A New Method Developed in Measuring the Dielectric Constants of Metallic Nanoparticles by a Microwave Double-Cavity Dielectric Resonator

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***Abstract*—A microwave double dielectric resonator is implemented to measure the dielectric constants of nano-metallic powders. The metallic nanoparticles prepared by vacuum evaporation in inert gas are collected and mixed with alumina powder to fill the inner hole of a sapphire disc by which the resonant frequency and Q factor are measured at the TE_{011} mode to derive the complex dielectric constant.**

***Index Terms*—Dielectric constants, metallic nano-particles, microwave dielectric resonator.**

I. Introduction

MICROWAVE dielectric resonators are generally exploited in the fabrication of resonators, filters and radar detectors etc. In basic research, they allow high precision measurements of the temperature dependence of the London penetration depth in oxide superconductors [1], [2]. Many techniques are developed [3], [4] to obtain more versatile applications by dielectric resonators.

The origins of dielectric constant of materials are attributed to the polarization change of the movements of electrons and ions and molecular vibrations depending on the frequency of incident waves. Within the microwave frequencies, the dielectric constant of metals mainly arises from the displacements of electrons and ions. For metallic nanoparticles, the quantum splitting arising from size effect intrigues inter- or intra- subband transitions, which also should contribute to the dielectric constants [5], [6]. In conventional method, the real and imaginary parts of a complex dielectric constant are, respectively, determined by the phase shift of a propagation length and the absorption magnitude at a given disc thickness. However, an accurate determination of the length and thickness for nanoparticles is not accessible. Till now, the measurements of dielectric constants of metallic particles near microwave frequencies have never been attempted.

In this work we endeavor to develop a new method to measure the dielectric constant of different-size metallic particles. A microwave ring dielectric resonator is designed in which the powder mixture is filled inside the inner hole of the resonator. The real part of the dielectric constant is derived from the resonant frequency, which can be solved from the Jocobian determinant resulting from the continuity of boundary conditions [2]. The imaginary part and the penetration depth of conducting wall are determined from the resonant Q-factor, which can be measured from the bandwidth at 3 dB attenuation.

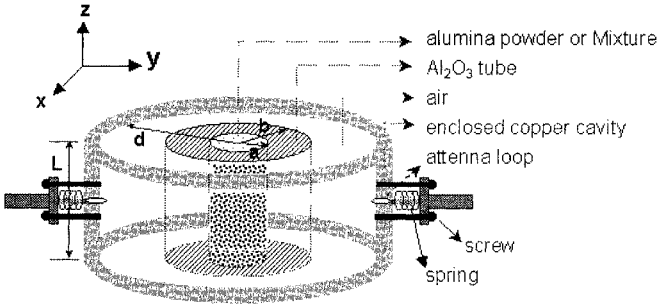

Fig. 1. Double dielectric resonator in which the diameters of the inner hole, the sapphire ring and copper cavity are a, b and d, respectively, with a length of L.

II. Construction Detail of Dielectric Resonator

To perform this experiment, as sketched in Fig. 1, a ring-shaped sapphire disc with a hole at the center is filled with a mixture of metallic particles and alumina powders with a proper ratio. The double dielectric resonator is installed at the center of a cylindrical cavity made of copper. Two straight coaxial loop antennas are inserted to excite the TE_{0n1} modes from the opposite sides of the cavity. The couplings of the microwave power into and out of the cavity can be readily obtained by adjusting the depth and loop orientations of the antenna for under-damped conditions.

Since metallic particles have exceptionally high microwave absorption, they cannot directly be filled inside the dielectric cavity [7]. A low ratio of metallic nanoparticles is mixed with a low dielectric loss Al_2O_3 powder and filled inside the hole. The dielectric constant of the powder mixture inside the sapphire ring disc is measured to be an effective dielectric constant ε_{eff} which is extracted from the EM field program that calculates resonant frequency by curve fitting. The dielectric constant ε_m of Al_2O_3 powder is different from its crystalline sapphire disc

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owing to its porous and isotropic properties. The dielectric constant ε of metallic particles is derived form the effective-medium theory [8] and is given by

$$\varepsilon = \varepsilon_m \frac{f(\varepsilon_{eff} + 2\varepsilon_m) + 2(\varepsilon_{eff} - \varepsilon_m)}{f(\varepsilon_{eff} + 2\varepsilon_m) - (\varepsilon_{eff} - \varepsilon_m)} \tag{1}$$

where ε_m is the dielectric constant of pure alumina powder and f is the filling factor specifying the volume ratio of metallic nanoparticles and Al_2O_3 powders.

