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On the generation of complex susceptibility data through the use of the Hilbert transform

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Abstract. *It is shown that, by the use of the Hilbert transform, it is possible to obtain values of the imaginary component, $\chi''(\omega)$, of the* AC *magnetic susceptibility of a colloidal suspension of single-domain magnetic particles, from experimental measurements of the real component, $\chi'(\omega)$.*

1. Introduction

The real and imaginary components of the frequency-dependent, complex magnetic susceptibility of a ferrofluid, $\chi'(\omega)$ and $\chi''(\omega)$ respectively, can usually be obtained using a measurement technique such as the toroidal technique of Fannin *et al* [1]. However, occasions arise when, due to the existence of a very low level of susceptibility, it is possible to obtain good quality data for $\chi'(\omega)$ but not for $\chi''(\omega)$. Also, other methods [2] may not lend themselves to a simple or accurate determination of $\chi''(\omega)$. In these two cases, an alternative analytical technique to determine $\chi''(\omega)$ is readily available since $\chi'(\omega)$ and $\chi''(\omega)$ are a Hilbert transform pair.

In this paper we show that the application of the Hilbert transform technique does generate accurate data for $\chi''(\omega)$ from a knowledge of $\chi'(\omega)$. The technique has been applied here to magnetic fluids, but, of course, it holds for any linear magnetic system. On a point of interest, for the case of a spin glass, the literature [3, 4] reports on the determination of $\chi''(\omega)$ from a knowledge of $\chi'(\omega)$, with measurements being a function of temperature at spot frequencies.

2. Complex susceptibility

The theory developed by Debye [5] to account for the anomalous dielectric dispersion in dipolar fluids has been used [6, 7] to account for the analogous case of magnetic fluids. Debye's theory holds for spherical particles when the magnetic dipole–dipole interaction energy, U, is small compared with the thermal energy kT.

The complex frequency-dependent magnetic susceptibility, $\chi(\omega)$, may be written in terms of its real and imaginary components, where

$$\chi(\omega) = \chi'(\omega) - \mathrm{i}\chi''(\omega). \tag{1}$$

According to Debye's theory the complex susceptibility, $\chi(\omega)$, has a frequency dependence given by the equation

$$\chi(\omega) = \chi_\infty + (\chi_0 - \chi_\infty)/(1 + \mathrm{i}\omega\tau) \tag{2}$$

where χ_0 and χ_∞ indicate the values of susceptibility at $\omega = 0$ and at very high frequencies respectively. τ is the effective relaxation time with

$$\tau = 1/\omega_\mathrm{m} = 1/2\pi f_\mathrm{m} \tag{3}$$

where f_m is the frequency at which $\chi''(\omega)$ is a maximum.

This may be expanded to show that

$$\chi'(\omega) = \chi_\infty + (\chi_0 - \chi_\infty)/(1 + \omega^2\tau^2) \tag{4}$$

and

$$\chi''(\omega) = \omega\tau(\chi_0 - \chi_\infty)/(1 + \omega^2\tau^2). \tag{5}$$

It follows that

$$\chi''(\omega) = \omega\tau(\chi'(\omega) - \chi_\infty). \tag{6}$$

For the Debye relationship, the relaxation time τ can be found by determining, from the $\chi'(\omega)$ plot, the value of ω_m at $\chi'(\omega) = (\chi_0 - \chi_\infty)/2$, for which $\omega_\mathrm{m}\tau = 1$. Alternatively this frequency, $\omega_\mathrm{m}/2\pi$, also corresponds

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to the point at which the slope of the $\chi'(\omega)$ curve is a maximum.

Thus it is a simple matter to generate the $\chi''(\omega)$ curve by multiplying values of $(\chi'(\omega) - \chi_\infty)$ by $\omega\tau$.

The above case for Debye is a particularly simple example. However for situations where we have a distribution of relaxation times, it is not possible to generate susceptibility curves by the method described above. In such cases, since the system is linear and causal, the real and imaginary components of $\chi(\omega)$ are Hilbert transform pairs [8, 9]. A brief explanation of the Hilbert transform follows.

