Acoustic radiation force on small spheres due to transient acoustic fields

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Acoustic radiation force is a net force experienced by an object under the action of an acoustic wave. Most theoretical models require the acoustic wave to be periodic, if not purely monofrequency, and are therefore irrelevant for the study of acoustic radiation force due to acoustic pulses. Here, we introduce the concept of finite-duration pulses, which is the most general condition to derive the acoustic radiation force. In the case of small spheres, we extend the Gor’kov formula to unsteady acoustic fields such as traveling pulses and interfering wave packets. In the latter case, our study suggests that the concept of acoustic contrast is also relevant to express the acoustic radiation force. For negative acoustic contrast particles, the acoustic trapping region narrows with shorter pulses, whereas positive contrast particles (such as biological cells) can fall in secondary traps when the pulse width deviates from an optimal value. This theoretical insight may help to improve the selectivity of pulsed acoustic tweezers.

I. INTRODUCTION

Acoustic radiation pressure is generally described as a steady force acting on surfaces exposed to acoustic waves. While radiation pressure has been extensively studied for almost two centuries, the steady nature of the force has been little discussed. Since the pioneering work of Rayleigh [1, 2], radiation pressure has mainly been studied in the monofrequency regime and been defined as an exchange of momentum between a wave and a particle. Integrating the acoustic radiation pressure over an object boundaries yields a net acoustic radiation force, which has been computed for incompressible spheres exposed to plane waves by King [3] and later on by taking the sphere compressibility into account by Yosioka and Kawasima [4]. In 1962, Gor’kov derived an elegant formula for the acoustic radiation force of spheres much smaller than the acoustic wavelength subject to arbitrary acoustic fields [5]. Even though these calculations have been extended to arbitrary large spheres in complex acoustic fields [6, 7], and to include viscous effects and even acousto-thermal effects [8, 9], the monofrequency dogma has remained essentially unchallenged.

Silva et al. have investigated the parametric oscillations of spheres of arbitrary size exposed to bichromatic and polychromatic acoustic waves in an inviscid fluid [10, 11]. They have shown that, unlike the monochromatic case, the amplitude of the parametric forcing depends on the nonlinearities in the fluid equation of state. A simplified expression of the acoustic radiation force was independently provided by Karlsen and Bruus for the average force experienced by small spheres exposed to polychromatic waves in a thermoviscous fluid [12]. In both cases, the polychromatic assumption refers to a discrete combination of modes, which is tantamount to periodic excitation signals. Although this allows the analysis of a combination of transducers [16, 17], the periodic assumption makes it a priori irrelevant for continuous combinations of frequencies [18] and acoustic pulses [19].

While the traditional use of acoustic radiation force for acoustic levitation and acoustic tweezing allows considering extremely long actuation signals, and is therefore well described by existing theories, using a continuous range of frequencies offers a superior flexibility in device operation. This enables finely tuning the spatial geometry of an acoustic field, which is tantamount to adjusting the acoustic radiation force landscape. For instance, Kang et al. have demonstrated such a multifrequency device [18] for the fine positioning of particles. However, one of the most interesting prospect of pulses is to increase the acoustic trapping selectivity, that is the ability to trap a particle within many others by confining the acoustic field around this particle. Unlike the complex transducers [20, 21] or transducer arrays [22–24] typically needed to achieve a high selectivity, Collins et al. have pioneered an alternative strategy using ultrashort acoustic pulses [19]. They assumed that, similarly to the monofrequency case, traveling acoustic pulses would generate much less acoustic force than standing acoustic pulses (obtained by the interference between two counter-propagating pulses). They demonstrated that acoustic pulse width as short as 10 acoustic periods yield an acoustic radiation force, and that acoustic interference and trapping regions overlap. Even though the limited bandwidth of their transducers restricted the pulse shortness to at least 10 periods, it not only challenges the monofrequency assumption, but also suggests that further reducing the pulse width may enable even higher selectivity.

