Improvement of the transmission/reflection method for dielectric and magnetic measurements on liquids between 0.1 and 20 GHz

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Abstract. We propose an appropriate calculation method, giving both the complex
permittivity ε^* and permeability μ^* of a liquid sample, from transmission/reflection
measurements obtained from a network analyser working on a coaxial line between
0.1 and 20 GHz. A 20 mm long specific coaxial cell has been constructed using two
polytetrafluoroethylene joints to ensure air tightness. Its small length allows rejection
towards the upper frequencies of the measurement anomalies given by the
Nicolson–Ross equations. Positions and thickness of the joints are taken into account by
the calculus. The whole system has been tested with the empty cell and then on several
alcohols and ferrofluids. The results on alcohols are in excellent agreement with all
previous results; measurement on one of the magnetic fluids (Fe_2MnO_4 in CCl_4) has
shown a Néel relaxation and also a gyromagnetic resonance.

1. Introduction

To obtain complex permittivity ε^* and permeability μ^*
of a material in the frequency range 0.1–20 GHz, the
transmission/reflection (TR) method is widely used. This
is due to its relative simplicity and to the development
of automatic network analysers. Measurement is made
on a sample within a coaxial line or a waveguide.
Scattering (S) parameters given by the analyser lead to
ε^* and μ^* [1] according to the Nicolson–Ross [2] and
Weir [3] relations.

This method is not well adapted to low-loss materials,
because there is a great inaccuracy when the half wave-
length of the signal is an integer multiple of the sample
length. Baker-Jarvis and co-workers have shown that it
is possible to bypass this difficulty for ε^* [4] and ε^* and
μ^* [5] with an appropriate calculus algorithm. Specific
methods are used when only the permittivity is measured.
The reflection method on an open end coaxial line
dipped into the liquid has led to various theoretical
models and different types of probes [6–8].

Only a coaxial cell allows use of the TR method in
the largest frequency range. To obtain simultaneously
ε^* and μ^* of a liquid we have to confine the sample by
two etancheity joints (whose positions and thickness
must be known and taken into account). Using a cell

of small length allows rejection towards the upper
frequencies of the measurement anomalies given by the
Nicolson–Ross equations [2].

In this paper, we propose a compromise between
solution of the etancheity problem and the broad band
frequency sweep, by an appropriate calculus of ε^* and
μ^* with a specific cell well adapted to our network
analyser.

2. Experimental

The measurement configuration is shown in figure 1.
The network analyser is the HP 8510 system: HP 8510 B
analyser, HP 8515 A S parameters test-set usable from
45 MHz to 26 GHz, HP 8341 B synthesizer sweeper
from 10 MHz to 20 GHz. We used APC 7 connectors.
This system is controlled by a personal computer via
an HPIB bus. From 100 MHz to 20 GHz we use 401
frequency points.

We have built a coaxial line cell of 20 mm length,
made of stainless steel. The surface irregularities are less
than 0.2 μm. To avoid any discontinuity in the propa-
gation mode, we have made two PTFE joints (Teflon
discs) encased in the cell (figure 2). To ensure etancheity
of the cell, the joints are placed on the inner conductor
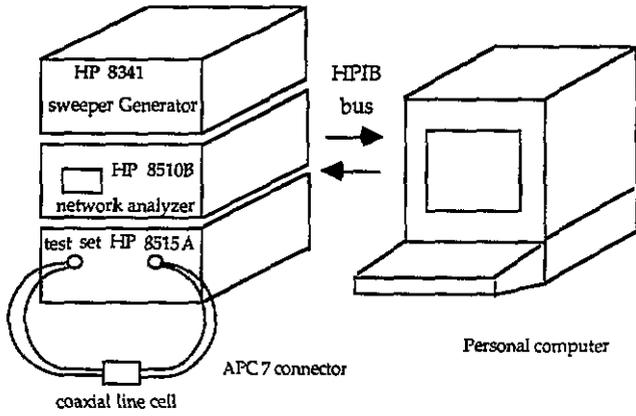
Figure 1. Measurement configuration.
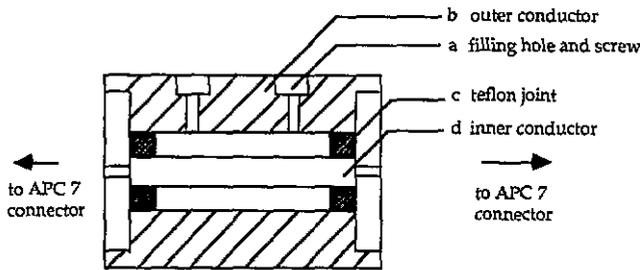
Figure 2. The coaxial line measurement cell.

