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Asymptotics of a time correlation function in multiple recurrent scattering of scalar waves

By N. M. Temme¹, F. Vitalis², B. A. van Tiggelen^{2,3}, and A. Lagendijk^{2,4}

1. Introduction

We consider the integral

$$G(\tau) := \int_0^\infty z^2 [1 - \cos(\tau e^{iz}/z)] dz. \quad (1.1)$$

This integral recently showed up [1] in the calculation of the time-dependent field-field correlation function of the electric field inside polarizable (dielectric) particles. In particular, the integral (1.1) describes the influence of resonantly induced dipole-dipole coupling (Van-der-Waals interaction) between small Mie-spheres. Without this coupling the field correlation decays exponentially, with the so-called *dwell time* [2] as characteristic time. This time is (in some respects) the classical-wave equivalent of the inverse Einstein spontaneous emission coefficient A^{-1} in quantum-mechanical light scattering [3]. The time-variable τ has been scaled with this time.

The purpose of this paper is to give representations of $G(\tau)$ which are more suitable for obtaining qualitative and quantitative information. We give tables of numerical values and we show how to obtain the large τ -behaviour from these new representations. We obtain the asymptotic result

$$G(\tau) \sim -\frac{\pi}{2} \ln^2 \tau - \frac{i}{3} \ln^3 \tau, \quad \text{as } \tau \rightarrow \infty. \quad (1.2)$$

In §5 we compare this with the results of numerical computations.

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2. Interpretation of the integral with respect to convergence

The integral (1.1) is not convergent in the classical sense when we integrate over real positive z -values. To make the integral convergent, we assume that at infinity the path of integration terminates in the upper complex plane, along a ray $\arg z = \theta$, with $0 < \theta < \pi$. Since scattering theory usually considers $\exp(iz)$ as $\exp[i(z + i0)]$ this interpretation is in agreement with the physical context. Near the origin $z = 0$ we integrate along real positive z -values. To be more specific, we assume that the path of integration in (1.1) consists of the interval $[0, 1]$ and the half line that starts at the point $z = 1$ and that makes an angle θ , $0 < \theta < \pi$, with the positive real z -axis. Later we define completely different paths of integration, which are more suitable for obtaining asymptotic information, and which also can be used for numerical quadrature.

3. Non-oscillating representations of $G(\tau)$

We first use integration by parts in order to obtain a representation that is more manageable for asymptotic analysis. We have

$$\begin{aligned} G(\tau) &= \frac{1}{3} \int_0^\infty [1 - \cos(\tau e^{iz}/z)] dz^3 \\ &= -\frac{1}{3} \tau \int_0^\infty z(iz - 1)e^{iz} \sin(\tau e^{iz}/z) dz \\ &= G_1(\tau) + G_2(\tau), \end{aligned}$$

where

$$\begin{aligned} G_1(\tau) &= \frac{i\tau}{6} \int_0^\infty z(iz - 1)e^{iz + i\tau e^{iz}/z} dz, \\ G_2(\tau) &= \frac{\tau}{6i} \int_0^\infty z(iz - 1)e^{iz - i\tau e^{iz}/z} dz. \end{aligned} \tag{3.1}$$

The function $G_2(\tau)$ is quite easy to handle.

To this end, we define a new path of integration for $G_2(\tau)$. The integrand is analytic in the complex z -plane, with exception of the origin. Taking into account the behaviour of $\exp[iz - i\tau \exp(iz)/z]$ at infinity in the upper half plane $\Im z > 0$, we can deform the original path into the positive imaginary axis. To avoid the essential singularity at the origin, we introduce first a small quarter circle that runs from the positive real axis to the positive imaginary axis. The contribution along this quarter circle vanishes when the radius of the circle vanishes. To verify this we consider the

singular part in the exponential function: $w(z) = -i\tau \exp(iz)/z$. Writing $z = x + iy$, we have

$$w(z) = \frac{\tau e^{-y}}{x^2 + y^2} [(x \sin x - y \cos x) - i(x \cos x + y \sin x)].$$

When $y > x \tan x$, the real part of $w(z)$ is negative. Hence, then $\exp[w(z)]$ is bounded near the origin. When $y \leq x \tan x$, we have $y = \mathcal{O}(x^2)$ when x is small. Hence,

$$\frac{x \sin x - y \cos x}{x^2 + y^2} = \mathcal{O}\left(\frac{x^2}{x^2 + y^2}\right),$$

which is bounded near the origin. It follows that the contribution along a quarter circle with radius δ of an integral with integrand as in the second line of (3.1) equals $\mathcal{O}(\delta^2)$ as $\delta \rightarrow 0$.

Integrating with respect to $z = iy$, $y > 0$, we obtain

$$G_2(\tau) = \frac{-i\tau}{6} \int_0^\infty y(y+1)e^{-y-\tau e^{-y/y}} dy. \quad (3.2)$$

The integrand is now non-oscillating and purely real. Moreover, the integrand is exponentially small at both end points of integration.

A similar approach for $G_1(\tau)$ is not possible. It would yield an integral as in (3.2), with a different sign of τ . But then the convergence at the origin is violated. Turning the path of integration of $G_1(\tau)$ to the negative imaginary axis would give a convergent integral (change the signs of both y and τ in (3.2)). But we have assumed that both integrals in (3.1) terminate in the upper half plane. When we turn the path of $G_1(\tau)$ into the lower half plane, convergence is violated when we pass the real positive z -axis at infinity. Turning around a small quarter circle, that runs from the positive real axis to the negative imaginary axis is possible, however. This follows from a similar analysis as is given above for the integral $G_2(\tau)$.