The resonant frequency can be determined from the Jocobian determinant [2], [3] which satisfies the continuity boundary conditions embodied in the dispersion relations at double cavities. Besides measuring the resonant frequency to derive the real part of the dielectric constant, we can also measure the quality factor Q to derive the loss tangent. The total energies stored inside the center hole, the sapphire ring and outside the ring are

$$\begin{aligned} W_T =& \frac{\varepsilon_0 \varepsilon_{r1}}{2} \int |E_{\phi 1}|^2 dV_1 + \frac{\varepsilon_0 \varepsilon_{r2}}{2} \int |E_{\phi 2}|^2 dV_2 \\ &+ \frac{\varepsilon_0}{2} \int |E_{\phi 3}|^2 dV_3 \\ \equiv& \varepsilon_{r1} W_1 + \varepsilon_{r2} W_2 + \varepsilon_{r3} W_3 \end{aligned} \tag{2}$$

where ε_{r1}, ε_{r2} and ε_{r3} are the real parts of the dielectric constants of the inner cavity, the sapphire disc and the outside air cavity, respectively and the W_i' are the stored energies in corresponding regions. In general, the dielectric loss of metallic particles is much larger than other parts and is the dominant term to be measured. The quality factor of nanoparticles is derived from

$$\begin{aligned} \frac{1}{Q_{nano}} \cong& \frac{1}{Q_{02}} - \frac{1}{Q_{01}} = \frac{1}{Q_{powder+nano}} - \frac{1}{Q_{powder}}, \\ \frac{1}{Q_{01}} =& \frac{1}{Q_c} + \frac{1}{Q_{powder}} + \frac{1}{Q_{2d}} + \frac{1}{Q_r}, \\ \frac{1}{Q_{02}} =& \frac{1}{Q_c} + \frac{1}{Q_{powder+nano}} + \frac{1}{Q_{2d}} + \frac{1}{Q_r} \end{aligned} \tag{3}$$

where Q_{01} and Q_{02} are the unloaded quality factor of alumina powder and mixed powder, respectively and the Q_r is that for the radiation loss. For a closed cavity with highly conducting walls, the Q_r approaches infinite and Q_c is that for total conducting walls of the cavity, Q_{2d} will be defined in (4). To obtain a higher accuracy in the determination of sample dielectric, the cavity length L should be much shorter than the radius b.

The dielectric losses of the metallic nanoparticles and the sapphire ring are respectively as given by

$$\begin{aligned} Q_{nano} \cong& \frac{\omega \varepsilon W_t}{\sigma_{nano} f W_1} = \frac{W_t}{f W_1} \frac{1}{\tan \delta_{nano}} = \frac{W_t}{f W_1} \frac{\varepsilon_r}{\varepsilon_i}, \\ \text{and } Q_{2d} \cong& \frac{\omega \varepsilon_{r2} W_t}{\sigma_2 W_2} \end{aligned} \tag{4}$$

where $W_t = W_1 + W_2 + W_3$ and the complex dielectric constant of the nanoparticles is specified as $\varepsilon = \varepsilon_r - j\varepsilon_i$, σ_{nano} is the conductivity of nanoparticles and $\tan \delta_{nano}$ is the loss tangent.

III. Performance and Discussion

The iron nanoparticles are prepared by thermal vacuum evaporation in an argon gas at a filled pressure of $30 \sim 60$ mtorr. The iron nanoparticles with nearly spherical shape are inert to be oxidized. The particle size crucially depends on the filled gas pressure and also depends on the distance between the source heater and the substrate. The grain size and the structure of the metallic particles can be checked by the x-ray powder diffraction method as shown in Fig. 2. Single crystalline iron structure is found to be maintained for each particle, the particle size can be determined by the well-known Sherrer's formula [9]. An Anritsu 37 347A network analyzer is implemented to measure the S_{21} transmission signal. The resonant frequency of the cavity before installing the sample has a dielectric constant of $\varepsilon_r = 9.73$ with the sapphire ring.