by warming, and into the outer conductor by freezing with liquid nitrogen.

Most cells of this type are built from standard air coaxial lines with etancheity joints placed just in contact with the right sides of the two conductors used as electrodes. Though these joints are obviously very thin to reduce discontinuity of conductors, they give rise to some errors in measurements at different frequencies; minimization of these errors is furthermore dependent on screwing the connections tight. Moreover, it is difficult to ensure etancheity at the time of removal and the liquid may spoil the APC 7 connectors.
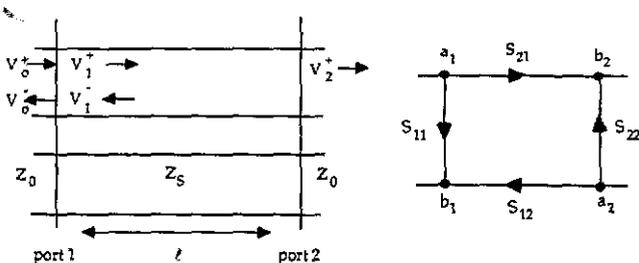
Figure 3. The air line filled with material. V^+ and V^- are respectively the incident and reflected voltages.

3. Theory

In this simple case of a material whose length is equal to that of the sample holder (figure 3), we have the equations

$$S_{11}(\omega) = \frac{(1 - T^2)\Gamma}{1 - T^2\Gamma^2} \qquad (1)$$

$$S_{21}(\omega) = \frac{(1 - \Gamma^2)T}{1 - T^2\Gamma^2} \qquad (2)$$

where Γ is the reflection coefficient between Z_0 and Z_S when the material length is infinite and is given by

$$\Gamma = \frac{Z_S - Z_0}{Z_S + Z_0} = \frac{(\mu_r^*/\varepsilon_r^*)^{1/2} - 1}{(\mu_r^*/\varepsilon_r^*)^{1/2} + 1} \qquad (3)$$

with Z_0 and Z_S being characteristic impedances of the coaxial line without and with the material, while ε_r^* and μ_r^* are the relative complex permittivity and permeability. The term T is the transmission coefficient of the material of finite length l:

$$T = \exp\left(-j\frac{\omega}{c}(\mu_r^* \varepsilon_r^*)^{1/2} l\right) \qquad (4)$$

where c is the speed of light in air.

From these equations, we find

$$S_{11}\Gamma^2 - (S_{11}^2 - S_{21}^2 + 1)\Gamma + S_{11} = 0. \qquad (5)$$

Resolution of this relation leads to

$$\Gamma = K \pm (K^2 - 1)^{1/2} \qquad (6)$$

with

$$K = \frac{S_{11}^2 - S_{21}^2 + 1}{2S_{11}}. \qquad (7)$$

From equations (1) and (2) we obtain

$$T = \frac{S_{21}}{1 - S_{11}\Gamma}. \qquad (8)$$

Hence we can calculate the complex permittivity ε^* and permeability μ^* from

$$\frac{\mu_r^*}{\varepsilon_r^*} = \left(\frac{1 + \Gamma}{1 - \Gamma}\right)^2 = x \qquad (9)$$

$$\mu_r^* \varepsilon_r^* = -\left[\frac{c}{\omega l} \ln\left(\frac{1}{T}\right)\right]^2 = y \qquad (10)$$

$$\mu_r^* = (xy)^{1/2} \qquad (11)$$

$$\varepsilon_r^* = (y/x)^{1/2}. \qquad (12)$$

The thickness of the joints is not negligible, so it must be taken into account to obtain the true value of ε_r^* or μ_r^* of the sample under test (figure 4). Then equations

