

# Response of granular superconductors to large amplitude microwave fields

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**Abstract.** We calculate the impedance of a granular superconductor for different levels of microwave field using a macroscopic nonlinear model. In this model Josephson junctions between grains are responsible for high-frequency current and field characteristics. The change in surface impedance at the frequency of weak amplitude wave, when perturbed by a strong amplitude wave of different frequency, is also calculated. Application of the obtained nonlinear properties to experimental data is discussed.

## 1. Introduction

It is now generally accepted that a granular superconductor should be considered as a Josephson medium, where superconducting grains are coupled by weak junctions. Josephson media were considered theoretically before the discovery of HTSC [1]. This approach was later applied to describing different properties of HTSC: critical state [2], linear microwave response [3], nonlinear response near  $T_c$  [4]. In a number of papers the Josephson medium is described in terms of circuit theory, using notions of specific resistance and inductance [5-9]. This approach enables one to describe many features of the microwave properties of HTSC.

In this paper we calculate the microwave surface impedance of the Josephson medium over a wide range of amplitudes of a high-frequency field using a variant of macroscopic equations from reference [10].

## 2. Basic equations

First, we outline the derivation of the macroscopic equations following [10]. Consider a three-dimensional medium consisting of superconducting grains with Josephson junctions between them.

The current through the  $k$ th junction, which has resistance  $R_k$  may be written as [11]

$$i_k = I_k \sin \phi_k + \frac{\Phi_0}{2\pi c} R_k \frac{d\phi_k}{dt} \quad (1)$$

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where  $\phi_k$  is the phase difference at the junction,  $I_k$  is the critical current,  $\Phi_0 = \pi \hbar c / e$  is the quantum of magnetic flux. In reference [10] it was argued that the phase difference  $\phi_k$  may be expressed through a macroscopic vector potential  $A$  in the following way

$$\phi_k = -\frac{2\pi}{\Phi_0} A a_k + 2\pi m_k \quad (2)$$

where  $m_k$  is an integer and  $a_k$  is a vector between centres of adjacent grains. Equation (3) is consistent with the quantization of magnetic flux

$$\frac{2\pi}{\Phi_0} \oint A ds + \sum_k \phi_k = 2\pi m \quad (3)$$

where the closed contour passes through Josephson junctions [12]. In writing equation (3) we have chosen some calibration of the vector potential.

Consider a volume  $V$  containing many junctions, but small enough to consider the vector potential  $A$  as a constant. Average current density in this volume is

$$j = \frac{\sum_l^N i_l a_l}{V} = \rho \langle i_l a_l \rangle \quad (4)$$

where  $\rho$  is the concentration of Josephson junctions. Assume that  $a_k$ ,  $I_k$ ,  $R_k$  are independent random variables with

$$\langle I_k \rangle = I_j \quad \langle R_k \rangle = R.$$

Also, we assume that all directions of  $a_k$  are equally probable and its modulus is distributed with probability distribution function  $W(a)$ . After averaging we obtain

$$j = -I_j \rho A G(A^2) - \frac{dA}{dt} \frac{\pi a_0^2 \rho}{8Rc}. \quad (5)$$

Here  $a_0 = \langle a_k \rangle$  and

$$G(A^2) = \frac{\Phi_0^2}{4\pi^2 A^3} \int_0^\infty W\left(\frac{\Phi_0 x}{2\pi A}\right) x^{-1} (\sin x - x \cos x) dx.$$

In particular, in reference [10] the Maxwell distribution function was used

$$W(a) = \frac{32}{\pi^2 a_0^3} a^2 \exp\left(-\frac{4a^2}{\pi a_0^2}\right)$$

which gives

$$G(A^2) = \frac{\pi^2 a_0^2}{\Phi_0} \exp\left(-\frac{\pi^3 a_0^2}{4\Phi_0^2} A^2\right). \quad (6)$$

In the stationary case, current density is limited by the value  $I_j \rho a_0 \sqrt{(\pi/16e)}$ . In the next section we will use this particular form of nonlinear current-field characteristic. Some generalizations will be discussed in section 4.

