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Theory of Temperature Modulation Studies of Ferromagnetic Materials

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Abstract

A theoretical account is given of temperature modulation experiments on a ferromagnet sample in which the resulting voltage induced in a pick-up coil is monitored by phase-sensitive detection. Calculations are made of the effects of magnetic relaxation and of the thermal skin depth on the signal amplitude and phase. The relaxation calculations are shown to be consistent with recent experimental data for gadolinium.

Introduction

Recently we (Chaplin *et al.* 1973) reported the application of a new technique involving temperature modulation and phase-sensitive detection to determine the temperature derivatives of the DC magnetization M and AC susceptibility χ of the ferromagnetic metal gadolinium near its Curie temperature. The differential susceptibility curves always exhibited a satellite peak corresponding to an inflection in the $\chi(T)$ curve a few degrees below the Curie point and, in a further study of the AC susceptibility and coercive field using conventional techniques (Sydney *et al.* 1974), it was shown that this satellite peak was associated with domain nucleation. A fuller description of the experimental results for gadolinium is being published elsewhere (Sydney *et al.* 1976). Below the Curie point, non-equilibrium effects associated with relaxation of domain properties are important. In the present paper we outline a general theory of temperature modulation experiments including the effects of magnetic relaxation and of the thermal skin depth.

To introduce the bases of the experiments we first neglect the effects of relaxation and of the skin depths, i.e. it is assumed that at any time every point in the sample is at the same temperature T and experiences the same applied field. The oscillatory term in the temperature is then given by the solution of

$$mC\,\mathrm{d}T/\mathrm{d}t + bT = P\cos(\omega t), \tag{1}$$

where m and C are the mass and specific heat respectively of the sample and b is the Newton's law of cooling constant for the sample in its environment. The oscillatory heat input $P\cos(\omega t)$ is provided by an electrical heater wound over the sample. If a sinusoidal heater voltage is used without DC offset then ω is at the second harmonic of the voltage, whereas by employing a DC offset a power term at the frequency of the voltage waveform is introduced. The solution of equation (1) leads in the steady state to a temperature modulation

$$T = \bar{T} + \Delta_T \cos(\omega t + \phi_T), \tag{2}$$

where $\bar{T}$ is the mean temperature. The modulation amplitude is

$$\Delta_T = P(b^2 + \omega^2 m^2 C^2)^{-\frac{1}{2}} \tag{3}$$

and there is a thermal phase lag given by

$$\phi_T = -\arctan(\omega m C/b). \tag{4}$$

The rise δT in the mean temperature above ambient is given by

$$\delta T = \bar{P}/b,$$

where $\bar{P}$ is the average heater power and, from this, values for b may be obtained by plotting the mean steady-state temperature versus $\bar{P}$.

In the DC experiments an applied DC magnetic field $\boldsymbol{H}$ is used to create a sample magnetization $M(T)$. The temperature modulation produces a small oscillation in M, and the resulting voltage induced in a pick-up coil is monitored by phase-sensitive detection. We treat this in terms of the oscillation in the susceptibility $\chi = M/H$, which is defined here for any field H. By means of a Taylor expansion, the time dependence of $\chi(t)$ may be expressed to first order as

$$\chi(t) = \chi(\bar{T}) + \Delta_T \chi^1(\bar{T}) \cos(\omega t + \phi_T), \tag{5}$$

where χ^1 is the temperature derivative. The signal (the voltage induced in the pick-up coil) is given by

$$v = -\xi V \mathrm{d}M/\mathrm{d}t,$$

where V is the sample volume and ξ is determined by the number of turns, the coil dimensions and the sample filling factor. Hence we have

$$v = \xi \Delta_T V \omega H \chi^1(\bar{T}) \cos(\omega t + \phi_T - \tfrac{1}{2}\pi). \tag{6}$$

When, as is usual, $b \ll \omega m C$, this becomes (from equation 3)

$$v = \{\xi V P H \chi^1(\bar{T})/mC\} \cos(\omega t + \phi_T - \tfrac{1}{2}\pi), \tag{7}$$

and the signal amplitude is independent of the modulation frequency. Generally only very small modulation amplitudes ($\sim$mK) are used, so that the expansion (5) is valid. If larger values are used, so that the signal leads to modulation-broadened curves, instead $\chi^1(\bar{T})$ of equation (5) should be replaced by a Fourier integral (Wilson 1963).

