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Nonlinear microwave response of a YBaCuO

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Abstract. — The review of modern situation concerning the studies of the generation of microwave harmonics in YBaCuO is given. Far from T_c the nonlinear response of ceramics YBaCuO has Josephson nature in a weak magnetic fields or is described in terms of the critical state model in a strong external magnetic field H. Near T_c, the signals of the second $P_{2\omega}(T)$ and the third $P_{3\omega}(T)$ harmonics have the form of spikes of generation. Yet the signal $P_{2\omega}(H)$ shows a hysteresis and is proportional to the square of incident power $P_{2\omega} \propto P_\omega^2$ while the signal $P_{3\omega} \propto P_\omega^3$ and doesn't depend on the field H. Such behavior of the nonlinear response near T_c is typical of both ceramics and single crystals of YBaCuO. We use the Portis model to describe the signal $P_{2\omega}$ and the model based on Eliashberg theory to describe the signal $P_{3\omega}$. The question about the model which can simultaneously explain the behavior of the second and the third harmonics remains open.

Introduction.

The papers devoted to the studies of nonlinear properties of high temperature superconductors have been published of late. It has been caused by both the possibility of new superconductors being employed for applications and general interest of physicists studying non-stationary dynamic processes in superconductors.

The measurement of the intensity of the highest harmonics occurring at electromagnetic wave reflection from the superconductor surface is a convenient method to study such processes. In particular the signals of double and triple frequencies have been observed in microwave range. To explain experiments various mechanisms of generation have been proposed.

In this review, we have tried to classify the already known [1-6] and the latest results of microwave harmonics studies. The detection processes and frequency mixing, analyzed for example in references [7-10], are not considered here. Moreover we will also omit experimental results from references [11-13] which concerned the nonlinear dependence of microwave absorption on modulating field amplitude.

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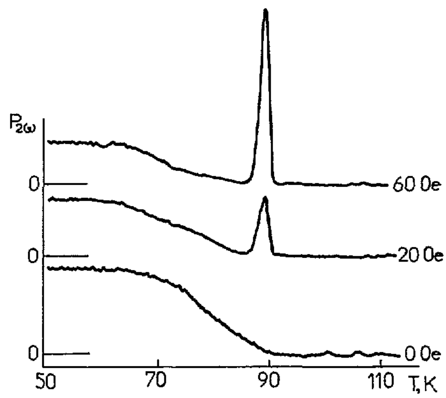

Fig. 1. — The second harmonic signal, $P_{2\omega}$, *versus* the temperature in various magnetic field H (the curve labels at the right). The maximum amplitude $H_{\sim}$ of the alternating magnetic field H_{ω} at the surface of the sample $H_{\sim} = 18$ Oe ; $\omega/2\pi = 9.3$ Ghz [2].

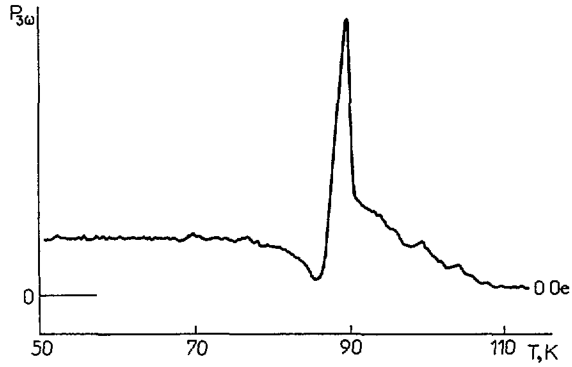

Fig. 2. — The third harmonic signal, $P_{3\omega}$, *versus* the temperature at $H = 0$; $H_{\sim} = 10$ Oe ; $\omega/2\pi = 9.4$ GHz [2].

Figures 1 and 2 represent the typical plots of harmonics powers : the second $P_{2\omega}$ and the third $P_{3\omega}$ as a function of temperature for ceramic sample of YBaCuO [1, 2]. A similar dependence $P_{2\omega}(T)$ has been observed in references [3, 4]. The curves in figures 1, 2 have three peculiarities :

1) absence of nonlinear signal in the normal state of a superconductor ;
2) maximum in the vicinity of the superconducting transition temperature T_c ;
3) substantial and almost constant signal level at $T < T_c$.

The measurements of $P_{2\omega}$ and $P_{3\omega}$ as a function of external magnetic field and amplitude of microwave incident on the sample showed that peculiarities 2) and 3) have different nature. We shall consider them separately.

Experiment.

The essence of the method of harmonics generation consists in irradiating YBaCuO sample by an electromagnetic wave of high frequency ω and detecting a signal of frequency 2ω (or 3ω) occurring in the sample.

The sample is placed in a bimodal resonator which may be simultaneously tuned to either frequencies ω and $2\,\omega$ or ω and $3\,\omega$. Rectangular and cylindrical resonators [14] were employed. The sample was exposed to an electromagnetic wave with frequency $\omega/2\,\pi = 9.4$ GHz from a magnetron, in pulsed operation with a pulse repetition frequency of 50 Hz and a pulse length of 2 μs. The wave power entering the resonator, could be varied from 1 W up to 2 kW, which corresponded to the change of the maximal amplitude $H_{\sim}$ of the alternating magnetic field H_{ω} on the sample surface within the interval $0.5\ \text{Oe} \leqslant H_{\sim} \leqslant 25$ Oe. A set of absorbing and reflecting filters (see Fig. 3) was employed for suppression of magnetron harmonics. The harmonics signal was directed from the sample to superheterodyne receiver with the sensitivity up to 10^{-12} W and then registered by boxcar SR250. The quantities measured were the pulse powers $P_{2\,\omega}$ or $P_{3\,\omega}$ of the radiated wave of frequencies $2\,\omega$ or $3\,\omega$. It was convenient to investigate weak signals by applying a microwave bias to the detector. In this case the 2nd (or 3d) harmonics was separated from the magnetron spectrum and was the reference wave. The presence of a phase shifter in the reference signal channel made it possible to measure the variation of the phase of the wave generated in the sample as a function of H.

In these experiments we used a static external magnetic field $-7\ \text{kOe} < H < 7$ kOe, which could be rotated in the plane parallel to the irradiated surface of the sample. The field $H < 200$ Oe was created by means of a set of Helmholtz coils with Earth field compensation.

The sample temperature was measured by germanium thermometer soldered to the external surface of resonator. In the experiment there was a possibility to carry out measurements at any fixed temperature, the adjustment accuracy of which was ∓ 0.01 K.

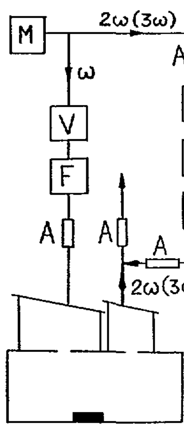

Fig. 3. — Block diagram of high-frequency channels ; M-magnetron, V-isolator, F-filter, A-attenuator, R-phase shifter.

Nonlinearity far from T_c.

At first let's consider the nonlinear signals observed at $T < T_c$ and weakly dependent on temperature (Figs. 1, 2).