Furthermore, non-conventional acoustic generation methods such as percussions and photoacoustic effects
FIG. 1. Scattering and interference of wave packets. (a)-(f) time-lapse of the diffraction of an acoustic wave by a wave packet. A particle (brown disk) surrounded by viscous and thermal boundary layers (blue and red) sits in a quiescent fluid. The fixed domain Ω is much larger than the acoustic wavelength and encompasses both boundary layers. At (a-b), a wave packet enters Ω, then (c) is scattered by the particle and (c,d,e) generates a scattered field (shown as circular waves) that (f) eventually exits Ω. Neglecting the particle motion, the state of Ω is identical in steps (a) and (f) which allows defining τ as the duration between these two steps. (g) A Gaussian acoustic pulse. The gray region indicates the pulse duration, which can be chosen arbitrarily larger than the pulse width. (h) Interference of two Gaussian pulses. The gray band indicates the pulse duration chosen in this particular example.

The small ε ≲ 0.01 for most acoustofluidic applications suggests that the above inequalities hold as long as τ ≲ 100 acoustic periods long. Assuming that the excitation signal bandwidth is bounded by [f_{min}, f_{max}], this finite-pulse theory will be relevant for f_{min} ≪ f_{max}^2 ∼ 10^{-4}f_{max}. More stringent conditions are foreseen when the particle can resonate with the incident field and emit the scattered field for a longer duration. This may occur for bubbles or particles with a size comparable to the acoustic wavelength. For periodic series of pulses, this has little consequences because the acoustic field variables will cycle over the pulse repetition period so that the acoustic radiation force of each pulse period can be evaluated by carefully including the steady-state scattered field due to this periodic excitation. For isolated or stochastic acoustic pulses however, the finite duration condition would require τ large enough to allow the scattered field to decay to negligible levels before evaluating the acoustic radiation force.

This finite duration allows to overlook the complex dynamics found by Longhorn and subsequent studies [14, 15, 27–30], which are difficult to resolve experimentally at present [30, 31], and to focus instead on the transformation of the particle from the initial state before wave packet scattering and a final state after the scattering event. This is illustrated in Fig. 1a-f: the domain Ω contains a particle (brown disc) with a non-viscous boundary layer (in blue and red). An incident wave packet falls on the particle and generates a scattered field that eventually escapes Ω at step (f). The finite-duration condition requires that the state of Ω at step (f) is the same as in step (a).
After generalizing the expression of the acoustic radiation pressure tensor to finite duration wave packets, we will re-derive the acoustic radiation force on small spheres while relaxing the monofrequency assumption. This will provide a generalized Gor’kov equation for acoustic wave packets of finite duration. Although our final expression could be derived from Karlsen et al. expression for periodic waves [12] using Parseval’s theorem, and even for non-periodic pulses using Plancherel theorem, (i) re-deriving the equation provides a deeper physical meaning of its origin, especially regarding the scattering coefficients, and (ii) our derivation has a broader set of validity than what these theorems alone would warrant. In particular, using the Plancherel theorem requires to integrate the time-domain function over \([ -\infty, +\infty] \) and would therefore not ensure, for instance, that successive pulses would not interfere. After deriving this general expression, we will then evaluate the acoustic radiation force of these packets in the simplified cases of traveling and standing waves, and use our analytical expression to optimize the selectivity of acoustic trapping by acoustic pulses.

