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Microwave propagation through superconductor-polymer composites

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We report on microwave and low-frequency properties of composites consisting of $YBa_2Cu_3O_{7-x}$ (YBCO) or $Bi_2Sr_2CaCu_2O_x$ grains embedded in a polymer matrix with strong emphasis on microwave transmission experiments through YBCO-polymer composites. The low-frequency conductivity vs the YBCO volume fraction f can be described by a three-dimensional percolation model with a percolation threshold of $f_p \cong 0.2$. The microwave phase velocity and the transmission coefficient are significantly larger in the superconducting state of the grains compared to their normal-state values. These parameters are sensitive to the external magnetic field in the superconducting state. The increase of the phase velocity in the superconducting state is attributed to the change of the effective magnetic permeability which is in conflict with previous theoretical predictions.

I. INTRODUCTION

The propagation of electromagnetic (EM) waves in disordered media is a subject of considerable interest both theoretically[1–8] and experimentally.[8–10] In contrast to bulk metals and superconductors in which the electromagnetic wave is highly attenuated due to the skin effect or to the Meissner effect, in metal-insulator composites the EM wave infiltrates between the grains and may travel rather large distances. The dissipation in such composites is determined mostly by eddy-current losses, which are negligibly small if the grains are superconducting. The propagation of the microwave through a composite consisting of decoupled superconducting grains in the lossless insulating support was considered theoretically by Pomerantz.[2] Using the dynamic effective-medium approximation (DEMA) of Stroud and Pan,[1] he demonstrates that the effective dielectric constant ϵ of the composite is real and frequency independent. For small concentration of superconducting grains $\epsilon > 0$, which makes this medium transparent to the EM wave, while for high concentration of the superconducting grains $\epsilon < 0$, so it is nontransparent. This model, similar to most of the effective-medium theories, assumes that the electrodynamic properties of the composite media are determined by the effective dielectric constant[1–3] and neglect the role of magnetic permeability. Only recently, the importance of the effective magnetic permeability of the composite media, especially at microwave frequencies, was recognized.[4–7]

This paper reports on the microwave propagation in a composite consisting of small (compared to wavelength) $YBa_2Cu_3O_{7-x}$ (YBCO) or $Bi_2Sr_2CaCu_2O_x$ grains embedded in the insulating matrix (with the emphasis on the YBCO). The measurements were carried out in the absence of and in the presence of an external magnetic field in both the normal and superconducting states of the grains. To the best of our knowledge, there is no previous experimental study of the microwave propagation through such composites, although the microwave conductivity of similar composites has been studied before by resonant methods.[11–13] To our surprise, we found that the microwave propagates with a low attenuation through the superconductor-insulator mixtures, even at high concentration of superconductor. The measured dielectric constant of superconductor-insulator composites is in disagreement with the DEMA predictions.[1–2] We provide evidence that the magnetic permeability plays a major role in the microwave propagation through superconductor-insulator mixtures.

II. SAMPLE PREPARATION AND LOW-FREQUENCY STUDIES

The samples were prepared by homogeneously mixing the superconducting powder (YBCO or $Bi_2Sr_2CaCu_2O_x$, average grain size of 20 μm, produced by Hoechst, Frankfurt) with dry polymethylmetacrylate powder. Adding a small amount of the liquid monomer results in a fast (5–10 min) polymerization of metamethylacrylate and solidification of the mixture. Only homogeneous composites with volume fraction f of superconductor less than $f = 0.5$ could be prepared by this method. The samples were cast into rectangular bars that fit the WR90 waveguide with a cross section of 2.3×1 cm^2. The length of the samples varied between 0.5 and 2 cm. Scanning electron microscopy pictures show that the YBCO particles have irregular forms with the sizes of 5 to 20 μm and tend to cluster into large loose aggregates with dimensions of 200 μm.

