A theoretical framework for acoustically produced luminescence: From thermometry to ultrasound pressure field mapping

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Acoustically produced luminescence (APL) can be used for fast and easy mapping of ultrasound pressure fields, allowing quantitative investigation of these fields for a wide range of acoustic frequencies and pressures. APL offers a fast and inexpensive alternative for the conventional point-by-point hydrophone scanning. This can benefit industrial and medical ultrasound applications that experience stringent certification and safety requirements on pressure field characterization. APL was shown to originate from absorption-mediated heating by ultrasound irradiation of a membrane material, which consists of a polymer binder and a luminescent material (or phosphor). This heating induces local thermoluminescence emission, which is proportional to the ultrasound pressure. However, a precise framework describing the physics of the APL process, allowing the retrieval of acoustic field information from the measured light emission has been lacking. Here, we present a full theoretical model of the APL phenomenon, allowing the reconstruction of both the pressure and temperature fields from the measured luminescence. The developed theoretical model is verified using finite-element modeling and experimental validation. We then demonstrate how APL can be used to obtain a 3D reconstruction of an ultrasound pressure field, in a fast and easy way. Finally, the general model demonstrated here can also prove useful for other applications, e.g. in luminescence-based thermometry using persistent phosphors.

1. Introduction

Ultrasound (US) imaging is the most widely used imaging modality in clinical practice [1]. Additionally, the use of US is well-established for non-destructive testing in industry [2,3] and for acoustic microscopy [4]. New developments are constantly broadening the application range of US. For instance, the introduction of microbubbles for therapeutic use in recent years has shown potential for single cell therapy, gene therapy, blood-brain barrier opening, local drug delivery and histotripsy [5–9]. Moreover, the development of high-speed plane wave imaging over the past decade has opened the door for different branches of ultrasonic research in testing [10,11], microscopy [12,13], super localization and functional imaging [4,14–16], particle manipulation [17–22] and non-linear US [2,10,23]. These advanced ultrasonic applications share the same bottleneck: they require detailed knowledge of the ultrasonic pressure field to produce quantitative results. Several two dimensional pressure mapping techniques exist, but they offer limited accuracy and are technically challenging [24–26]. Schlieren imaging maps the diffraction of light from a collimated light source as it passes through the US field. This integrative technique is affected by out-of-plane waves, difficult to quantify and restricted to visualizing planes orthogonal to the transducer [24]. Thermal imaging was also proposed to image pressure fields, but this method suffers from the limitation of infrared imaging in terms of penetration through water for immersed samples and its relatively high cost [26]. Therefore, US field characterization is commonly achieved by scanning the entire field point-by-point with a calibrated hydrophone. Hydrophone scanning requires physical motion between each acquisition point, making it time consuming (1 h for a limited 2D scanning to 1 day for a precise 3D scan). Furthermore, a calibrated optical hydrophone (with an increased spatial resolution of 10 μm over

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piezo or PVDF (polyvinylidenfluorid) hydrophones) is expensive and requires additional equipment to operate [27,28]. Any improvement in the time constraints quantifying these pressure fields will have a significant impact on the development of US innovations in academia, as well as in industry and healthcare.

We have recently presented an alternative approach to map US pressure fields by means of acoustically produced luminescence (APL) [29,30]. APL takes advantage of the sound-to-heat-to-light conversion properties of specifically designed semi-transparent luminescent membranes to obtain a quantitative cross-sectional visualization of the US beam in a matter of seconds [30]. The membrane consists of a thin slab of PDMS (polydimethylsiloxane) polymer embedded with microparticles of a luminescent material, like BaSiO$_2$:Eu$^{2+}$ (BaSiON). This is an oxynitride luminescent material, or phosphor, that features persistent luminescence with an afterglow duration of over 1 h at room temperature. Prior to the US irradiation, the membrane is exposed with blue light or UV radiation, which fills (or charges) certain defects in the phosphor [31,32]. When an acoustic wave insonifies such a charged PDMS/BaSiON membrane, the high acoustic absorption leads to local heating, increasing the probability of the release of the trapped charge carriers and, subsequently, persistent luminescence emission from the BaSiON particles. This effect of ultrasonically induced light emission was discovered some years ago, and at first it was only investigated phenomenologically [29,33-39]. In fact, acoustically induced thermoluminescence was initially (mistakenly) assumed to be mechanoluminescence linked to the acoustic radiation pressure or compression [34,35,37]. Previously, we have shown that the physical mechanism underlying the APL phenomenon is in fact driven mainly by thermoluminescence, rather than mechanoluminescence [30]. Note that the acronym APL stands for Acoustically Produced Luminescence in this work, in contrast to the acronym used in Ref. [29], which used a wrong assumption of the physical mechanism. The contribution of mechanically induced emission is negligible compared to the thermally induced emission, which follows from the excellent agreement between experimental APL emission and thermoluminescence simulations, and the slower emission dynamics compared to mechanoluminescence (which peaks very quickly, whereas APL emission exhibits a slower build-up) [40]. Furthermore, we previously determined an empirical calibration curve that provides pressure information based on the APL emission [30]. However, this curve depends on a series of calibration parameters, including US frequency and the specific spatio-temporal pressure field. Practical implementation of the APL technique thus requires a generalization of the calibration which, in turn, requires a thorough physical understanding of heat deposition and light generation, both in space and time. In this paper, we develop an analytical acoustothermal (AT) model to predict the transient and spatial behavior of the acoustically induced heating. This AT model is validated by comparing it to a 3D finite-element method (FEM) simulation and to experimental data. The subsequent APL emission is then calculated using an established thermoluminescence (TL) model [30]. The combined model (AT and TL) is shown to accurately predict the full APL phenomenon. We also propose an (approximate) inverse model, which allows to analytically reconstruct the spatio-temporal pressure maps of the US generation, directly from the APL emission. Finally, we demonstrate a practical example of how the AT model can generate a fast and quantitative 3D visualization of an entire ultrasonic beam.