II. MODEL

A. Definition of the acoustic radiation force

Using the summation of repeated indices, mass and momentum conservation in a Newtonian fluid of shear viscosity \(\mu\) and bulk viscosity \(\mu_b\) read:

\[
\begin{align*}
\partial_1 \rho + \partial_j \rho v_i &= 0, \\
\partial_1 \rho v_i + \partial_i \rho v_j v_j &= \partial_j (-p \delta_{ij} + \Sigma_{ij}), \\
\Sigma_{ij} &= \mu \left( \partial_j v_i + \partial_i v_j \right) + \left( \frac{\mu}{3} + \mu_b \right) \delta_{ij} \partial_t v_j, 
\end{align*}
\]

(2a, 2b, 2c)

with \(p, \rho\) and \(v\) the fluid pressure, density and velocity, and \(\delta_{ij}\) is the Kronecker delta function. These equations are complemented with the isentropic equation of state up to the second order in \(\rho - \rho_0\):

\[
p - p_0 = c_0^2 (\rho - \rho_0) + \frac{1}{2} \Gamma (\rho - \rho_0),
\]

(3)

with \(\Gamma = \frac{b}{\rho_0} \frac{\rho_0^2}{\rho} \), where \(b\) is an important parameter in nonlinear acoustics [14, 32].

A particle with boundaries located at \(\partial \Omega_p(t)\) immersed in this fluid will experience a force \(F_i = \oint_{\partial \Omega_p(t)} p \delta_{ij} - \Sigma_{ij} dS_j\) [12, 32]. When the fluid motion is solely due to acoustic waves, the time-average of this force is the acoustic radiation force. However, the particle position and shape are affected by the acoustic field, meaning that the integration boundary \(\partial \Omega_p\) varies with time. This problem is addressed by using the Gauss integral theorem on Eq. (2b) to get the Reynolds transport equation. This allows to evaluate the stress from a fixed boundary \(\partial_1\) located arbitrarily far away from the particle:

\[
\partial_1 \int_\Omega \rho v_i dV + \oint_{\partial \Omega} (\rho v_i v_j + p \delta_{ij} - \Sigma_{ij}) dS_j = -F_{\text{tot}}^i
\]

(4)

with \(F_{\text{tot}}^i\) the reaction force of the fluid to all the external forces acting on it.

The main caveat of using the transport theorem is that it operates a momentum balance over the entire domain \(\Omega\), such that acoustic momentum transfer from the wave to the fluid inside \(\Omega\), and the steady flow generated by the acoustic wave outside this domain (acoustic streaming [32]) are included in the left-hand term [32]. It is therefore customary to distinguish the variations of momentum due to the particle itself (acoustic scattering, microstreaming and so on) and the effects that arise from the acoustic wave attenuation that would have occurred even in the absence of any particle. Conveniently, the spatial scale of acoustic attenuation \(\Lambda_{\text{ac}} = \frac{\rho c_0^3}{\mu (4/3 + b) \rho_0^2}\) is often many orders of magnitude larger than the scale of the viscous \(\Lambda_{\text{visc}} = \sqrt{2 \mu / \rho_{\text{visc}} G}\) boundary layers (with \(D_T\) the thermal diffusivity in the fluid), which suggests that the radiation force is well approximated by considering an inviscid and adiabatic fluid, provided that the local effects of microstreaming and heat conduction are well resolved in the direct vicinity of the particle [12]. Using this inviscid and adiabatic approximation to compute the acoustic radiation force, the viscous effects can be recovered by computing the acoustic streaming and adding it as a drag force acting on the particle [34].

B. Perturbation expansion in the far field

Based on the discussion above, we now omit the viscous effects in the fluid, and use the small Mach number \(\epsilon\) to expand the evolution of the pressure, velocity and density fields by perturbation of increasing orders in \(\epsilon\). 0-order quantities are denoted \(x_0\), first-order \(\hat{x}\) and second-order \(\tilde{x}\):

\[
\begin{align*}
\rho &= \rho_0 + \epsilon \hat{\rho} + \epsilon^2 \tilde{\rho}, \\
p &= \rho_0 + \epsilon \hat{p} + \epsilon^2 \tilde{p}, \\
v_i &= 0 + \epsilon \hat{v}_i + \epsilon^2 \tilde{v}_i.
\end{align*}
\]

(5a, 5b, 5c)