The samples were characterized by measuring their room-temperature low-frequency complex conductivity using an HP-4141 impedance meter in the frequency range $\omega/2\pi = 100$ Hz–1 MHz. Figure 1 shows a Cole-Cole plot (dependence of the imaginary part of the complex resistivity on its real part at varying frequency) for some YBCO samples measured at room temperature in this frequency range. The shape of this diagram resembles a slightly flattened semicircle, suggesting a relatively narrow distribution of relaxation times in each sample. We note that the data for all the samples nearly fall on the same curve. This suggests that all the samples can be

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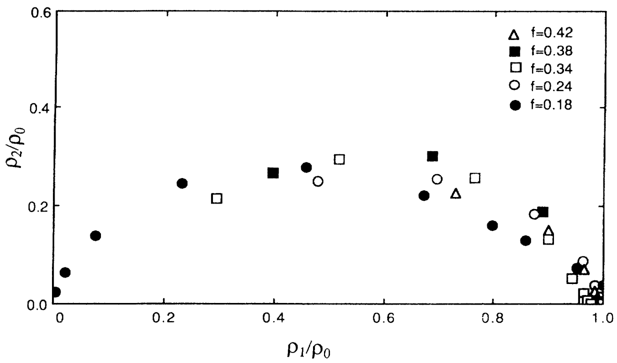

FIG. 1. Cole-Cole plot of the imaginary part ρ_2 vs real part ρ_1 of the complex resistivity $\rho=\rho_1-i\rho_2$ at varying frequency (100 Hz–1 MHz) at $T=300$ K and for composites with different volume fraction f of YBCO. ρ_0 is the real part of the complex resistivity at 100 Hz.

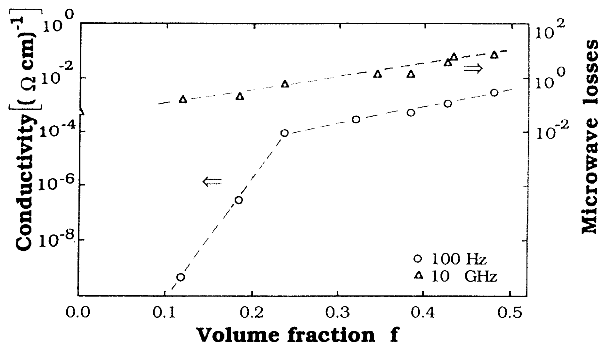

FIG. 3. Low-frequency ($\omega/2\pi=100$ Hz, $T=300$ K) conductivity σ and microwave losses $2nk$ ($\omega/2\pi=10$ GHz, $T=97$ K) vs YBCO volume fraction. In nonmagnetic materials ($\mu'=1$ and $\mu''=0$) the parameter $2nk$ is directly related to the microwave conductivity $\sigma(\omega)$, e.g., $2nk=4\pi\sigma(\omega)/\omega$. The dashed lines are guides to the eye.

approximated by the same equivalent electric circuit. A complete analysis of the Cole-Cole diagram for these composites will be given elsewhere. Figure 2 shows the frequency dependence of the real part of the conductivity for different samples at room temperature. The conductivities of the samples are fairly frequency independent at low frequencies $\omega/2\pi<500$ Hz and increase at higher frequencies. Figure 3 demonstrates the conductivity at 100 Hz (which we take as a measure of the dc conductivity) as a function of the volume fraction f of YBCO. Below $f=0.2$ the conductivity is exceedingly low ($\sigma<10^{-6}\ \Omega^{-1}\,\mathrm{cm}^{-1}$), while above $f=0.2$ it is several orders of magnitude higher ($\sigma\approx10^{-2}\ \Omega^{-1}\,\mathrm{cm}^{-1}$). However, it is still much lower than the dc conductivity of YBCO single crystals at 300 K ($\sigma\approx10^{3}-10^{4}\ \Omega^{-1}\,\mathrm{cm}^{-1}$). The dielectric constant for the same samples was determined from the imaginary part of the complex conductivity. It decreases with increasing frequency in the range $10^{3}-10^{6}$ Hz approximately as $\epsilon\sim\omega^{-n}$ (not shown here),