2. Material and methods

2.1. Sample preparation

BaSiO$_2$:Eu$^{2+}$ powders were synthesized using a solid state reaction method as described before [30]. PDMS membranes were produced by thoroughly mixing the powder in a 1:4 ratio with PDMS resin. The curing agent was then added, and the mixture was stirred. Finally, the mixture was drop-casted on a PMMA substrate. The thickness of the layer was controlled by the razor blade method. The substrate carrying the layer was left in a dry stove at 50 °C for 24 h, after which the deposited and cured layer was removed. This produced a standalone, semi-transparent composite PDMS/BaSiON membrane with a thickness of 285 μm, as measured in a scanning electron microscope (Hitachi S-3400 N) at a chamber pressure 25 Pa and an electron acceleration voltage of 20 kV.

2.2. Measurement setups

A physiotherapeutic Gymna 200 piston transducer was used to produce US waves at a frequency of 3.3 MHz and acoustic power ranging from 0.1 to 2.0 W cm$^{-2}$. The resulting APL emission was recorded by a Ximea MC031MG-SY-UB camera equipped with a Sony CMOS sensor chip (3.1 MPix) and an Edmund Optics lens with a focal length of 3.5 mm (TechSpec C Series). Unless specified otherwise, the exposure time set for each recording was 250 ms. The validation experiments for the prediction and inverse reconstruction models were carried out with a GE H5K immersion transducer (aperture of 10 mm) with a center frequency of 5 MHz. The pressure references requiring a hydrophone scan were recorded with an Onda HGL-0400 lipstick hydrophone (aperture of 400 μm) equipped with an AH-2010-100 pre-amplifier (bandwidth 50 kHz–100 MHz), operated with a step size of 75 μm, taking about 30 min per scan. Laser Doppler Vibrometer experiments were performed with a 3D infrared scanning machine (Polytec PSV-500-3D XTRA).

2.3. Measurement protocol

A full APL measurement is composed of three phases. First, a few reference dark images of the membrane are recorded. Secondly, the membrane is excited with blue light or UV radiation until it reaches saturation. After the optical excitation is turned off, the camera starts to record the afterglow (AG) emission. Thirdly, after a certain time delay chosen by the experimenter, the transducer is switched on for a certain duration, typically between 10 s and 30 s. Additionally, after recharging the membrane, an AG recording in the absence of US irradiation is acquired in the same manner (same optical excitation conditions, camera settings etc.) and both the APL and AG recording are corrected for stray light by subtracting an average dark frame. Then, the standard AG normalization processing procedure is performed, where each frame of the APL recording is divided by the corresponding frame of the AG recording, thus correcting for local loading inhomogeneities in the membrane [30].

2.4. Modeling/simulation methods

APL was modeled through an analytical model implemented in MATLAB$^{(R)}$, and verified by finite-element (FE) modeling using the standard implicit solver of Abaqus software. For the FE simulation, the insonified region (containing both membrane and water) was meshed with 25,000 8-node quadratic axisymmetric quadrilateral heat transfer elements (DCAX8) with a size of 4 μm in the r-direction and 6 μm in the r-direction. The rest of the membrane and the surrounding water were modeled using 7500 linear elements (DCAX4) in a graded mesh (bias ratio of 250), fine near the heat source and coarser further away from it. This mesh structure is shown in Figure S1b of the Supporting Information.

2.5. PDMS/BaSiON membrane characterisation

The US attenuation of the PDMS/BaSiON membranes was characterized by repeated insonation with an US pulse, consisting of 30 cycles and slightly tapered at the start and at the end of the pulse, with frequencies ranging from 0.5 to 9.5 MHz. The measured attenuation, recorded with a second transducer, originated from both reflections at the PDMS/water interfaces and from US absorption. Since the acoustic
reflection coefficients do not depend on the frequency \([41]\), transmission through the thin membrane resulted in interferences which translated into a regular modulation of the attenuation as a function of frequency \(f\). The acoustic absorption coefficient of the membrane can be described using an empirical law \(a(f) = a_0 f^b\) (for reference, \(b = 2\) for water and \(b \approx 1\) for soft tissue) \([42]\). This allows the separation of the contributions of reflection and absorption to the attenuation, as only the absorption leads to heat dissipation within the membrane. Since the thickness of the membrane is comparable to (or less than) the acoustic wavelength at these frequencies, the transient times for interference are on the order of the acoustic period. Therefore, the comparatively long US pulses provided results that are representative of a continuous exposure, as used for APL. Acoustic scattering from the particles was assumed to be negligible and was not considered in this procedure, and thus the acoustic attenuation was a considered a result of acoustic absorption alone.