3. Hilbert transform

The Hilbert transform of a function $x(t)$ is defined as

$$\hat{x}(t) = 1/\pi \int_{-\infty}^{\infty} x(\tau)/(t-\tau)\,\mathrm{d}\tau \tag{7}$$

$$= (1/\pi t)^{*} x(t) \tag{8}$$

where $*$ represents a convolution operation. This operation corresponds to phase-shifting all frequency components of $x(t)$ by 90° and it may be represented by a linear system with an impulse response of

$$h(t) = 1/\pi t \tag{9}$$

and transfer function

$$H(f) = -\mathrm{i}\,\mathrm{sgn}(f) \qquad f = \omega/2\pi. \tag{10}$$

As convolution in the time domain is equivalent to multiplication in the frequency domain, equation (8) may be written as

$$\hat{X}(f) = H(f)X(f) \tag{11}$$

where $\hat{X}(f)$, $H(f)$ and $X(f)$ are the Fourier transforms of $\hat{x}(t)$, $h(t)$ and $x(t)$ respectively. $\hat{X}(f)$ and $\hat{x}(t)$ are related by the expression

$$\hat{x}(t) = \int_{-\infty}^{\infty} \hat{X}(f)\mathrm{e}^{\mathrm{j}2\pi f t}\,\mathrm{d}f \tag{12}$$

and from equation (11), assuming that $X(f)$ is an even function, it follows that

$$\hat{x}(t) = 2\int_{0}^{\infty} X(f)\sin 2\pi f\,\mathrm{d}f \tag{13}$$

Consider the example of the function [10]

$$x(t) = 1/(1+t^2). \tag{14}$$

This function has a Fourier transform of

$$X(f) = \pi\mathrm{e}^{-|2\pi f|} \tag{15}$$

and from equation (13) the corresponding Hilbert transform is

$$\hat{x}(t) = t/(1+t^2). \tag{16}$$

It is of interest to note that equations (14) and (16) have the same form as the frequency-dependent term of $\chi'(\omega)$ in equation (4) and $\chi''(\omega)$ in equation (5) respectively. The components χ_0 and χ_∞ are constants, and since the Hilbert transform of a constant is zero, it is clear that $\chi''(\omega)$ of equation (5) is indeed the Hilbert transform of $\chi'(\omega)$.

3.1. Realization of the Hilbert transform

The Hilbert transform may be implemented in the frequency domain by direct computation of equation (8). For discrete samples, this becomes a matrix multiplication:

$$X[\hat{K}] = h[K] \times [K]. \tag{17}$$

$h[K]$ is an n by n matrix, whose elements are given by $1/(u+v-1)\pi\Delta f$, where u and v are the row and column indices respectively, and Δf is the frequency increment which cannot be greater than the start frequency. The computation of this formula is very time consuming, since it requires of the order of n^2 multiplications and additions. Such a computational burden is greatly reduced by implementing equation (11). This involves calculating the inverse fast Fourier transform (IFFT) of $\chi'(f)$, multiplying by $-\mathrm{i}\,\mathrm{sgn}(t)$ and calculating the Fourier transform of the result, as illustrated in the following flow chart:

$$|\chi'(f)| \xrightarrow{\text{IFFT}} |\chi'(t)| \xrightarrow{-\mathrm{i}\,\mathrm{sgn}(t)} |\hat{\chi}'(t)| \xrightarrow{\text{FFT}} |\hat{\chi}'(f)|.$$

This above method was used to obtain the Hilbert transforms presented in this paper.

In a practical measurement, $\chi'(f)$ is measured at evenly spaced intervals from a start frequency of Δf (Hz) to a frequency f_∞ (Hz); the latter being the frequency where χ_∞ is deemed to have been reached. Also, the start frequency must be equal to, or be a multiple of, the incremental frequency. The technique also requires data at zero Hz and so it is necessary to either make a measurement at DC or to assume that the zero-frequency data are the same as those obtained at the first measurement point. Furthermore, since the FFT technique requires the signal to be transformed to be periodic, the negative of $\chi'(f)$ from 0 to $-f_\infty$ (Hz) must be generated. This is a simple matter since $\chi'(f)$ is an even function of f and is easily implemented in software.

A factor which should be considered when choosing the incremental measurement frequency Δf and the final frequency f_∞ is the fact that the limits of integration of the integral in equation (7) are from $\pm_\infty$. In practice this is limited to $\pm f_\infty$, which is equivalent to multiplying $\chi'(f)$ by a rectangular window. Other types of windows, e.g. Hamming or Triangular, may be used in order to minimize the effects of the rectangular window.