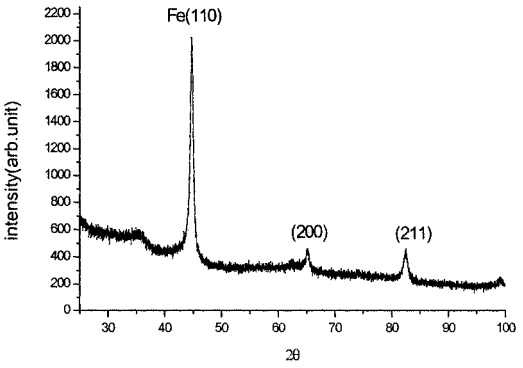

Fig. 2. Powder x-ray diffraction of iron nanoparticles prepared by thermal evaporation under an argon gas filling pressure of 60 mtorr.

TABLE I
Measured Q Factors and the Calculated Dielectric Constant of Iron Nanoparticles Where f_0 (GHz) is the Resonate Frequency, $f = 1/200$ is the Filling Factor, Q_0 is the Q Factor of Empty Inner Hole, Q_{01} is That for Pure Alumina Powder and Q_{02} is That for the Alumina and Iron Nanoparticle Mixture

Particle size (Å)	f_0 (GHz)	Q_0	Q_{01}	Q_{02}	ε
117	14.1816	3061	2288	1305	-26.52+31.67j
150	14.1910	3061	2288	1518	-33.03+26.56j

To obviate the microwave absorption due to moisture absorbed in the alumina powder, the pressed powder should be baked in an oven for several hours. The resonant frequency f_0 and the bandwidth of TE_{011} line yield the Q factors. The Q value changed from 2200 toward 1200 when the alumina powder was stored at ambient in the room showing that the absorption of moisture is a serious concern. A typical experimental result for various Q factors at two specified particle sizes to derive the dielectric constants of iron nanoparticles is depicted in Table I.

The dielectric constants of iron nanoparticles measured in this experiment are $-26.52.06 + 31.67j$ and $-33.03 + 26.56j$ for grain sizes of 117 Å and 150 Å, respectively, revealing that the magnitude decreases with the particle size. These values are comparable to the values of silver films measured in the visible optical wavelength, which changes from $-33 + 10.0j$ to $-67.03 + 2.44j$ for particle sizes varying from 100 Å to bulk value [10], respectively. Unexpectedly, we found that a large fluctuation in the determination of the dielectric constant of metallic particles occurs when the filling factor in the powder

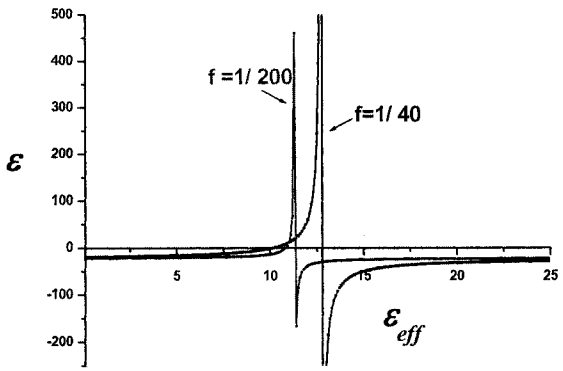

Fig. 3. Relation of ε and ε_{eff} at the filling factors to be 1/40 and 1/200.

mixture is small. This embodies the fact that to derive the dielectric constant ε of metallic powder from the effective ε_{eff} as dictated in (2) protrudes a singular point as the filling factor f approaches to the value $(\varepsilon_{eff}-\varepsilon_m)/(\varepsilon_{eff}+2\varepsilon_m)$. Fig. 3 indicates that for small values of the factor f, the measured ε_{eff} will be located more on the left side of the singular peak. A choice of $f = 1/150$ is more germane to obtain a conclusive result. The relevant sources of the measurement uncertainty are most attributed to the determination of true ε from the measured effective ε_{eff} which can possibly be overcome by replacing the alumina powder by a nonabsorbing powder with larger ε_m (e.g., Y_2O_3). Baking the powder to obviate moisture absorption can certainly also increase the measurement accuracy.

IV. Conclusion

In this scenario, we have designed a microwave dielectric resonator, which is easily detachable for filling the powder sample and changing the coupling coefficients to measure the dielectric constant of powders. The measured magnitude of the negative constant of metallic powder increases with the particle size and approaches to the bulk value. With different design of cavity sizes and selection of higher modes, the dielectric constants of metallic powders at various frequencies can be measured.

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