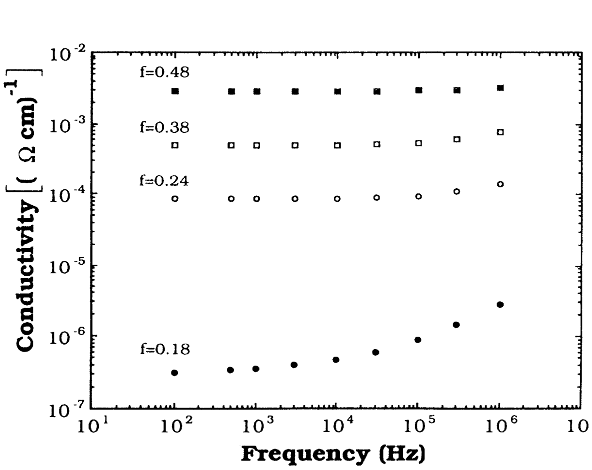

FIG. 2. Frequency dependence of the real part of the conductivity for composites with different YBCO volume fraction f at $T=300$ K.

with n varying from 0.2 to 0.3 when the volume fraction f changes from $f=0.24$ to 0.42. Such concentration and frequency dependencies of the conductivity and dielectric constant are characteristic for percolating systems.[14,15] Similar frequency dependencies of the conductivity and dielectric constant were obtained recently for percolating polymer composites[16] and for metal colloids in an insulator.[10] The percolation threshold as estimated from Fig. 3 is $f_p\cong0.2$. We have measured also the temperature dependence of the dielectric constant of YBCO samples with volume fractions $f=0.24$ and $f=0.18$ at $\omega/2\pi=1$ kHz. Within the accuracy of the experiment (2%) there is no variation of ϵ upon superconducting transition of the grains ($T_c=91$ K).

III. MICROWAVE STUDIES

The microwave studies were carried out in the frequency range 8–10 GHz by measuring the amplitude and phase of the transmitted and reflected waves from the rectangular bar of the superconductor-insulator composite, which completely fills the waveguide. The peripheries of the front and rear surfaces of the sample were painted with the silver paint to prevent microwave leakage between the sample surface and the walls of the waveguide.[17] The microwave system consists of the HP-83623A synthesized sweeper, the reference channel, and two channels for transmitted and reflected signals. By means of a variable attenuator and phase shifter, the mixer's output is balanced and the amplitude and phase of the transmitted and reflected waves are measured. The system was calibrated with an open end and with a short circuit.

Figure 4 shows the temperature dependence of the amplitude and the phase of the transmitted microwave through a YBCO composite ($f=0.38$). Figure 5 demonstrates similar results for the $Bi_2Sr_2CaCu_2O_x$ composite ($f=0.44$). Above T_c of the grains both the amplitude and the phase are fairly temperature independent. Upon

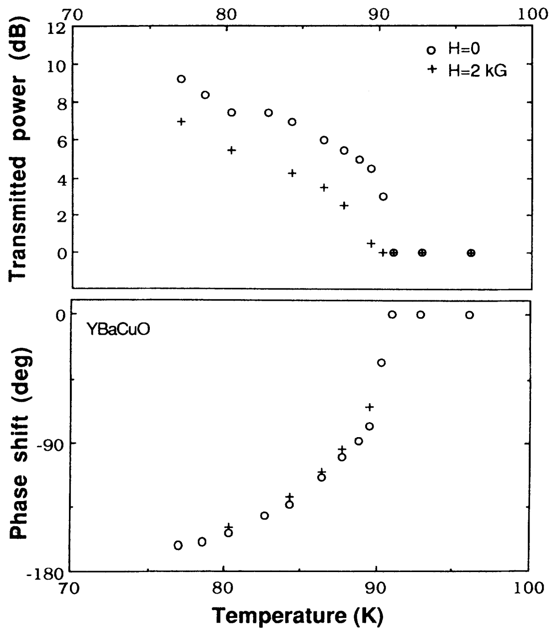

FIG. 4. Temperature dependence of the amplitude and of the phase shift of the microwave (relative to those at $T=97$ K) transmitted through a YBCO-polymethylmetacrylate composite with a volume fraction of $f=0.38$ and length of $d=1.46$ cm in the presence and absence of magnetic field ($H=2$ kG).