3.2. Application of Hilbert transform technique

To test the accuracy of the proposed technique it was firstly applied to theoretical Debye curves, where one knew what the resultant Hilbert transform should be. It was then tested on data obtained in a dynamic situation.

Initially equation (2), with $\chi_\infty = 0$, was plotted for $n = 8192$ points and the resultant plots of $\chi''(\omega)$, $\chi'(\omega)$ and its Hilbert transform $\hat{\chi}'(\omega)$ are shown in figure 1. It can be seen that $\hat{\chi}'(\omega)$ is indeed a good approximation to $\chi''(\omega)$, with some error existing in the low-frequency region of the plots. This error was reduced significantly

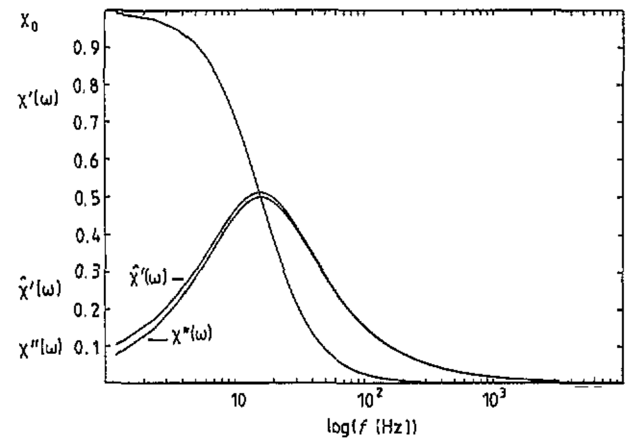

Figure 1. A plot of $\chi''(\omega)$, $\chi'(\omega)$ and its Hilbert transform $\hat{\chi}'(\omega)$, against log(f in Hz), for the Debye case with $\chi_\infty = 0$ and $n = 8192$ points.

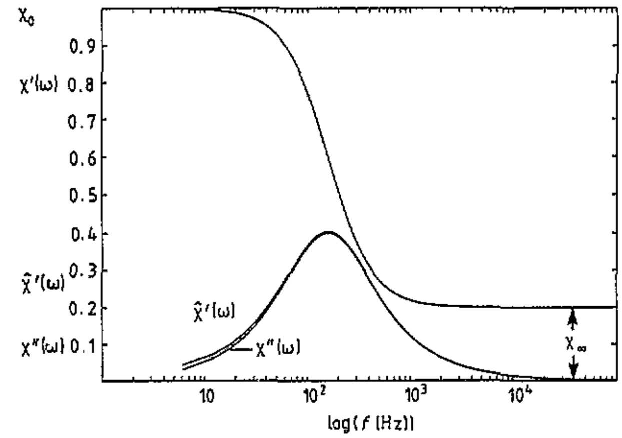

Figure 3. A plot of $\chi''(\omega)$, $\chi'(\omega)$ and its Hilbert transform $\hat{\chi}'(\omega)$, against log(f in Hz), for the Debye case with $\chi_\infty = 0.2$ and $n = 8192$ points.

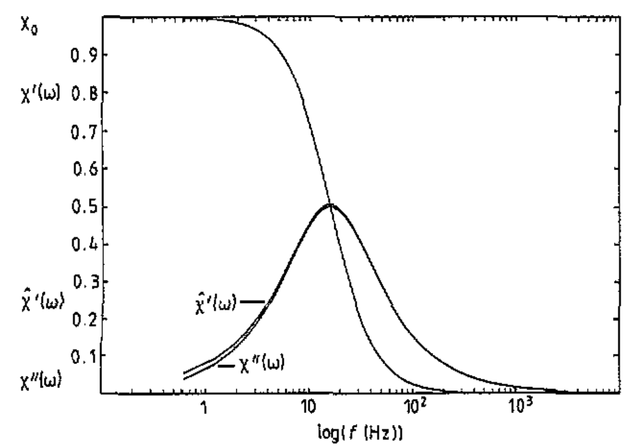

Figure 2. A plot of $\chi''(\omega)$, $\chi'(\omega)$ and its Hilbert transform $\hat{\chi}'(\omega)$, against log(f in Hz), for the Debye case with $\chi_\infty = 0$ and $n = 16\,384$ points.

