Magneto-thermal-acoustic differential-frequency imaging of magnetic nanoparticle with magnetic spatial localization:
A theoretical prediction
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ABSTRACT
The magneto-thermo-acoustic effect that we predicted in 2013 refers to the generation of acoustic-pressure wave from magnetic nanoparticle (MNP) when thermally mediated under an alternating magnetic field (AMF) at a pulsed or frequency-chirped application. Several independent experimental studies have since validated magneto-thermo-acoustic effect, and a latest report has discovered acoustic-wave generation from MNP at the second-harmonic frequency of the AMF when operating continuously. We propose that applying two AMFs with differing frequencies to MNP will produce acoustic-pressure wave at the summation and difference of the two frequencies, in addition to the two second-harmonic frequencies. Analysis of the specific absorption dynamics of the MNP when exposed to two AMFs of differing frequencies has shown some interesting patterns of acoustic-intensity at the multiple frequency components. The ratio of the acoustic-intensity at the summation-frequency over that of the difference-frequency is determined by the frequency-ratio of the two AMFs, but remains independent of the AMF strengths. The ratio of the acoustic-intensity at the summation- or difference-frequency over that at each of the two second-harmonic frequencies is determined by both the frequency-ratio and the field-strength-ratio of the two AMFs. The results indicate a potential strategy for localization of the source of a continuous-wave magneto-thermal-acoustic signal by examining the frequency spectrum of full-field non-differentiating acoustic detection, with the field-strength ratio changed continuously at a fixed frequency-ratio. The practicalities and challenges of this magnetic spatial localization approach for magneto-thermo-acoustic imaging using a simple envisioned set of two AMFs arranged in parallel to each other are discussed.

Keywords: alternating magnetic field, magnetic nanoparticle, magnetothermoacoustic, thermoacoustic imaging, second-harmonic, magnetic localization.

1. INTRODUCTION
Inductive heating of conductive materials by using an alternating magnetic field (AMF) has allowed some special industrial processes including non-contact heat-processing to be done effectively. When magnetic nanoparticles (MNPs) such as ferrofluids are exposed to steady AMF, the MNPs also experience significant heating, which has been implemented for various industrial and medical applications [1, 2]. When exposed to AMF, the MNPs undergo relaxation processes including hysteresis [3], Brownian relaxation [4], and Néel relaxation [5, 6] that all contribute to heating of the MNPs with different dependence upon the size of the particles. The localized and highly efficient heating of MNPs using steady AMF with a frequency tailored to the MNPs, has been employed for targeted tumor hyperthermia [7] and thermally sensitive payload release [8].

The end-result of MNP relaxation under AMF is the conversion of electromagnetic energy to heat. Similarly, heat conversion is the end-result when laser light is absorbed by chromophores, microwave energy is absorbed by dielectric tissue constituents, and ionizing radiation is absorbed by tissue of non-negligible stopping power. Whenever the absorption of an electromagnetic energy by a target tissue varies rapidly, the time-varying heat dissipation will result in transient thermos-elastic expansion that in turn may induce acoustic wave—a mechanism broadly referred to as thermo-acoustic effect. When light is the activation source of energy to be absorbed by the target or tissue, the acoustic generation as a result of light activation is commonly referred to as photoacoustic effect.
Thermo-acoustic wave generation by rapid time-varying deposition of electromagnetic energy other than laser light has been demonstrated with microwave irradiation [11–13] and X-ray irradiation [14, 15].

A new mechanism for thermos-acoustic wave generation—referred to as “magneto-thermo-acoustics”, was proposed by us [16] that directed to the feasibility of employing the known heating effect of AMF-mediation of MNPs to produce acoustic signal for theranostic application. It has been predicted that acoustic-pressure wave will be generated by MNPs when thermally mediated under an AMF at a pulsed or frequency-chirped application. Several independent experimental studies [17–19] have since validated magneto-thermo-acoustic effect, whereby the AMF was applied in one of two temporal patterns, 1) pulsed, or 2) the amplitude varied at a chirped envelope pattern. Applying AMF in a pulsed mode is straightforward for generating the thermos-acoustic-wave from MNPs. The AMF with the amplitude modulated by a chirped envelop function produces an AMF that is continuously applied at a fixed frequency, but with the amplitude varies according to an envelope function whose frequency increases or decrease between two values and repeats the change (a chirped application). That pattern of AMF application is not identical to the one described in our original prediction [16]—the AMF is continuously applied at the fixed amplitude and it is the frequency of AMF, not the envelop of AMF amplitude, chirps. Regardless of this difference, however, applying an AMF whose frequency chirps or whose amplitude envelop chirps will cause the time-averaged heating applied upon MNPs to vary according to the cycles of the chirp-modulation of AMF, providing a foundation for frequency-domain detection of the thermos-acoustic wave.