WEAK EXTERNAL FIELD. — Figure 4 represents the dependence of the 2nd harmonics $P_{2\,\omega}$ signal on the external magnetic field $\mathbf{H} /\!/ \mathbf{H}_{\omega}$ in ceramic YBaCuO sample, cooled down to $T = 4.2$ K in the field $H = 0$. The similar dependences were also observed at other temperatures $T < T_c$. Figure 5 depicts the curve $P_{3\,\omega}(H)$ at $T = 60$ K [2].

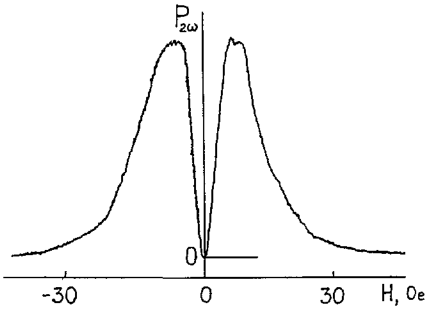

Fig. 4.

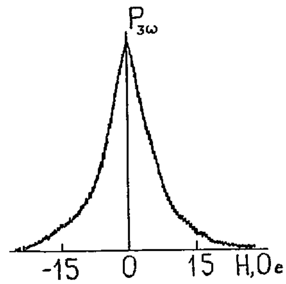

Fig. 5.

Fig. 4. — Typical plot of the second harmonic $P_{2\omega}$ as a function of the magnetic field H: $\mathbf{H} /\!/ \mathbf{H}_\omega$; $H_{\sim} = 2$ Oe ; $T = 4.2$ K ; $\omega/2\pi = 9.4$ GHz.

Fig. 5. — Dependence of $P_{3\omega}(H)$; $H_{\sim} = 3$ Oe, $T = 60$ K ; $\omega/2\pi = 9.4$ GHz.

The signals $P_{2\omega}(H)$ and $P_{3\omega}(H)$ disappear in a sufficiently weak magnetic field. The shape of curves $P_{2\omega}(H)$ and $P_{3\omega}(H)$ is reproduced in figures 4, 5 in changing sweep direction.

Note that the disappearance of the signal $P_{2\omega}(H)$ is not observed on any ceramic YBaCuO sample, e.g. in the experiments [1-3] $P_{2\omega}$ is not equal to zero even at $H > 100$ Oe. In these cases maximum of $P_{2\omega}(H)$ in weak fields is reproduced if the magnetic field increases or decreases within the approximate interval -100 Oe $< H < 100$ Oe. The case of strong fields will be considered below.

Figure 6 shows the dependence of the power $P_{2\omega}$ on the power P_ω incident on the sample, obtained at $H = 15$ Oe and $T = 55$ K [2]. This dependence is not monotonic, the quadratic regime $H_{\sim}^{2\omega} \propto (H_{\sim}^{\omega})^2$ taking place only at the least levels of P_ω (in Fig. 6 up to $P_\omega = 3$ dB).

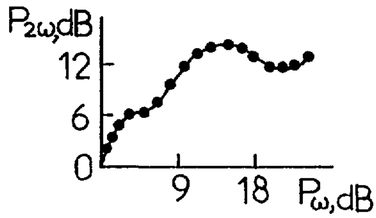

Fig. 6. — Dependence of the radiated power $P_{2\omega}$ on the power P_ω incident on the ceramic YBaCuO sample ; $H = 15$ Oe ; $T = 55$ K ; $\omega/2\pi = 9.4$ GHz [2].

The displayed dependences of harmonics intensity on the external field H and on the amplitude of incident microwave are indicative of Josephson generation mechanism at $T < T_c$ [1, 2].

Such mechanism was considered in detail in the paper [15] before the discovery of the high temperature superconductivity (*). Then Jeffries and co-workers [16, 17] proposed a similar model for explanation of the observed dependences of harmonics intensity on a weak field at $T < T_c$ in YBaCuO.

(*) The authors are indebted to W. L. McLean who drew our attention to this paper.

The superconducting granules in a ceramic sample are supposed to be weakly linked with each other by means of Josephson junctions with the area s. In the presence of the magnetic fields current loop with the area S containing the junction is formed. A superconductor has quite a complete set of current loops (similar to the set of high-frequency squids), which consists of superconducting rings shunted by Josephson junction. The current in the loop is

$$I(t) = I_c \sin\left(\frac{2\pi\Phi}{\Phi_0}\right) \tag{1}$$

where I_c is the critical transition current, $\Phi_0 = hc/2e$ is the flux quantum, Φ is full magnetic flux through the loop. The critical current can be written as [18] :

$$I_c = \left[\frac{\pi\Delta(T)}{2eR_n} \cdot \text{th}\left(\frac{\Delta(T)}{2kT}\right)\right] \cdot \frac{\sin(x)}{x} = I_c(T) \cdot \frac{\sin(x)}{x} \tag{2}$$

where $\Delta(T)$ is the gap, R_n is normal resistance, $x = sH/\Phi_0$. Magnetic flux is

$$\Phi = \mathbf{SH} + \mathbf{SH}_\omega + LI(t). \tag{3}$$

Here L is the self-inductance of the loop, $H_\omega = H_\sim \sin(\omega t)$ is the microwave magnetic field. Introducing the terms

$$\alpha \equiv \frac{2\pi SH}{\Phi_0}, \quad \beta \equiv \frac{2\pi SH_\sim}{\Phi_0}, \quad \delta \equiv \frac{2\pi L}{\Phi_0},$$

we find from (1), (2) and (3)

$$I(t) = I_c \sin[\alpha + \beta\sin(\omega t) + \delta I(t)]. \tag{4}$$

Following paper [3], let's suppose, the term $\delta I(t)$ is small as compared to the other terms. In the first order approximation on $\delta I(t)$ we obtain

$$I(t) \simeq I_c[\sin(\alpha + \beta\sin(\omega t)) + \delta I(t)\cos(\alpha + \beta\sin(\omega t))]. \tag{5}$$

From (5) it is not difficult to select the current component $I_{2\omega}$ in one loop at the frequency 2ω :

$$I_{2\omega} = 2I_c \sin\alpha\,[J_2(\beta) + \delta I_c\cos\alpha\,(J_1^2 + 2J_0J_2 - 2J_1J_3)]\cos(2\omega t) \tag{6}$$

where $J_i(\beta)$ is the Bessel function of i-th order.

