Enhanced deep-tissue photoacoustics by using microcomposites made of RF-metamaterials and soft polymers: double- and triple-resonance phenomena

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Photoacoustics + ultrasound-transducer systems offer an imaging platform with high resolution to explore body tissues, food, and valuable artwork. Plasmonics constitutes one source of resonant heating and thermal expansion necessary to generate acoustic waves. However, its associated techniques are seriously limited to the laser penetration and the non-specific hyperthermia in the sample. Besides, there is a compromise between the nanoparticle size and the pressure generated since the photoacoustic efficiency depends on the scatterer’s surface-to-volume ratio. This work adopts a paradigm shift in photoacoustics. By simulating microparticles made of some random composites, the pressure calculated can be similar or superior to the one obtained with plasmonic optoacoustics. The improvement is due to a phenomenon called double or triple resonance: the excitation of one or both electric and magnetic "radio-plasmons" and the simultaneous excitation of the particle’s acoustic mode. As electromagnetic pulses restrict to nanosecond widths and MHz frequencies, the proposed method avoids the low penetration in tissues and the thermal damage, constituting a non-invasive technique of theragnosis. Moreover, the "resonant" pressure obtained lasts longer in time than conventional photoacoustic pressures, providing another central feature to enhance detection. To fully comprehend the multi-resonance framework, this work develops a complete photoacoustic solution. The proposed approach paves the way to thermo-acoustic imaging/manipulation methods for sensitive materials/tissues reaching micrometer resolution.

INTRODUCTION

Photoacoustics date back to 1880, when A. G. Bell heard a "pure musical tone" in a closed gas volume that had absorbed a modulated light beam. However, the interest in this field dropped off due to the lack of enough technology until relatively recently. Around 1980, many experiments about photochemical reactions and sensitive gas detection followed. In the '90s, with the advent of newer technology, photoacoustic (PA) imaging was considered a promising technique for biomedical applications. Since then, various contrast agents have enabled image reconstruction from body tissues. But it was not until the 2000’s that the plasmonic nanoparticles made of noble metals were introduced in Photoacoustics. Plasmonic nanoparticles have the advantage over previous contrast agents, as molecular dyes, that they possess high absorption efficiencies with a tunable spectral response due to their surface plasmon (SP) excitation in the optical range. A few nanoparticles show low cytotoxicity to biological tissues, are chemically inert, and manifest environment sensitivity. They also can be easily functionalized for different purposes. In this way, plasmonic-based techniques are still considered optimal for photothermal treatment of cancer cells because of the high-temperature rise that occurs during optical absorption.

However, this is the first main obstacle when non-invasive techniques are required. The temperature rise can be significant to thermally damage some non-targeted tissues, even using short laser pulses of a few nano or femtoseconds. In addition, the effect leads to various undesirable phenomena like nano/microbubble formation or particle-shape modifications, both phenomena changing the sensitivity to the environment and the spectral properties of the particle. The second main obstacle when using plasmonic-based methods is the relatively poor penetration in biological tissues because optical beams are needed. Besides, the PA effect with nanoparticles strongly depends on the surface-to-volume ratio because the major component of the acoustic pressure generated is due to the embedding medium’s layer that surrounds the particle. Thus, slight variations in the nanosources strongly affect the PA efficiency, causing instabilities of the technique.

This paper proposes a new paradigm for PA pressure generation to sort out the obstacles that concern optical plasmonics. Here, Radioplasmonic  is considered as an extension of optical plasmonics to radiofrequency (RF) range. Surface plasmons in RF can be excited in microparticles made of random metamaterials that have been recently reported as radiofrequency-metamaterials (RF-MMs). Electromagnetic (EM) radiation at low energies allows for large penetration in tissues and resonant absorption due to plasmon excitation. Here, the simulation of microparticles made of RF-MMs and soft polymers shows that EM and acoustic resonances can match. A new concept enters the discussion about Photoacoustics: double and triple resonance. In the former one, one electric/magnetic SP matches the monopolar acoustic mode of the particle. In the latter one, both the electric and magnetic SPs match the particle’s acoustic mode. Then, the PA pressure results higher in intensity and time duration than what optical fields and metal nanoparticles obtain.

For the first time, a complete analytical PA solution shows the new multi-resonance framework using an arbitrary EM source. The leading example consists of a simulation of a Gaussian pulse exciting "realistic" metamaterials. The pressure calculated enhances with neglectable temperature rises.

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A double-resonance effect was already predicted in Ref. [13] using examples of excitation of the usual electric SP + the acoustic monopole. Here the concept includes a novel "magnetic" photoacoustics and triple-resonance phenomena.

The following section explains the electric and magnetic resonances that can occur in RF-MMs to define the novel framework.

The methodology used to simulate realistic composites is detailed. As their EM, thermal, and mechanical properties are essential, adequate "effective medium" theories are applied when needed. They are described in the Supplementary Material (SI) for the interested reader.