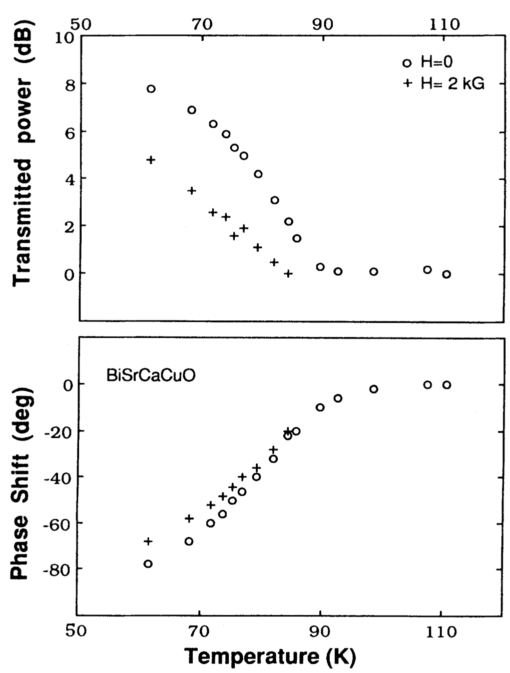

FIG. 5. Temperature dependence of the amplitude and of the phase shift of the microwave (relative to those at $T=110$ K) transmitted through a $Bi_2Sr_2CaCu_2O_x$ polymethylmetacrylate composite with a volume fraction of $f=0.44$ and length of $d=2$ cm in the presence and absence of magnetic field ($H=2$ kG).

transition to the superconducting state of the grains the phase of the transmitted wave dramatically decreases and the transmittance increases. Far below T_c these parameters are nearly temperature independent. Figures 4 and 5 demonstrate also the effect of a magnetic field of 2 kG on the phase shift and on the transmittance of the sample. The magnetic field does not produce any effect for $T>T_c$. However, when the grains are superconducting it increases the transmission coefficient and decreases the phase shift. The field-induced phase shift in $Bi_2Sr_2CaCu_2O_x$ composites is observed far below T_c (Fig. 5), while in YBCO composites it is observed only near T_c (Fig. 4). In a composite with the volume fraction $f=0.48$ of YBCO (grain size of 20 μm) and length $d=1.4$ cm, the phase shift due to a field of $H=2$ kG is $\Delta\Theta_t=10^0$ near T_c. In a composite with the volume fraction $f=0.44$ of $Bi_2Sr_2CaCu_2O_x$ and length $d=2$ cm the field-induced phase shift is $\Delta\Theta_t \approx 12^0$ at $T=63$ K and $H=2$ kG (Fig. 5).

The wave propagation in a uniform medium with negligible scattering is determined by the effective complex refraction index $N=n+ik$, where the real part n yields the ratio of the phase velocity in vacuum to that in the medium, while the imaginary part k is directly related to attenuation. The size of the YBCO particles in our composites ($2r=20$ μm) is small compared to the wavelength ($\lambda=1-3$ cm), and therefore such a medium may be treated as uniform. The values of n and k can be estimated from the complex transmission $T\exp(i\Theta_t)$ and reflection $R\exp(i\Theta_r)$ coefficients of a TE_{10} wave in the dielectrically filled rectangular waveguide through the following equations:

$$Te^{i\theta_t}=\frac{4q_s}{q_0\mu}\,\frac{e^{-iq_0 d}}{\left[1+\dfrac{q_s}{q_0\mu}\right]^2 e^{-iq_s d}-\left[1-\dfrac{q_s}{q_0\mu}\right]^2 e^{iq_s d}}\,, \tag{1a}$$

$$Re^{i\theta_r}=\frac{\left[1-\left[\dfrac{q_s}{q_0\mu}\right]^2\right](e^{-iq_s d}-e^{iq_s d})}{\left[1+\dfrac{q_s}{q_0\mu}\right]^2 e^{-iq_s d}-\left[1-\dfrac{q_s}{q_0\mu}\right]^2 e^{iq_s d}}\,, \tag{1b}$$