In the AC experiments the temperature modulated sample is used as the core of an AC transformer operating at a carrier frequency $\omega_c \gg \omega$. If the amplitude of magnetic field produced by the primary coil is H_0, the sample magnetization becomes

$$M(t) = \chi(T) H_0 \cos(\omega_c t) \tag{8}$$

and the Taylor expansion leads to

$$M(t) = H_0\{\chi(\overline{T}) + \Delta_T \chi^1(\overline{T})\cos(\omega t + \phi_T)\}\cos(\omega_c t). \tag{9}$$

Neglecting the contribution $\mu_0\, \mathrm{d}H/\mathrm{d}t$, which is not modulated, the secondary voltage is given by

$$\begin{aligned} -\xi V \mathrm{d}M/\mathrm{d}t &= \xi V H_0\, \omega_c\, \chi(\overline{T}) \sin(\omega_c t) \\ &\quad + \xi V H_0\, \Delta_T\, \chi^1(\overline{T})\{\omega_c \cos(\omega t + \phi_T)\sin(\omega_c t) + \omega \sin(\omega t + \phi_T)\cos(\omega_c t)\}, \end{aligned}$$

for which the amplitude-modulation signal after detection is, for $\omega_c \gg \omega$,

$$v = \xi V H_0\, \Delta_T\, \omega_c\, \chi^1(\overline{T})\cos(\omega t + \phi_T) \tag{10}$$

or, assuming that $b \ll \omega m C$,

$$v = \{\xi P V H_0\, \omega_c\, \chi^1(\overline{T})/mC\omega\}\cos(\omega t + \phi_T). \tag{11}$$

A comparison between equations (7) and (11) shows the main advantage of the AC technique—a signal enhancement of order ω_c/ω.

By measuring the modulation amplitude Δ_T as a function of $\overline{T}$, for example, by detection of the oscillatory e.m.f. from a thermocouple, the contribution of the specific heat to the signal amplitude may be allowed for and the derivative $\chi^1(T)$ obtained. Before carrying out the experiments on gadolinium we had expected simultaneously to obtain accurate data for the specific heat anomaly near the Curie temperature from measurements of the phase lag ϕ_T. However, in our application of modulation and conventional techniques to study the AC and DC susceptibility in the region of the Curie point, it was observed that relaxation effects dominated the observed signal phase, producing a phase lead. These effects are treated in the next section.

Equations (7) and (11) for v give the signals for DC and AC experiments in which phase-sensitive detection at the modulation frequency is used. In general, detection at the nth harmonic of the modulation frequency yields the nth derivative of $\chi(T)$ because of the expansion (Russell and Torchia 1962)

$$\chi(T) = \chi(\overline{T}) + \sum_{n=1}^{\infty} \Delta_T^n (2^{1-n}/n!)\, \chi^{(n)}(\overline{T}) \cos\big(n(\omega t + \phi_T)\big), \tag{12}$$

where $\chi^{(n)}$ represents the nth derivative. Another method of determining the temperature modulation amplitude is as follows: If experiments are performed by detecting at both the first and second harmonics, the amplitude of the second-harmonic signal versus $\overline{T}$ will be equal to $\frac{1}{4}\Delta_T$ multiplied by the derivative of the corresponding first-harmonic curve.

Relaxation Effects

Here we treat the effects of magnetic relaxation on the signals. It is still assumed that there are a uniform temperature and an applied field at all points in the sample at any given time. The treatment of relaxation effects on both the DC and AC experiments is identical, involving only the time dependence of the susceptibility.

Simple Relaxation

For 'simple relaxation' it is assumed that, as the temperature is modulated, χ is always relaxing exponentially towards its equilibrium value $\chi_e(T)$ with a relaxation time τ, so that we have

$$d\chi/dt = -\{\chi(t)-\chi_e(T)\}/\tau. \tag{13}$$

Such simple behaviour would not be expected for a ferromagnet below the Curie temperature, but a comparison of the observed behaviour with that expected for simple relaxation will still be worth while. Simple relaxation would be expected in temperature modulation experiments on low temperature nuclear magnetizations.

By substituting a Taylor expansion of $\chi_e(T)$ into equation (13) it follows that

$$\chi(t) = \chi_e(\bar{T}) + \sum_{n=1}^{\infty} \Delta_T^n(2^{1-n}/n!)\alpha_n \chi_e^{(n)}(\bar{T})\cos\bigl(n(\omega t+\phi_T)-\phi_n\bigr), \tag{14}$$

where $\phi_n = \arctan(n\omega\tau)$ and $\alpha_n = (1+n^2\omega^2\tau^2)^{-\frac{1}{2}}$. Comparing this result with equation (12), we see that the relaxation will result in a reduction of the nth-harmonic signal amplitude by the factor α_n and in an additional phase lag ϕ_n.