The current loops and junctions in the sample may have various areas and directions with respect to the fields $\mathbf{H}$ and $\mathbf{H}_\omega$. For calculation of non linear response $P_{2\omega}$ it is necessary to select the suitable distribution of areas S and s. In references [3, 16] Gaussian distribution is employed :

$$F(A) = \exp[-(A-1)^2/2\sigma^2]/\sigma\sqrt{2\pi}$$

where σ is a mean quadratic deviation. Then, supposing the significant values of the areas s and S change from zero to their two-fold average values, we get for the power $P_{2\omega}$.

$$\begin{aligned} P_{2\omega} \propto (I_{2\omega})^2 \propto \Bigg\{\iint & I_c(T) \,.\, \sin(Ax)/(Ax)/(Ax) \,.\, \sin(A\alpha) \times \\ & \times \{J_2(A\beta) + I_c(T) \,.\, A\delta\sin(Ax)/(Ax) \,.\, \cos(A\alpha) \\ & \times [J_1^2(A\beta) + 2J_0(A\beta)J_2(A\beta) - 2J_1(A\beta)J_3(A\beta)]\}\, F(A)\,\mathrm{d}A\Bigg\}^2 \end{aligned} \tag{7}$$

Expression (7) containing the phenomenological parameters s, S, σ and δ (or $\delta I_c(T)$ at the present temperature) gives a good description of the experimental results on the second harmonics generation in the weak magnetic field and at low input power levels. As an example figure 7 shows dependences $P_{2\omega}(H)$ at $\mathbf{H} /\!/ \mathbf{H}_\omega /\!/ \mathbf{H}_{2\omega}$, two temperatures $T =$ 4.2 K, $T = 77$ K and at the various amplitudes $H_\sim$ of the microwave field H_ω [3]. Different symbols correspond to experimental data, solid lines are $P_{2\omega}$ calculated from expression (7) at the parameter values s, S, $\delta I_c(t)$ and σ indicated in the caption. The values s, S and σ correspond to ones published earlier in references [16, 19] for low frequencies.

Ciccarello *et al.* [3] indicate the satisfactory coincidence of experiment and calculation may be obtained for each separate curve $P_{2\omega}$ by the change of only three parameters s, S and σ, supposing $\delta = 0$ everywhere. However to describe the set of curves in figure 7 obtained at various levels of incident power, it is necessary to take into account the self-inductance $L \propto \delta$.

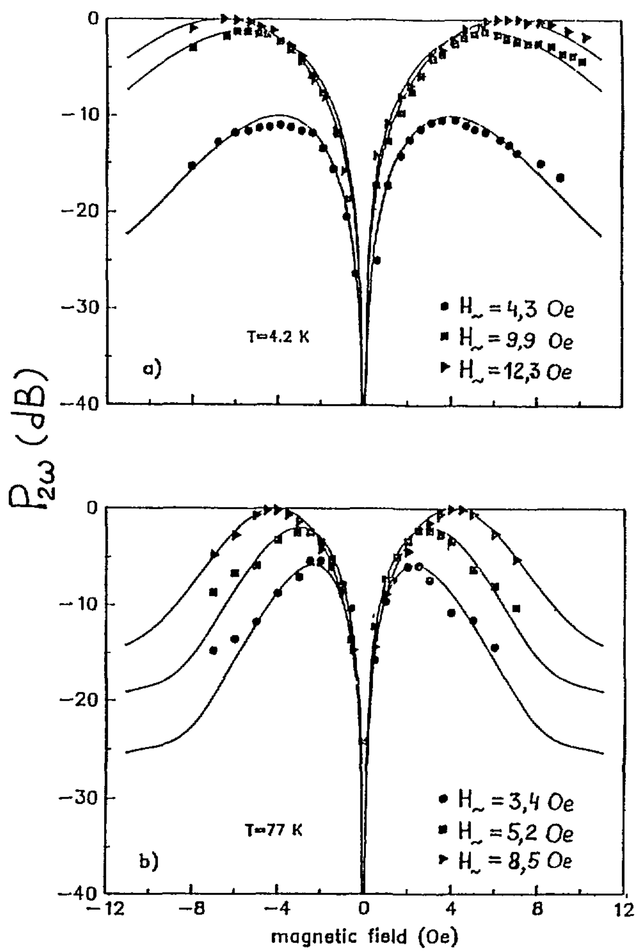

Fig. 7. — $P_{2\omega}$ signal as a function of the applied field H at different microwave field amplitudes $H_\sim$ at a frequency $\omega/2\pi = 3$ GHz [3]. Symbols are experimental data ; continuous lines are plots of equation (7) with : a) $s = 8 \times 10^{-9}$ cm^2 ; $S = 4.8 \times 10^{-9}$ cm^2 ; $\delta I_c(T) = 0.48$; $\sigma = 0.5$; b) $s = 1.54 \times 10^{-8}$ cm^2 ; $S = 9.2 \times 10^{-9}$ cm^2 ; $\delta I_c(T) = 0.4$; $\sigma = 0.5$.

At larger amplitudes $H_{\sim} > 10$ Oe it is impossible to reach agreement between the experimental and calculated according to (7) results, even varying all the parameters. This fact follows from figure 8 of the same reference [3], where the dependences $P_{2\omega}(P_{\omega})$ are shown.

Thus the Jeffries model [16, 17] is valid for description of the nonlinear response of high temperature superconductors placed in rather weak electromagnetic fields.

With the increase of the external magnetic field in ceramics YBaCuO at $T < T_c$ the hysteresis phenomena were observed. Now we are coming over to their consideration.

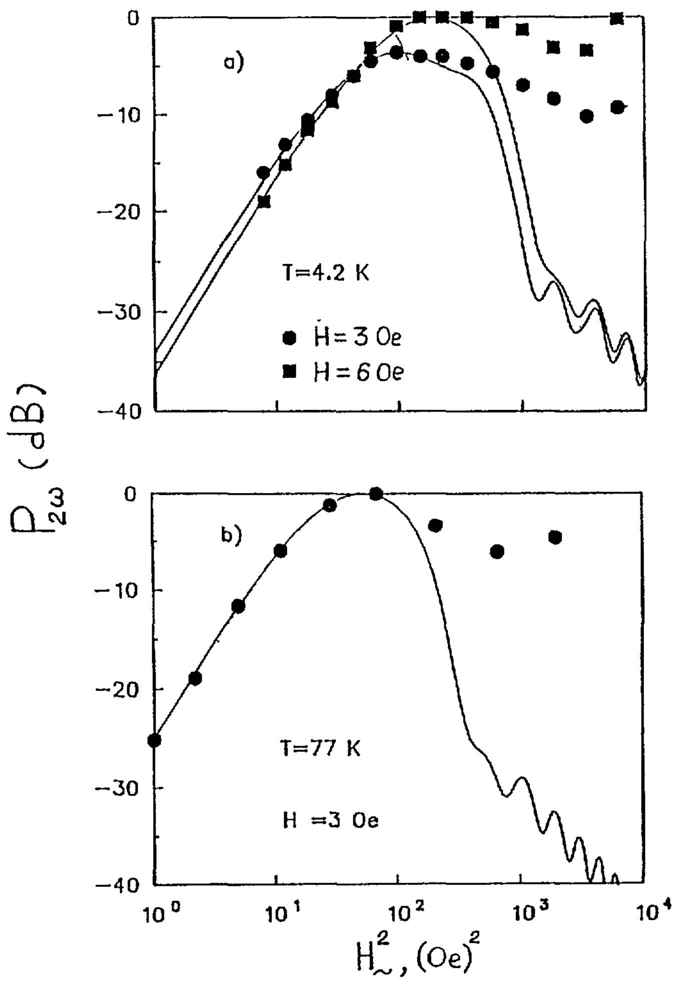

Fig. 8. — The $P_{2\omega}$ dependences as a function of the square of the microwave field amplitude $H^2_{\sim} \propto P_{\omega}$; $\mathbf{H} /\!/ \mathbf{H}_{\omega}$; $\omega/2\pi = 3$ GHz [3]. Symbols are experimental points ; continuous lines are plots of equation (7) in which for a) and b) is made use of the same values of s, S, $\delta I_c(T)$ and σ as for figure 7a) and 7b), respectively.