A latest study [20] has discovered that acoustic-wave at the second-harmonic frequency of the AMF could be generated from MNPs when exposed to AMF operating continuously. This second-harmonic frequency was in fact embedded in the analytical preparations that had led to our prediction of magneto-thermo-acoustic effect. In this work, we propose that applying two AMFs with differing frequencies to MNP will produce acoustic-pressure wave at the summation and difference of the two frequencies, in addition to the two second-harmonic frequencies. We will show that, analysis of the specific absorption dynamics of the MNP when exposed to two AMFs of differing frequencies produces some interesting patterns of acoustic-intensity at multiple frequency components. The initial theoretical derivation based on some realistic assumptions of AMFs and magnetic susceptibilities has shown that, the ratio of the acoustic-intensity at the summation-frequency over that of the difference-frequency is determined by the frequency-ratio of the two AMFs, but remains independent of the AMF strengths. The derivation also shows that, interestingly, the ratio of the acoustic-intensity at the summation- or difference-frequency of the two AMFs over that at each of the two second-harmonic frequencies of the two AMFs is determined not only by both the frequency-ratio but also the field-strength-ratio of the two AMFs. These results point to a potentially interesting strategy for localization of the source of a continuous-wave magneto-thermal-acoustic signal generated by using two AMFs with differing frequencies but variable field intensities, by examining the frequency spectrum of the acoustic signal that is acquired without discriminating the position from the volume of AMF application. By changing the field-strength ratio for two AMFs with a fixed frequency-ratio, it is also possible to magnetically steer the position for signal detection. This method, which will be called “magneto-thermo-acoustic differential-frequency imaging”, may also be applied to thermo-acoustic signal generation from conductive objects that can be inductively heated, for non-destructive evaluation.

2. THEORY

2.1 Specific absorption dynamics of magnetic nanoparticles when exposed to two alternating magnetic fields with different frequencies

For a constant density system, the first law of thermodynamics governs that [21]

\[
\frac{dU}{dt} = \frac{dQ}{dt} + \frac{dW}{dt}
\]

(1)

where \( U \) [unit: J] is the internal energy, \( Q \) [unit: J] is the heat added, and \( W \) [unit: J] is the magnetic work done on the system. Under a collinear magnetic field, the differential magnetic work applied to the system is \( dW = \vec{H} \cdot d\vec{B} = \vec{H} \cdot dB \), where \( \vec{H} \) [unit: A m \(^{-1}\) or \( 4\pi \times 10^{-3}\) Oe] is the magnetic field intensity and \( \vec{B} \) [unit: T or V s A \(^{-1}\) m \(^{-2}\)] is the magnetic induction. Because \( B = \mu_0 (H + M) \), where \( M \) [unit: A m \(^{-1}\)] is the magnetization
and $\mu_0 = 4\pi \times 10^{-7}$ [unit: V s A⁻¹ m⁻¹] is the permeability of free space, the differential internal energy when the constant-density system is also confined in an adiabatic process, i.e. $\partial Q = 0$, becomes
\[
\frac{dU}{dt} = \mu_0 H \cdot \left( \frac{dH}{dt} + \frac{dM}{dt} \right)
\] (2)

We denote the dimension-less complex magnetic susceptibility of MNPs as $\chi = \chi' - i\chi''$, of which the real part is $\chi'$ and the imaginary part is $\chi''$. When the system is subjected to a time-varying magnetic field with an instant angular frequency $\omega_0$, the real and imaginary part of the magnetic susceptibility become respectively
\[
\left\{ \begin{array}{l}
\chi' (\omega) = \frac{\chi_0}{1 + [\omega \tau_\rho]^2} \\
\chi'' (\omega) = \chi_0 \frac{\omega \tau_\rho}{1 + [\omega \tau_\rho]^2}
\end{array} \right.
\] (3)