Substituting expressions (5) and (6) in the Maxwell equation (we neglect displacement current here)

$$\text{curl curl } A = \frac{4\pi\mu j}{c}$$

(where the magnetic permeability accounts for Meissner currents of individual grains) we obtain

$$\text{curl curl } A = -\frac{1}{\lambda_j^2} A F(A^2) - \frac{\tau}{\lambda_j^2} \frac{dA}{dt} \quad (7)$$

where

$$F(A^2) = \exp(-A^2 \lambda_j^{-2} B_j^{-2}).$$

Here the quantities

$$\lambda_j^2 = \frac{c\Phi_0}{I_j \rho \pi^3 \mu a_0^2} \quad \text{and} \quad B_j^2 = \frac{4\Phi_0 I_j \rho \mu}{c}$$

have the physical meaning of penetration depths of a weak static magnetic field in the granular medium and the first critical current respectively [10]. The characteristic time scale

$$\tau = \frac{\lambda_j^2 \pi^2 a_0^2 \rho \mu}{2Rc^2}$$

will be assumed to be much less than the period of the microwave field.

### 3. Nonlinear surface impedance

In many experiments on microwave losses in a granular superconductor, a sample (ceramic or film) is part of a resonant cavity wall. It is not difficult to find the amplitude of the microwave field on the surface of the sample if input power and total losses in the cavity are known. Thus, we can study the problem assuming that

the magnetic field at the surface of the superconductor is given.

Consider a halfspace  $x \geq 0$  filled with the granular superconductor. We assume that all fields depend on the coordinate  $x$  only, magnetic field is directed along  $y$  and the vector potential  $A$  has only  $z$  component  $A(x)$ . The problem is in solving equations (6) and (7) with boundary conditions

$$\left. \frac{dA}{dx} \right|_{x=0} = -H_0 \cos \omega t \quad A|_{x \rightarrow \infty} = 0. \quad (8)$$

Consider first the linear case  $H_0 \ll B_j$ . We can set  $F(A^2) = 1$  in (7) and with the usual substitution

$$A = \frac{1}{2}(A_0 \exp(ikx - i\omega t) + \text{c.c.}) \quad (9)$$

obtain

$$k(\omega) = -(\lambda_j^{-2} - i\omega\tau\lambda_j^{-2})^{1/2} \approx \lambda_j^{-1}(i + 1/2\omega\tau). \quad (10)$$

One can see from equation (10) that for the static field ( $\omega = 0$ )  $\lambda_j$  is just the penetration length. From the solution of (9) and (10) the linear surface impedance  $\zeta_l$  is easily expressed:

$$\zeta_l(\omega) \approx \frac{\omega^2 \lambda_j \tau}{2c} - i \frac{\omega \lambda_j}{c} \quad (11)$$

In the nonlinear case we have to solve nonlinear equation (7). We will assume field  $A$  to be nearly sinusoidal and apply the harmonic balance method. Representing the vector potential in the form

$$A(x, t) = \frac{1}{2}(A_0(x) e^{i\omega t} + \text{c.c.}) \quad (12)$$

and substituting this in equation (7), we get from the main harmonic balance condition

$$i\omega\tau A_0 = \lambda_j^2 \frac{d^2 A_0}{dx^2} - A_0 f(|A_0|^2 \lambda_j^{-2} B_j^{-2}) \quad (13)$$

where

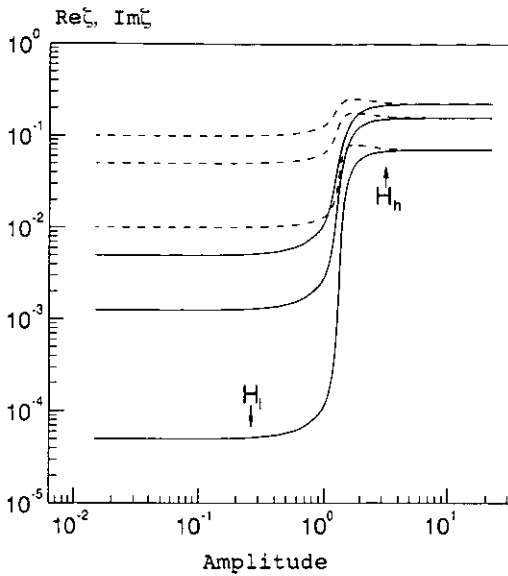
$$f(x) = e^{-x/2}(I_0(x/2) - I_1(x/2)).$$

$I_0$  and  $I_1$  are modified Bessel functions. Thus we have reduced the partial differential equation (7) to the ordinary differential equation (13), which can be easily solved numerically with the boundary condition  $dA_0(0)/dx = H_0$ . The surface impedance resulting from this solution is given by