(1) and (2) become

$$S_{11} = \frac{(1 + \Gamma_x \Gamma_0 T_2^2)(T_1^2 \Gamma_x + \Gamma_0) - T_x^2(\Gamma_0 T_2^2 + \Gamma_x)(T_1^2 + \Gamma_x \Gamma_0)}{(1 + \Gamma_x \Gamma_0 T_2^2)(1 + \Gamma_x \Gamma_0 T_1^2) - T_x^2(\Gamma_0 T_2^2 + \Gamma_x)(\Gamma_0 T_1^2 + \Gamma_x)} \tag{13}$$

$$S_{21} = \frac{T_1 T_2 T_x (1 - \Gamma_0^2)(1 - \Gamma_x^2)}{(1 + \Gamma_x \Gamma_0 T_2^2)(1 + \Gamma_x \Gamma_0 T_1^2) - T_x^2(\Gamma_0 T_2^2 + \Gamma_x)(\Gamma_0 T_1^2 + \Gamma_x)} \tag{14}$$

where Z_0 is the characteristic impedance of the coaxial air line (50 Ω), Z_t is the characteristic impedance of the coaxial line filled with Teflon and Z_x is the characteristic impedance of the coaxial line filled with the studied dielectric. $\Gamma_0 = (Z_t - Z_0)/(Z_t + Z_0)$ is the coefficient of reflection between Z_0 and Z_t when the length of Teflon is infinite. $\Gamma_x = (Z_x - Z_t)/(Z_x + Z_t)$ is the coefficient of reflection between Z_t and Z_x when the length of studied material is infinite. $Z_t = Z_0/\sqrt{\varepsilon_t}$, where ε_t is the real permittivity of Teflon, its losses being negligible.

Hence the coefficient of reflection Γ_x is

$$\Gamma_x = \frac{(\mu_r^*/\varepsilon_r^*)^{1/2} - 1/\sqrt{\varepsilon_t}}{(\mu_r^*/\varepsilon_r^*)^{1/2} + 1/\sqrt{\varepsilon_t}}. \tag{15}$$

The transmission coefficients are

$$T_1 = \exp\left(-j\frac{\omega}{c}\sqrt{\varepsilon_t}l_1\right)$$

through joint 1

$$T_2 = \exp\left(-j\frac{\omega}{c}\sqrt{\varepsilon_t}l_2\right)$$

through joint 2 and

$$T_x = \exp\left(-j\frac{\omega}{c}(\mu_r^*\varepsilon_r^*)^{1/2}l\right)$$

through the material. From Γ_x and T_x we can obtain the values of μ_r^* and ε_r^* from the relations

$$\frac{\mu_r^*}{\varepsilon_r^*} = \left(\frac{1 + \Gamma_x}{1 - \Gamma_x}\frac{1}{\sqrt{\varepsilon_t}}\right)^2 \tag{16}$$

$$\mu_r^*\varepsilon_r^* = \left[\frac{c}{\omega l}\ln\left(\frac{1}{T_x}\right)\right]^2. \tag{17}$$

To calculate Γ_x we must resolve the equation

$$A\Gamma_x^2 + B\Gamma_x + C = 0 \tag{18}$$
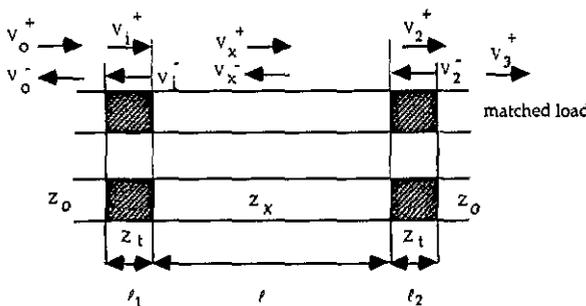
Figure 4. The air line filled with material and Teflon joints.