We analyze the specific absorption dynamics of MNPs when exposed to two independent AMFs that have the same starting phase, differ slightly in frequency, and have independent field amplitude. For simplicity, the susceptibility is assumed the same at the two AMF frequencies. A steady-state magnetic field that is composed of two AMFs with different frequencies is represented by
\[
H(t) = H_1 \cos(\omega_1 t) + H_2 \cos(\omega_2 t) = \Re \left[ \tilde{H}(t) \right] = \Re \left[ \tilde{H}_1(t) + \tilde{H}_2(t) \right]
\] (4)

under which the magnetization of MNP is
\[
M(t) = \Re \left\{ \chi \cdot \tilde{H}(t) \right\} = \Re \left\{ \chi \cdot \left[ H_1 \exp(i\omega_1 t) + H_2 \exp(i\omega_2 t) \right] \right\}
\]

\[
= \Re \left\{ \chi' H_1 \cos(\omega_1 t) + H_2 \cos(\omega_2 t) \right\} + \chi'' \left[ H_1 \sin(\omega_1 t) + H_2 \sin(\omega_2 t) \right]
\] (5)

Then Eq. (2) becomes
\[
\frac{dU}{dt} = \mu_0 H \cdot \left( \frac{dH}{dt} + \frac{dM}{dt} \right)
\]

\[
= \mu_0 \left[ H_1 \cos(\omega_1 t) + H_2 \cos(\omega_2 t) \right].
\]

\[
\left\{ \frac{d}{dt} \left[ H_1 \cos(\omega_1 t) + H_2 \cos(\omega_2 t) \right] + \frac{d}{dt} \left[ \chi' \left[ H_1 \cos(\omega_1 t) + H_2 \cos(\omega_2 t) \right] + \chi'' \left[ H_1 \sin(\omega_1 t) + H_2 \sin(\omega_2 t) \right] \right] \right\}
\]

\[
= -\mu_0 \left( 1 + \chi' \right) \left[ \omega_1 H_1 \sin(\omega_1 t) + \omega_2 H_2 \sin(\omega_2 t) \right] \left[ \omega_1 H_1 \sin(\omega_1 t) + \omega_2 H_2 \sin(\omega_2 t) \right] + \mu_0 \chi'' \left[ \omega_1 H_1 \cos(\omega_1 t) + \omega_2 H_2 \cos(\omega_2 t) \right] \left[ \omega_1 H_1 \cos(\omega_1 t) + \omega_2 H_2 \cos(\omega_2 t) \right]
\] (6)

After some algebraic manipulations, Eq. (6) will become the following:
\[
\frac{dU}{dt} = \frac{1}{2} \mu_0 \chi' \left[ \omega_1 \left| H_1 \right|^2 t + \omega_2 \left| H_2 \right|^2 t \right] + \frac{1}{2} \mu_0 \omega_1 \left( H_1 \right)^2 \sqrt{(1 + \chi')^2 + (\chi'')^2} \left[ \frac{\chi''}{\sqrt{(1 + \chi')^2 + (\chi'')^2}} \cos(2\omega_1 t) + \frac{1 + \chi'}{\sqrt{(1 + \chi')^2 + (\chi'')^2}} \sin(2\omega_1 t) \right] \\
+ \frac{1}{2} \mu_0 \omega_2 \left( H_2 \right)^2 \sqrt{(1 + \chi')^2 + (\chi'')^2} \left[ \frac{\chi''}{\sqrt{(1 + \chi')^2 + (\chi'')^2}} \cos(2\omega_2 t) + \frac{1 + \chi'}{\sqrt{(1 + \chi')^2 + (\chi'')^2}} \sin(2\omega_2 t) \right] \\
+ \frac{1}{2} \mu_0 \left( \omega_1 + \omega_2 \right) H_1 H_2 \sqrt{(1 + \chi')^2 + (\chi'')^2} \left[ \frac{\chi''}{\sqrt{(1 + \chi')^2 + (\chi'')^2}} \cos[(\omega_1 + \omega_2) t] - \frac{1 + \chi'}{\sqrt{(1 + \chi')^2 + (\chi'')^2}} \sin[(\omega_1 + \omega_2) t] \right] \right\}.
\]
Further we arrive at the following equation
\[
\frac{dU}{dt} = \frac{1}{2} \mu_0 \chi^2 \left[ \omega_1 (H_1)^2 + \omega_2 (H_2)^2 \right] + \frac{1}{2} \mu_0 \omega_1 (H_1)^2 \sqrt{(1 + \chi')^2 + (\chi'')^2} \left[ \cos(2 \omega_1 t + \alpha) \right] + \frac{1}{2} \mu_0 \omega_2 (H_2)^2 \sqrt{(1 + \chi')^2 + (\chi'')^2} \left[ \cos(2 \omega_2 t + \alpha) \right] + \frac{1}{2} \mu_0 H_1 H_2 \sqrt{((\omega_1 - \omega_2)(1 + \chi'))^2 + ((\omega_1 + \omega_2)(\chi'')^2} \left[ \cos((\omega_1 - \omega_2) t + \beta) \right]
\]
(8)