Relaxation of Domain Properties

In applying temperature modulation techniques to study a ferromagnetic material near the Curie temperature T_c, applied fields which are small in comparison with the exchange fields will generally be used so that the intrinsic magnetic ordering is then observed. Above T_c exchange-enhanced paramagnetism with a field-independent susceptibility is then expected, while below T_c the response to the applied field will be determined by the intrinsic magnetization $\sigma(T)$ and by the domain properties. We treat here the case where the response of σ to the temperature modulation is fast enough for it to be always in thermal equilibrium, but with relaxation effects arising from the domain properties.

We write, for the equilibrium susceptibility,

$$\chi_e(H,T) = G_e(H,T)\,\chi_N(H,T), \tag{15}$$

where χ_N is the susceptibility which would be observed if there were no domains present. Hence G_e represents the effects of the domains upon the thermal equilibrium susceptibility. Writing $\chi_N(H,T) = \sigma(T)/H$ it follows that the equilibrium magnetization is

$$M_e(H,T) = \sigma(T)\,G_e(H,T). \tag{16}$$

When the applied field is large enough to magnetically saturate the sample, i.e. there are no domains present, equation (16) shows that $G_e = 1$. When, on the other hand, H is sufficiently small for the linear reversible domain-wall displacement to dominate the susceptibility, then M_e will be proportional to H and we may write, by equation (16): $G_e(H,T) = g_e(T)H$, where $g_e(T)$ represents the temperature dependence of the domain wall mobility. Then, defining in the same way an instantaneous g, it follows that in a temperature modulation experiment the time dependence of χ is given by $\chi(t) = g(t)\,\sigma(T)$.

We assume that, when the temperature is changing, g undergoes exponential relaxation with a time constant τ, that is, we have

$$\mathrm{d}g/\mathrm{d}t = -\{g(t) - g_{\mathrm{e}}(T)\}/\tau$$

and hence, as for χ in the preceding subsection, we obtain

$$g(t) = g_{\mathrm{e}}(\bar{T}) + \sum_{n=1}^{\infty} \Delta_T^n (2^{1-n}/n!)\, g_{\mathrm{e}}^{(n)}(\bar{T})\, \alpha_n \cos\bigl(n(\omega t + \phi_T) - \phi_n\bigr). \tag{17}$$

Assuming that σ is always in thermal equilibrium, we have

$$\sigma(t) = \sigma(\bar{T}) + \sum_{n=1}^{\infty} \Delta_T^n (2^{1-n}/n!)\, \sigma^{(n)}(\bar{T}) \cos\bigl(n(\omega t + \phi_T)\bigr). \tag{18}$$

For nth-harmonic detection we consider only the terms in the product $g(t)\sigma(t)$ which have frequency $n\omega$. For first-harmonic detection this leads to

$$\chi_1(t) = \Delta_T\{g\sigma' \cos(\omega t + \phi_T) + \sigma g' \alpha_1 \cos(\omega t + \phi_T - \phi_1)\}, \tag{19}$$

where the values of σ, g and their temperature derivatives all correspond to the equilibrium functions for the mean temperature $\bar{T}$. For slow modulation ($\omega\tau \to 0$) we have $\chi_1(t) \to \Delta_T \chi_{\mathrm{e}}^1(\bar{T}) \cos(\omega t + \phi_T)$, which as expected is the same as is given by equation (5) for the case of no relaxation effects. For fast modulation ($\omega\tau \to \infty$) we have

$$\chi_1(t) \to \Delta_T g_{\mathrm{e}}(\bar{T})\, \sigma'(\bar{T}) \cos(\omega t + \phi_T), \tag{20}$$

and here also there is no phase shift caused by the relaxation, with g remaining equal at all times to $g_{\mathrm{e}}(\bar{T})$ and with the signal being produced solely by the oscillation in σ. Equation (19) may also be written as

$$\chi_1(t) = \Delta_T\{(g\sigma' + \sigma g' \alpha_1^2)^2 + (g'\sigma\omega\tau\alpha_1^2)^2\}^{\frac{1}{2}} \cos(\omega t + \phi_T + \phi_{\mathrm{r}}), \tag{21}$$

where the phase shift ϕ_{r} due to the relaxation is given by

$$\tan(\phi_{\mathrm{r}}) = -\sigma g_{\mathrm{e}}' \omega\tau / (\chi_{\mathrm{e}_{\mathrm{i}}}^1 + g\sigma'\omega^2\tau^2). \tag{22}$$