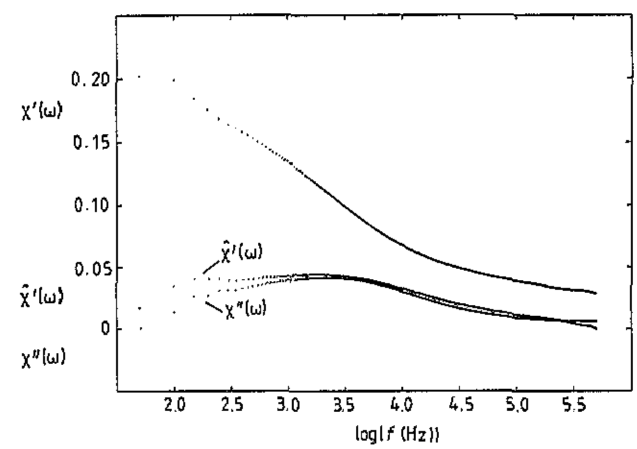

Figure 4. A plot of $\chi''(\omega)$, $\chi'(\omega)$ and its Hilbert transform $\hat{\chi}'(\omega)$, against log(f in Hz), for a ferrofluid sample.

by doubling the number of points to 16 384, as shown by the corresponding curves in figure 2.

The technique was then applied to equation (2) with $\chi_\infty = 0.2$ and $n = 8192$; the corresponding plots of $\chi''(\omega)$, $\chi'(\omega)$ and its Hilbert transform $\hat{\chi}'(\omega)$ are shown in figure 3. Again the Hilbert transform, $\hat{\chi}'(\omega)$, proved to be an excellent approximation to $\chi''(\omega)$.

Thus, having shown the technique to be successful for theoretically generated data, it was then applied to data obtained for a ferrofluid.

Measurements were made on a ferrofluid consisting of a colloidal suspension of single-domain cobalt ferrite particles with a log–normal volume distribution of median diameter 10.3 nm and standard deviation 0.54, dispersed in a perfluoro carrier. The process of relaxation of the magnetic moment of these particles is dominated by the Brownian relaxation mechanism

$$\tau_B = 4\pi\eta r^3/kT \tag{18}$$

where r is the hydrodynamic radius of the particle and η is the dynamic viscosity of the carrier liquid. This arises because the magnetic moment of most of the particles is not free to rotate within the particles i.e. it is 'blocked' because of the high-energy barrier (KV) to rotation. $K (= 2 \times 10^5$ J m^{-3}) is the effective anisotropy constant (a combination of shape and crystalline anisotropies) and V is the median magnetic volume of the particles.

Measurements were performed over the approximate frequency range 50 Hz to 500 kHz in steps of 50 Hz and the resulting plots of $\chi''(\omega)$, $\chi'(\omega)$ and $\hat{\chi}'(\omega)$ are shown in figure 4. $\hat{\chi}'(\omega)$ proved to be a very good approximation to $\chi''(\omega)$, with ω_{max} for both curves being identical. As with the case of the theoretical data, some error does exist, particularly in the low-frequency region, however this error can be reduced by reducing the size of the incremental frequency, Δf. At high frequencies the onset of the 'window' effect can be seen with $\hat{\chi}'(\omega)$ crossing and becoming smaller than $\chi''(\omega)$.

Regarding the experimental data, one should note the difference in broadness (or bandwidth) of the $\chi''(\omega)$ peak in figure 4, and that of the Debye profile in figure 1. This difference is due, in part, to the fact that the colloidal suspension consists of a distribution of relaxation times; it is also indicative of the level of system damping prevalent, with the former profile representing the more heavily damped system.

4. Conclusion

It has been demonstrated, both in a theoretical and practical situation (involving a system of single-domain particles), that the use of the Hilbert transform does indeed generate accurate data for $\chi''(\omega)$ from a knowledge of $\chi'(\omega)$, thus verifying the usefulness of the technique, particularly in a dynamic situation. However, should the need arise, the technique can also be used to generate data on $\chi'(\omega)$ from a knowledge of $\chi''(\omega)$, simply by taking the inverse Hilbert transform of $\chi''(\omega)$. This is due to the fact that the Hilbert transform of a Hilbert transform returns the original signal with a change in sign [11].

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