where $q_0=[\omega^2/c^2+(\pi/a)^2]^{1/2}$ and $q_s=[\omega^2(n+ik)^2/c^2+(\pi/a)^2]^{1/2}$ are the wave vectors for a TE_{10} wave in the empty and in the dielectrically filled waveguide, respectively; d is the sample length, $\omega/2\pi$ is the frequency, a is the width of the waveguide, and μ is the magnetic permeability of the sample. The complex transmission and reflection coefficients were measured in the frequency range of 8–10 GHz and were fitted to Eqs. (1a) and (1b) with n, k, and μ as *independent* fitting parameters that were assumed to be frequency independent in the above range. Generally, we found a better fit to the transmittance than to the reflectance. In our analysis we have compared the transmittance and reflectance for a whole bar ($f=0.34$) to those of different pieces cut out of the same bar. We found the values of n and k, determined using either the whole bar or each of its parts, to be the same and thus be independent of the sample length. We found that the complex transmission and reflection coefficients are very sensitive to n and k and rather insensitive to μ [Eq. (1)]. Indeed, we varied μ from $\mu=1$ to $\mu=0.6$ and found that the corresponding variations of n and k, as obtained from the fitting procedure, do not exceed 3% and 10%, respectively. Therefore, we were unable to obtain μ from such fits and have estimated it through the dependence of n and k on μ (see discussion).

Figure 6 demonstrates the temperature dependence of n and k for a sample with a YBCO volume fraction of $f=0.38$. These values of n and k were obtained using the data in Fig. 4 and our analysis. Both n and k are fairly temperature independent in the normal state. These parameters decrease upon superconducting transition (in contrast to the low-frequency dielectric constant) and are temperature independent far below T_c. Figure 7 demonstrates the dependence of n and k on the volume fraction of YBCO as measured far above and far below T_c. Both n and k steadily increase with the YBCO volume fraction in the normal as well as in the superconducting states. We note that k in the superconducting state is significantly lower than its normal-state value. Therefore, an EM wave penetrates through larger distances in such a mixture when the grains are superconducting. For example, in the material with volume fraction $f=0.4$ of YBCO and $d=1$ cm the amplitude of the propagating microwave in the normal state is attenuated by 50% (6.3 dB/cm), while in the superconducting state it is attenuated only by 30% (3.1 dB/cm). We believe that the attenuation in the superconducting state may be further decreased by decreasing the losses in the polymer matrix.

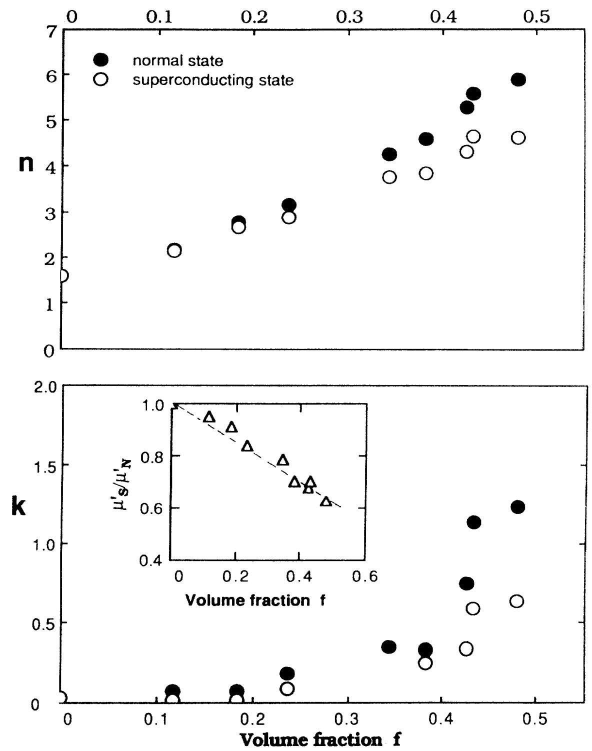

FIG. 7. Dependence of the real and imaginary parts of the complex refraction index $N=n+ik$ on the YBCO volume fraction f in the normal ($T=97$ K) and superconducting ($T=70$ K) states. Inset shows the ratio of the effective magnetic permeabilities in the normal and superconducting states μ'_S/μ'_N as obtained using Eqs. (7) and (8) (see text). Dashed line is the linear interpolation $\mu'_S/\mu'_N=1-0.8f$.

