin \Phi_{ik}. \tag{4}$$

Here i and k are nearest neighbors and $J_j = 2e\varepsilon_j/\hbar$.

Since we shall be interested below only in self-consistent equations that describe the electrodynamics of JM at average values of the fields and currents, it is convenient to change to a continual limit. Φ_{ik} takes then the form $\mathbf{Q}(\mathbf{r}_i - \mathbf{r}_k)$, where

$$\mathbf{Q}=\nabla\chi-(2\pi/\Phi_0)\mathbf{A}, \tag{5}$$

and $\mathbf{r}_i - \mathbf{r}_k$ for nearest adjacent granules can be replaced in this limit by a continuous vector $\mathbf{n}(\mathbf{r})a$, where $\mathbf{n}(\mathbf{r})$ is defined as the mean value of the unit vector $(\mathbf{r}_i - \mathbf{r}_k)/|\mathbf{r}_i - \mathbf{r}_k|$ with $(\mathbf{r}_i + \mathbf{r}_k)/2 = \mathbf{r}$.

The vector potential $\mathbf{A}$ in (5) determines the magnetic induction $\mathbf{B}$ resulting from the action of the external field $\mathbf{H}$ applied to the sample in conjunction with the screening currents in the granules and the screening Josephson currents flowing through the entire bulk of the sample. If the Josephson contribution to the magnetization $\mathbf{M}_j$ is determined by the traditional approach[11] from the relation $c\,\mathrm{curl}\mathbf{M}_j = \mathbf{J}_j$ and $\mathbf{B}$ is expressed in the form

$$\mathbf{B}=\mathrm{rot}\,\mathbf{A}=\mathbf{H}+4\pi\mathbf{M}, \tag{6}$$

then $\mathbf{M} = \mathbf{M}_g + \mathbf{M}_j$, where the magnetization $\mathbf{M}_g$ due to the screening currents flowing in the granules is linearly connected with the effective field $\mathbf{H} + 4\pi\mathbf{M}_j$. As a result, the first Maxwell equation takes the form

$$\mathrm{rot}\,\mathbf{B}=\frac{4\pi}{c}\mu_c\frac{J_j}{a^2}\mathbf{n}(\mathbf{r})\sin[a\mathbf{n}(\mathbf{r})\mathbf{Q}]+\mu_c\frac{\bar{\varepsilon}}{c}\frac{\partial\mathbf{E}}{\partial t} \tag{7}$$

where $\bar{\varepsilon}$ is the effective dielectric constant of the JM and takes into account the fact that the electric field is produced only in dielectric regions (with constant ε) through which the Josephson bonds between the granules are established ($\bar{\varepsilon}\approx dl^2\varepsilon/a^3$, d is the distance between the contacting surfaces of the granules, and $l<a$ is the effective transverse dimension of the bond). The field $\mathbf{E}$ in (7) is connected with $\mathbf{Q}$ by the Josephson relation

$$\partial\mathbf{Q}/\partial t=2e\mathbf{E}/\hbar. \tag{8}$$

If a weak alternating field of frequency ω is applied to the sample together with the constant external field, the nonlinearity of Eq. (7) leads to generation of higher harmonics of this frequency. The amplitude $\mathbf{B}_2$ of the second harmonic of the magnetic induction is proportional to the constant magnetization of the sample by the Josephson screening currents flowing through the entire sample:

$$\mathbf{B}_2=4\pi\mu_c(eaE_0/\hbar\omega)^2\mathbf{M}_j, \tag{9}$$

where E_0 is the electric field amplitude ($eaE_0/\hbar\omega \ll 1$).