3. Results and discussion

3.1. APL fundamentals

Fig. 1 shows an example of a typical APL measurement performed in a water tank. The PDMS/BaSiON membrane is at a fixed position and the US transducer is placed with its central axis (x-axis) orthogonal to the membrane. After charging to saturation with either blue light or UV radiation, the membrane shows decay AG emission with wavelengths between 450 and 550 nm (black curve in Fig. 1a) \([30,31,43]\). After a certain time delay (15 s in this example), the US transducer is switched on, producing a 2D cross section of the US beam on the membrane, visible to the naked eye (Fig. 1b). The light intensity is recorded by a camera placed outside the water tank, imaging the light emission of the membrane in transmission. Switching on the US transducer results in as a sharp rise in the light emission intensity (Fig. 1a, green curve), which quickly reaches a peak before decaying at a faster rate than the reference AG (Fig. 1a, black curve). Once the transducer is switched off again, the light emission drops below the reference AG level. The initial optical excitation of the membrane fills up specific electronic defects (called traps) with charge carriers originating from the excited luminescent centers (electrons in the case of the europium-doped BaSiON). Optically saturating the membrane at the beginning of the measurement ensures a good reproducibility. This saturated state corresponds to a dynamic equilibrium between optical trapping and thermal detrapping at room temperature and can be identified by the stabilization of the light emission intensity from the membrane (\(t < -15\) s).

When the US is switched on (between \(t = 0\) s and \(t = 5\) s, with constant US intensity), the emission intensity above the AG intensity, due to increased thermal detrapping as an effect of US-induced heating of the BaSiON phosphor. The luminescent particles are homogeneously dispersed within the binder matrix and the thermal properties of the membrane are assumed to be isotropic in a first order approximation \([30]\). Acoustic extinction when the US wave passes through the membrane mainly results from absorption and reflection, with only the former giving rise to heating and thus to APL emission. The dynamics of both heat generation through absorption and heat transfer through diffusion determine the local temperature field and thus the light emission. Being the key material parameter in the APL phenomenon, the absorption coefficient of the membranes was experimentally characterized as a function of frequency (see Section 2 of the Supporting Information), revealing an absorption of the APL membrane of approximately 10 times that of unloaded PDMS \([44]\).

Fig. 2 shows the scheme which was adopted to develop a theoretical framework to describe the APL phenomenon, and serves as a guideline in this paper. Two theoretical models were developed. The first one describes the AT conversion of US waves into heat, while the second describes the thermally induced light (TiL) emission based on a given temperature field. The AT model is validated by finite-element simulations in this work, while the TiL model was validated before \([30]\). The forward prediction model (combination of steps 1 and 2) predicts the measured APL features. In practice, however, interest lies mainly in the inverted models (steps 3 and 4) which allow the recovery of the acoustic pressure field directly from the observed APL emission intensity. These steps are combined in the inverse reconstruction model.

Here, it should be noted that any time dependency described in this work should be viewed at the timescale of global variations of the ultrasound pulse, and not of the oscillation period of its pressure amplitude. APL thus only provides temporally averaged information about the US pressure, and no phase information can be extracted.

3.2. The acoustothermal model

In the AT model, we consider the homogeneously loaded PDMS/BaSiON membrane as a thermally isotropic plate immersed in a stagnant water bath at temperature \(T_w\). This plate is insonified by an axially symmetric US beam, corresponding to the cross-sectional profile of most common single-element transducers. The typical geometric size scale of the pressure field in the radial direction is in the order of \(\lambda_p\), with \(\lambda_p\) the numerical aperture of the transducer and \(\lambda\) the acoustic wavelength. For example, \(\lambda_p\) varies between 3 and 0.6 mm for a frequency range of 1–5 MHz and a numerical aperture of 0.5. The geometrical features in the axial direction are typically several times larger. Therefore, and since (i) the sensing membrane is thin compared to the US wavelength (thickness \(d_m = 285 \mu m\)), and (ii) most of the acoustic intensity is transmitted through the membrane (70% at a frequency of 5 MHz for the PDMS/BaSiON membrane discussed here, see Section 2 of the Supporting Information), we assume no significant pressure amplitude variations within the thickness of the membrane.

In stagnant water, thermal diffusion is considered the dominant mechanism for heat transfer, as radiative transfer can be neglected owing to the small temperature differences at play. The typical time scale for thermal diffusivity over a distance \(L\) inside a medium with a diffusivity \(D\) is given by the characteristic time \(t_c = L^2/D\). The ratio of the characteristic time for thermal diffusion from the center of the membrane towards the edge (the radial direction) to the one for thermal diffusion along the axis of the US beam (the axial direction) can then be

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**Fig. 1.** a) APL experiment with a physiotherapeutic transducer (\(f = 3.3\) MHz, \(I_{US} = 2.0\) W/cm\(^2\)) insonifying the PDMS/BaSiON membrane. The excitation light is switched off at \(t = -15\) s, and the membrane is irradiated with US between \(t = 0\) s and \(t = 5\) s. b) Optical recording of the APL emission intensity at \(t = 1\) s.