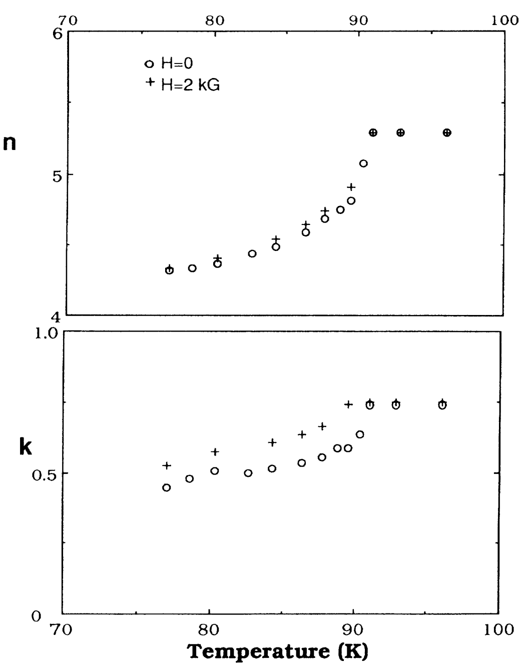

FIG. 6. Temperature dependence of the real and imaginary parts of the complex refraction index $N=n+ik$ at a frequency of 10 GHz in the presence and absence of magnetic field ($H=2$ kG) for the same YBCO sample as in Fig. 4.

IV. DISCUSSION

Generally speaking, the complex refraction index of the uniform medium $N=n+ik$ is determined by the complex dielectric constant $\epsilon=\epsilon'+i\epsilon''$ and by the complex magnetic permeability $\mu=\mu'+i\mu''$ as follows:

$$N^2=(n^2-k^2)+2ink=\epsilon\mu \ . \tag{2}$$

A similar expression was suggested by Mahan[4] for the complex refraction index of a metal-insulator composite medium in the long-wavelength limit. Choy and Stoneham[6,7] argue that ϵ and μ of the composite medium are not independent and so the accuracy of Eq. (2) in this case is questionable. However, these authors do not give any close expression for the complex refraction index of the composite. In the absence of a complete theory we use Eq. (2), where ϵ and μ represent, respectively, the effective dielectric constant and magnetic permeability for the composite medium. Assuming $\epsilon''\mu''\ll\epsilon'\mu'$, Eq.(2) yields

$$n^2-k^2=\epsilon'\mu'-\epsilon''\mu''\approx\epsilon'\mu'; \quad 2nk=\epsilon'\mu''+\epsilon''\mu' \ . \tag{3}$$

The microwave losses are characterized by the parameter $2nk$, which in nonmagnetic materials ($\mu'=1$ and $\mu''=0$) is directly related to the microwave conductivity

($\sigma=\epsilon''\omega/4\pi$) as $\sigma=nk\omega/2\pi$ [Eq.(3)]. In Fig. 3 we plot the microwave losses $2nk$ versus the YBCO volume fraction. Clearly, no dramatic change in the microwave losses is seen near the dc percolation threshold $f_p \cong 0.2$ in contrast to the low-frequency conductivity. The microwave refraction index n also does not show any singularity at $f_p \cong 0.2$ (Fig. 7). This is consistent with the predictions of the percolation theory[15] and means that the capacitive coupling between the grains prevails over the resistive coupling at microwave frequencies.