THEORETICAL CONSIDERATIONS

A metamaterial (MM) particle consists of some conductive nanoelements immersed in a matrix or host medium, Fig. 1a. It constitutes a microcomposite, and the host medium has a low speed of sound for later purposes. These nanoelements can be carbon nanotubes, graphene sheets, nanofibers, or even nanoparticle clusters, whether they create preferential conduction directions. When zoomed in, the particle shows that there may exist three possibilities to function as an EM scatterer, depending on the concentration (CC) of the conductive elements in the matrix. A low CC of elements (in orange, online version, Fig. 1b) in the matrix (in light blue) creates a composite below the so-called percolation threshold, with poor electric conduction due to hopping-electron mechanism (see inset below 1b). As a result, the whole MM behaves as a pure dielectric, with both real and imaginary parts of the permittivity function positive, Fig. 1c. It is what happens in any typical material in the RF domain. On the other hand, a high CC of the conductor phase can produce a composite near or above the percolation threshold, Fig. 1d, with high electric conduction due to free-electron currents (see inset below 1b). As a consequence, a conduction net establishes inside the MM, and one or two possibilities may arise. In the first case, the free-electron conduction creates a metal-like composite in the RF domain, and it shows a negative real part of the permittivity, Fig. 1d. An oscillating electric field \( \mathbf{E}_0 \), red arrow in Fig. 1d, can move the electrons in the particle and induce an electric dipole moment \( \mathbf{p} \). In this way, an electric surface plasmon (ESP) can be excited at the appropriate wavelength, Fig. 1h, as it occurs in the usual plasmonics with the optical fields when the pole of the electric polarizability \( \alpha_E \) is reached (see Electromagnetic response for details). The spots in red correspond to the dipole electric field of the ESP (Fig. 1h). In the second case, another effect can happen besides the electric conduction. Magnetic response of the MM may arise as current loops around the net paths appear (green arrows). This induced field in the MM responds to the magnetic field \( \mathbf{H}_0 \) of the incident wave, and couples with the electronic circulation (like in eddy currents); see the blue crossed symbols in Fig. 1d indicating a downward induced field according to the current loops. In a form analogous to the negative-real permittivity, negative-real permeability can also occur as an attempt of the MM to oppose the disturbing external fields, Fig. 1d. A magnetic dipole moment appears as a response to the incident magnetic field, blue arrow in Fig. 1h. Then, a possible magnetic surface plasmon (MSP) can be excited at the appropriate wavelength, see in Electromagnetic response the condition to reach the pole in the magnetic polarizability \( \alpha_M \). The blue spots, in this case, correspond to the dipole magnetic field of the MSP, Fig. 1h. In addition, the particle can heat up by the electric+magnetic Joule effect. This phenomenon is schematized with a spread-out cloud around the particle (f+h).

To give a few examples of realistic MMs, the microparticles simulated are composites made of three types of materials; an electric MM or EMM, a magnetic MM or MMM, and an acoustic material or A-material. In general, the A-material is simply a dielectric with a low speed of sound to allow for the acoustic resonances in the MHz frequencies.

### Electromagnetic response

A similar procedure to the one reported in [13] is generalized to include magnetic response. A plane wave characterizes the spatial dependence of the EM radiation with electric and magnetic fields \( \mathbf{E}_0 \) and \( \mathbf{H}_0 \), respectively. Given that the minimum penetration depth is several times the particle size, the fields are constant inside the particles and the quasistatic approximation can be used [23] provided that \( R << \lambda \), being \( R \) the particle radius and \( \lambda \) the wavelength of the incident wave. Then the particle is represented by their electric and magnetic dipole moments

\[
\mathbf{p} = \varepsilon_0 \varepsilon_r \alpha_E \mathbf{E}_0, \quad (1)
\]

\[
\mathbf{m} = \alpha_M \mathbf{H}_0, \quad (2)
\]

where \( \varepsilon_r \) is the relative dielectric function of the embedding medium. The polarizabilities are defined by [23]

\[
\alpha_E = \left( \frac{\alpha_{E,0} - \frac{i k_b^3}{6 \pi}}{ \alpha_{E,0} + \frac{i k_b^3}{6 \pi}} \right)^{-1}, \quad \alpha_{E,0} = 3 V \frac{\varepsilon_r - \varepsilon_{rb}}{\varepsilon_r + 2 \varepsilon_{rb}}, \quad (3)
\]

\[
\alpha_M = \left( \frac{\alpha_{M,0} - \frac{i k_b^3}{6 \pi}}{ \alpha_{M,0} + \frac{i k_b^3}{6 \pi}} \right)^{-1}, \quad \alpha_{M,0} = 3 V \frac{\mu_r - \mu_{rb}}{\mu_r + 2 \mu_{rb}}, \quad (4)
\]

where \( k_b = \sqrt{\varepsilon_r \mu_r \omega^2 / c}, \quad \omega \) is the angular frequency, and \( c \) the speed of light. \( k_b \) and \( \mu_{rb} \) are the wavenumber and relative permeability of the surrounding medium, respectively.

\( V = 4 \pi R^3 / 3 \) the particle volume. \( \varepsilon_r \) and \( \mu_r \) are the relative dielectric and magnetic functions of the particle. \( \alpha_{E,0} \) and \( \alpha_{M,0} \) are called the quasi-static polarizabilities.

The permittivities and permeabilities of the MMs used can fulfill one or both conditions \( \varepsilon_r (\omega') + 2 \varepsilon_{rb} = 0 \), \( \mu_r (\omega') + 2 \mu_{rb} = 0 \) at the desired spectral positions if the composite is appropriately designed, i.e. the complex denominators of eqs. [3,4] to be zero. In this way, one obtains a single- or double-resonant behavior, one for each particle polarizability and, consequently, a resonant absorption in the corresponding spectrum. Each electric and magnetic resonance corresponds respectively to the excitation of the dipolar ESP and MSP in
FIG. 1. (Color online) “Radioplasmonics” and mechanism to obtain electric and magnetic surface plasmons by using RF-metamaterials. (a) The particle is a composite representing one or several metamaterials. In a simple case, a dielectric medium or host (light blue) contains a concentration (CC) of conductive nanoelements (orange lines). Depending on this CC, one can obtain (b) a disperse “solution” and hopping-electron conduction (inset in b), which results in a whole dielectric behavior (c) for the composite; or (d) a concentrated solution, near or beyond the percolation threshold, which forms a conductive network with metal-like conduction (free-electrons are depicted by the inset in d). (e) This metal-like behavior translates into a negative-real part of the dielectric function for the whole composite. (f) An incident wave with electric field $E_0$ (red arrow) induces a dipole moment $p$. The resonance associated with the electric polarizability $\alpha_E$ is the dipolar electric surface plasmon, represented as a red spot in the scheme. (g) Negative-real permeability can also appear as an attempt of the composite to oppose the incident magnetic fields, generating an inner magnetic response represented by the blue symbols and the green current loops in (d). (h) As a consequence, an incident wave with magnetic field $H_0$ (blue arrow) can induce a magnetic dipole moment $m$. The resonance associated with the pole of the magnetic polarizability $\alpha_M$ is the dipolar magnetic surface plasmon, represented as a blue spot in the scheme. Moreover, the particle can heat up by the electric+magnetic Joule effect when irradiated with radio waves. This effect is schematized as a pink cloud around the particle in (f+h).