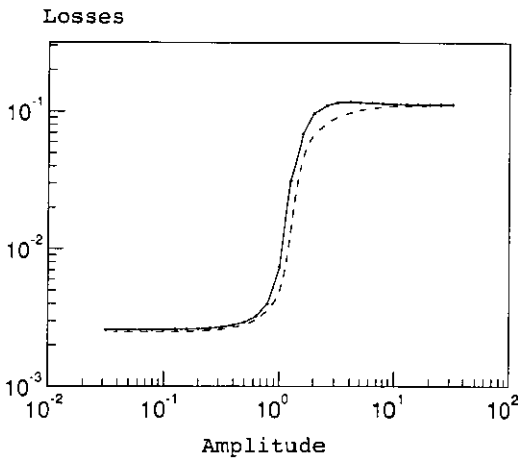
$$\zeta = \frac{E(0)}{H(0)} = \frac{i\omega A_0(0)}{cH_0}$$

and is presented in figure 1. One can see that the real part of  $\zeta$ , which is responsible for losses, is approximately a step function

$$\text{Re}\zeta(H_0) \approx \begin{cases} \text{Re}\zeta_l(\omega) & H_0 \ll B_j \\ \text{Re}\zeta_0(\omega) & H_0 \gg B_j \end{cases} \quad (14)$$



**Figure 1.** Real (full curve) and imaginary (broken curve) parts of the surface impedance versus amplitude of microwave field  $H_0$  (in units of  $B_j$ ). From bottom to top:  $\omega\tau = 0.01; 0.05; 0.1$ .



**Figure 2.** The total losses in the bulk of the superconductor (arbitrary units) versus boundary field amplitude. Broken curve—harmonic balance method; full curve—solution of the full partial differential equation;  $\omega\tau = 0.1$ .

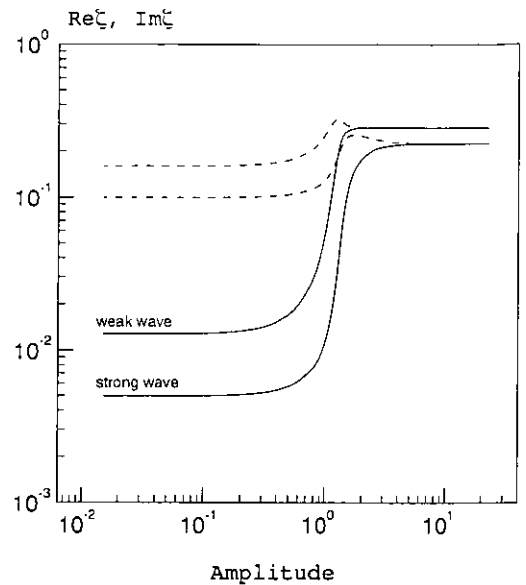
where

$$\zeta_0(\omega) = \frac{\lambda_j \omega^{1/2} i + 1}{c\tau^{1/2} 2}$$

is the linear impedance in the absence of Josephson currents ( $I_k \equiv 0$ ).

In order to check the validity of the harmonic balance method in our problem, we compared the solution of (13) with that of the full equation (7) for one particular frequency  $\omega$  (see figure 2). There is some discrepancy in the data in the region of moderate external fields, where it is possible that higher harmonics are important. Nevertheless, for large fields equation (13) gives a good approximation to solutions of (7), at least for estimating losses.

In some experiments losses were measured, not for the large amplitude microwave field, but for a small



**Figure 3.** The real (full curve) and imaginary parts of the surface impedance for a small amplitude wave with  $\Omega\tau = 0.16$  versus amplitude of microwave field with  $\omega\tau = 0.1$ .

amplitude test field with frequency different from that of a large amplitude field [13]. This case may be treated in the same way as before, if, instead of (12), we take

$$A(x, t) = \frac{1}{2}(A_0(x)e^{i\omega t} + B_0(x)e^{i\Omega t} + c.c.)$$

where  $|B_0| \ll |A_0|$ . Again, using the harmonic balance method, we obtain equation (13) for  $A_0$ . For  $B_0$  we obtain

$$i\Omega\tau B_0 = \lambda_j^2 \frac{d^2 B_0}{dx^2} - B_0 g(|A_0|^2 \lambda_j^{-2} B_j^{-2}) \quad (15)$$