STRONG EXTERNAL FIELD. — Figure 9 shows the curves $P_{2\omega}(H)$ for a YBaCuO sample cooled down to $T = 4.2$ K in the field $H = 0$ [3]. In increasing H the maximum of $P_{2\omega}$ is observed then the signal drops, the line shape being reproduced if at $H < 100$ Oe the field sweeping is reversed. The curves of such a type were discussed above (Figs. 4, 5, 7). If

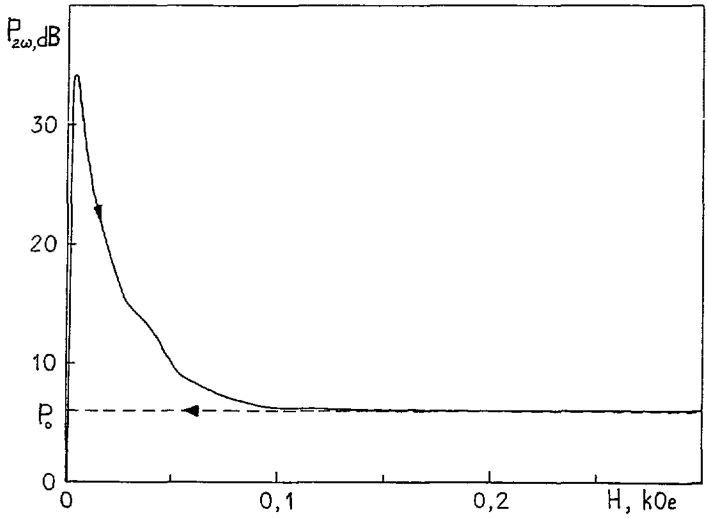

Fig. 9. — Dependence of $P_{2\omega}$ signal on the external field $0\ \text{Oe} \leq H \leq 300\ \text{Oe}$; $\mathbf{H} /\!/ \mathbf{H}_{\omega} /\!/ \mathbf{H}_{2\omega}$; $H_{\sim} = 3.5$ Oe ; $\omega/2\pi = 3$ GHz [3].

the field sweep direction is reversed at $H = 200\text{-}300$ Oe, when the signal $P_{2\omega}$ is very small, then at any further changes of field the signal is equal to P_0 (dashed line in Fig. 9). The value of this « remanent » signal may be increased only by increasing the level of the wave power incident on the sample. In this case the dependence $P_{2\omega}(H)$ would be similar to that observed on the samples cooled down to the temperatures $T < T_c$ in a sufficiently strong field $H = 1\,000\text{-}3\,000$ Oe [5].

Figure 10a demonstrates an example of the dependence $P_{2\omega}(H)$ for such a case. The nonlinear signal practically does not depend on an external field as long as H increases (or decreases) monotonically. Sharp minima are observed at the two end points of the diagram where the magnetic field sweep reverses its direction. At the same time the value of the field H wherein the sweep changes its direction, is insignificant.

For further comprehension of the generation mechanism it is important to measure the phase of the radiated signal as a function of the field H (Fig. 10b). In increasing magnetic field from D to A the phase remains constant. It does not vary also in decreasing field from A to B, but at the moment of passing through B it alters abruptly by 180°. At further decrease of the field H the phase keeps on its new value until the point D has been passed through where the phase alters abruptly by 180° again.

The experimental results, presented in figure 10, are accounted for in reference [5] on the basis of the Bean critical state model [20]. Let's consider a cylindrical superconductor of radius R, placed in the magnetic field $\mathbf{H}_0$ parallel to its axis. The magnetic flux density inside the superconductor is

$$\textbf{rot}\,\mathbf{B} = 4\pi \mathbf{j}_c/c \tag{8}$$

where j_c is the critical current density. The Bean model supposes that j_c does not depend on magnetic field induction and it may have only three values 0, $\pm j_c$ inside the superconductor. Then the field distribution in the cylinder will be

$$B(r) = H_0 \pm 4\pi j_c(R-r)/c\,. \tag{9}$$

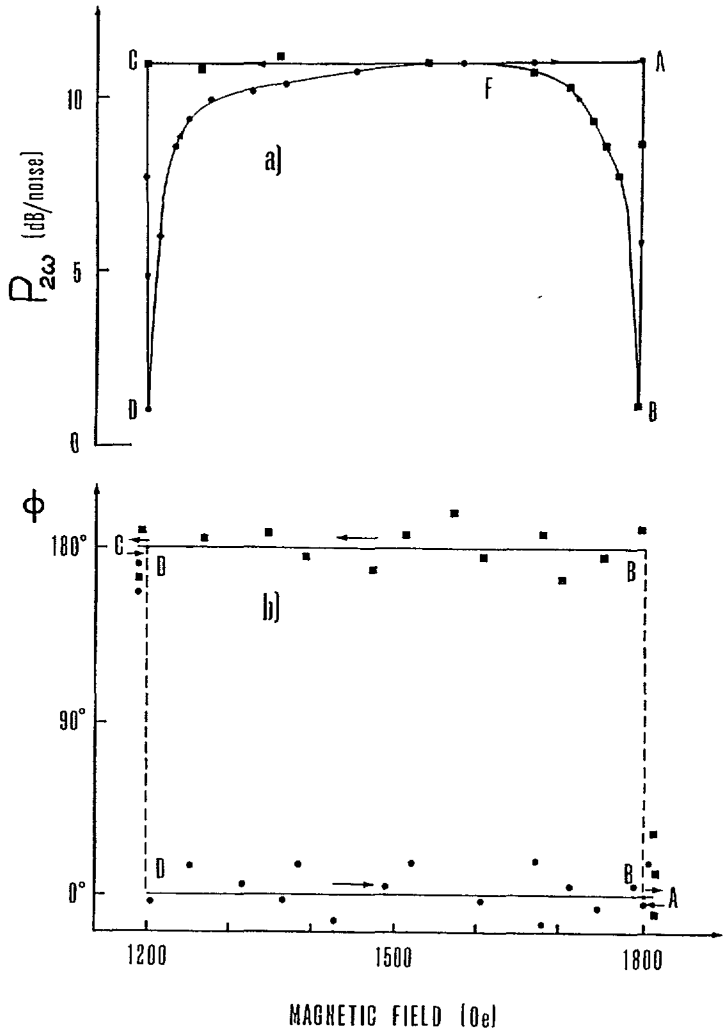

Fig. 10. — a) $P_{2\omega}$ signal as a function of the applied magnetic field in a ceramic YBaCuO sample, cooled down to the $T = 4.2$ K in the field $H > 1\ 000$ Oe ; $\mathbf{H} /\!/ \mathbf{H}_\omega$, $\omega/2\pi = 2$ GHz, $P_\omega \simeq 1.5$ W. b) Phase of the harmonic signal as a function of the applied magnetic field. Measurements performed at increasing field (•) and decreasing field (■) [5].