In order to get non-trivial values for the Mach number, we now solve each perturbation order independently. The order 0 in \(\epsilon\) is the hydrostatic equilibrium:

\[
\begin{align*}
\partial_t \rho_0 &= 0, \\
\partial_t p_0 &= 0,
\end{align*}
\]

(6a, 6b)

which suggests a uniform pressure \(p_0\) and density \(\rho_0\) in the fluid at rest. Then, the 1st order in \(\epsilon\) describes the
acoustic field:
\[
\partial_t \tilde{\rho} + \rho_0 \partial_t \tilde{v}_i = 0, \quad (7a)
\]
\[
\rho_0 \partial_t \tilde{v}_i = -\partial_i \tilde{p}. \quad (7b)
\]
Taking the divergence of Eq. (7b) and using the equation of state (Eq. (3)) up to the first order in \(\epsilon\) yields the d’Alembert equation:
\[
\partial_t^2 \tilde{p} - c_0^2 \partial_x^2 \tilde{p} = 0. \quad (8)
\]

The second order in \(\epsilon\) unveils the main nonlinear effects:
\[
\partial_t \tilde{p} + \rho_0 \partial_t \tilde{v}_i + \rho_0 \partial_i \tilde{v}_j = 0, \quad (9a)
\]
\[
\partial_t \rho_0 \tilde{v}_i + \partial_i \rho_0 \tilde{v}_j + \rho_0 \partial_j \tilde{v}_i \tilde{v}_j = -\partial_i \tilde{p}, \quad (9b)
\]
\[
\tilde{p} = c_0^2 \tilde{\rho} + \frac{1}{2} \Gamma \tilde{\rho}^2. \quad (9c)
\]

To facilitate comparison with earlier works \([5, 12, 13]\), we will consider the average of the quantities of interest, instead of the time-integral as suggested by the physical setting: \(\langle x \rangle = \frac{1}{t} \int t/2 \ x(t) dt\). Due to the finite duration of the pulse, any 1st order quantity \(\hat{x}\) satisfies:
\[
\langle \partial_t \hat{x} \rangle = \hat{x}(\tau/2) - \hat{x}(\tau/2) = 0, \quad (10)
\]

This allows to simplify Eqs (11a) (11b) (11c):
\[
\partial_t \langle \tilde{\rho} \tilde{v}_i \rangle + \rho_0 \partial_t \langle \tilde{v}_i \rangle = 0, \quad (11a)
\]
\[
\rho_0 \partial_t \langle \tilde{v}_i \tilde{v}_j \rangle = -\partial_i \langle \tilde{p} \rangle , \quad (11b)
\]
\[
\tilde{p} = c_0^2 \tilde{\rho} + \frac{1}{2} \Gamma \langle \tilde{p} \rangle^2. \quad (11c)
\]

Substituting Eqs (11a) (11b) into \(\rho_0 \langle \partial_j \tilde{v}_i \tilde{v}_j \rangle \) and then using integration by part, we get:
\[
\rho_0 \langle \partial_j \tilde{v}_i \tilde{v}_j \rangle = \partial_i \langle \mathcal{L} \rangle , \quad (12)
\]
with \(\mathcal{L} = \mathcal{K} - \mathcal{V}\) the Lagrangian density of the wave, with the acoustic kinetic energy \(\mathcal{K} = \frac{1}{2} \rho_0 \tilde{v}_j \tilde{v}_j\) and the acoustic potential energy \(\mathcal{V} = \frac{\tilde{\rho}^2}{2 \rho_0 c_0^2}\).

C. Lagrangian pressure and Brillouin tensor

In order for the fluid to be at mechanical equilibrium, one must satisfy Eq. (11b). Substituting Eq. (12) in Eq. (11b) yields the Lagrangian pressure:
\[
\langle \tilde{p} \rangle = \mathcal{L} - \langle \mathcal{L} \rangle , \quad (13)
\]
with \(\mathcal{C}\) a constant independent of the position in the fluid \([35]\). Similarly to the monofrequency case \([32, 35]\), a consequence of Eq. (13) is to set the value of the density \(\tilde{\rho}\) to fulfill Eq. (11c) so that the nonlinearities in the equation of state play no role in the average acoustic radiation pressure of acoustic pulses. We note that this assertion is valid only over the whole duration of the pulse, while it was shown that \(\Gamma\) was important for the detailed dynamics \([30]\).