where

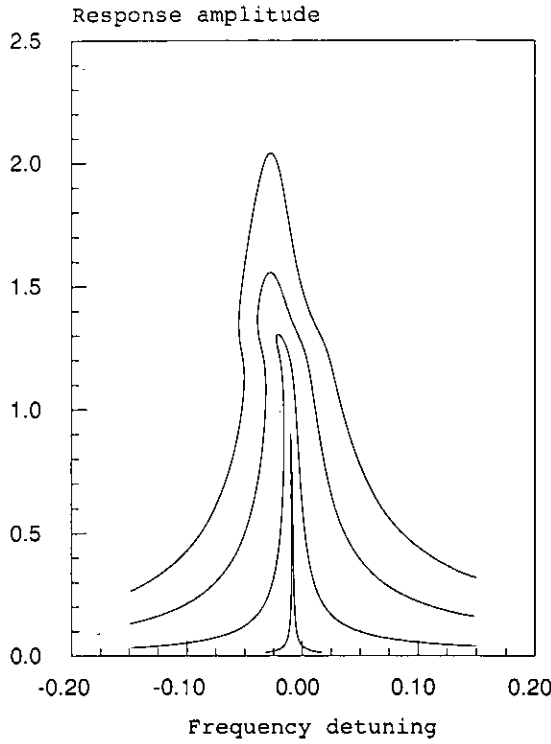
$$g(x) = \exp(-x/2)(I_0(x/2) + xI_1(x/2)).$$

The solutions of equation (15), presented in figure 3, give surface impedance similar to figure 1.

Nonlinear dependence of real and imaginary parts of the surface impedance of the superconductor on the amplitude of the microwave field implies nonlinear properties of the resonant cavity [14]. Because impedance is small, shift of the resonant frequency is small and in the first approximation we may write the equation describing the amplitude–frequency dependence in the form

$$C = I \left[ \omega_0^2 - \omega^2 - \frac{\omega_0^3}{\pi\omega} \text{Im} \zeta(|C|^2 \omega_0^2 \omega^{-2}) + i \frac{\omega_0^3}{\pi\omega} \text{Re} \zeta(|C|^2 \omega_0^2 \omega^{-2}) \right]^{-1}. \quad (16)$$

Here  $C$  is the amplitude of the field in the cavity, excited by the force, proportional to  $I$ ;  $\omega_0$  is the resonant frequency of the ideal cavity (where all walls are ideal conductors). We neglect here the losses due to non-superconducting walls. The resonant curves  $C$  against



**Figure 4.** Nonlinear resonance curves according to equation (16);  $\omega\tau = 0.05$ .

$\omega$  are presented in figure 4. These curves are skewed in a complicated manner, due to complicated dependence of  $\text{Im } \zeta$  on the field amplitude. The main effect of nonlinearity is in the drastic increase of resonance width because of a strong dependence of  $\text{Re } \zeta$  on the microwave field.

#### 4. Application to experiment

There are many references where the surface impedance of granular superconductors in a high-amplitude microwave field has been investigated experimentally. There are some on low-temperature superconductors, in particular on the films of NbTiN [15], but many more on high-temperature superconductors. Nonlinear microwave response of YBaCuO samples has already been mentioned in initial experiments. In references [16, 17] dependence of impedance for ceramic samples on the field amplitude was presented, while in some references harmonics of the basic frequency were investigated (see [18]). Most thoroughly investigated has been the nonlinear dependence of the surface impedance on the amplitude for films [19–24]. In all these experiments a cavity or a stripline resonator was used and their resonant frequency and quality factor were measured as functions of external field. For higher sensitivity one can use a two-frequency method with a sample being placed in a bimodal cavity [13]. Using pulsed large amplitude waves and continuous small amplitude waves it is possible to measure how a large wave produces small changes of the impedance at small-wave frequency (cross-modulation).

The recent results show that the granular structure of superconductors plays a crucial role in the nonlinear response. However, it is difficult to extract universal properties for all samples. We can say that the nonlinear dependences of impedance on the microwave field with frequency 821 MHz, as given in reference [19], for a large number of granular samples, are qualitatively similar to those presented in figure 1. In order to obtain quantitative agreement as well, we must modify equation (5), which is not valid for films. It is more appropriate to use a model where all vectors  $\mathbf{a}$  are in the film's plane and there are no Josephson currents in the transverse direction. This model is described with probability distribution function

$$W(\mathbf{a}) = \delta(a_x/\Delta)w(r) \quad (17)$$

where  $\Delta$  is the characteristic width of the film and  $\delta$  is Dirac's function,  $r = (a_y^2 + a_z^2)^{1/2}$ . After averaging with distribution function (17) we obtain, instead of (6), a hypergeometric function [25]

$$F(A^2) \sim {}_1F_1(2, 2; -\frac{\pi^3 a_0^2 A^2}{4\Phi_0^2}).$$

Qualitatively, this function is similar to that of (6).