The negative sign in (9) corresponds to the external field H increase. If the external field is increased up, to the value H_0 ($H_0 > H_{c1}$), magnetic flux enters the sample. At $H = H_0$ critical state occurs, in which the Lorentz force is balanced by that of fluxons pinning on inhomogeneities, current density equaling j_c. In this case the average magnetic flux density is determined by the relation

$$\langle B \rangle = \int B \, \mathrm{d}V / \int \mathrm{d}V = H_0^2/H_* - H_0^3/3\, H_*^2 \tag{10}$$

where $H_* = 4\,\pi j_c/c$ — entire penetration field at which the whole sample is in critical state.

If then the external field is decreased from the H_0 value to some value of H, the current alters its direction at the sample surface, retaining the same direction inside and the average field will equal :

$$\langle B \rangle = H_0 H/H_* - (H^2 - H_0^2)/2 H_* - (H_0^3 + H^3/3 + H_0^2 H - H_0 H^2)/4 H_*^2 . \tag{11}$$

Now let's include the alternating field $\mathbf{H}_\omega \, /\!/ \, \mathbf{H}$. If the field modulation frequency is high, the superconductor response in critical state will change within the wave period.

Let's consider the sample in critical state arising in increasing field up to the value H_0. We shall refer to such a state as a direct critical state. Within the semiperiod when the field H_ω is positive, it is very difficult to change the induction flux in a sample. Indeed, as soon as the current density value is stable in critical state, the induction flux change may be caused only by the shift of the whole fluxons lattice so that the region with gradient of fluxons density will be increased. But due to short time of the wave semiperiod such an event is hardly probable. On the other hand during the semiperiod with the negative value H_ω, the flux density may change since the fluxons situated on the sample surface or in its vicinity can easily leave the sample.

The opposite situation will take place when the critical state is established in decreasing external field. Such a state will be called a reverse critical state.

The presented considerations are valid as long as the sample is in a critical state. If the latter is absent (at a very small field change the critical state may not occur at the sample surface due to the current redistribution) the sample responds equally both to the field increase and decrease.

Thus, the superconductor, being in direct or reverse critical state, acts as a « rectifier » of a high frequency field, and does not if the critical state has not been established. Moreover detection does not occur even in critical state if the frequency of alternating field is so small that the induction flux is able to follow the field alteration during the whole wave period.

Following this model for the sample in critical state arises in the external field increasing up to H_0 we suppose that in the sample the induction flux does not change during the positive high-frequency field semiperiod $H_\omega = H_\sim \sin(\omega t)$, while it does during the negative one. Then for the induction flux it is not difficult to obtain from (10), (11) :

$$\langle B \rangle = B(H_0) - H_\sim^2 \sin^2(\omega t)/2 H_* - H_\sim^3 \sin^3(\omega t)/12 H_*^2 \tag{12}$$

for $-\pi < \omega t < -\pi/2$ and

$$\langle B \rangle = B(H_0) + (H_\sim^2/H_* - H_\sim^3/4 H_*^2) \sin(\omega t) + \\ + (H_\sim^2/2 H_* - H_\sim^3/4 H_*^2) \sin^2(\omega t) - (H_\sim^3/12 H_*^2) \sin^3(\omega t) \tag{13}$$

for $-\pi/2 < \omega t < 0$. Finally $\langle B \rangle = B(H_0)$ at $0 < \omega t < \pi$.

If now Fourier transform of the function $\langle B(t) \rangle$ is fullfilled we can find the magnetization component, oscillating at the frequency 2ω at $H_\sim \ll H_*$ [5] :

$$M_{2\omega} \propto (H_\sim^2/3 \pi H_*) \cos(2 \omega t) + (H_\sim^2/6 \pi H_*) \sin(2 \omega t) . \tag{14}$$

Note that $M_{2\omega}$ depends on $H_\sim^2$. Though H_0 does not explicitly enter the expression (14) it is the field H_0 that provides the critical state and, consequently, harmonics generation effect in the sample.

According to (14) the expression for power at doubled frequency will have the form

$$P_{2\omega} \propto (M_{2\omega})^2 \propto (a_2 \cos 2 \omega t + b_2 \sin 2 \omega t) = 5 H_\sim^4 \cos^2(2 \omega t + \varphi)/36 \pi^2 H_*^2 \tag{15}$$

$$\operatorname{tg} \varphi = b_2/a_2 = 0.5 . \tag{16}$$

The similar calculations, performed in the case of reverse critical state result in the same expression (15) with

$$\operatorname{tg} \varphi = (-b_2)/(-a_2) = 0.5 . \tag{17}$$

Consequently the phase φ does not depend on external field as long as the superconductor is in the critical state (e.g. in direct one). At the transition to the reverse critical state the phase either remains the same or undergoes rapid change by 180°. The relations (15), (16) and (17) agree well with the experimental results.

Firstly the observed harmonic signal is proportional to the square of incident power, $P_{2\omega}$, as it follows from expression (15).

Secondly as figure 10a and (15) show the value $P_{2\omega}$ does not depend on the external field H as long as the H is monotonically increased (or decreased). If at the point A the sample is in direct critical state at the transition to the point B the generation drops because of changing field sweep direction. The same occurs at the transition from C to D. One can make conclusion that within these transitions critical state disappears and the difference of fields at points A and B defines the initial field H_{c1}^* of critical state establishment [11, 12]. In the experiment $H_{c1}^* \simeq 10$ Oe. The reverse critical state itself is completely formed at considerably large field changes from the point B to the point F.

Thirdly the phase measurement results in figure 10b correspond to conditions (16) and (17). The phase of signal $P_{2\omega}(H)$ in the given field H_0 depends on pre-history of H_0 establishment. The phase remains the same until the critical state is reversed. If the critical state reverses occurs, the harmonic signal phase alters by 180°.

Nonlinearity near T_c.

Let's consider maxima of $P_{2\omega}(T)$ and $P_{3\omega}(T)$ in the vicinity of superconducting transition temperature T_c (Figs. 1, 2).

THE 2nd HARMONIC GENERATION. — In this part we are reporting the $P_{2\omega}$ signal generation mechanism near T_c in a YBaCuO single crystal [21].

The sample had the dimensions $4 \times 4 \times 0.1$ mm^3, resistivity $\rho \simeq 50$ μΩ.cm at $T \simeq T_c$ and the superconducting transition width $\simeq 0.5$ K according to susceptibility measurements.

The temperature dependence of $P_{2\omega}(T)$ is shown in figure 11. In contrast to the results on ceramic YBaCuO samples (Figs. 1, 2) the $P_{2\omega}(T)$ and $P_{3\omega}(T)$ signals were observed only near T_c. All the rest measurements have been performed at the temperature $T \simeq T_c$ corresponding to the peak of maximum on the curve of $P_{2\omega}(T)$.