**Fig. 2.** Conceptual picture of the different energy conversions at play during an APL measurement. On the left side is an US transducer, in the center a temperature field of the PDMS/BaSiON membrane caused by the ultrasonic heating, and on the right the resulting APL emission pattern. The blue arrows indicate the steps of the forward prediction model, while the red arrows are showing the inverse reconstruction model.
estimated: \( \frac{2\pi}{\lambda} \frac{d^2 P_{\text{obs}}}{d\omega d\Omega} \). Since the diffusivities of water (\( D_w = 1.46 \times 10^{-7} \) m\(^2\) s\(^{-1}\) and PDMS [45] (\( D_{\text{PDMS}} = 1.06 \times 10^{-7} \) m\(^2\) s\(^{-1}\)) are comparable, this can be further approximated as \( \frac{2\pi}{\lambda} \frac{d^2 P_{\text{obs}}}{d\omega d\Omega} \). This ratio lies between 0.06% (at a frequency of 1 MHz) and 1.50% (at a frequency of 5 MHz) for the membrane and transducer used here. This means that the heat transfer occurs predominantly in the axial direction, justifying the use of a heat flux boundary condition, which entails that the heat flux \( \Phi(r,t) \) is directly linked to the thermal energy deposited in the membrane by the US beam. Initially, the membrane is at thermal equilibrium \( T = T_w \). When the transducer is switched on (at \( t = 0 \)) the insonified area of the membrane starts to heat up. The temperature field of the membrane/water system, according to these approximations, can be calculated by solving the heat diffusion equation in cylindrical coordinates:

\[
\frac{\partial T(r,z,t)}{\partial t} = D_w \nabla^2 T(r,z,t).
\]

(1)

Here, \( r \) indicates the radial direction in the membrane and \( z \) the axial one, along the beam axis. As we consider an acoustically thin membrane and therefore assume a homogeneous pressure through the thickness of the membrane, the pressure wave is, locally, a plane wave. The general solution of Equation (1) is found in the Fourier-Hankel space, considering the imposed heat flux at \( z = 0 \) (the membrane/water interface). It is given by

\[
T(r,z,t) = T_w + \int_{-\infty}^{\infty} \int_{0}^{\infty} H_0(k,\omega) J_0(kr) \exp \left\{ -ikz \left[ 1 + \left( \frac{\omega}{\Omega_D} \right)^2 \right] + i\alpha t \right\} \exp(i\omega t) dk d\omega,
\]

with \( \omega_B \) the thermal conductivity of the water and \( J_0(kr) \) the zeroth-order Bessel function of the first kind. \( G(r,\omega) \) is the Fourier transform of the heat flux \( \Phi(r,t) \) at the membrane/water interface, and \( H_0(k,\omega) \) is the zeroth-order Hankel transform of \( G \):

\[
H_0(k,\omega) = \int_{0}^{\infty} G(r,\omega) J_0(kr) r \, dr.
\]

(3)

The derivation of this general solution can be found in Section 1 of the Supporting Information. When applied to the specific case of APL, the acoustic intensity \( I_{\text{US}} \) of a plane wave (in W m\(^{-2}\)) as it travels a distance \( \Delta m \) through the membrane can then be written as:

\[
I_{\text{US}}(r,z,t) = \frac{P(r)^2}{2\rho_w c_w} 10^{\frac{z_B}{10}} f(t).
\]

(4)

Here, \( P(r) \) is the pressure field in the radial direction and \( f(t) \) is the time envelope of the transducer operation. \( \rho_wc_w \) is the specific acoustic impedance \( \zeta_m \) of the membrane and \( \Omega \) is the acoustic absorption of the PDMS/BaSiON membrane in dB m\(^{-1}\). The total drop in acoustic intensity due to absorption after passing through the membrane is then

\[
\Delta I_{\text{US}}(r,z = \Delta m, t) = \frac{P(r)^2}{2\rho_w c_w} \left( 1 - 10^{\frac{z_B}{10}} \right) f(t).
\]

(5)

As the heat flux at each membrane/water boundary is given by \( \frac{2\pi}{\lambda} \frac{d^2 P_{\text{obs}}}{d\omega d\Omega} \), the temperature maps \( T(r,z,t) \) can be computed by inserting the Fourier Transform of Equation (5) into Equation (3), and then computing Equation (2). Note that the acoustic absorption of the membrane is the only material parameter needed in this model. This allows the PDMS/BaSiON membranes to be tailored to specific experimental needs (e.g., higher emission intensities, different waiting times, different acoustic frequencies or pressures, etc.) by tuning their composition ratio, and thereby their acoustic absorption.