with

$$A = C = T_1^2 T_2^2 [S_{21}^2 \Gamma_0 - (1 - S_{11}\Gamma_0)(\Gamma_0 - S_{11})] \tag{19}$$

$$B = S_{21}^2 T_1^2(1 + \Gamma_0^2 T_2^4)$$
$$- T_2^2[T_1^4(1 - S_{11}\Gamma_0)^2 + (S_{11} - \Gamma_0)^2]. \tag{20}$$

As in the preceding simplified case, we have from equation (6)

$$\Gamma_x = K \pm (K^2 - 1)^{1/2}$$

with $K = -B/2A$ and

$$T_x = \frac{S_{21}(1 + \Gamma_x \Gamma_0 T_2^2)T_1}{[\Gamma_x(\Gamma_0 - S_{11}) + T_1^2(1 - S_{11}\Gamma_0)]T_2}. \tag{21}$$

4. Tests and results

4.1. Tests

This method has been tested in the case in which our experimental cell was empty. The obtained values of $\varepsilon_r^*(\varepsilon_r', \varepsilon_r'')$ and $\mu_r^*(\mu_r', \mu_r'')$ are given in figures 5(a) and (b). ε_r' and μ_r' are close to 1 and ε_r'' and μ_r'' are close to 0 in the frequency range 0.1–10 GHz.

The maximum obtainable frequency is limited to 10 GHz by the usable length of the cell. This maximum frequency can be increased, if we reduce this length by using longer Teflon joints.

The accuracy of this method has been studied in two different ways. On the one hand we have compared the obtained results with the theoretical values ε^* and μ^* of air; this way leads to an accuracy of 6% in ε_r' and μ_r' and to data not exceeding 5×10^{-2} for ε_r'' and μ_r'', throughout the frequency range. On the other hand we have compared the measurements with data obtained when the permittivity of Teflon ε_t and the three lengths l_1, l_2 and l vary within 2% (figure 6(a) and (b)) of their known values. This second method leads to an accuracy better than 5% for ε_r' and μ_r' and to the extreme value 3×10^{-2} for ε_r'' and μ_r''.