the particle. The EM functions $\varepsilon(\omega')$ and $\mu(\omega')$ are taken from the experimental data, or their corresponding fits using Drude-Lorentz functions, as reported.

In this way, the absorption cross-section for the particle follows from the absorption by the electric and magnetic dipole moments

$$\sigma_{abs} = \sigma_{abs,E} + \sigma_{abs,M},$$

where

$$\sigma_{abs,E} = k_b \left( Im(\alpha_E) - \frac{k^3}{6\pi} |\alpha_E|^2 \right),$$

$$\sigma_{abs,M} = k_b \left( Im(\alpha_M) - \frac{k^3}{6\pi} |\alpha_M|^2 \right).$$

The field maps in this paper are calculated by a Mie code\textsuperscript{25}. Mie theory is also used to check the quasistatic approximation in the results.

From here on, all the results correspond to an environment that mimics body-tissue surroundings; the temperature is $T_0 = 36.6^\circ C$, and the permittivity of water is $\varepsilon_{rb} = 73.3$ for low salinity\textsuperscript{26}. The acoustic/mechanical properties also correspond to water and fit many water-like tissues. Refer to the SI for more details.

Photoacoustic phenomenon: Full solution.

The complete problem for the thermoacoustic generation of pressure is described by the following system of coupled Eqs.
in every medium involved.\(^\text{22}\)

\[
\frac{\partial}{\partial t} \left( T - \gamma \frac{1}{\gamma a} \rho \right) = \frac{\nabla \cdot (\kappa \nabla T)}{\rho C_p} + \frac{Q}{\rho C_p},
\]

(8)

\[
\left( \nabla^2 - \gamma \frac{\partial^2 T}{\partial t^2} \right) T = -\frac{\alpha}{v_s^2} \frac{\partial^2 T}{\partial t^2},
\]

(9)

where \( T \) is the temperature, \( p \) is the pressure, \( \gamma \) is the specific heat ratio, \( a = \left( \frac{\partial^2 p}{\partial T^2} \right)_\nu \) is the pressure expansion coefficient at constant volume, \( \kappa \) is the thermal conductivity, \( \rho \) is the density, \( C_p \) is the heat capacity at constant pressure, \( v_s \) is the speed of sound, and \( Q \) is the energy deposited per unit time and unit volume in the particle, which transforms into heat. As a simplification, it is common to assume \( \gamma \approx 1 \) for any typical water-like fluid. Besides, the condition \( \nabla \cdot (\kappa \nabla T) \ll \frac{\partial T}{\partial t} \) is a reasonable approximation for fast excitation in all media involved. Assuming also that the heating process occurs only in the particle’s medium through the relation \( \frac{\partial T}{\partial t} = \frac{\partial T}{\partial r} \), the PA generation due to an arbitrary EM field is reduced to solve the following system of uncoupled Eqs.

\[
\nabla^2 \rho_s - \frac{1}{v_{ss}^2} \frac{\partial^2 \rho_s}{\partial t^2} = -\frac{\beta}{C_p} \frac{\partial Q}{\partial t},
\]

(10)

\[
\nabla^2 \rho_f - \frac{1}{v_{sf}^2} \frac{\partial^2 \rho_f}{\partial t^2} = 0,
\]

(11)

where \( p_s (p_f) \) is the pressure inside (outside) the particle, \( \beta \) is the volumetric thermal expansion, and \( v_{ss} (v_{sf}) \) is the longitudinal speed of sound in the particle’s (the embedding) medium. Here, transversal waves are not considered. The solution of the system\(^\text{10}\) is bound to the boundary conditions, i.e. continuity of the pressure and radial velocity for each medium, taking the spherical symmetry into consideration. In particular, as the particle is small enough, let’s assume that the heating source \( Q \) is homogeneous inside the particle and that the process is fast, i.e. \( \nabla^2 \rho_s \ll \frac{1}{v_{ss}^2} \frac{\partial^2 \rho_s}{\partial t^2} \). Then the inhomogenous solution of Eq.\(^\text{10}\) i.e. \( \rho_0 \), is obtained from

\[
\frac{1}{v_{ss}^2} \frac{\partial^2 \rho_0}{\partial t^2} = \frac{\beta}{C_p} \frac{\partial Q}{\partial t},
\]

(12)

such that \( \rho_s = \rho_{sh} + \rho_0 \) is the complete solution of Eq.\(^\text{10}\) where \( \rho_{sh} \) is the homogeneous part of the solution. \( \rho_f \) is the homogeneous solution of Eq.\(^\text{11}\).