Taking the time-average of Eq. (11) and expanding up to the second order in \(\epsilon\), we get:
\[
\int_{\partial \Omega} \langle B_{ij} \rangle \ dS_j = -F_i, \quad (14)
\]

with \(\langle B_{ij} \rangle = \langle \rho_0 \tilde{v}_i \tilde{v}_j - L \delta_{ij} \rangle\) the Brillouin tensor, which remains the same as in the monofrequency regime. Eq. (14) is the starting point to compute the acoustic radiation force on arbitrarily-shaped objects \([36]\) without additional restrictions of size other than satisfying the finite-duration conditions detailed previously.

D. Acoustic radiation force on small spheres

It is worth noting that, had Eq. (12) been valid everywhere inside \(\Omega\), Eq. (14) would vanish. This suggests to decompose the acoustic field quantities \(\hat{x}\) into a background incident acoustic field, denoted by the subscript \(\hat{x}_{in}\), (that fulfills Eq. (12) and therefore generates no force); and a scattered acoustic field due to the particle, denoted by the subscript \(\hat{x}_{sc}\):
\[
\hat{\rho} = \hat{\rho}_{in} + \hat{\rho}_{sc}, \quad (15a)
\]
\[
\tilde{\rho} = \tilde{\rho}_{in} + \tilde{\rho}_{sc}, \quad (15b)
\]
\[
\tilde{\rho}_{sc} = -f_1(\omega) \frac{a^3}{3 \rho_0} \frac{\partial_i \hat{\rho}_{in} \big|_p}{r} - f_2(\omega) \frac{a^3}{2} \partial_i \left[ \frac{\hat{\rho}_{in} \big|_p}{r} \right], \quad (17)
\]

where \(a\) indicates that \(x\) is evaluated at the location of the particle \((0)\) at the retarded time \((t-r/c_0)\). We note that these functions still depend on space due to the time-retarded argument. The monopole and dipole scattering
coefficients $f_1$ and $f_2$ are complex numbers which depend on the chosen convention $\hat{x} = \hat{x}(\omega)e^{-i\omega t}$. They can be found in the comprehensive study by Karlsen and Bruus.

Substituting Eq. (19) in (14), and neglecting squares of $\hat{F}_{in}$ as they model a wave without any interaction with a particle, therefore yielding no momentum exchange, and the squares of $\hat{F}_{sc}$ proportional to $\omega^0$ and therefore negligible for a small particle, we get:

$$F_i = -\int_\Omega \rho_0 \left( \dot{v}_{i,in} \left( \frac{\partial^2}{\partial t^2} - \frac{1}{c_0^2} \frac{\partial^2}{\partial r^2} \right) \delta_{sc} \right) dV.$$  

Similarly to the previous studies [5, 12, 13], we recall that the D’Alembert equation acting on the monopole and dipole terms [3] result in a singular density point $\hat{p}_{in}|_p \delta(r)$ and a singular velocity point $\hat{v}_{i,in}|_p \delta(r)$, with $\delta$ the Dirac distribution.

$$\left( \frac{\partial^2}{\partial t^2} - \frac{1}{c_0^2} \frac{\partial^2}{\partial r^2} \right) \hat{\phi}_{sc} = f_1(\omega) \frac{4\pi a^3}{3p_0} \dot{\hat{p}}_{in}|_p \delta(r) + f_2(\omega) \frac{2\pi a^3}{3} \partial_j \hat{v}_{j,in}|_p \delta(r).$$  