What is the physical mechanism for the decrease of the refraction index n in going from the normal to the superconducting state (Figs. 6 and 7)? According to Eqs. (2) and (3) this decrease may be attributed either to a change in ϵ' or to a change in μ'. There are several models for the dielectric constant of the composite media. Pomerantz[2] has calculated the dielectric constant $(\epsilon')_{\text{sup. ins.}}$ of the superconductor-insulator composites using the dynamic effective-medium theory of Stroud and Pan[1] together with the dielectric function for a London superconductor (which at $T=0$ can be written as $\epsilon_S=1-c^2/\omega^2\lambda_p^2$, where ω is the electromagnetic wave frequency and λ_p is the penetration depth of superconductor). For small volume fraction f, he obtains the following expression for the dielectric constant of a composite in which the grains are in the superconducting state

$$(\epsilon')_{\text{sup. ins.}}=\epsilon_0[1+3f(1-r^2/30\lambda_p^2)] , \tag{4a}$$

while for the same composite in the normal state,

$$(\epsilon')_{\text{norm met ins}}=\epsilon_0(1+3f) . \tag{4b}$$

Here r is the radius of the superconducting particles and ϵ_0 is the dielectric constant of the insulator. We note that the dielectric constants as given by Eqs. (4a) and (4b) are frequency independent. This is a consequence of the ω^{-2} frequency dependence of the dielectric function of the London superconductor. Therefore, according to the model of Pomerantz [Eqs. (4a) and (4b)], any change of ϵ' upon transition to the superconducting state should be the same at low and at high frequencies. However, our low-frequency results do not show any change in ϵ', while we see a clear change of the refraction index at microwave frequencies (Figs. 6 and 7). This is in contrast to the model of Pomerantz.[2]

Certainly, one can argue that the frequency dependence of the dielectric function of high-T_c materials might be different from ω^{-2}. In fact, the frequency-dependent dielectric function of high-T_c superconductors as obtained from the infrared transmission studies[18] deviates considerably from the ω^{-2} dependence at frequencies higher than 100 cm^{-1}. However, an estimate of the corresponding deviation at microwave frequencies $\omega/2\pi\approx 10^{10}$ Hz does not exceed 0.01%.[18] We note also that the direct measurement of the change of the effective dielectric constant of the high-T_c superconducting ceramics by the resonant method[19] at 8.8 GHz is extremely small (0.8%). This would imply a change of the refraction index of ceramics ($f=0.8$–0.9) of 0.4% in contrast to the significant change observed experimentally [which is as high as 30% (Figs. 6 and 7) even at lower volume fractions of superconductor $f=0.4$]. We thus conclude that the change of the refraction index n upon transition to the superconducting state is probably not related to the change of the dielectric constant.

We make an alternative assumption that the change of the refraction index n (Figs. 6 and 7) is due entirely to the change of magnetic permeability μ. Then we can write, using Eq. (3), the following relation:

$$\frac{(n^2-k^2)_S}{(n^2-k^2)_N}=\frac{\mu_S'}{\mu_N'} , \tag{5}$$

where the indices S and N refer to the superconducting and normal states, respectively. Using the experimental values of n and k (Fig. 7) and Eq. (5), the dependence of μ_S'/μ_N' on the YBCO volume fraction f may be roughly approximated as $\mu_S'/\mu_N'=1-\beta f$ with $\beta=0.8$ (see inset to Fig. 7). We compare this result with an estimate based on the Clausius-Mossotti approximation for magnetic composites[20]

$$\frac{\mu-1}{\mu+2}=\frac{4\pi}{3}\sum_j \alpha_j c_j , \tag{6}$$

where $\mu=\mu'+i\mu''$ is the effective magnetic permeability, c_j is the concentration of magnetic particles in the nonmagnetic insulator, and $\alpha_j=\alpha_j'+i\alpha_j''$ is the magnetic polarizability of the j components. This approximation [Eq. (6)] successfully describes the static magnetic properties of granular superconducting systems.[21] For an isotropic conducting sphere with radius r, the magnetic polarizability α can be expressed as[6,22]

$$\alpha=-\frac{r^3}{2}\left[1+\frac{3}{qr\tan(qr)}-\frac{3}{(qr)^2}\right] . \tag{7}$$

Here $q=(1+i)/\delta$ in the normal state; $q=i/\lambda_p$ in the superconducting state, δ is the skin depth, and λ_p is the penetration depth. The polarizability α varies from $\alpha=0$ for a poorly conducting metal particle to $\alpha=-r^3/2$ for a highly conductive particle with the skin depth (or penetration depth if the particle is superconducting) small compared to the radius of the grain. For a composite consisting of isotropic conducting spheres in a nonmagnetic insulator, the magnetic permeability is derived from Eqs. (6) and (7) as

$$\mu=\frac{1+\dfrac{f\alpha}{r^3}}{1-2\dfrac{f\alpha}{r^3}} . \tag{8}$$

Here f is the volume fraction of conductor. For $f \ll 1$ Eq. (8) reduces to the effective-medium result $\mu\approx 1+3\alpha f/r^3$.