The complete solution for any EM excitation with source intensity \( I(t) \) can be built from the convolution product between a PA pressure generated by a monochromatic plane wave (MPW) and \( I(t) \)

\[
p_s(r,t) = p_s(r,t)|_{\text{MPW}} * \frac{I(t)}{I_0} = \int_{-\infty}^{\infty} p_s(r,t-t')|_{\text{MPW}} \frac{I(t')}{I_0} \, dt',
\]

(13)

\[
p_f(r,t) = p_f(r,t)|_{\text{MPW}} * \frac{I(t)}{I_0} = \int_{-\infty}^{\infty} p_f(r,t-t')|_{\text{MPW}} \frac{I(t')}{I_0} \, dt',
\]

(14)

where \( I_0 \), the initial intensity, is known. In general, the solutions \( p_s(r,t-t')|_{\text{MPW}} \) and \( p_f(r,t-t')|_{\text{MPW}} \) are not straightforward to get. However, making use of the Fourier transform, this convolution product can be easily calculated. By taking the parity properties of the complex fields into account, the solution can be built from the frequency-domain functions as follows

\[
p_s(r,t) = \Re \left\{ \mathcal{F}^{-1} \left[ \hat{p}_s(r,\omega) \frac{I(\omega)}{I_0} \right] \right\},
\]

(15)

\[
p_f(r,t) = \Re \left\{ \mathcal{F}^{-1} \left[ \hat{p}_f(r,\omega) \frac{I(\omega)}{I_0} \right] \right\},
\]

(16)

where the Fourier transform is defined as \( \mathcal{F}[f(t)] = \hat{f}(\omega) = \int_{-\infty}^{\infty} f(t) e^{-i\omega t} \, dt \), such that \( \mathcal{F}^{-1}[\hat{f}(\omega)] = f(t) = \frac{1}{\sqrt{2\pi}} \int_{-\infty}^{\infty} \hat{f}(\omega) e^{i\omega t} \, d\omega \). Specifically, the functions \( \hat{p}_s(r,\omega) \) and \( \hat{p}_f(r,\omega) \) are obtained from the frequency-domain solution to the PA generation under a MPW. To do this, the illumination takes form \( I(t) = I_0 e^{-i\omega_0 t} \) such that \( Q(t) = \sigma_{abs} I(t)/V \). Using this \( Q(t) \) into Eq.\(^\text{12}\), \( \hat{p}_s(r) \) and \( \hat{p}_f \) are obtained from the corresponding Helmholtz equations derived from Eqs\(^\text{10}\) and\(^\text{11}\) under spherical symmetry. In general, they are expressed as series of the spherical Bessel functions and the Legendre Polynomials, but let’s focus only on the monopolar solutions, the ones that matter in the present study. The solution is constrained to the following boundary conditions\(^\text{23}\)

\[
\hat{p}_s|_{r=R} = \hat{p}_f|_{r=R},
\]

(17)

\[
\frac{1}{\rho_s} \frac{\partial \hat{p}_s}{\partial r} \bigg|_{r=R} = \frac{1}{\rho_f} \frac{\partial \hat{p}_f}{\partial r} \bigg|_{r=R}.
\]

(18)

Solving the system, it is found that\(^\text{24}\)

\[
\hat{p}_s(r,\omega) = \hat{p}_f \frac{1}{\rho_f} \frac{\partial \hat{p}_f}{\partial r},
\]

(19)

\[
\hat{p}_f(r,\omega) = \hat{p}_f \frac{\partial \hat{p}_f}{\partial r}
\]

(20)

where \( \hat{p}_f = \frac{\hat{p}_f}{\hat{p}_f} \) and \( \hat{p}_f \). From Eqs.\(^\text{19}\)\(^\text{20}\) it is clear that the acoustic monopole has resonance when the denominator of these expressions goes to zero. This is the acoustic resonance to be exploited in this work. Assuming an MPW as the
illumination, the time-domain solution is then given by

\[
p_s(r,t)_{\text{MPW}} = -\frac{\Gamma_{\text{abs}} l_0}{\omega V} \Im \left\{ e^{-i\omega t} \right\} \left[ 1 + \frac{\hat{v}_{\hat{P}} h_1^{(1)}(kR) j_0(kr)}{h_0^{(1)}(kR) j_1(kR) - \hat{v}_{\hat{P}} h_1^{(1)}(kR) j_0(kR)} \right],
\]

(21)

so that the products in time-domain given in Eqs. 13-14 are now defined. Note that the formulation is written in terms of \(\Gamma = \frac{\rho \kappa}{\epsilon_0 c}\), the well-known Grüneisen coefficient.\(^{[3]}\)

Observe that the expressions \(^{[5]}^{[6]}\) are general. For example, in the limit of an extremely fast pulse, \(I(t) = I_0 \delta(t)\), then \(I(\omega)/I_0 = 1\), and \(p_s(r,t)_{\text{MPW}}, p_f(r,t)_{\text{MPW}}\) are recovered by the definition of the inverse Fourier transform of an MPW.

At this point, a Gaussian pulse becomes the EM source \(I(t)/I_0\) corresponding to the normal distribution

\[
g(t) = 2\sqrt{\ln(2)} \frac{e^{-\frac{(\ln(2)/\tau)^2}{\tau^2}}}{\sqrt{\pi} \tau},
\]

(23)

where \(\tau\) is the full width at half maximum and \(t_0\) the time to reach the pulse’s maximum. If the Fourier transform of the function \(^{[22]}\) is considered, a full time-domain solution for the PA pressure generated is obtained from the following convolution product

\[
p_s(r,t) = \frac{\tau}{\pi} \Re \left\{ \int_0^\infty \hat{p}_s(r,\omega) e^{-\frac{\omega^2}{\omega^2_{\text{abs}}}} e^{-i\omega t} d\omega \right\},
\]

(24)

\[
p_f(r,t) = \frac{\tau}{\pi} \Re \left\{ \int_0^\infty \hat{p}_f(r,\omega) e^{-\frac{\omega^2}{\omega^2_{\text{abs}}}} e^{-i\omega t} d\omega \right\}.
\]

(25)

Note that in the limit \(\tau \to 0\), \(I(t) = I_0 \delta(t)\), and the solutions \(^{[24]}^{[25]}\) become zero because the pulse has a finite energy (\(g(t)\) is a normalized function).