Integrating Eq. (19) over all angular frequencies $\omega$, we get the time-dependent scattered field:

$$\left( \frac{\partial^2}{\partial t^2} - \frac{1}{c_0^2} \frac{\partial^2}{\partial r^2} \right) \hat{\phi}_{sc} = \frac{4\pi a^3}{3} \left[ \frac{1}{p_0 c_0^2} \partial_t \delta(r) \hat{p}_{in}|_p + \partial_j \delta(r) \hat{v}_{j,in}|_p \right],$$

with the monopole and dipole scattered fields:

$$\hat{p}_{in}|_p = \frac{1}{2} \int_0^\infty f_1(\omega) \hat{p}_{in}|_p + f_1^*(\omega) \hat{p}_{in}|_p \omega^2 d\omega, \quad \hat{v}_{j,in}|_p = \frac{3}{4} \int_0^\infty f_2(\omega) \hat{v}_{j,in}|_p + f_2^*(\omega) \hat{v}_{j,in}|_p \omega^2 d\omega,$$

where the $\partial_t$, $\partial_j$ operators and the $\delta$ distribution commute with the integral over angular frequencies.

Substituting Eq. (19) in Eq. (14) and using Gauss theorem (see [12] for additional details), the time-retarded argument in the scattered fields is simplified by the Dirac function, so that Eqs. (23) can now be evaluated at the location of the particle. We obtain a first expression of the acoustic radiation force:

$$F_i = -\frac{4\pi a^3}{3} \left[ \hat{v}_{i,in} \left( \frac{1}{c_0^2} \partial_t \hat{p}_{in} \right) - \langle \rho_0 \hat{v}_{i,d} \partial_j \hat{v}_{i,in} \rangle \right].$$

Integrating Eq. (23) by part then yields:

$$F = -\frac{4\pi a^3}{3} \left[ \frac{1}{p_0 c_0^2} \hat{p}_{in} \nabla \hat{p}_{in} - \langle \rho_0 \hat{v}_d \cdot \nabla \hat{v}_{in} \rangle \right],$$

which in the monofrequency regime simplifies into the Gor’kov equation [5, 12, 13].

III. RESULTS AND DISCUSSION

In the following, we consider the effect of pulses on the motion of particles. While Eq. (23) is valid for any finite-duration pulse, the general calculation is complicated by the convolution products in Eqs. (21) [22]. These equations can be simplified (i) by considering narrow-bandwidth acoustic beams so the variation of all quantities but the wave spectrum can be neglected over the integration bandwidth, or (ii) by considering inviscid fluids where the scattering coefficients can be factored out of the integrals.

When a particular example is needed, we will use Gaussian plane wave packets propagating along the $x$ direction $p_{in}(\theta) = p_{max} w(\theta)$ with:

$$w(\theta) = \exp(-\sigma\theta^2) \cos(\theta\theta),$$

with $\theta = t - s \cdot r$ the retarded time of the wave, where $s = \frac{1}{c_0} \epsilon_x$. Advantageously, calculations with these waves are relatively straightforward but still allow tuning the pulse width ($\Theta \propto \sigma^{-1/2}$) by changing $\sigma$. For such traveling plane waves, the velocity field is given by $\hat{v}_{in} = \hat{p}_{in} / p_{max} \epsilon_x$. An important quantity when studying the acoustic radiation force is the mean energy density $\langle E \rangle$, defined as the total energy flux per unit length $\langle E \rangle = s \cdot \langle \hat{p} \hat{v} \rangle$. In the case of a Gaussian wave packet, it reads:

$$\langle E \rangle = \langle E_\infty \rangle \left( 1 + e ^{-\frac{\theta^2}{\Theta^2}} \right),$$

with $\langle E_\infty \rangle = \frac{p_{max}^2}{\rho_0 c_0^2} \frac{\pi}{8\sigma} \frac{1}{\tau}$. The time and frequency spread of the wave packet (defined as the standard deviation in time and frequency space) will also be useful, and read (respectively):