Certainly, the high-T_c superconductors are highly anisotropic and this anisotropy should be taken into consideration. Let us assume, first, that all the grains are uniformly oriented relative to the microwave magnetic field such that the screening currents are in the a-b plane. In this case the skin depth of YBCO at 10 GHz is $\delta\cong 5$ μm (assuming a normal-state conductivity of 10^4 $\Omega^{-1}\,\text{cm}^{-1}$) and the penetration depth is $\lambda_p \cong 0.13$ μm.[23]

Thus, far from T_c and for $f \ll 1$, Eqs. (7) and (8) yield for the composite containing YBCO grains with the average radius $r = 10$ μm the following effective permeabilities: $\mu'_N \approx 1\text{-}0.4f$, $\mu'_S \approx 1\text{-}1.4f$, and $\mu'_S/\mu'_N \approx 1\text{-}\beta f$ with $\beta = 1$. For the other limit, when all the grains are oriented such that the screening currents are in the a-c (b-c) planes, we estimate $\delta \cong 50$ μm, $\lambda_p \cong 0.45$ μm.[23] The corresponding permeabilities are $\mu'_N \approx 1$, $\mu'_S \approx 1\text{-}1.3f$, and $\mu'_S/\mu'_N \approx 1\text{-}\beta f$ with $\beta = 1.3$. For a composite with randomly oriented grains we expect that the ratio μ'_S/μ'_N will be described by a similar expression: $\mu'_S/\mu'_N \approx 1\text{-}\beta f$, where β lies between the two extreme limits, i.e., $\beta = 1$ (screening currents in the a-b plane) and $\beta = 1.3$ (screening currents in the a-c or b-c plane). The experimental data are indeed described by such linear dependence (see Fig. 7, inset) with $\beta = 0.8$. In view of the approximate nature of the above calculations, such a difference is not surprising. We relate the temperature and magnetic-field variations of the refraction index n in the superconducting state to the changes in the penetration depth [Eqs. (7) and (8)] due to the temperature and field dependence of the concentration of the superconducting pairs and due to the ac response of the pinned vortices and weak links.[24]

The decrease of the attenuation constant k in the superconducting state according to Eq. (3) may be due to the dielectric losses ϵ'' as well as to the magnetic losses μ''. Both arise from dissipative electric currents in the grains. This dissipation is greatly reduced in the superconducting state, resulting in low attenuation of the wave (Figs. 4–6). The effect of magnetic field on the attenuation constant in the superconducting state (Fig. 6) may arise from the dissipation due to the motion of vortices introduced by the external magnetic field. The finite attenuation in the superconducting state in the absence of magnetic field may be due also to enhanced backward scattering. In fact, for the small ideally conducting metal particles, the backward scattering of EM waves dominates.[22]

In summary, the superconductor-insulator composites have unusual electromagnetic properties. They differ from bulk superconductors in that the electromagnetic wave (with a below-gap frequency) propagates in such a medium even when the volume fraction of superconductor exceeds 40%, while in bulk superconductors such a wave is fully reflected. However, these composites are similar to a bulk superconductor in that they are virtually lossless. The wave velocity in the superconducting state of these composites depends on the concentration, temperature, and magnetic field (due to the penetration length change). This may have a practical application in the construction of high-frequency phase shifters and modulators from these materials. The effect of the superconducting transition of the grains on the phase velocity is due to the change of the effective magnetic permeability and not to the change of the effective dielectric constant as predicted by the dynamic effective-medium theory.[1,2] A study of the electromagnetic wave propagation through the superconducting composites may provide useful information about the electrodynamical properties of the superconducting component and may serve as a tool for the determination of the rf penetration length.

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