It is instructive to examine the effects of this relaxation at temperatures where $\chi_{\mathrm{e}}^1 = 0$. Here, in the absence of any relaxation effects ($\omega\tau = 0$), the first-harmonic signal should be zero. Zero values of χ_{e}^1 will occur when the contributions from the temperature dependences of σ and g cancel, i.e. when $g\sigma' = -\sigma g'$. Relaxation of $g(t)$ will introduce an imbalance, and the pair of equations (21) and (22) shows that the signal amplitude will then be nonzero with a phase shift given by

$$\tan(\phi_{\mathrm{r}}) = 1/\omega\tau.$$

For slow modulation ($\omega\tau \ll 1$) small signal amplitudes with a large phase lead will result. For fast modulation ($\omega\tau \gg 1$) there will be large, almost in-phase ($\phi_{\mathrm{r}} \approx 0$), signals.

Fig. 1 shows experimental curves for gadolinium which give the temperature dependence of (*a*) the initial field-independent AC susceptibility and (*b*) the signal amplitude and (*c*) phase of the AC first-harmonic modulation signal. These curves were obtained for a monotonically increasing temperature; near the Curie temperature there is considerable temperature hysteresis in the domain nucleation (Sydney *et al.* 1974). The form of the modulation curves is well explained in terms of the relaxation of domain properties. The decrease in susceptibility as the temperature is lowered from 291·9 to 290·0 K has been ascribed to the onset of domain nucleation (Sydney

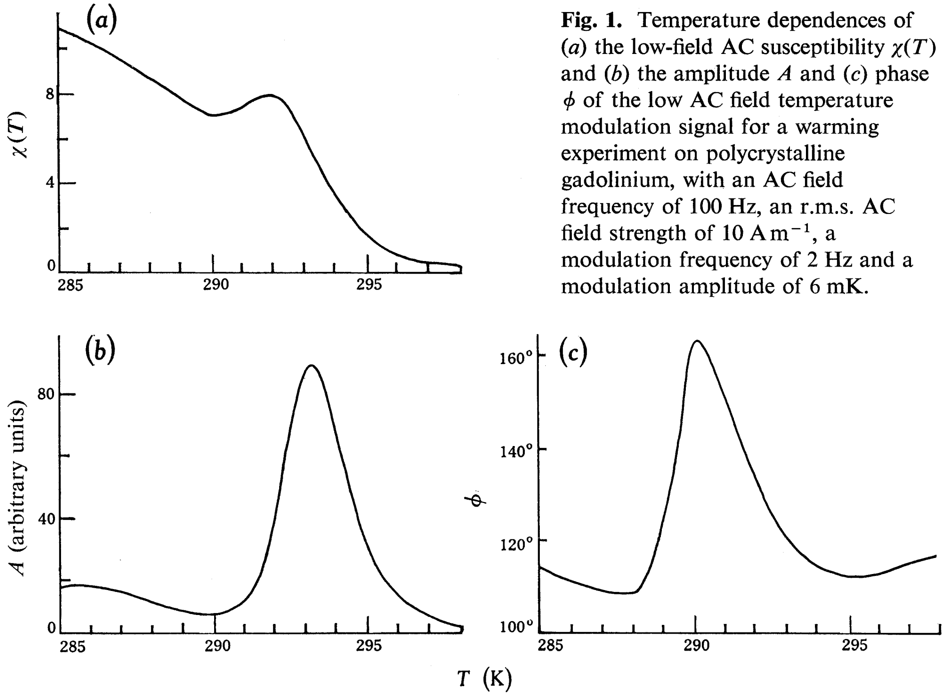

Fig. 1. Temperature dependences of (*a*) the low-field AC susceptibility $\chi(T)$ and (*b*) the amplitude A and (*c*) phase ϕ of the low AC field temperature modulation signal for a warming experiment on polycrystalline gadolinium, with an AC field frequency of 100 Hz, an r.m.s. AC field strength of 10 $\mathrm{A\,m^{-1}}$, a modulation frequency of 2 Hz and a modulation amplitude of 6 mK.