$$\delta t = \frac{1}{\sqrt{2\sigma}}, \quad \delta f = \frac{\sigma}{2\pi^2}.$$  

A. Narrow-band waves in a thermoviscous fluid

We first consider an acoustic wave packet with a bandwidth $\delta f = \frac{\Delta f}{2\pi}$ narrow enough to neglect the variations of $f_1$ and $f_2$ over this frequency band. Before discussing the force, we note that this assumption poses some constraints on the pulse duration. According to Settnes, Karlsen and Bruus, this requires that the thermoviscous boundary layer thickness $\Lambda_{tv}$ to particle radius ratio do not change much over $\delta \omega$ [12, 13], which yields $\delta \omega \ll 2\omega / \Lambda_{tv}$, with $\Lambda_{tv} = \min(\Lambda_{visc}, \Lambda_{therm})$. According to the Gabor limit, $\delta t \delta f \geq \frac{1}{2\pi} \frac{\pi}{\Theta}$, with the equality being true when using Gaussian wave packets. Combining these inequalities with Eq. (27a) yields a lower limit for
the number of periods $n_T$ of the pulse:

$$n_T \simeq \sqrt{\frac{2 \omega^2}{\sigma}} \gg \frac{\Lambda_{\text{vis}}}{a}. \quad (28)$$

While this inequality does not contradict Eq. (11), their combination restricts this thermoviscous case to low-amplitude long-duration wave packets.

Separating the real and imaginary parts of $f_1 = f_1^r + i f_1^i$ and $f_2 = f_2^r + i f_2^i$ and noting that in the current complex convention for the scattering coefficients we have $i(\tilde{x} - \tilde{x}^*) = -\frac{1}{\omega} \partial_t (\tilde{x} + \tilde{x}^*)$, yields:

$$\tilde{p}_m|_p = f_1^r \tilde{p}_m|_p - \frac{f_1^i}{\omega} \partial_t \tilde{p}_m|_p, \quad (29)$$
$$\tilde{v}_{j,d}|_p = \frac{3}{2} \left( f_2^r \tilde{v}_{j,\text{in}}|_p - \frac{f_2^i}{\omega} \partial_t \tilde{v}_{j,\text{in}}|_p \right), \quad (30)$$

where, consistently with the narrow-band hypothesis, the variations of $1/\omega$ over the frequency interval were neglected.

Substituting Eqs. (29) (30) in Eq. (24), and omitting the real part of $f_1$ and $f_2$ that do not contribute to the acoustic radiation force for traveling waves up to $O(a^6)$ [38], we get the force due to a narrow-band traveling wave $\tilde{p}(\theta)$ in a thermoviscous fluid:

$$F = -s \frac{4 \pi a^3}{3} \frac{1}{\omega \rho_0 c_0^2} \left[ f_1^r - \frac{3 f_2^i}{2} \right] \langle (\partial_\theta \tilde{p}_m)^2 \rangle, \quad (31)$$

which agrees with [12] except for a minor typo in the sign of $f_1^r$.

In the case of Gaussian wave packets, the force simplifies into:

$$F = -s \frac{4 \pi a^3}{3} \frac{1}{\omega \rho_0 c_0^2} \left[ f_1^r - \frac{3 f_2^i}{2} \right] \langle E_{\infty} \rangle. \quad (32)$$

Careful inspection of Eq. (32) reveals that the acoustic radiation force decreases in $1/\tau$. This mathematical artifact is due to the time-averaging of the force over the time $\tau$ chosen arbitrarily. Hence, a better measure of the effect of the acoustic radiation force of finite-duration pulses is the transmitted acoustic radiation momentum $q = \tau F$, which depends on the physically-relevant pulse width ($\Theta \propto \sigma^{-1/2}$) but is independent of the pulse duration.