Figure 12 presents the dependence of $P_{2\omega}$ on the power P_ω incident on a YBaCuO single crystal. The same dependence has been obtained also for ceramic. In the whole of the P_ω change region the quadratic mode of $P_{2\omega} \propto P_\omega^2$ occurs, the latter basically differs from nonmonotonic dependence of $P_{2\omega}(P_\omega)$ in ceramic at $T < T_c$ and approximately within the similar interval of the P_ω change (Fig. 6).

Near T_c the dependence of $P_{2\omega}$ on weak external magnetic field (Fig. 13) is also different from the function type $P_{2\omega}(H)$ at $T < T_c$ (Figs. 4, 7). The curve of $P_{2\omega}(H)$ in figure 13 is of a hysteresis character both in a single crystal and ceramics.

A sample was cooled in the field $H = 0$ and the signal of $P_{2\omega}$ equalled zero. With increasing field the $P_{2\omega}$ grows rapidly and becomes saturated at $H \simeq 5$ Oe. At further magnetic field growth the $P_{2\omega}$ level remains practically the same. At the A point corresponding to $H = H_0$ (Fig. 13) the direction of a field sweep is switched over to the opposite one. At this moment the $P_{2\omega}$ signal drops and then decreases smoothly almost down to zero and only in a very weak field begins to grow. Then in the range of $-H_0 H < H < H_0$

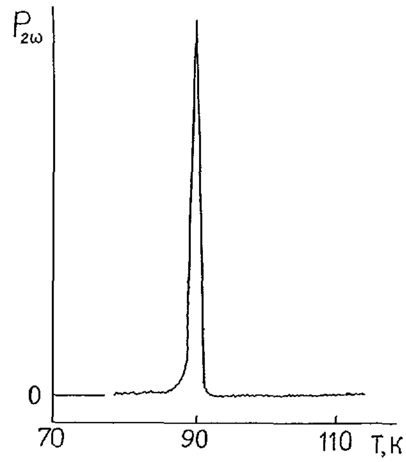

Fig. 11. — Dependence of the second harmonic $P_{2\omega}$ power on the temperature in a YBaCuO single crystal ; $\mathbf{H} /\!/ \mathbf{H}_{\omega}$, $H = 20$ Oe ; $H_{\sim} = 15$ Oe ; $\omega/2\pi = 9.4$ GHz.

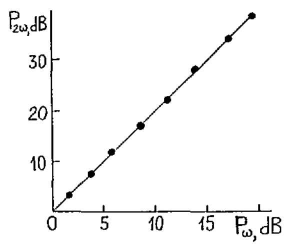

Fig. 12. — Dependence of the radiated power $P_{2\omega}$ on the power P_{ω} incident on the YBaCuO single crystal near $T \simeq T_c$; $\mathbf{H} /\!/ \mathbf{H}_{\omega}$, $H = 15$ Oe, $\omega/2\pi = 9.4$ GHz [21].

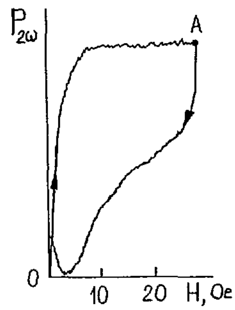

Fig. 13. — Plot of $P_{2\omega}(H)$ for the YBaCuO single crystal at $T \simeq T_c$; $H_{\sim} = 10$ Oe ; $\omega/2\pi = 9.4$ GHz. At the A point magnetic field sweep reverses its direction [21].

the $P_{2\omega}(H)$ curve does not vary and remains symmetric about the ordinate axis $H = 0$.

The measurements of the $P_{2\omega}(H)$ signal phase have demonstrated that the latter does not change within the whole $-H_0 < H < H_0$ region.

Thus, near T_c the superconductor microwave response at the doubled frequency is the same for both ceramics and single crystal. Probably the origin of this response is not connected with the Josephson generation mechanism occurring in weak fields at $T < T_c$.

To explain experimental results we use the Portis model [22, 11], which depicts low-field nonresonant absorption of high temperature superconductors in critical state [12].

According to this model a microwave response arises during the interaction of superconducting current with free or weakly pinned fluxons. Pinned fluxons do not contribute to the absorption. The equation of motion of a fluxon is

$$M \frac{\mathrm{d}V_\omega}{\mathrm{d}t} + \eta V_\omega + k\xi = \frac{1}{c} \cdot j_\omega \varphi_0 . \tag{18}$$

Here M is the fluxon mass per unit length, η is the fluxon viscous damping constant, k is the force constant, restraining fluxon displacement, j_ω is the superconducting current density, ξ is the displacement and φ_0 is the flux quantum.

Let the incident wave have temporary dependence $\exp(- i\omega t)$. Then the fluxon velocity amplitude will equal

$$V_\omega = \frac{j_\omega \varphi_0}{c} / (- i\omega M + \eta + ik/\omega) . \tag{19}$$

The moving fluxon creates an electric field at frequency ω

$$E_\omega^\varphi = - V_\omega fH/c \tag{20}$$

where f is the fraction of free or weakly pinned fluxons. Under the experiment conditions in [22] $f \simeq 0.1$.

In its turn the field (20) together with the applied microwave field $\mathbf{E}_\omega$ define the vector potential $\mathbf{A}_\omega$ and self-consistently the $\mathbf{j}_\omega$ current from the London equation :

$$\mathbf{j}_\omega = - (c\mathbf{A}_\omega)/(4\pi\lambda_{\mathrm{mw}}^2) , \quad \mathrm{d}\mathbf{j}_\omega/\mathrm{d}t = c^2(\mathbf{E}_\omega + \mathbf{E}_\omega^\varphi)/(4\pi\lambda_{\mathrm{mw}}^2) \tag{21}$$

where λ_{mw} is the depth of microwave field penetration.

By means of Maxwell equations the expression for a surface impedance can be found out [22] with the use of the j_ω current and E_ω field connection (21).

Taking into account a magnetic field H_ω of the microwave the second harmonic of a fundamental frequency will occur in the spectrum of an electric field created by a fluxon

$$E_{2\omega}^\varphi \propto fH_\omega V_\omega . \tag{22}$$

From the London equation (21) we shall obtain the expression for « nonlinear source » [23] at the doubled frequency :

$$\mathbf{j}_{2\omega}^{\mathrm{nL}} = ic^2 \mathbf{E}_{2\omega}^\varphi/(8\omega\pi\lambda_{\mathrm{mw}}^2) . \tag{23}$$

This source defines the radiated power, registered in the experiment

$$P_{2\omega} \propto \left| \int \mathbf{j}_{2\omega}^{\mathrm{nL}}(z) \cdot \mathbf{E}_{2\omega}(z)\,\mathrm{d}z \right|^2 \tag{24}$$

where $\mathbf{E}_{2\omega}(z)$ describes the field dependence on the z coordinate at the doubled frequency in linear approximation. Axis z is directed into volume of a superconductor.