The items at the right-hand-side of Eq. (8) as separated to four lines correspond respectively to the following four components:
1) the DC component or the steady-state part
2) the composite 2\textsuperscript{nd}-harmonic components associated with each of the two AMF fields
3) The summation-frequency component whose frequency is the summation of the two AMF frequencies
4) The difference-frequency component whose frequency is the difference of the two AMF frequencies.

These four frequency components are respectively represented in the following
\[
\left( \frac{dU}{dt} \right)_{DC} = \frac{1}{2} \mu_0 \chi^2 \left[ \omega_1 (H_1)^2 + \omega_2 (H_2)^2 \right]
\]
(9a)
\[
\left( \frac{dU}{dt} \right)_{2nd} = \frac{1}{2} \mu_0 \omega_1 (H_1)^2 \sqrt{(1 + \chi')^2 + (\chi'')^2} \left[ \cos(2 \omega_1 t + \alpha) \right] + \frac{1}{2} \mu_0 \omega_2 (H_2)^2 \sqrt{(1 + \chi')^2 + (\chi'')^2} \left[ \cos(2 \omega_2 t + \alpha) \right]
\]
(9b)
\[
\left( \frac{dU}{dt} \right)_{summ} = \frac{1}{2} \mu_0 (\omega_1 + \omega_2) H_1 H_2 \sqrt{(1 + \chi')^2 + (\chi'')^2} \left[ \cos((\omega_1 + \omega_2) t + \alpha) \right]
\]
(9c)
\[
\left( \frac{dU}{dt} \right)_{diff} = \frac{1}{2} \mu_0 H_1 H_2 \sqrt{((\omega_1 - \omega_2)(1 + \chi'))^2 + ((\omega_1 + \omega_2)(\chi'')^2} \left[ \cos((\omega_1 - \omega_2) t + \beta) \right]
\]
(9d)

The energy deposition at any given duration of \([t, t + \Delta t]\) thus becomes
\[
Q_{MNP}(t) = \int_{t}^{t + \Delta t} \left( \frac{dU}{dt} \right)_{DC} dt + \frac{1}{2} \rho \Delta T \int_{t}^{t + \Delta t} \left( \frac{dU}{dt} \right)_{2nd} dt + \frac{1}{2} \rho \int_{t}^{t + \Delta t} \left( \frac{dU}{dt} \right)_{summ} dt + \frac{1}{2} \rho \int_{t}^{t + \Delta t} \left( \frac{dU}{dt} \right)_{diff} dt
\]
(10)

So the local pressure rise within the magnetic nanoparticle (usually in an aqueous host medium) is [20]
\[
P_{MNP}(t) = \frac{\beta}{k \rho C_v} Q_{MNP}(t)
\]
(11)

Where \(\beta\) is the thermal coefficient of volumetric expansion, \(\kappa\) the isothermal compressibility, \(\rho\) the mass-density, and \(C_v\) the volumetric heat capacity. It can be appreciated from Eqs. (9) to (11) that the spectrum of the local acoustic pressure wave carries the individual and summation/difference frequency components associated with the two AMF fields.
2.2 The dependence of the relative intensity of the frequency components of MNPs when exposed two AMFs of different frequencies--------magnetic encoding of the source of the magneto-thermo-acoustic signal.