**B. Arbitrary waves in an inviscid fluid**

When the particle is much larger than the visco-acoustic or thermo-acoustic boundary layers, the scattering coefficients $\hat{f}_1(\omega) = f_1^r(\omega)$ and $\hat{f}_2(\omega) = f_2^r(\omega)$ are purely real and independent of the frequency [3], and therefore can be factored out of the integrals in Eqs. (21) (22), which yields:

$$F = -s \frac{4 \pi a^3}{3} \hat{\nabla} \langle U \rangle \quad (33)$$

with the dynamic Gor’kov potential:

$$U = \frac{f_1}{2 \rho_0 c_0^2} \tilde{p}_m^2 + \frac{3 \rho f_2}{4} \tilde{v}_m^2. \quad (34)$$

**1. Traveling waves**

In the case of traveling waves, $\tilde{p}_m$ and $\tilde{v}_{j,\text{in}}$ are only functions of $\theta = t - s \cdot r$ with $s$ the wave slowness. Therefore, $U$ is a function of $\theta$ only, such that:

$$\nabla \langle U \rangle = \langle \partial_\theta U \nabla \theta \rangle = -s \langle \partial_\theta U \rangle = O(a^6), \quad (35)$$

which vanishes up to the small terms in $O(a^6)$ that were neglected in Eq. (18). Extrapolating from the monofrequency regime, one may reasonably expect that higher order terms called scattering force will dominate the acoustic radiation force in this case.

**2. Interference of two plane wave packets**

We next consider two interfering plane wave packets $\tilde{p} = \tilde{p}(\theta^+) + \tilde{p}(\theta^-)$, as illustrated in Fig. 1c. For the sake of generality, the two packets intersect with an angle $2\eta_R$, so that $\theta^+ = t + s_x x - s_z z$ and $\theta^- = t - s_x x - s_z z$, with $s_x = \frac{\sin \eta_R}{c_0}$ and $s_z = \frac{\cos \eta_R}{c_0}$. While $\eta_R = \pi/2$ is relevant for a frontal interference [38] [10], the general case applies to surface-acoustic-wave-based tweezers such as the ones proposed by Collins et al. [40], where $\eta_R$ plays the role of the Rayleigh angle that the acoustic radiation makes with the substrate [41].

Assuming that the incident wave reads $\tilde{p}_\text{in} = p_{\text{max}} \langle \tilde{w}(\theta^+) + \tilde{w}(\theta^-) \rangle$, with $\tilde{w}$ given in Eq. (25), we have

$$\tilde{v}_{\text{in},x} = -\frac{p_{\text{max}}}{\rho \omega c_0} [\tilde{w}(\theta^+) - \tilde{w}(\theta^-)] \quad \text{and} \quad \tilde{v}_{\text{in},z} = \frac{p_{\text{max}}}{\rho_0 \omega c_0}.$$

This yields the Gor’kov potential and the acoustic radiation
Acoustic radiation force has long been considered a steady-state time-averaged phenomenon. Here, we derive an expression of the acoustic radiation force acting on spheres for arbitrary acoustic pulses, as long as the pulse bandwidth satisfies (i) the small sphere condition $f_{\text{max}} \ll \frac{\omega_s}{a}$ and (ii) pulse durations are short enough to neglect the sphere displacement compared to the wavelength $f_{\text{min}} \simeq \frac{1}{\tau} \gg f_{\text{max}}^2$. We provide an extension of the Gor’kov formula in the case of wave packets and other unstable acoustic fields. Even in the time-domain, traveling waves in an inviscid fluid do not generate a net force up to $a^6$, while standing waves do yield a force proportional to $a^4$. Based on the complex monopole and dipole scattering coefficients, our model can account for thermoviscous effects that can considerably enhance the acoustic radiation force of traveling waves. This model provides a theoretical foundation for the use of pulsed acoustic waves to enhance acoustic tweezers selectivity, and clarifies that an acoustic radiation force exists even for a single acoustic period.
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