Relation (9) was derived under conditions with a given alternating electric field. In a real experiment it is impossible as a rule to determine the electric field. The simplest situation, when the external alternating magnetic field is specified, leads to a much more complicated connection between the constant Josephson magnetization and the amplitude of the second harmonic of the induction. If, however, the constant field is $H \gg H_{c1}{}^*$, the corresponding connection reduces to Eq. (9). Here $H_{c1}{}^*$ is the field of vortex penetration into the JM and not into the granules ($H_{c1}{}^* \ll H_{c1}$):

$$H_{c1}{}^*\approx\frac{\Phi_0}{4\pi\mu_c\lambda_c{}^2}\ln\frac{\lambda_c}{a}=\frac{2\pi J_j}{ca}\ln\frac{\lambda_c}{a}, \tag{10}$$

where λ_c is the depth of penetration of the field into the JM,[12] and can be determined from Eqs. (5) and (7):

$$\lambda_c{}^2=\hbar c^2 a/8\pi e\mu_c J_j.$$

Thus, an investigation of the response to the second harmonic makes it possible to separate in the total magnetization the contribution of the Josephson currents.

Obviously, relation (9) with allowance for the remarks above can also be used to determine the magnitude of the detected amplitude-modulated signal.

3. EXPERIMENT

Our experiment was the following. An amplitude-modulated electromagnetic wave was applied to the sample, and a useful frequency-modulated signal was received. The incident-wave and modulation frequencies were $\omega_0/2\pi\approx 1.2$ MHz and $\Omega/2\pi\approx 811$ Hz, respectively. This difference between the pump frequency ω_0 and the modulation frequency Ω makes it relatively easy to construct passive high-pass and low-pass filters, a very important factor in experiment, since the useful signal is usually quite weak. The ceramic $YBa_2Cu_3O_{7-\delta}$ sample used by us was a cylinder 3.5 mm in diameter and 23 mm long. The high-frequency field was produced by a copper coil having approximately 30 turns. The useful signal was received with a coil wound above the high-frequency coil and having approximately 150 turns. Both coils were ≈ 12 mm long and were placed around the center of the sample. The earth's magnetic field was compensated for, accurate to $\pm$ 10 mOe, by a system of Helmholtz coils. The constant magnetic field H was produced by additional Helmholtz coils. It was directed along the sample axis and could be varied from 0 to 100 Oe. The rf pump field was excited through a high-pass filter by a G4-102A oscillator. Its amplitude h_1 could reach 1.5 Oe. The useful signal was received through a low-pass filter by a UPI converter-amplifier. The minimum recorded signal was ≈ 10 nV. The sample temperature was measured with a copper-constantan thermocouple fastened to the surface of the receiving coil. The temperature was stable to within $\pm$ 0.03 K.

4. RESULTS

It is known that a significant role in the behavior of ceramic HTSC is played by the method of cooling the sample inside and outside a magnetic field.[6] The cause is the trapping of the flux by such granulated systems. If the investigated sample traps a magnetic fluid, then a useful signal at the modulation frequency Ω will not vanish in a zero magnetic field.

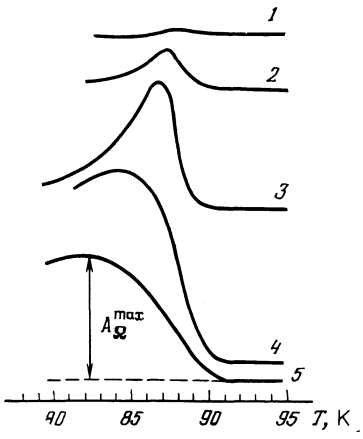

FIG. 1. Temperature dependence of A_Ω for different values of the constant magnetic field H: *1*—$H = 0$; *2*—0.1 Oe; *3*—0. 4 Oe; *4*—10 Oe; *5*—20 Oe.

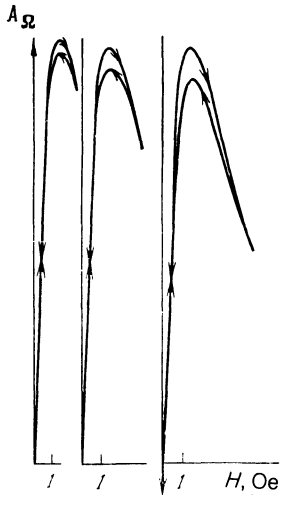

FIG. 3. Example of determination of the field at which trapping of the magnetic flux begins at $T = 84.5$ K.