It follows from (22), that the nonlinear current (23) is proportional to the fraction of free (or weakly pinned) fluxons which is defined by the module of the field H gradient. The free fluxons concentration averaged with regard to microwave field penetration equals

$$\langle f \rangle \propto \int_0^{\infty} \left| \frac{\mathrm{d}H}{\mathrm{d}z} \right| \exp(-3\, z/\lambda_{\mathrm{mw}})\, \mathrm{d}z \tag{25}$$

and varies as a function of the $H(z)$ field distribution in a sample. The Portis model proposes the following :

1. The rising magnetic field $\mathbf{H}_0$, parallel to the sample surface, penetrates inside a sample according to the exponential law $H(z) = H_0 \exp(-z/\lambda)$ (λ is the depth of a static field penetration) up to $H_{\mathrm{cl}}^* = 4\, \pi j_{\mathrm{c}}\, \lambda/c$ (j_{c} is the critical current).

2. If magnetic field rises later field gradient at a sample surface remains constant and equals $4\, \pi j_c/c = H_{\mathrm{cl}}^*/\lambda$ (the Bean critical state).

The field decreases linearly

$$H(z) = H_0 - zH_{\mathrm{cl}}^*/\lambda$$

at $z < z_1$ and exponentially

$$H(z) = H_0 \exp[-(z - z_1)/\lambda]$$

at $z > z_1$, where $z_1 = \lambda\, (H_0 - H_{\mathrm{cl}}^*)/H_{\mathrm{cl}}^*$.

3. Let's assume that the applied magnetic field, after having reached some maximal value of H_{m}, starts to decrease. According to [24] within the interval $H_{\mathrm{m}} - 2\, H_{\mathrm{cl}}^* \leqslant H_0 \leqslant H_{\mathrm{m}}$ the dependence of $H(z)$ has a form :

$$H(z) = H_{\mathrm{m}} - (H_{\mathrm{m}} - H_0) \exp(-z/\lambda) - zH_{\mathrm{cl}}^*/\lambda$$

so

$$\left| \frac{\mathrm{d}H}{\mathrm{d}z} \right| = \frac{1}{\lambda} [H_{\mathrm{cl}}^* - (H_{\mathrm{m}} - H_0) \exp(-z/\lambda)]$$

at $H_{\mathrm{m}} - H_{\mathrm{cl}}^* < H_0 \leqslant H_{\mathrm{m}}$ and at $H_{\mathrm{m}} - 2\, H_{\mathrm{cl}}^* \leqslant H_0 \leqslant H_{\mathrm{m}} - H_{\mathrm{cl}}^*$

$$\left| \frac{\mathrm{d}H}{\mathrm{d}z} \right| = \begin{cases} (H_{\mathrm{m}} - H_0) \exp(-z/\lambda)/\lambda - H_{\mathrm{cl}}^*/\lambda & 0 \leqslant z \leqslant z_2 \\ H_{\mathrm{cl}}^*/\lambda - (H_{\mathrm{m}} - H_0) \exp(-z/\lambda)/\lambda & z > z_2 \end{cases}$$

where $z_2 = \lambda \ln [(H_{\mathrm{m}} - H_0)/H_{\mathrm{cl}}^*]$.

4. In the fields $H_0 < H_{\mathrm{m}} - 2\, H_{\mathrm{cl}}^*$ a reverse critical state develops at a sample surface and field distribution is

$$H(z) = \begin{cases} H_0 + zH_{\mathrm{cl}}^*/\lambda & z \leqslant z_3 \\ H_{\mathrm{m}} - 2\, H_{\mathrm{cl}}^* \exp[-(z - z_3)/\lambda] - (z - z_3)\, H_{\mathrm{cl}}^*/\lambda & z > z_3 \end{cases}$$

where $z_3 = \lambda\, (H_{\mathrm{m}}/H_{\mathrm{cl}}^* - H_0/H_{\mathrm{cl}}^* - 2)$.

The reduced expressions for $H(z)$ enable to calculate the value of $\langle f \rangle$ from (25) in the entire interval of the external magnetic field change.

The measured dependences of $P_{2\omega}(H)$, $P_{2\omega}(P_{\omega})$ and $P_{2\omega}(T)$ are determined by the integral (24). Both factors in the integral expression change in changing magnetic field. In contrast with the intensity of the 2nd harmonic the impedance has the nonzero value [25] in a zero field. Therefore linear electrodynamic characteristics of YBaCuO may be considered independent on a field value in sufficiently weak fields. Hence the basic dependence of the

$P_{2\omega}$ signal on an external field is determined by the function $\mathbf{j}_{2\omega}^{\mathrm{nL}}(z, H)$ in (24). Taking into account (22), (23) one can consider that in weak fields

$$P_{2\omega} \propto P_{\omega}^{2} \cdot \langle f \rangle^{2} \tag{26}$$

This conclusion is confirmed by the phase measurements of a harmonic signal. In accordance with (26) the $P_{2\omega}$ phase does not depend on a magnetic field.

The proportionality of the radiated $P_{2\omega}$ power to the square of the incident power from (26) is confirmed experimentally as well (Fig. 12).

Let's consider the curve $P_{2\omega}(H)$ in figure 13. The quick growth and saturation of the observed signal, as an applied field increases, correspond to the establishment of the critical current in a sample skin-layer. When switching over field sweep direction the current value drops retaining the same sign. This leads to abrupt decrease of the $P_{2\omega}$ signal.

This part of the $P_{2\omega}(H)$ curve is well described by the correlation (26), in which the $\langle f \rangle$ function was computed from the formulae (25) with the field distribution $H(z)$, indicated at points 1 to 3. The only fitting parameter was the ratio $\lambda_{\mathrm{mw}}/\lambda$. For the curve in figure 13 the best coincidence was obtained at $\lambda_{\mathrm{mw}}/\lambda \simeq 1, 5$.

The final part of the curve obtained in decreasing field applied (left of the arrow in Fig. 13) corresponds to the establishment of a reverse critical state, when the current in a skin-layer changes its sign. One can not describe it by means of the formula (26).

Note that the attempt to reproduce the hysteresis loop shape can hardly be reasonable because of the necessity to account for a specific sample shape.

The temperature dependence of $P_{2\omega}(T)$ in a YBaCuO single crystal (Fig. 11) is primarily defined by strong dependence of an impedance on temperature. When temperature is decreased below T_{c} the impedance drops abruptly, diminishing wave interaction space in a superconductor, i.e. the integration region in (24). In YBaCuO single crystal the impedance is 50 times less than in ceramic (at $T \simeq 77$ K) [26], which takes the nonlinear effects far from T_{c} practically unobservable in single crystals.

THE 3d HARMONIC GENERATION. — The magnetic field dependence of the microwave response of a YBaCuO at the doubled frequency was explained in previous part. As it has been shown in [6] the $P_{3\omega}$ response at $T \simeq T_{\mathrm{c}}$ does not depend on magnetic field.