Explicitly, inserting the expressions \(^{[19]}^{[20]}\) into Eqs. \(^{[24]}^{[25]}\), the PA pressure in time-domain for a Gaussian pulse can be rewritten as

\[
p_s(r,t) = -\frac{\sigma_{\text{abs}}(\omega') \Gamma F}{\pi V} \Im \left\{ \int_0^\infty e^{-iqt} e^{-\frac{\omega^2}{\omega^2_{\text{abs}}}} \right\} \left[ \frac{1}{q} + \frac{\hat{P} \sin(q) / q}{q^2 - i\hat{v} \hat{P} \sin(q) / q} \right] \left[ (1 - \hat{P}) \frac{\cos(q) - q \sin(q)}{q} + i\hat{v} \hat{P} \sin(q) \right] dq \right\}.
\]

(26)

\[
p_f(r,t) = -\frac{\sigma_{\text{abs}}(\omega') \Gamma F R}{\pi V \hat{r}} \Im \left\{ \int_0^\infty e^{-iqt} e^{-\frac{\omega^2}{\omega^2_{\text{abs}}}} p(q) dq \right\},
\]

(27)

\[
P(q) = \frac{\sin(q) - q \cos(q)}{q} \left[ (1 - \hat{P}) \frac{\cos(q) - q \sin(q)}{q} + i\hat{v} \hat{P} \sin(q) \right],
\]

(28)

where \(r \neq 0\), and the acoustic variable \(q = \alpha R/v_{\text{as}}\) is defined like in Ref\(^{[19]}\) which involves the acoustic angular frequency \(\omega\). In this complete version of the PA response under Gaussian illumination, note that \(\sigma_{\text{abs}}(\omega')\) in general depends on the driving frequency \(\omega'\) (in general \(\neq \omega\)) of the EM field. \(F\) is the pulse fluence, \(r\) is the radial variable from the particle’s center, and \(\hat{r} = \frac{\omega}{\omega'} \left[ t - t_0 - \frac{r}{v_{\text{as}}} \right]\).

From this complete solution, it is clear that at least two kinds of resonances can couple; one is any implicit EM resonance in \(\sigma_{\text{abs}}\), the other is the acoustic resonance corresponding to the excitation of any monopolar mode. The concept of mixing different resonance types in a novel PA phenomenon is the core of the present study.

**NUMERICAL RESULTS AND DISCUSSION**

**Excitation of Radioplasmons**

In what follows, a 10\(\mu\)m-particle is taken as the example for discussion. This particle can be a composite made of three types of realistic materials. The first material is a magnetic-electric composite, a Y\(_3\)Fe\(_5\)O\(_{12}\) matrix (Yttrium Iron Garnet, YIG) that is filled with Ag nanoparticles. This is the MMM that facilitates magnetic properties to the particle.\(^{[31]}\) The second compound is the EMM, namely an Al\(_2\)O\(_3\) matrix with MWCNT as inclusions in it;\(^{[17]}\) it facilitates the electric properties. The last material is a high dielectric that has a low speed of sound to act as an A-material (or simply material "A") for later purposes. Its dielectric constant is \(\varepsilon_{A} = 250\) with negligible imaginary part, and \(\mu_{A} = 1\). As a realistic example, it can be a PANI-based polymer.\(^{[31]}\) For more details of the data used in the simulations, see the SI.

It is not the purpose of the paper to discuss how to fabricate this particle but to show the consequences of assuming such a scatterer. As a starting point, the EM absorption by the particle is studied as a function of the particle composition and driving frequency \(\hat{r}F\). The normalized absorption \(\eta\) is the absorption cross-section \(\sigma_{\text{abs}}\) scaled by the factor \(\sigma_{\text{abs, max}} = 0.42 \mu m^2\), Fig. 2. For convenience, half of the composite corresponds to the A-material which acts as a host or matrix. The rest 50% of the composite is left for the EMM whose properties are varied. The MMM can be written as [Ag(X)/YIG]\(_{0.5}\), where \(D\) is its CC in %, acting as the guest that fills into the host [MWCNT(11%)/Al\(_2\)O\(_3\)]\(_{0.5}\). Here, \(X\) is the concentration in % of Ag fillers in YIG. As reported in Ref\(^{[31]}\) \(X = 5\%\) is below the percolation limit in Ag/YIG but shows positive permeability (\(\mu_{X} \approx 5\)) for this MMM, Fig. 2. Note how an ESP appears around 355 MHz for \(D = 0\%\) due to the EMM. As \(D\) increases, the ESP "redshifts" and vanishes since this MMM does not support EM resonances. In Fig. 2, the EMM is calculated with \(\mu_{X} = 1\).

Very different behavior is found for \(X = 50\%\) in the magnetic composite, which is beyond the percolation limit for Ag/YIG, Fig. 2. In this case, the ESP due to the presence of EMM is "blueshifted". Besides, the MMM satisfies the condition of MSP; it appears as a spot having a maximum of around
432 MHz for $D = 100\%$. But more importantly, the maximum absorption is found for $(D = 43.6\%, 499.5\text{MHz})$ because both the ESP and MSP overlap when the three materials coexist. In Fig. 2b, the EMM is calculated with $\mu_r(MWCNT(11\%)/Al_2O_3) = 1.0$. The maximum absorption is found for $D=43.6\%$ and Frequency=499.5 MHz. (c) The absorption of this composite is compared with the one for pure MWCNT(11\%)/Al$_2$O$_3$ and pure Ag(50\%)/YIG. (d) Electric and magnetic contributions to case $D=43.6\%$, showing the addition of the electric and magnetic surface plasmons. The particle composition is schematized with a pie chart, where E (M) stands for EMM (MMM) for simplicity. This is also used in the legends of (c+d).