In experiment we are able to carry out measurements on a sample cooled in a zero and in a finite magnetic field. Figure 1 shows the temperature dependences of the signal $A_\Omega \propto \Omega B_2$ for different magnetic fields. It is seen from Fig. 1 that cooling in a field 0.1 Oe increases A_Ω approximately tenfold compared with the signal in the residual field of our facility.

It is evident from Fig. 1 that the temperature dependence of A_Ω has a maximum whose magnitude and position in temperature depend on the field H in which the sample was cooled. A plot of $A_\Omega^{max}(H)$ is shown in Fig. 2. The presence of a second-harmonic amplitude was noted in Ref. 3. The simple model proposed in Ref. 3 does not explain the presence of this maximum. The maximum is well accounted for qualitatively by Eq. (9). In fact, the temperature dependence of the magnetization in a fixed field takes the form of a curve that rises monotonically when T is lowered and reaches saturation at temperatures below a $T_{c1}(H)$ value determined from the relation $H_{c1}^*(T) = H$. The value of μ_c, on the contrary, decreases here from $\mu_c = 1$ at $T = T_c$ to a value $\approx \rho_n$. The resultant curve has therefore a maximum. In fields $H \ll \mu_c(0) H_{c1}(0)$ the maximum follows practically linearly the growth of the field (see Fig. 2). In relatively strong fields, however, when $H/H_{c1}(0) > \mu_c(0)$, the signal at the maximum depends little on the field H.

In addition to the described experiment, alternative measurements can be made, namely, track the value of A_Ω as a function of the field H at a fixed temperature. As noted above, A_Ω vanishes in a zero field. This simple fact can be used to measure the temperature dependence of the magnetic field value at which trapping of the magnetic flux begins. To this end, the sample is first cooled in a zero field to the specified temperature, and the appearance of a residual signal noted after the magnetic field was built up and dumped. Figure 3 shows an example of the experimental plots used to determine the field at which magnetic-flux trapping sets in. The temperature dependence obtained is shown in Fig. 4.

Of greatest interest, in our opinion, are the measured field dependences of A_Ω for various temperatures. Figure 5 shows a typical $A_\Omega(H)$ plot for $T = 86.3$ K. If the field in which the maximum of A_Ω is observed is comparable with $H_{c1}^*(H_{c1}^* \gtrsim H_{max}$, where H_{max} is the field at which A_Ω is a maximum (see Fig. 3), it can be assumed that $H \gg H_{c1}^*$ on the descending section of the $A_\Omega(H)$ curve. We have assumed that the $A_\Omega(H)$ has a power-law dependence at $H \gg H_{c1}^*$, i.e. $A_\Omega(H) \propto H^{-q}$. The exponent q was determined from a log-log $A_\Omega(H)$ plot drawn with the zero point of $A_\Omega(H)$ naturally chosen in the experiment to correspond to cooling in a zero field. The log-log plots of $A_\Omega(H)$ in Fig.

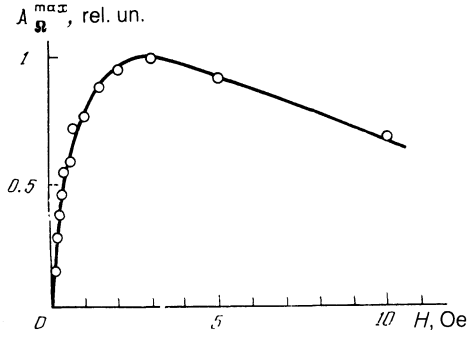

FIG. 2. Dependence of A_Ω^{max} on the field H.

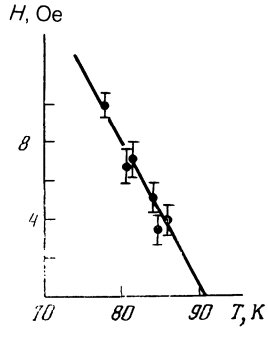

FIG. 4. Temperature dependence of the field at which trapping of the magnetic flux begins.