The experimental temperature dependence of $P_{3\omega}(T)$ near T_{c} of YBaCuO single crystal (the same crystal has been employed as that in paper [21]) is shown by the solid line in figure 14 [6]. Note that the $P_{3\omega}(T)$ signal is asymmetric. We observed the same $P_{3\omega}(T)$ line shape near T_{c} in other YBaCuO single crystals. The $P_{3\omega}(T)$ signal did not change when we applied a static magnetic field in the direction parallel to the surface of the sample with a strength of up to 7 kOe. It was established experimentally that the amplitude of the alternating field corresponding to that recorded in figure 14 ($H_{\sim}^{\omega} = 5$ Oe) is within the interval for which the amplitude of the third harmonic field, $H_{\sim}^{3\omega}$, is proportional to the cube of the field at the fundamental frequency :

$$H_{\sim}^{3\omega} \propto (H_{\sim}^{\omega})^{3} . \tag{27}$$

This situation means that one can use perturbation theory to describe the interaction of an intense electromagnetic field with the superconductor.

When studying nonstationary processes in superconductors in the vicinity of T_{c} an important question arises concerning inertial properties of the order parameter Δ and of the relaxation frequency Ω_0 which is characteristic of Δ. This question cannot be resolved on the basis of linear electrodynamics, in which Ω_0 does not arise, and another frequency, Ω_1, is important. The latter frequency separates the region in which the field penetrates into

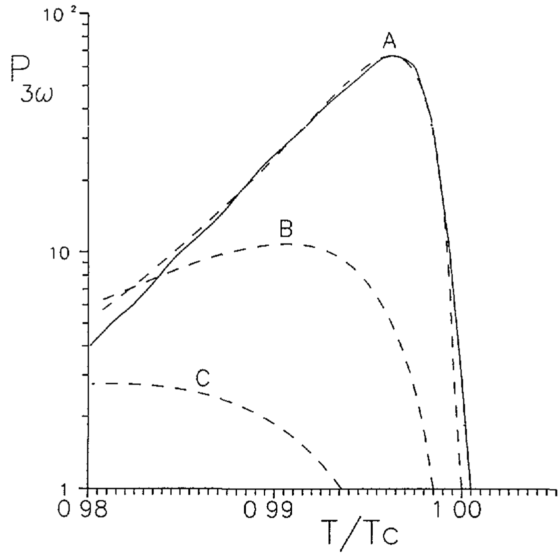

Fig. 14. — The third harmonic signal $P_{3\omega}$ *versus* the temperature T/T_c. Solid line : experimental ; dashed lines : theoretical for various values of $\omega\tau$; a) $\omega\tau = 0.003$, b) 0.008 ; c) 0.015 [6].

superconductor as a result of a skin effect from the region of the Meissner effect. The nonlinear response of a superconductor to an r.f. field, on the other hand, depends on both of these frequencies, Ω_0 and Ω_1. Gor'kov and Eliashberg showed that the relation $\Omega_0 = \Omega_1$ holds in a superconductor having a high concentration of paramagnetic impurities [27]. This situation was studied experimentally by Amato and McLean [28], who found the relaxation time Ω_0^{-1} near the critical temperature T_c from the lineshape of the nonlinear response of an $La_{1-x}Gd_xSn_3$ alloy.

In YBaCuO single crystals, as it will be shown below, $\Omega_0 < \Omega_1$ and the curve $P_{3\omega}(T)$ in figure 14 cannot be described by the theory derived in, for example, references [27, 29] on the basis of the BCS model.

A more appropriate approach for the high T_c superconductors starts from Eliashberg's equations [30]. From these equations we find for the frequencies the following expressions, which are quite accurate near T_c [6] :

$$\Omega_0 \simeq 6\,T_c(1+\lambda)\left(1-\frac{T}{T_c}\right), \quad \Omega_1 \simeq \frac{2}{\tau(1+\lambda)}\left(1-\frac{T}{T_c}\right) \tag{28}$$

where $\lambda = \lambda(0)$ is a coupling constant, $\tau = m^*/\rho n e^2$, ρ is the resistivity, n is the density of holes, and m^* is the band mass of the holes.

For a numerical analysis of the Eliashberg's equations, we selected the model of a phonon spectrum of a YBaCuO consisting of three Einstein oxygen modes [31] with frequencies of 200, 600, and 1 000 K. As a result, we found both the correct value of T_c (at $\lambda = 1, 4$) and the behavior $P_{3\omega}(T)$ shown by dashed line A in figure 14 which is the best fit of the experimental curve.

Generally speaking the dependence $P_{3\omega}(T)$ is very complicated but schematically it may be presented as follows :

$$P_{3\omega}(T) \propto \frac{t^2\ .}{(\Omega_1^2+\omega^2)^4\,.\,(\Omega_0^2+\omega^2)} \tag{29}$$

where $t = (T_c - T)/T_c$. According to (28) the relation

$$\Omega_0/\Omega_1 \simeq 3(1 + \lambda^2)\,\tau T_c \gg 1$$

does not depend on temperature and is determined by single crystal parameters. It is clear from (29) that the $P_{3\omega}(T)$ peak width in the vicinity of T_c is determined by the frequency Ω_1. And indeed, changing the parameter τ, we obtained practically complete coincidence of the experimental curve and that calculated from the Eliashberg's equations. Corresponding to line A in figure 14 is the parameter value $\omega\tau = 0.003$. With $\omega/2\pi = 9.4$ GHz and $\rho = 50$ μΩ.cm we find the ratio $m^*/nm_0 = 0.8 \times 10^{-21}\ \mathrm{cm}^{-3}$ (for $\lambda = 1.4$). From (28) we find

$$\Omega_0 \simeq 1.8 \times 10^{14}\left(1 - \frac{T}{T_c}\right), \quad \Omega_1 \simeq 1.6 \times 10^{13}\left(1 - \frac{T}{T_c}\right).$$

Lines B and C in figure 14 correspond to $\omega\tau$ values of 0.008 and 0.015.

An alternative to the generation mechanism discussed above might be a mechanism based on a strong dependence of the impedance of the sample on the strength of the external magnetic field, $Z(H)$. A manifestation of this mechanism would be most understandable in type-I superconductors [32], in which the resultant applied field $(H + H_\omega)$ is comparable to the critical field H_c, and the sample is in either the normal state or the superconducting state during a period of the incident wave. If the dependence $Z(H)$ involves a threshold, the reflected signal should have a nonharmonic shape, so a response should arise at all multiple frequencies with an amplitude proportional to H_ω. This conclusion does not correspond to experimental result (27). Furthermore, if the observed nonlinear effect were due exclusively to a $Z(H)$ dependence, then the amplitude of the $P_{3\omega}$ signal would vary with the strength of the external field, in contradiction of experiment.

We thus believe that the nonlinear response of a YBaCuO single crystal near T_c is a consequence of a time variation of the order parameter Δ under the influence of the r.f. field. The measured relaxation time Δ is $\Omega_0^{-1} \simeq 5.6 \times 10^{-15}(1 - T/T_c)^{-1}$

Conclusion.

A few of the first steps have been made in the study of high temperature superconductors nonlinear microwave electrodynamics. In the nearest future the experiments will probably be carried out in a large interval of frequencies and fields, and also on different subjects : films, powders, single crystals, high temperature superconductors of another compositions. The up-to-date comprehension level is well characterized in paper [17] : « The microscopic models presented here are sufficient to explain much of our data, but we have yet to show that they are also necessary ; simple empirical or phenomenological models may also suffice. »

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Proofs not corrected by the authors.