In order to verify the approximation made (i.e., treating the membrane as an interface with no thickness), the AT model was compared to an axisymmetric finite-element (FE) model. A schematic of the model structure is shown in Fig. 3a, and the mesh structure is shown in Figure 3b of the Supporting Information. The outer boundary condition \( T = T_w \) is enforced 33.5 mm away from the membrane in the \( z \)-direction, and 10 mm in the \( r \)-direction. The volumetric heat generation in the FE model obeyed Equation (5), with a Gaussian width consistent with our single-element 5-MHz transducer (FWHM of 1.44 mm). The full list of parameters used in the FE modeling is given in Section 2 of the Supporting Information. Fig. 3b shows the temporal temperature profiles \( T(r) \) of the membrane, calculated with both the analytical AT model and the FE simulation (evaluated at the center of the membrane for the FE simulation) at three different radial distances. Fig. 3c shows the radial temperature profiles \( T(r) \), calculated with both methods (evaluated at the center of the membrane for the FE simulation). The FE simulation and the AT model are in excellent agreement, which validates the physical assumptions and approximations used in the analytical model.

The analytical AT model can also be experimentally validated, using a thermographic recording of the insonated PDMS/BaSiON membrane. The membrane was insonated at \( I_{\text{US}} = 2.0 \) W cm\(^{-2}\) with a physiotherapeutic Gymna 200 piston transducer placed (in water) at a distance \( x = 6 \) cm. The resulting temperature field of the membrane was recorded by a FLIR A35 thermographic camera. Fig. 3d shows the experimental and analytical temporal temperature profiles \( T(r,t) \) at three different radial distances (evaluated at the water/membrane interface for the FE simulation). The AT model shows excellent agreement with this experimental data. Note that a non-axisymmetric pressure field can be readily handled by the proposed model by means of higher order Bessel functions and Hankel transforms, sacrificing simplicity and readability. This extension would allow to model more complex beam shapes created by, e.g., linear arrays.

### 3.3. The forward prediction model

The second modeling step of Fig. 2 consists of the TL model, of which the details and validation can be found in Ref. [30]. In short, the TL emission is calculated according to the Randall-Wilkins equation for TL [46,47]:

\[
I_{\text{TL}}(t) = \sum_{i} n_{\text{Ti}} \exp \left( -\frac{E_i}{k_B T(t)} \right) \exp \left( -s \int_{0}^{t} \exp \left( -\frac{E_i}{k_B T(t')} \right) \, dt' \right).
\]

(6)

This model assumes first-order kinetics, i.e., each detrapped electron leads to recombination and thus TL emission. In this equation, \( n_{\text{Ti}} \) is the number of trapped electrons (per unit volume) at time \( t = 0 \), for the traps characterized by a trap depth \( E_i \), \( s \) is the frequency factor of the material (also described as the escape frequency), which is a measure for the rate (in Hz) at which a trapped electron interacts with a lattice phonon and thus “attempts to escape”. \( E_i \) denotes the energy (in eV) of the thermal barrier for a specific trap level \( i \). This thermal barrier, also called the trap depth, is the energy which is required for the trapped charge to be released from the trap, after which recombination at the luminescent center can occur. The \( E_i \) parameters used in the TL model are not necessarily related to the physical trap level energies of BaSiON, as they were not derived from measurements but rather fitted to the persistent luminescence behavior of BaSiON. A set of 9 of these trap depths provide a good representation of both the glow curve and the AG intensity profiles of BaSiON [30]. Adding more trap levels does not significantly improve the TL model. Equation (6) describes the emission of a single, free-standing phosphor particle. In a PDMS/BaSiON membrane, however, optical scattering can occur at neighboring particles, and Equation (6) cannot, in principle, be simply applied to each location of the membrane to predict the actual, observed emission pattern. The effect of optical scattering on the spatial resolution of an APL characterization
was previously shown to be less than 200 μm (see Section 3 of the Supporting Information), and the loss of spatial resolution is therefore neglected [30]. Thus, the emission behavior described by Equation (6) is considered to be consistently applicable for the whole membrane.

The accuracy of the full forward prediction model (steps 1 and 2 in Fig. 2) can be demonstrated by predicting a full APL experiment, including afterglow and US insonation. An APL experiment was performed using a GE H5K single-element unfocused immersion transducer (f = 5 MHz) placed at a distance x = 10 cm from the membrane. The transducer was driven in continuous mode for 10 s, providing a step function as the US application profile. The US pressure field $P(r,t)$ in water, 10 cm away from the transducer, was measured using a needle hydrophone and fed into the forward prediction model, which then delivered both the generated heat map and the TiL emission intensity map. Fig. 4 shows the calculated temperature map (first column, step 1 of the prediction), the calculated TiL intensity map (second column, step 2 of the prediction) and the experimentally recorded APL intensity map (third column), at four different times during the APL process. The last column of Fig. 4 gives a direct comparison between cross sections (horizontal scan line (y = 0) through the center) of the measured and calculated intensity maps.