et al. 1974). For $T \gtrsim 292 \cdot 5$ K, the sample is paramagnetic without any domains present. In this region the modulation amplitude varies as the derivative $\chi^1(T)$ with a constant phase angle in a manner characteristic of fast relaxation. At 291·9 K, where the derivative $\chi^1(T) = 0$, there is a large modulation signal but only a small phase lead over that in the paramagnetic region. This is characteristic of extremely slow relaxation so that the signal is produced almost solely by the modulation of σ. At 290·0 K the derivative $\chi^1(T)$ is again zero but here there is a small signal amplitude and a large phase lead corresponding to relatively fast relaxation. We conclude that the domain properties responsible for these effects relax very slowly at the Curie point where nucleation is commencing, with much faster relaxation at lower temperatures. Full details of the modulation and susceptibility measurements for gadolinium are being published elsewhere (Sydney *et al.* 1976).

For second-harmonic detection, collection of terms in $g(t)\sigma(t)$ with frequency 2ω leads to

$$\chi_2(t) = \tfrac{1}{4}\Delta_T^2\{\alpha_2\,\sigma g''\cos(2(\omega t+\phi_T)-\phi_2) + 2\alpha_1\,g'\sigma'\cos(2(\omega t+\phi_T)-\phi_1) + g\sigma''\cos(2(\omega t+\phi_T))\}. \qquad (23)$$

For slow modulation ($\omega\tau \to 0$) we have

$$\chi_2(t) \to \tfrac{1}{4}\Delta_T^2 \chi_e^{(2)}(\bar{T}) \cos\big(2(\omega t + \phi_T)\big)$$

in agreement with equation (12). For fast modulation ($\omega\tau \to \infty$) we have

$$\chi_2(t) \to \tfrac{1}{4}\Delta_T^2 g(\bar{T})\, \sigma''(\bar{T}) \cos\big(2(\omega t + \phi_T)\big)\,.$$

Note that, as the mean temperature is varied, a phase reversal of the second-harmonic signal is expected near the Curie point in the limits of both slow and fast modulation.

Effects of Thermal Skin Depth

We consider here the effects of the thermal skin depth δ upon the signal amplitude and phase for both DC and AC experiments. The role of the electromagnetic skin depth can be eliminated in AC experiments by choosing an AC frequency ω_c sufficiently low so that this skin depth is large compared with the sample dimensions. The effect of the thermal skin depth will be to produce a variation of the temperature-modulation amplitude and phase with position in the sample. The signal for a DC experiment is then given by

$$v = -\xi H \chi'(\bar{T})\, \mathrm{d}\langle T - \bar{T}\rangle/\mathrm{d}t \tag{24}$$

and for an AC experiment by

$$v = -\xi H_0\, \omega_c\, \chi'(\bar{T}) \langle T - \bar{T}\rangle\,, \tag{25}$$

where

$$\langle T - \bar{T}\rangle = \int (T - \bar{T})\, \mathrm{d}V\,. \tag{26}$$

As was seen in the Introduction, the effect of the time derivative in equation (24) is to introduce an additional factor of ω and a phase shift of $\frac{1}{2}\pi$ so that we now consider only the average modulation term $\langle T - \bar{T}\rangle$. The treatment of the Introduction applies to the case where δ is much greater than the sample radius.

Before treating the more general case of cylindrical symmetry we first consider the plane wave solution which should apply whenever δ is much smaller than the sample radius. The equation for planar geometry is

$$\frac{\partial T}{\partial t} = \frac{K}{\rho C} \frac{\partial^2 T}{\partial x^2}\,, \tag{27}$$

where x is the distance from the surface, and K and ρ are the thermal conductivity and density respectively. Using phasor notation, the steady state oscillatory term in T may be written as

$$\boldsymbol{T}(x,t) = \boldsymbol{\theta} \exp\big(\mathrm{i}\omega t - (\mathrm{i}+1)kx\big)\,, \tag{28}$$

where

$$k = (\rho\omega C/2K)^{\frac{1}{2}} = 1/\delta\,. \tag{29}$$

The phasor $\boldsymbol{\theta}$ gives the amplitude and phase of the oscillation and is determined by the boundary condition

$$P' \cos(\omega t) = b'\, T(0,T) - K(\partial T/\partial x)_{x=0}\,, \tag{30}$$

which relates the heater power to the heat lost to the surroundings and the heat entering unit surface area of the sample. Here P' and b' are normalized so that $P' = P/A$ and $b' = b/A$, where A is the surface area. By substituting the phasor (28) into equation (30) we obtain the steady state solution