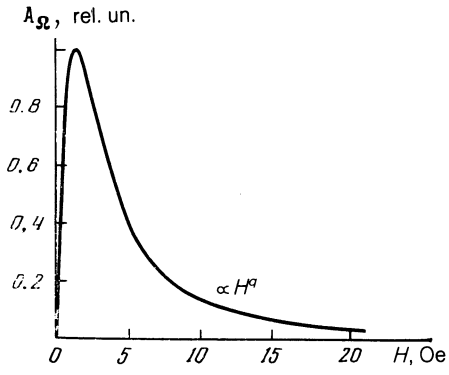

FIG. 5. Form of the $A_\Omega(H)$ dependence when a sample is magnetized after cooling to $T = 86.3$ K in a zero field.

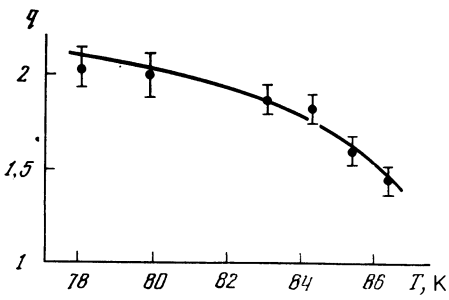

FIG. 7. Temperature dependence of the exponent q.

6 show that $A_\Omega(H)$ for $H \gg H_{c1}$* is indeed a power-law function of the magnetic field. This experiment has shown that the exponent q depends on temperature, as shown in Fig. 7.

5. CONCLUSION

We note in conclusion that the experimental $M_0(H) \propto H^{-q(T)}$ dependence in the region $H \gg H_{c1}$* [where $q(T)$ is a function that varies little with temperature] was to be expected from the following considerations. In an ideal superconductor, the decrease of the magnetization in a strong field is logarithmic (see, e.g., Ref. 13), i.e., $q = 0$. The effective dimensionality of the system is here $d = 2$. For ideal extended Josephson junctions[14] we have $d = 1$ and $q = 3$. Since onset of disorder lowers the effective dimensionality of the system,[15] the expected range of q must be $0 < q < 3$. Usually, however, lowering the effective dimensionality of the system (see also Ref. 16) is not accompanied by violation of universality. The obtained $q(T)$ dependence can attest to violation of universality. This circumstance agrees with the results of Ref. 17, where it is shown that universality in ceramic HTSC is lost in magnetic fields. This conclusion is, of course, not final and requires additional confirmation, both experimental and theoretical.

The nonlinear properties of bulky YBaCuO samples investigated here can be understood, as shown in Sec. 2, in the context of the self-consistent averaged equations that describe the JM dynamics. Unfortunately, however, a quantitative comparison of theory with experiment is beyond the scope of the simplest theory, since calculation of the magnetization $M_J(H)$ for a random network of Josephson bonds remains to this day a complicated theoretical problem. Nonetheless, the experiments have shown that an investigation of the nonlinear properties of HTSC makes can yield nontrivial physical information.

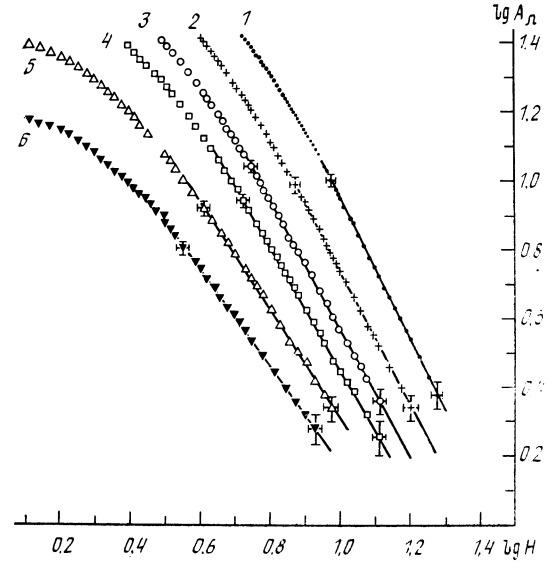

FIG. 6. Log-log plot of the $A_\Omega(H)$ dependence: *1*—$T = 78$ K; *2*—79.6 K; *3*—83.2 K; *4*—84.5 K; *5*—85.2 K; *6*—86.3 K (the field H is in arbitrary units).

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