432 MHz for $D = 100\%$. But more importantly, the maximum absorption is found for $(D = 43.6\%, 499.5\text{MHz})$ because both the ESP and MSP overlap when the three materials coexist. In Fig. 2b, the EMM is calculated with $\mu_r = 0.8$ since some magnetic activity for this composite can take place. This maximum value corresponds to $\sigma_{\text{abs,max}} = 0.42 \mu W/m^2$ so that $\eta = 1.0$.

By comparing the absorption for the optimal condition $D = 43.6\%$ with the ones for pure EMM and MMM, Fig. 2c, the particle resonances can be tuned by their composition.

The particle can be electric, magnetic, or electric-magnetic as required for different purposes. It is worth mentioning that Ag(50\%)/YIG is already an electric-magnetic material; observe that the red line with square symbols has also a low energy peak around 100 MHz. However, this peak is so weak and out of resonance compared with the MSP that another EMM is needed to strengthen the whole composite at the desired spectral location. To appreciate the overlapping effect of both the ESP and MSP at the optimal condition $D = 43.6\%$, the individual absorptions due to the electric and magnetic dipoles are calculated from Eqs. 6 and 7. The maximum absorption is found for $D=43.6\%$ and Frequency=499.5 MHz. This kind of superposition can be designed for any MM particle at any frequency if the parameters of the mixture are carefully chosen. In this example, the materials are proven to exist and are easily fabricable. In the rest of the work, the study focuses on [Ag(50\%)/YIG] as the MMM.

Now the nature of these resonances is analyzed by calculating near-field maps at the spectral locations of the SPs, Fig. 3. The maps show the magnitude of each field scaled to the respective values of the incident wave. The left (right) column corresponds to the electric (magnetic) field. The incident wave approaches from the bottom, as the inset arrows indicate for the incident wavevector. Figs. 3a and b show the fields’ patterns in the case of $D = 0\%$ or pure EMM; the ESP is excited, showing the typical distribution of an SP for the particle (a), and there is no appreciable magnetic activity (b). On the other hand, for a pure MMM, namely $D = 100\%$, there are both electric and magnetic responses (Figs. 3c and d). This phenomenon occurs because, as said, Ag(50\%)/YIG is indeed an electric-magnetic MM. Although the electric response is not resonant at 431.5 MHz, the magnetic one is. Figs. 3e and f show the results for the optimal condition $(D = 43.6\%, 499.5\text{MHz})$. In this case, both kinds of patterns clearly show dipolar resonances corresponding to the ESP and MSP, respectively. Note in all the maps that the fields inside the particle are constant and the fields outside it have dipolar patterns; this confirms the particle as a dipolar scatterer given its small size compared with the underwater wavelength in the RF domain.
First, a novel concept of magnetic PA generation is illustrated. The results for a pure magnetic particle made of [Ag(50%)/YIG]10%[A]50% are shown in Figs. 3 and 4. Moreover, a double-resonance phenomenon is explored in Fig. 5.

FIG. 3. (Color online) Near-field maps around a 10μm-particle showing plasmonic resonances under plane-wave illumination. The particle composite is [Ag(X)/YIG]10%/YIG/MWCNT(11%)/Al2O3, 50%/50% p2d(A)10%. (a+b+d-f) electric field, (b+d+f) magnetic field. The arrows indicate the wavevector $k$ and field direction. (a+b) electric surface plasmon for $D = 10\%$ or filling fraction$=0$; (c+d) magnetic surface plasmon for $D = 100\%$ or filling fraction$=1$; (e+f) electric and magnetic surface plasmons for $D = 43.6\%$ or filling fraction$=0.436$. The frequencies calculated appear above each panel.

FIG. 4 shows two situations; one calculated using a relatively long pulse of width $\tau = 15$ ns and fluence $F = 60$ J/m$^2$ (a+b), and the other using a relatively short pulse of $\tau = 0.1$ ns and $F = 0.4$ J/m$^2$ (c+d). Figs. 4 and c show frequency spectra while Figs. 4b and d show time signals.

The coupling between the electromagnetic and the acoustic resonances can be visualized through Figs. 4 and c. From Fig. 4a, it can be noted how out of resonance are the excitation pulse and the acoustic oscillations since the monopole mode has a much higher frequency than the pulse width in the frequency domain, compare the black-solid line with the red line with squares. The acoustic resonance is shown by the adimensional magnitude $|P|^2 = \left|\frac{\tilde{P}(\omega)}{P_0}\right|^2$, which means the relative intensity of the acoustic mode. The fact that $|P|^2$ scales to $P_0^2$ makes the result independent of the PA source of excitation. The value of $\tau = 2\sqrt{2ln2}\sigma$, being $\sigma$ the standard deviation of the pulse (a normal distribution), has an associated width in frequency-domain $\sigma_\omega = 1/\sigma$ that does not practically include the particle’s acoustic resonance in the integration. Fig. 4a. As a consequence, the product $\tilde{I} \cdot \tilde{P}$ between the pulse and the acoustic resonance is relatively weak, see the green line with circles in 4a. As a result, the time-domain pressure is relatively low and damped, see the black-solid line in Fig. 4b, resembling the typical shape of the PA pressure generated in optical range with usual plasmonics. The result is independent of the type of SP being excited.

To compare the relative phases between the incoming pulse and the PA pressure, the red line with symbols shows the EM optical range with usual plasmonics. The result is independent of the PA source of excitation. The value of $\tau = 2\sqrt{2ln2}\sigma$, being $\sigma$ the standard deviation of the pulse (a normal distribution), has an associated width in frequency-domain $\sigma_\omega = 1/\sigma$ that does not practically include the particle’s acoustic resonance in the integration. Fig. 4a. As a consequence, the product $\tilde{I} \cdot \tilde{P}$ between the pulse and the acoustic resonance is relatively weak, see the green line with circles in 4a. As a result, the time-domain pressure is relatively low and damped, see the black-solid line in Fig. 4b, resembling the typical shape of the PA pressure generated in optical range with usual plasmonics. The result is independent of the type of SP being excited.