If we replace our encased Teflon joints by thin mica joints (0.05 mm) close to the right sides of the two conductors, we obtain more inaccurate measurements of ε_r^* and μ_r^*, as it can be seen in figures 5(a) and (b). Under the measurement conditions we observe that the discontinuity of conductors provides a discrepancy between the two recordings. Anomalies appear in ε_r^* and μ_r^* at 3.2 GHz. The accuracy of this method is less than 8% throughout the frequency range.
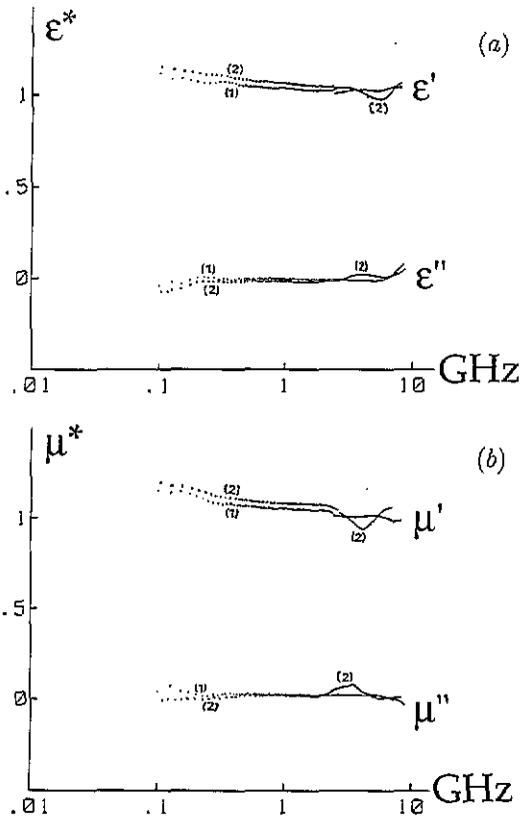
Figure 5. (a) Complex permittivity ε^* measured with an empty cell: (1) our method; (2) with thin mica joints. (b) Complex permeability μ^* measured with an empty cell: (1) our method; (2) with thin mica joints.
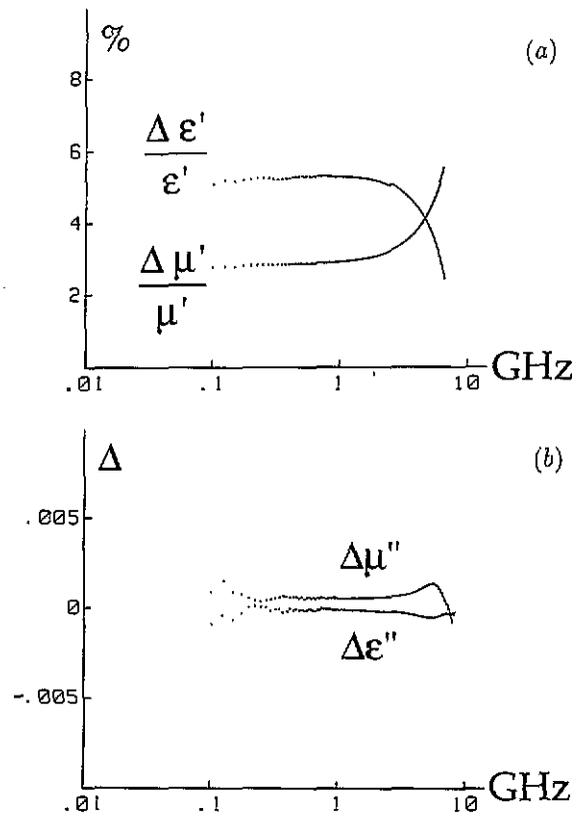
Figure 6. (a) Relative (percentage) deviations $\Delta\varepsilon'/\varepsilon'$ and $\Delta\mu'/\mu'$ when the permittivity of Teflon ε_t and the three lengths l_1, l_2 and l are varied by $\pm2\%$. (b) Errors in ε'' and μ'' when ε_t, l_1, l_2 and l are varied by $\pm2\%$.

4.2. Results

This method may be illustrated by the study of different liquids such as some alcohols or magnetic fluids, which are new technological materials.

The two studied alcohols are ethanol and propan-1-ol, measurements being made at 21.5 °C. The corresponding curves (figure 7) display evidence of the well-known dielectric relaxation of these polar liquids. These data on permittivity, static permittivity and position of the relaxation peak are in excellent agreement with those obtained by other authors from a time domain spectroscopy (TDS) method [9], and also with our previous results at lower frequencies and temperature [10]. We can quote as evidence the accordance between our measurements and data obtained from the TDS method, by plotting the normalized curves $\varepsilon_n'' = \varepsilon''/(\varepsilon_S - \varepsilon_\infty)$ versus $\varepsilon_n' = (\varepsilon' - \varepsilon_\infty)/(\varepsilon_S - \varepsilon_\infty)$ (figures 8(a) and (b)).

Magnetic fluids or ferrofluids are colloidal suspensions of magnetic particles in a carrier liquid. Our samples were prepared in Professor Massart's laboratory by the ionic method [11], using carbon tetrachloride (which is a non-polar solvent) as carrier liquid. We have recorded the relative complex permeability of the two following ferrofluids.