By implementing the following symbols:

$$\tan(\alpha) = \frac{1 + \chi'}{\chi^*}$$  \hspace{1cm} (12a)

$$\rho_o = \frac{\omega_2}{\omega_1}$$  \hspace{1cm} (12b)

$$\rho_H = \frac{H_2}{H_1}$$  \hspace{1cm} (12c)

$$\beta = \tan^{-1}\left[\frac{\omega_2 - \omega_1}{(\omega_1 + \omega_2)\chi^*}\right] = \tan^{-1}\left[\frac{1 - \omega_2}{\omega_1(1 + \chi')}\right] = \tan^{-1}\left[\frac{(1 - \rho_o)(1 + \chi')}{(1 + \rho_o)\chi^*}\right]$$  \hspace{1cm} (12d)

Eq. (9) will become

$$\left(\frac{dU}{dt}\right)_{dc} = \frac{1}{2} \mu_o \chi^* \omega_1 (H_1)^2 \left[1 + \rho_o (\rho_H)^2\right]$$  \hspace{1cm} (13a)

$$\left(\frac{dU}{dt}\right)_{2nd} = \frac{1}{2} \mu_o \omega_1 (H_1)^2 \sqrt{(1 + \chi')^2 + (\chi^*)^2} \left[\cos(2\omega_1 t + \alpha)\right] + \rho_o (\rho_H)^2 \left[\cos(2\omega_2 t + \alpha)\right]$$  \hspace{1cm} (13b)

$$\left(\frac{dU}{dt}\right)_{summ} = \frac{1}{2} \mu_o \omega_1 (H_1)^2 (1 + \rho_o) \rho_H \sqrt{(1 + \chi')^2 + (\chi^*)^2} \left[\cos((\omega_1 + \omega_2) t + \alpha)\right]$$  \hspace{1cm} (13c)

$$\left(\frac{dU}{dt}\right)_{diff} = \frac{1}{2} \mu_o \omega_1 (H_1)^2 \rho_H \sqrt{[(1 - \rho_o)(1 + \chi')]^2 + [(1 + \rho_o)\chi^*]^2} \left[\cos((\omega_1 - \omega_2) t + \beta)\right]$$  \hspace{1cm} (13d)

Selected ratios of the magnitudes of the time-varying components as defined in the set of Eq. (13) are given in the following:

$$\left|\frac{dU}{dt}\right|_{diff} \left/ \left|\frac{dU}{dt}\right|_{summ}\right| = \sqrt{[(1 - \rho_o)(1 + \chi')]^2 + [(1 + \rho_o)\chi^*]^2} \left/ (1 + \rho_o)\sqrt{(1 + \chi')^2 + (\chi^*)^2}\right.$$  \hspace{1cm} (14)

$$\left|\frac{dU}{dt}\right|_{diff} \left/ \left|\frac{dU}{dt}\right|_{2nd}\right| = \rho_H \sqrt{[(1 - \rho_o)(1 + \chi')]^2 + [(1 + \rho_o)\chi^*]^2} \left/ \sqrt{(1 + \chi')^2 + (\chi^*)^2}\right.$$  \hspace{1cm} (15a)

$$\left|\frac{dU}{dt}\right|_{diff} \left/ \left|\frac{dU}{dt}\right|_{2nd}\right| = \frac{1}{\rho_o \rho_H} \sqrt{[(1 - \rho_o)(1 + \chi')]^2 + [(1 + \rho_o)\chi^*]^2} \left/ \sqrt{(1 + \chi')^2 + (\chi^*)^2}\right.$$  \hspace{1cm} (15b)

$$\left|\frac{dU}{dt}\right|_{summ} \left/ \left|\frac{dU}{dt}\right|_{2nd}\right| = (1 + \rho_o)\rho_H$$  \hspace{1cm} (16a)
At a constant frequency ratio of $\rho_\omega$, the amplitude ratios of the frequency components of Eqs. (15a), (15b), (16a), and (16b) are all determined uniquely by the intensity ratio $\rho_H$ between the two AMF fields. These analyses, when extended to considering the frequency-dependence of the magnetic susceptibility, will result in similar results—-the amplitude ratios between the summation or difference frequency component and either of the 2nd harmonic frequency component is dependent upon the ratio of the two AMF fields.