The width of the transitional region from low to high fields depends mainly on the precise form of the nonlinear current function  $F(A^2)$ . From figure 1  $H_b/H_1 \approx 10$  ( $H_b$  and  $H_1$  are amplitudes for which one can use approximation (14) (see figure 1), while in the experiments [19] this quantity is  $10^2$ – $10^3$ . Using other distribution functions of the type (6) (in particular,  $F(A^2) = (1 + A^2 \lambda_j^{-2} B_j^{-2})^{-1}$ ) this ratio may be increased up to 30–40.

It is worth noting that epitaxial films show very small nonlinearity, their real part of the surface impedance remains almost constant up to amplitudes of 100 Oe [20] and larger [22].

We also mention here that it is reasonable to compare experiments with the screened earth's magnetic field. This is important because although microwave magnetic fields influence surface resistance much more strongly than static ones, the latter may produce hypervortices [26], whose pinning may drastically change the equilibrium properties.

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#### References

- [1] Rosenblatt J et al 1988 *Physica B* 152 95

- [2] Dersch H and Blatter G 1988 *Phys. Rev. B* **38** 11391
- [3] Sonin E B and Tagantsev A K 1989 *Sov. Phys. JETP* **68** 572
- [4] Ivanchenko Yu M, Lisiansky A A and Tsindlekht M I 1990 *Sov. Phys. JETP* **70** 187
- [5] Hylton T L *et al* 1988 *Appl. Phys. Lett.* **53** 1343
- [6] Vendyk O G, Kozyrev A B and Popov A Yu 1990 *Rev. Phys. Appl.* **25** 255
- [7] Portis A M 1989 *Microwaves and Superconductivity Processes in the Intergranular Medium* ed J G Bednorz and K A Müller (Springer Series in Solid State Sciences) (Berlin: Springer)
- [8] Portis A M and Cooke D W 1992 *Supercond. Sci. Technol.* **5** 395
- [9] Bonin B and Sofa H 1991 *Supercond. Sci. Technol.* **4** 257
- [10] Belodedov M V and Ignatjev V K 1990 *Supercond. Phys. Chem. Technol.* **3** S24
- [11] Licharev K K 1986 *Dynamics of Josephson Junctions and Circuits* (New York: Gordon and Breach)
- [12] Schmidt V V 1982 *Introduction to the Physics of Superconductors* (Moscow: Nauka) (in Russian)
- [13] Koshelev A E, Leviev G I and Papikjan R S *Sov. Phys. JETP* (at press)
- [14] Halbritter J 1970 *J. Appl. Phys.* **41** 4581
- [15] Bosland P, Guemas F and Juillard M 1990 *ICM AS 90 Conf., Grenoble* ed A Niku-Lari
- [16] Bielsky M, Vendik O G, Gaidukov M M, Golman E K, Karmanenko S F, Kozyrev A B, Kolesov S G and Samoiloova T B 1987 *JETP Lett. Suppl.* **46**
- [17] Rezende S M and de Aguiar F M 1989 *Phys. Rev. B* **39** 9715
- [18] Trunin M R and Leviev G I 1992 *J. Phys. III (France)* **2** 355
- [19] Delayen J R, Bohn C L and Roche C T 1990 *J. Supercond.* **3** 243
- [20] Hein M *et al* submitted to *J. Less-Common Metals*
- [21] Hammond R B, Negrete G V, Bourne L C, Strother D D, Cardona A H and Eddy M M 1990 *Appl. Phys. Lett.* **57** 825
- [22] Oates D E, Anderson A C, Sheen D M and Ali J M 1991 *IEEE Trans. on Microwave Theory and Techniques* **39** 1522
- [23] Portis A M *et al* 1991 *Appl. Phys. Lett.* **58** 307
- [24] Cooke D W *et al* 1991 *Appl. Phys. Lett.* **58** 1329
- [25] Gradshteyn and Ryzhik 1981 *Tables of Series, Products and Integrals* (Frankfurt am Main: Harri Deutsch)
- [26] Sonin E B 1989 *JETP Lett.* **47** 496