(i) Fe_2MnO_4 in CCl_4 (figure 9(a)); the mean diameter of these manganese ferrite particles is
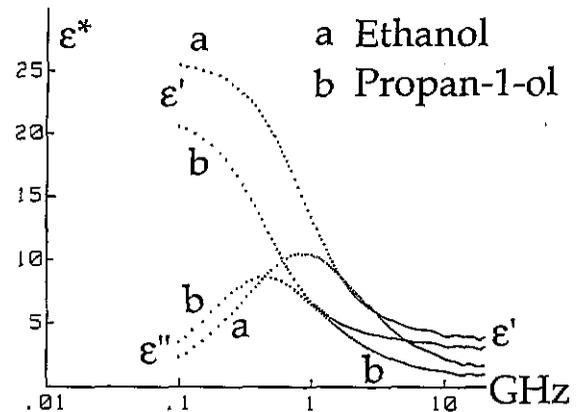
Figure 7. The complex permittivity of ethanol and propan-1-ol at 21.5 °C.

$d_m = 9.8$ nm and the distribution is Gaussian with a standard deviation $\sigma = 0.35$ nm.

(ii) Fe_2CoO_4 in CCl_4 with $d_m = 8.5$ nm and $\sigma = 0.5$ nm for the distribution of cobalt ferrite particles (figure 9(b)).

We detected two magnetic absorption peaks for the first sample (i) but none for the second (ii). The first peak indicates a Néel relaxation because, for a Brownian one, the relaxation frequency f_B would be less than 1 MHz for the considered particle sizes, and this relaxation
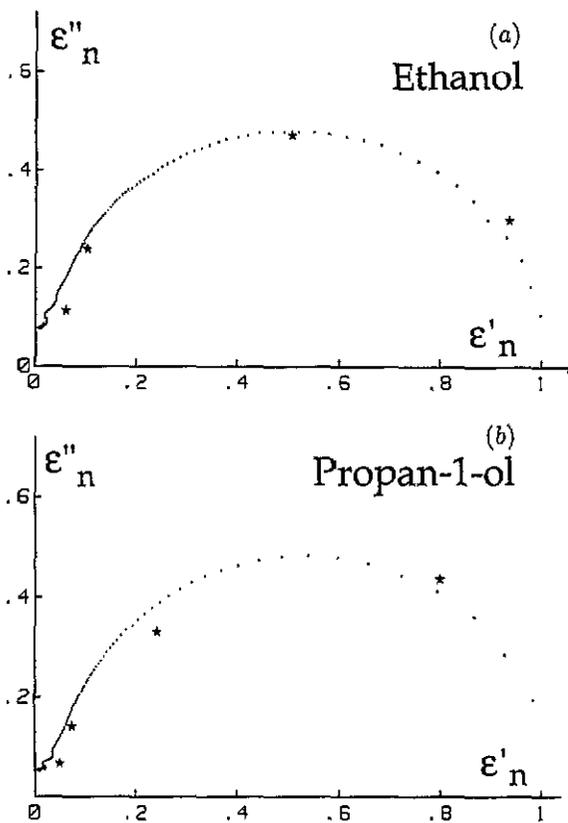
Figure 8. Normalized diagram of ε'' (ε'_n) for (a) ethanol and (b) propan-1-ol (★), data from [8].
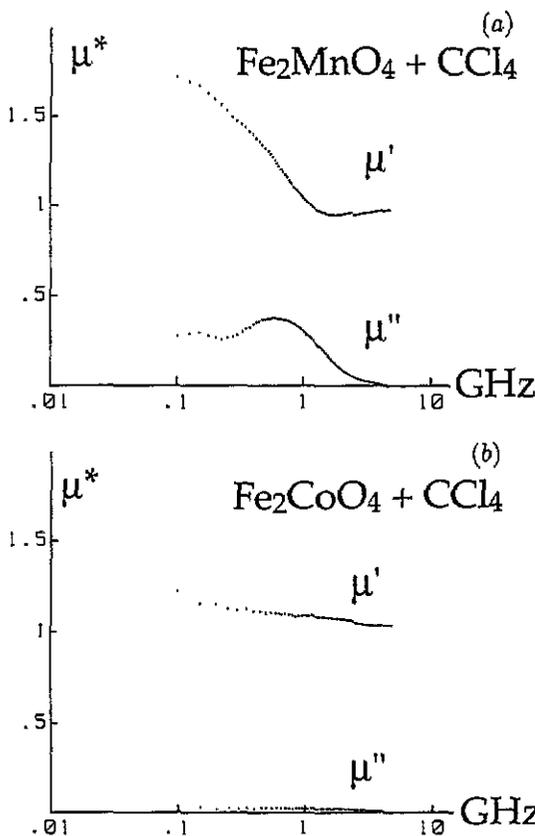
Figure 9. Recording of complex permeability for (a) Fe_2MnO_4 and (b) Fe_2CoO_4, in CCl_4 at 21.5 °C, showing for (a) a Néel relaxation and then a gyromagnetic resonance.