The fourth row of Fig. 4, at $t = 12$ s (i.e. 2 s after the transducer was turned off), shows a depletion effect at the location of the US spot. This effect is a 2D representation of the dip below the AG intensity profile after ending the US irradiation, which is also visible in Fig. 1a, and is due to the enhanced detraping during the US irradiation [30]. During insonation, ultrasonic heating induces a temperature rise $\Delta T(t)$ above room temperature. Since the detraping probability $p$ of a trap of depth $E$ follows a Boltzmann-like rule $p(T) = p_0 e^{-\frac{E}{k_B T}}$, an increase in temperature leads to an increase in the probability that this trap contributes to the emission. When the transducer is switched off, part of the traps have been emptied and cannot contribute to the AG emission anymore. Note that the prediction model succeeds in accurately describing this effect. Furthermore, the developed forward prediction model works for all insonation parameters, i.e. US frequency, duration of the insonation and delay between the charging and the insonation, thereby removing any calibration restrictions. Finally, the APL visualization method should also work for very short ultrasound durations (e.g. order of ms), as long as the energy deposited in the membrane is sufficiently high to induce an appreciable temperature rise (in the order of 0.1 °C). This requires high acoustic powers, accessible with certain transducers (e.g. used in HIFU [48–50]). Short timescale APL brings the advantage of avoiding thermal diffusion effects to a large extent, likely providing a more linear relationship between pressure and temperature, over the entire irradiated area. However, these high acoustic powers typically induce second order effects such as acoustic streaming, nonlinear propagation and possibly cavitation, and high-speed cameras are needed for the collection of emission light. Thus, while short timescale APL visualization is possible in theory, it will be challenging in practice.
3.4. The inverse reconstruction model

3.4.1. Analytical inversion

Step 3 in Fig. 2 corresponds to the calculation of the temperature map from an APL experiment, which can be obtained by inverting Equation (6) in the Fourier domain, resulting in

$$F(k, \omega) \approx j\omega H(k, \omega) j\omega G_g$$  

(7)

where $F(k, \omega)$ is the Fourier transform of $T(r, t)$, the two dimensional temperature field of the membrane. $H(k, \omega)$ is the Fourier transform of the function $H(r, t) = \frac{T(t)^2 \ln \left( \frac{I_{APL}(r,t)}{I_{AG}(r,t)} \right)}{\sum_{i=1}^N E_i \exp \left( \frac{-E_i}{T_w} \right) I_{AG}(r,t) I_{AG}(r,t)}$  

(8)

with $I_{APL}(r,t)$ and $I_{AG}(t)$ the observed APL and AG intensity, respectively, at time $t$ and location $r$ with respect to the center of the membrane. $G_g$ is $G(r, t = t_g)$, with

$$G(r, t) = \frac{\sum_{i=1}^N E_i \exp \left( \frac{-E_i}{T_w} \right) I_{AG}(r,t) I_{AG}(r,t)}{\sum_{i=1}^N E_i \exp \left( \frac{-E_i}{T_w} \right)}$$  

(9)

$I_{AG}(r,t)$ is the contribution to the overall AG intensity profile $I_{AG}(t)$ from charge carriers released from the trap $i$ with trap depth $E_i$ (see
Equation (2)), $t_g$ is the point in time around which we want to perform the inversion and recover the temperature field of the membrane. The approximate inversion of the model is optimal around $t_g$ and degrades as $|t - t_g|$ increases. The calculation of the US pressure experienced by the membrane from its temperature field (Step 4 in Fig. 2), is found by inverting Equation (2):

$$P^2(r, t) = \frac{4\rho c_v}{1 - 10 \frac{\kappa}{\rho}} \int_0^\infty R e^{-k} \left[ 1 + \left( \frac{\omega}{kD} \right)^2 \right] \exp \left( \frac{i \cdot \text{atan} \left( \frac{\omega^2}{k^2} \right)}{2} \right) J_0(kr) dk,$$

(10)

with

$$R_e = k \int_0^\infty \int_0^\infty rT(r, t) J_0(kr) \exp(-i\omega t) dr dw$$

(11)

and $\mathcal{F}(\omega)$ the Fourier Transform of the time envelope $f(t)$ of the ultrasound application (see Equation (4)). The full derivation of Equations (9) and (10) can be found in Section 4 of the Supporting Information. The upper row of Fig. 5 shows the procedure leading up to and including the reconstruction through analytical inversion. This is shown for an actual APL measurement (green curve in Fig. 5b) as well as for a simulated TiL event, for which the US generation is shown in Fig. 5a. The final reconstructed temperature and pressure profiles for both cases are shown in Fig. 5c.

The simulated pressure profile (blue dashed line in Fig. 5a) mimics the experimental conditions of a simple on-off use of a GE H5K transducer with a peak pressure $P_0 = 642$ kPa at the membrane. This pressure profile was used as input for the AT model to produce a temperature profile (red dashed line) and, from this temperature profile, the TiL emission of Fig. 5b (black dashed line). Additionally, the emission of an APL experiment is shown (full green line), which was obtained with the same ultrasound parameters (as verified with a hydrophone measurement). Fig. 5c shows the result of the reconstruction calculation for both cases, performed at $t_g = 33.3$ s (see Equation (9)). This timepoint is close to half of the insonation duration, and was found to give the best reconstruction results (see Section 5 of the Supporting Information). In this example, the expected pressure profile output of the simulated reconstruction procedure is a square shaped pulse, i.e. the input shown in Fig. 5a. The reconstructed pressure profiles (Fig. 5c) indeed reach the correct maximum value but, instead of showing the initial square shaped pulse, exhibit a build-up and keep rising slowly until the US transducer is turned off. Then, the pressure profiles do not reach zero immediately but show a more gradual decrease, following the typical heat loss features of the temperature profile. However, this gradual decrease happens much faster for the simulated reconstructed pressure profiles (approximately 2 s, see Fig. 5c) than for the temperature profiles (more than 5 s), which agrees with the input. This behavior is visible for the reconstruction of both the simulational and experimental input, and is a result of the approximations made in the derivations of Equation (2) and its inversion, Equation (10). For instance, for the assumption of a homogenous, isotropic membrane, the heavier BaSiON particles will transfer more of the acoustic energy into vibrational energy, thus heating up more than the surrounding polymer chains. This may induce small local temperature variations, meaning the membrane cannot be modeled as a zero-thickness interface. In practice, when we have knowledge of the transducer application profile, the reconstructed pressure profile can be fine-