A different situation is presented in Figs. 4(c+d). The shorter the pulse in time, i.e. $I(t) \rightarrow I_0\delta(t)$, the wider the pulse $\tilde{I}(\omega)$ in frequency-domain. In Fig. 4a, $\tilde{I}(\omega)$ is very wide and "contains" several monopolar modes so they are excited. However, as the pulse is normalized, some finite energy is distributed over all the modes. As a result, the coupling with the first monopole mode is weak, and the product $\tilde{I} \cdot \tilde{P}$ is relatively low and spread in all the first modes, see the green line with circles in 4a. Fig. 4a shows again an inadequate phase matching between the signals and the superposition of higher-order monopoles in the pressure (small ripples). It was already mentioned in Photoacoustic phenomenon: Full solution that short pulses result useless since the pressures weaken as $\tau \rightarrow 0$.

On the other hand, a perfect coupling between the resonances is shown in Fig. 5. The condition for the best phase matching between the pulse and the acoustic mode is that the pulse width in the time domain should equal the half period of the mode. In other words,

\[
\Delta t_{\text{pulse}} = \frac{T_1}{2}
\]

\[
\approx 2\sigma = \frac{1}{2F\text{Freq}_A}
\]

\[
\tau = \frac{\sqrt{2ln2}}{2F\text{Freq}_A}
\]  

(29)
In this expression, $\Delta t_{\text{pulse}}$ is the total pulse duration, which approximates to $2\sigma$, $\tau_A$ is the period of the acoustic monopole, and $\text{Freq}_A$ is its frequency. As in Fig. 4, the acoustic monopole is "prepared" to resonate at the same spectral location as the MSP. Using the condition Eq. 29, the value achieved is $\tau = 1.36$ ns so the fluence is $F = 5.46$ J/m$^2$. In this case, the pulse width in the frequency domain is large enough to include only the location of the first acoustic monopole, Fig. 5a. Then, the maximum possible coupling of the pulse with the strongest monopole occurs; see the relatively high values of $|\vec{I} \cdot \vec{P}|$ at low frequencies, green line with circles. Note also from Fig. 5a, that another monopolar resonance is being excited, see the red line with squares around 850 MHz. However, this secondary resonance is poorly included in the convolution and does not count for the effect. As a result of following Eq. 29 a resonant signal for the PA pressure appears in the time domain, Fig. 5b. Fig. 5c also shows the perfect phase-matching between the pulse and the acoustic mode to most efficiently excite the mode. The signal is enhanced and reaches the top value of the scale, namely $P_{\text{max},M} = 1016.6$ Pa, being in the order of PA generation due to gold nanoparticles using optical (electric) plasmonics, even though the pulse fluence is quite low.

Naturally, a comparison of the above case with a "pure electric" case is interesting, since the latter one corresponds to the typical Photoacoustics. Following the example of this work, a particle made of [MWCNT(11%)/Al$_2$O$_3$]$\text{50\%}_\text{A}$$\text{50\%}$ is simulated with the conditions to achieve the double-resonance phenomenon by matching the ESP with the acoustic monopole, Figs. 5a and b. Thus, the spectral location of the first acoustic mode is set at 355.6 MHz, the same location as the ESP, see red line with squares in Fig. 5c. The particle parameters for the calculations performed in Fig. 5a and b are $C_{\text{p,eff}} = 786.77$ J/K/kg, $\rho_{\text{eff}} = 1934$ kg/m$^3$, $\nu_{\text{eff}} = 0.476$, $E_{\text{eff}} = 3.397$ GPa, $\alpha_{\text{eff}} = 1.56 \times 10^{-4}$ K. To achieve these parameters for the particle, the acoustic material is assumed with the same thermo-mechanical properties as the one used in Figs. 2 and 3 except for $\nu_A = 0.4837$, $E_A = 1.543$ GPa.

Three acoustic monopoles of decreasing intensities for increasing energies exist in the range plotted. However, the problem is concerned with the first monopole. Following Eq. 29, $\tau \approx 1.66$ ns is obtained, meaning a fluence of $F = 6.62$ J/m$^2$. Hence $|\vec{I} \cdot \vec{P}|$ is high at low frequencies, green line with circles in Fig. 5c. Consequently, the PA signal in the time-domain is resonant and enhanced compared with the case for "magnetic double-resonance". As the fluence is similar to that for the "pure magnetic" case but a little higher, both results are comparable.

Finally, a triple-resonance phenomenon is illustrated in Figs. 6a and d. The composite [Ag(50\%)/YIG]$_{\text{50\%}_\text{A} \text{50\%}}$ is assumed for the particle with $D = 43.6\%$ as studied above. Now the particle is designed to have the first acoustic monopole at 499.5 MHz, the same spectral location as both the MSP and ESP of the composite, Fig. 6c. The particle parameters for the calculations performed in Fig. 6a and d are $C_{\text{p,eff}} = 559.15$ J/K/kg, $\rho_{\text{eff}} = 3018.71$ kg/m$^3$, $\nu_{\text{eff}} = 0.475$, $E_{\text{eff}} = 10.588$ GPa, $\alpha_{\text{eff}} = 1.558 \times 10^{-4}$ K. To achieve these parameters for the particle, the acoustic material is assumed with the same thermo-mechanical properties as before, except for $\nu_A = 0.4842$, $E_A = 4.93$ GPa.