$$T = \bar{T} + \Delta_T \exp(-kx)\cos(\omega t - kx + \phi_T), \tag{31}$$

where the surface modulation amplitude and phase are given by

$$\Delta_T = P'(b'^2 + 2b'kK + 2k^2K^2)^{-\frac{1}{2}} \tag{32}$$

and

$$\phi_T = \arctan\big(kK/(b' + kK)\big). \tag{33}$$

The average modulation is given by

$$\langle T - \bar{T} \rangle = A \int_0^\infty (T - \bar{T})\,\mathrm{d}x,$$

leading to

$$\langle T - \bar{T} \rangle = (A\delta/\sqrt{2})\Delta_T \cos(\omega t + \phi_T - \tfrac{1}{4}\pi). \tag{34}$$

The main differences between this and the result (2) for the case of a large skin depth are the effective volume factor $(A\delta/\sqrt{2})$, the difference between equations (4) and (33) for ϕ_T, and the additional phase lag of $\frac{1}{4}\pi$ resulting from the integration over the thermal wave. The dependence of phase upon the specific heat is now such that, as $\rho\omega CK/b'^2 \to 0$ and ∞, the total phase $(\phi_T - \frac{1}{4}\pi) \to -\frac{1}{4}\pi$ and $-\frac{1}{2}\pi$ respectively, compared with the case of a large skin depth where the phase $\phi_T \to 0$ and $-\frac{1}{2}\pi$ as $\omega mC/b \to 0$ and ∞ respectively.

For the more general case of cylindrical symmetry, with a cylindrical or toroidal sample, the differential equation is then

$$\frac{\partial T}{\partial t} = \frac{K}{\rho C}\frac{\partial^2 T}{\partial r^2} + \frac{1}{r}\frac{\partial T}{\partial r}, \tag{35}$$

where T is the temperature at a radial distance r from the centre of the cylinder of radius R. The steady state oscillatory term in T may be written as (e.g. Arparci 1966):

$$\boldsymbol{T}(r,t) = \boldsymbol{\theta} \exp(\mathrm{i}\omega t)\, \mathrm{I}_0\big(\sqrt{(2\mathrm{i})}\,kr\big)/\mathrm{I}_0\big(\sqrt{(2\mathrm{i})}\,kR\big), \tag{36}$$

where the I_j are modified Bessel functions. The phasor $\boldsymbol{\theta}$ is determined from the boundary condition

$$P' \exp(\mathrm{i}\omega t) = b'\,\boldsymbol{T}(R,t) + K(\partial \boldsymbol{T}/\partial r)_{r=R} \tag{37}$$

leading to

$$\boldsymbol{T}(r,t) = P' \exp\big(\mathrm{i}(\omega t + \phi)\big)\, \mathrm{I}_0\big(\sqrt{(2\mathrm{i})}\,kr\big)\,(X^2 + Y^2)^{-\frac{1}{2}}. \tag{38}$$

On writing

$$\mathrm{I}_j(\sqrt{\mathrm{i}}\,y) = R_j(y) + \mathrm{i}M_j(y),$$

we have

$$X = b'R_0 + Kk(R_1 - M_1) \qquad \text{and} \qquad Y = b'M_0 + Kk(M_1 + R_1),$$

with the R_j and M_j being calculated for $y = \sqrt{2}\,kR$. The phase is given by

$$\phi = -\arctan(Y/X),$$

with a surface oscillation phase $\phi_T = \phi + \arctan(M_0/R_0)$.

The average modulation is then

$$\langle T-\bar{T}\rangle = L\int_0^R (T-\bar{T})\,2\pi r\,\mathrm{d}r,$$

where L is the sample length. Hence

$$\langle T-\bar{T}\rangle = \sqrt{2}\,\pi RL\delta P'\{(R_1^2+M_1^2)/(X^2+Y^2)\}^{\frac{1}{2}}\cos(\omega t+\phi_T+\phi'), \tag{39}$$

where ϕ' is the additional phase shift associated with the integration and is given by

$$\phi' = \arctan\big((M_1-R_1)/(M_1+R_1)\big) - \arctan\big(M_0/R_0\big).$$

In the limit $y \to 0$ (that is, $\delta \gg R$) the expression (39) for $\langle T-\bar{T}\rangle$ becomes identical to (2) in both amplitude and phase. For $y \to \infty$ (that is, $\delta \ll R$) it becomes identical to the plane wave solution (31), as is also expected.

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