phenomenon would also appear for the second ferrofluid. Indeed, for a Brownian mechanism f_B is given by

$$f_B = \frac{1}{2\pi\tau_B} \qquad \tau_B = \frac{4\pi r^3 \eta}{kT} \qquad (22)$$

where r is the hydrodynamic radius of the particle, $\eta = 0.91 \times 10^{-3}$ Pa s^{-1} is the solvent viscosity, k is Boltzmann's constant and T the temperature in kelvins.

Consequently, the relaxation observed at 100 MHz and 21.5 °C for the manganese ferrite is probably of Néel type, the relaxation time then being given by

$$\tau_N = f_0^{-1} \exp\left(\frac{KV}{kT}\right) \qquad \frac{KV}{kT} \ll 1 \qquad (23)$$

$$\tau_N = f_0^{-1} \exp\left(\frac{KV}{kT}\right)\left(\frac{KV}{kT}\right)^{-1/2} \qquad \frac{KV}{kT} > 2 \quad (24)$$

where $f_0 \approx 10^{-9}$ s, V is the magnetic volume of the particle and K the magnetic anisotropy constant.

For Fe_2MnO_4, the constant K is about 4×10^3 J m^{-3} [11] giving $KV/kT \approx 0.47$, so to a first approximation using relation (23) we have $f_N = 1/(2\pi\tau_N) \approx 99$ MHz, which is close to the experimental value (about 110 MHz). For Fe_2CoO_4 we have $K \approx 2 \times 10^5$ J m^{-3} giving by relation (24) $f_N \approx 112$ Hz, which is outside the scanned frequency range.

The origin of the second absorption peak at about 600 MHz is probably the gyromagnetic resonance as suggested by Laurent *et al* [12], who have observed the same phenomenon with a sample γ-Fe_2O_3. The small increase in magnetic dispersion at the highest frequencies is quite in accordance with the presence of a resonance instead of a relaxation.

5. Conclusion

Results on ε^* or μ^* may be obtained, by this method, with only one coaxial line cell throughout the frequency range 0.1–2.0 GHz for polar liquids. For non-polar liquids we find the classical problem of uncertainty at some specific frequencies when the material length is a multiple of one half wavelength (that is, when $S_{11} \rightarrow 0$). For example, measurements on $CCl_4 + Fe_2CoO_4$, a previously studied ferrofluid, whose permittivity is about 3, also drop out at 7.5 GHz. Without particular treatment of the data, we can say that the frequency spectrum is limited to 7 GHz for non-polar liquids ($\varepsilon < 4$). To extend this range, without modification of the cell length, we should have to conceive an algorithm using an iterative calculus to bypass this difficulty that appears at the same frequency for some values of ε_r^* and μ_r^*.

Improvement of the Nicolson–Ross calculus for ε^* and μ^*, associated with a specific coaxial line cell, is a simple method of dielectric and magnetic characterization of liquids, which is well adapted to network analysers. Without a particular algorithm, it allows one to study most liquids up to 7 GHz.

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