![Fig. 5](image-url)

Fig. 5. (a) Simulated pressure input (blue dashed line), corresponding to an APL experiment at a frequency of 5 MHz, and the resulting calculated temperature rise (red dashed line). This generates (b) Experimentally recorded APL emission (green line) and the analytically calculated TiL emission (black dashed line), which is used for the reconstruction calculation in (c). On the one hand, the temperature and pressure profiles are recovered (dashed lines). Additionally, the full lines show the reconstruction calculation results for the experimentally recorded APL intensity profile. (d) Hydrophone recording of the US beam at the membrane location. (e) Reconstructed US pressure distribution. (f) Comparison of the vertical scan line ($x = 0$) through the center of the US beam profile, for the hydrophone recording and for the beam profile resulting from the reconstruction method.
tuned following this knowledge. In the example of Fig. 5a–c, the start and end times of US insonation are known, and the correct maximum pressure is recovered. Thus, the complete pressure profile can be simply reconstructed by stating that $P_0 \approx 642$ kPa between 27 s and 37 s, and zero otherwise. In most practical cases, a step profile is used for insonation, and the recovery of the maximum pressure is thus sufficient. In case the US insonation follows a more complex timing scheme, the reconstruction timepoint $t_g$ can be (iteratively) moved to the time of interest.

The procedures for the reconstruction in Fig. 5a–c are demonstrated for the center of the US beam profile ($r = 0$). However, the full APL intensity map is available, allowing the reconstruction of the full pressure map $P(r)$ at the membrane surface. Fig. 5d shows a hydrophone recording of the US pressure field at the position of the PDMS/BaSiON membrane and Fig. 5e shows the same field as resulting from the reconstruction procedure. Fig. 5f shows a vertical scan line through the beam profile obtained by the hydrophone scan, as well as through the reconstructed pressure map obtained by reconstruction calculation. Note that the vertical direction is chosen for comparison because of the slight asymmetry in the x-direction of the two maps, which is attributed to a small misalignment during recording. The absolute value of $P_0 = 642$ kPa (the pressure at the center of the map) can be retrieved with an error margin of roughly 5%, which is smaller than the typically best calibration error for needle hydrophones. One main advantage of the reconstruction through analytical inversion is that it can retrieve the pressure field from any arbitrary temporal profile of the acoustic pressure amplitude. This is demonstrated in Section 6 of the Supporting Information, for a repeated insonation profile. The retrieval of the

![Graphs](image-url)
temperature field of the membrane by using the inverted AT model can be seen as a special type of luminescent thermometry. Where traditional techniques probe direct changes in luminescent characteristics (such as peak position [51,52], peak intensity [53-55] or lifetime [56-58]) to monitor temperature, APL thermometry can reveal the temperature rise caused by insonation.

3.4.2. Calibration curves

The approximate inverse model only offers a good precision around a specific reconstruction timepoint \( t_r \) and requires some computation that uses the full experimental time duration. As an alternative method, the validated forward prediction model can be used to simulate the experimental conditions and, potentially, generate precise calibration curves. This kind of procedure requires a high level of control on the experimental conditions and timing (e.g. charging into saturation, delay time, insonation duration) in order to accomplish an accurate match with the intensity profiles calculated by the forward prediction model. Additionally, given the complex interplay of heat transfer and light generation involved in the APL process, it is not trivial to determine which parameter of the intensity profile can best function as the base of a calibration curve to provide accurate pressure reconstruction. The following section investigates empirically which feature of the intensity profiles is most suited for generating calibration curves based on both experimental data and predictions from the analytical model. Fig. 6a shows a set of experimentally recorded APL intensity profiles \( I_{APL}(t) \) resulting from insonation with a GE HSK transducer (center frequency 5 MHz), operated at different driving voltages, thus generating different pressures \( P_0 \) at the center of the PDMS/BaSiON membrane. Note that, since APL does not provide spectral information on the acoustic frequencies, the reported values of \( P_0 \) correspond to the total pressures experienced by the membrane at the center of the US spot as measured by a hydrophone, averaged over 9 pixels (\( \sim 0.3 \text{ mm}^2 \)). This then also includes the higher harmonics generated by nonlinear propagation of the ultrasound in the water tank (see Section 7 of the Supporting Information).