3. MAGNETIC SPATIAL LOCALIZATION—ONE DIMENSIONAL CASE

We analyze the spatial pattern of AMF field applied by two single-loop coils placed in parallel to each other, to illustrate the spatial dependence of the summation or difference frequency components of the acoustic pressure waves from MNPs when exposed to two AMFs of differing frequencies. The analysis of the concept of the spatial encoding is limited to one-dimension, which is readily expandable to three-dimensional analyses.

Assuming that two identical single-loop coils are placed face-to-face at a distance of 2d apart, as shown in Figure 1. Now consider a field point whereupon there is a concentration of MNP at the axis that connects the centers of both loops. Because the acoustic pressure for a given concentration of the MNP depends upon the field intensity at the given frequencies, the following analysis focuses on the field intensity patterns. We consider a field point that is at a distance of z from the center of the coil 1, and a distance of 2d–z from the center of coil 2. The field intensity at the on-axis point has no laterally-directional components. The z-directional components are as the following

$$H_1(z) = \frac{I_1 R^2}{2} \frac{1}{\left[R^2 + z^2\right]^{3/2}} \quad (17)$$

$$H_2(z) = \frac{I_2 R^2}{2} \frac{1}{\left[R^2 + (2d - z)^2\right]^{3/2}} \quad (18)$$

Which together give the following ratio of the field intensity at an an-axis point or a distance z from the coil 1:

![Figure 1. The geometry of two identical single-loop coil of radius R facing each other at a distance of 2d. We consider the field point along the coaxial line.](Image)
The field intensities and the ratio between them as defined by Eq. (17)-(19) are plotted in Figure 2 for the case of the two single-loop coils having the same coil current, a 25mm coil diameter, and a 20mm center distance between the two coils. It can be shown that there is a unique ratio between the intensities of the two fields for any on-axis point between the coils. The spatial dependence of the field-intensity-ratio indicates the feasibility of changing/setting the field-intensity ratio at a specific point, by changing the current ratio between the two coils. An example is shown in Figure 3, where the current of coil 1 changes from 0.5 times to 1.5 times of the current of coil 2, at a step of 0.1 times. As the current of coil 1 increases, the position of a specific field-intensity ratio (e.g., 1) shifts towards the coil 2. As the specific field-intensity-ratio determines the intensities of the acoustic pressure waves of individual frequency components as they originating from the MNP under the two fields, the feasibility of changing the field-intensity-ratio refers to the possibility of encoding the position of the pressure wave magnetically or identification of the source of the pressure wave by analyzing the spectrum of the acoustic pressure wave. When the analysis applied to on-axis point is extended to off-axis point, the field-amplitude ratio can be found to have more complicated spatial dependence. The 3-dimensional spatial dependence of the field-ratio, however, still render the possibility of uniquely and magnetically determining the position of a source of a thermos-acoustic wave generation due to the heating of MNPs under two AMFs, by applying two AMFs of differing frequencies in three orthogonal orientations.
4. CONCLUSIONS

The magneto-thermo-acoustic effect that we predicted in 2013 refers to the generation of acoustic-pressure wave from magnetic nanoparticle (MNP) when thermally mediated under an alternating magnetic field (AMF) at a pulsed or frequency-chirped application. We propose that applying two AMFs with differing frequencies to MNP will produce acoustic-pressure wave at the summation and difference of the two frequencies, in addition to the two second-harmonic frequencies. Analysis of the specific absorption dynamics of the MNP when exposed to two AMFs of differing frequencies has shown some interesting patterns of acoustic-intensity at the multiple frequency components. The ratio of the acoustic-intensity at the summation-frequency over that of the difference-frequency is determined by the frequency-ratio of the two AMFs, but remains independent of the AMF strengths. The ratio of the acoustic-intensity at the summation- or difference-frequency over that at each of the two second-harmonic frequencies is determined by both the frequency-ratio and the field-strength-ratio of the two AMFs. The results indicate a potential strategy for localization of the source of a continuous-wave magneto-thermal-acoustic signal by examining the frequency spectrum of full-field non-differentiating acoustic detection, with the field-strength ratio changed continuously at a fixed frequency-ratio.

REFERENCES