This time, $|\vec{I} \cdot \vec{P}|$ is maximum for $\tau \approx 1.18$ ns in agreement with the law given in Eq. 29 see green line with circles in Fig. 6c. This value of $\tau$ means a pulse fluence of $F = 4.71$ J/m$^2$. The function $|\vec{I} \cdot \vec{P}|$ is lower in intensity at low energies than that function for Fig. 5a (the scaling factor is practically equal in both cases). However, the PA pressure in the time domain for the triple-resonant particle is enhanced, Fig. 6d, even though the pulse fluence is 29% of 6.62 J/m$^2$. The maximum for this signal is more than doubled compared with the maximum for the "pure magnetic" particle. Besides, the signal lasts longer than the signal for the pure magnetic particle.
Comparing the resonant values of $\tau$ and given an almost constant fluence around 5 J/m$^2$, the triple-resonance signal at the particle surface is about 1.1 times higher than the magnetic double-resonance signal and 35.6 times higher than the electric double-resonance signal for a time $t \approx 29\tau$. The resonant signal for the electric particle lasts shorter in time because the energy of the acoustic mode is the shortest one of all cases, namely $F_{eqA} = 355.6$ MHz. On the other hand, the lower the frequency, the higher the body penetration. However, a pressure lasting for more time supposes an advantage to detect the signal with an ultrasound transducer, except for the possible attenuations due to ultrasound absorption in the outer medium.

As a final comment, let’s compare the results coming from the usual Plasmonics with the present results. In ref\textsuperscript{[12]}, a PA signal generated by a 40-nm gold nanoparticle under plasmon excitation at 532 nm was calculated with a value of $\tau$ in the ns range (5 ns), using a fluence equivalent to $F = 10$ J/m$^2$. The absorption cross-section reported was $\sigma_{abs} = 3.3 \times 10^{-15}$ m$^2$. The pressure signal was calculated at a distance of 1 mm from the particle’s center, giving a maximum in the order of 0.01 Pa. In the example given here due to triple-resonance, the pressure 1 mm away from the particle reaches $\approx 10.6$ Pa, i.e. more than a thousand times the maximum achieved in\textsuperscript{[12]} with conventional plasmonics. It is also known that this kind of nanoparticles may reach several Celsius degrees even for pulsed lasers, possibly damaging the tissues where applied\textsuperscript{[18]}. In addition to the advantage of having high penetration in tissues for RF, the maximum temperature rise in the case of Figs.\textsuperscript{[5]-d} does not reach 2.6 mK. This maximum temperature rise is calculated with a convolution solution based on the EM pulse and Green’s function of the thermal problem\textsuperscript{[22]}. The value was also checked with the approximation of point-absorber model given in Refs.\textsuperscript{[13]-[15]} since the condition $\tau \ll \frac{c}{2\omega}$ is valid, being $a_t$ the thermal diffusivity of the particle. Both calculations gave very similar results. All the mentioned improvements based on RF-MMs and fast RF pulses highlight the design of PA microtransducers. With the current technologies, this design may find applications in PA imaging of single cells\textsuperscript{[20]}, in-vivo monitoring of drug delivery\textsuperscript{[21]} and food treatment\textsuperscript{[21]} among others.

**CONCLUSION**

An unconventional photoacoustic concept was proven to be possible by simulating microparticles made of special random metamaterials mixed with soft polymers. These microcomposites have nanoelements at concentrations near the percolation limit and become conductors in the MHz range, allowing for the excitation of surface plasmons in radiofrequency. The soft polymer allows for the excitation of acoustic modes in the same range so both the electromagnetic and acoustic resonances can couple. This coupling gives an enhancement of the photoacoustic pressure if the materials are carefully chosen. To fully understand the basis of the phenomenon, a solution to a general photoacoustic problem is fully developed by means of convolution products. Neither this complete solution nor their consequences were reported before. The key concepts in this novel paradigm are the double-resonance and triple resonance phenomena to enhance the ultrasound pressures and make them last longer in time. More signal duration permits more detection efficiency than in the current approaches because the pressures become also resonant. The results illustrated here improve those obtained from the optical plasmonics with gold nanoparticles. The problems related to usual optoacoustics as thermal damage and poor tissue penetration are avoided, in particular using nanosecond electromagnetic pulses and MHz frequencies. In this way, a real non-invasive technique is suggested with enhanced body penetration. The proposed approach paves the way to realize thermo-acoustic imaging devices with a high resolution. These could be used for sensitive materials as organic tissues, ancient artwork, or food, among other underwater applications.

**SUPPLEMENTARY MATERIAL**

See the supplementary material for the effective medium formulas used, thermal and mechanical properties of the materials, and a calculation of the PA pressure inside the particle for the case given in Figs.\textsuperscript{[5]-d}. 

FIG. 6. (Color online) Electric double-resonance (a+b) vs. triple-resonance phenomenon (c+d). In (a+b), the particle is made of [MWCNT(11%)/Al$_2$O$_3$]$_{35\%}$ [A$_{35\%}$] at the electric plasmon resonance. The incoming pulse obeys the acoustic-resonance condition \textsuperscript{[29]} ($\tau \approx 1.66$ ns) where $F_{eqA} = 355.6$ MHz is the resonance acoustic frequency that matches the electric surface plasmon. In (c+d), the particle is made of [Ag(50%)/YIG]$_{21.8\%}$ [MWCNT(11%)/Al$_2$O$_3$]$_{28.2\%}$ [A$_{35\%}$]. Now Eq.\textsuperscript{[29]} gives $\tau \approx 1.18$ ns, and $F_{eqA} = 499.5$ MHz matches the resonance of the electric and magnetic surface plasmon.
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DATA AVAILABILITY

The data that support the findings of this study are available within the article and its Supplementary Material.

CONFLICT OF INTEREST

The authors have no conflicts to disclose.

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