Fig. 6a shows that the build-up time needed to reach the peak emission increases as the pressures decreases. Note that, since the transient time of typical US transducers is on the order of a period (e.g. 0.2 \( \mu \text{s} \) at a frequency of 5 MHz), this slow initial rise corresponds to the response of the system to a near-perfect step function. For higher pressures, the APL intensity profiles start decreasing after reaching the peak emission, while at lower pressures, the profiles continue to rise until the transducer is switched off. This is consistent with the trap depletions model: the fraction of shallow traps depletes increases exponentially with the temperature and thus with the pressure. Fig. 6b shows the TIL intensity profiles produced by the forward prediction model for the same frequency (\( f = 5 \text{ MHz} \)) and pressure \( P_0 \). Fig. 6c depicts six obvious candidates for parameters to describe the APL and TIL intensity profiles, namely the initial slope, the maximum emission intensity, the area under the profile integrated over 2, 5 and 10 s from the beginning of the insonation, and finally, the area post-insonation, where the profile goes below zero (depletion region), integrated for 10 s (the determination of these parameters is described in Section 8 of the Supporting Information).

Fig. 6d–i shows the evolution of these 6 parameters as a function of the central pressure \( P_0 \), thus making up potential calibration curves. All these parameters display a slow onset as a function of the acoustic pressure followed by a nearly linear behavior. The exact shape results from the quadratic dependency of the absorbed acoustic energy on the pressure combined with the nonlinear nature of light emission as a function of temperature and is therefore not trivial to predict. Although these 6 parameters show a remarkably good agreement with the calculated \( I_{TL}(P) \) intensity profiles, they do not all offer the same accuracy. Given the nature of the measurement, we define the accuracy using (i) the sensitivity with respect to a pressure change (i.e. the slope of the calibration curve) and (ii) the robustness against noise and experimental error. Given the small thickness of the membrane, heat diffusion is considered to occur only in the axial direction at early times. The initial rate of temperature rise is therefore linear with the absorbed power and quadratic with the pressure amplitude (Fig. 6d). The calibration curve determined with the short integration time of 2 s suffers from experimental noise, which is visible in the oscillations of the experimental curve around the calculated response in Fig. 6f. The initial slope and the peak emission show an excellent agreement with the model. However, the low sensitivity of the initial slope for acoustic pressures below 200 kPa favors the use of the peak emission intensity to generate more robust calibration curves.

3.5. Case study: full pressure field reconstruction

The chosen calibration curve based on the peak emission intensity was further evolved to reconstruct a quantitative 3D pressure map for a Gymna Pulsion 200 physiotherapeutic transducer (\( f = 3.3 \text{ MHz} \), \( IUS = 2.0 \text{ W cm}^{-2} \)). The 3D map was generated by recording a series of APL cross sections of the beam. Fig. 7b shows a simulated lateral XY view of the pressure field of the transducer calculated with a Fast Fourier Transform Beam Propagation Model (FFT-BPM). This method decomposes the plane waves in space and time, following the holographic principle, and couples them to a propagator term \( [29] \). Fig. 7a shows the source term for this numerical propagation model, which was recorded with Laser Doppler Vibrometry (LDV), in air. Note that the absence of water loading on the transducer surface may cause deviations in the measured oscillations compared to the APL case. However, only the general shape of the transducer surface mode is of interest here, for which the propagation medium plays a secondary role. Fig. 7d shows the same field of view, as reconstructed through the proposed APL scanning procedure (using 60 calibrated cross sectional views) in combination with the peak emission calibration procedure. Fig. 7c shows the first APL recording of the same field, at \( x = 5 \text{ mm} \). Finally, Fig. 7e shows two orthogonal planes of the entire 3D beam (which can be viewed online at www.apl.UGent.be). The complex spatial features of the beam, including the near-field to far-field transition, are well represented in the APL reconstruction and the pressure field is recovered quantitatively.

4. Conclusions and outlook

The characterization method of acoustically produced luminescence (APL) is proposed as a novel approach for the quantification of 3D acoustic pressure fields. We combine two analytical models describing firstly the heat generation via US absorption and subsequent diffusion (the AT model) and secondly the APL emission as thermoluminescence emission from the heated APL membrane (the TIL model). The complete model provides a full theoretical understanding of the APL phenomenon that enables accurate prediction of APL emission patterns based on the acoustic parameters. Finite element modeling was used to validate the AT model for the ultrasonic heating caused by insonation with an unfocused 5 MHz transducer, showing excellent agreement. The combination of the AT and TIL models was validated by accurately predicting an APL emission induced by the same 5 MHz transducer. Additionally, the inversion of the analytical models allows a quantitative inverse reconstruction of those acoustic parameters starting from an observed APL emission pattern upon insonation. The reconstruction method was validated by simulating an APL experiment (using the validated prediction models) and then retrieving the square wave pressure input, with an error margin within 5% on the retrieved maximum pressure.

The improved understanding of the APL phenomenon allows the development of PDMS/BaSiON membranes tailored to specific applications. APL allows for a fast, easy and quantitative 3D pressure field reconstruction and can play an important role in the development of new US techniques and technologies by offering an alternative to the expensive and time consuming conventional hydrophone scans. Finally, given its generality, the proposed theoretical framework can be
beneficial for other thermoluminescence applications. For example in thermometry, where the retrieval of a temporal temperature map from the observed emission can, in principal, be very useful.

Author statement

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Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supplementary data

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References