We use the method of saddle points (see [4]) to derive a new path of integration for $G_1(\tau)$. The dominant part of the integrand in the first line of (3.1) is the function $\phi(z) := ie^{iz}/z$. It has a saddle point at the point where the derivative $\phi'(z)$ vanishes. We have

$$\frac{d}{dz} \phi(z) = i \frac{d}{dz} \frac{e^{iz}}{z} = \phi(z) \left[i - \frac{1}{z} \right].$$

It follows that there is one saddle point, which is located at $z = -i$. In the saddle point method one tries to modify the original path of integration into a new path, such that the new path runs through the saddle point; several aspects should be taken into account: the original end points of the contour and preservation of convergence of the integral during this operation. Furthermore, one tries to obtain a contour along which the imaginary part

of the phase function, in our case $\phi(z)$, is constant. Considering this final point, one tries to solve the equation $\Im\phi(z) = \Im\phi(-i)$; the right-hand side is the value at the saddle point, which happens to be zero in our case. Writing $z = x + iy$, we obtain the equation

$$\Im\phi(x + iy) = \frac{e^{-y}}{x^2 + y^2} [x \cos x + y \sin x] = 0.$$

Hence, the equation $\Im\phi(z) = 0$ has the solutions $x = 0$ and $y = -x \cot x$. The latter defines a parabola shaped curve, $-\pi < x < \pi$, with minimal point at $z = -i$, the saddle point. Along this curve we have

$$\Re\phi(x + iy) = -e^{-y} \frac{\sin x}{x}.$$

By using these results the path of integration for the integral defining $G_1(\tau)$ in (3.1) is composed by two parts (see Figure 1):

- the path from the origin to the saddle point $z = -i$;
- the path starting at the saddle point and running to ∞ along a curve defined by the equation $y = -x \cot x$, ($0 \leq x < \pi$).

During the deformation of the original path into the new contour convergence of the integral is preserved, and both end points are maintained. The integrals along the two components of the paths are called $G_1^{(1)}(\tau)$, $G_1^{(2)}(\tau)$, respectively. By Cauchy's theorem: $G_1(\tau) = G_1^{(1)}(\tau) + G_1^{(2)}(\tau)$, where

$$G_1^{(1)}(\tau) = \frac{i\tau}{6} \int_0^1 y(1-y)e^{y-\tau e^{y/y}} dy, \quad (3.3)$$

$$G_1^{(2)}(\tau) = \frac{\tau}{6} \int_0^\pi f(x)e^{-y-\tau e^{-y} \sin(x)/x} dx, \quad (3.4)$$

Figure 1
Path of integration for $G_1(\tau)$ of (3.1),
giving the integrals in (3.3) and (3.4).

where

$$f(x) = i(x + iy)(ix - y - 1)e^{ix} \left(1 + i \frac{dy}{dx}\right)$$

and the relation between x and y is given by $u = -x \cot x$.

4. Asymptotic behaviour of $G(\tau)$

We derive first approximations for the real and imaginary parts of the functions given in (3.2), (3.3) and (3.4).

An estimate of $G_2(\tau)$

We recall (3.2):

$$G_2(\tau) = \frac{-i\tau}{6} \int_0^\infty y(y+1)e^{-y-\tau e^{-y}/y} dy$$

and take as new variable of integration $u := e^{-y}/y$. It follows that, by using $du/dy = -u(y+1)/y$ and a few other straightforward manipulations,

$$G_2(\tau) = \frac{-i\tau}{6} \int_0^\infty y^3 e^{-\tau u} du. \quad (4.1)$$

When τ is large, the main contributions to this integral come from a small neighbourhood of $u = 0$. When u is small the equation $u = e^{-y}/y$ can be inverted: (see [4, p. 25])

$$y = -\ln u - \ln(-\ln u) + \mathcal{O}\left(\frac{\ln(-\ln u)}{\ln u}\right). \quad (4.2)$$

Taking the first term in this expansion, we obtain

$$G_2(\tau) \sim \frac{-i\tau}{6} \int_0^\infty (-\ln u)^3 e^{-\tau u} du. \quad (4.3)$$

Using

$$\tau \int_0^\infty (-\ln u)^3 e^{-\tau u} du = \int_0^\infty (-\ln x/\tau)^3 e^{-x} dx = -\ln^3 \tau + \mathcal{O}(\ln^2 \tau),$$

as $\tau \rightarrow \infty$, we see that

$$G_2(\tau) \sim \frac{-i}{6} \ln^3 \tau, \quad \text{as } \tau \rightarrow \infty. \quad (4.4)$$

Remark. We can compute the $\mathcal{O}(\ln^2 \tau)$ term in this estimate, but we already neglected a term that is of higher order: going from the exact

relation (4.1) to (4.3), we neglected in y^3 , among others, the term $-3 \ln^2 u \ln(-\ln u)$.

An estimate of $G_1(\tau)$

Next we consider $G_1(\tau)$. The function $G_1^{(1)}(\tau)$ defined in (3.3) is exponentially small when τ is large. This follows from the behaviour of the dominant part of the integrand: $\exp(-\tau e^y/y)$. This function is maximal at the end point $y = 1$, where its value is $\exp(-\tau e)$. Hence, $G_1^{(1)}(\tau)$ is exponentially small compared with $G_2(\tau)$ and can be neglected in the asymptotic expansion of the function $G(\tau)$.

In the integral (3.4) defining the function $G_1^{(2)}(\tau)$ we take as new variable of integration $v := e^{-y} \sin(x)/x$ with $y = -x \cot x$. A straightforward manipulation of $f(x) dx/dv$ finally gives the representation

$$G_1^{(2)}(\tau) = \frac{i\tau}{6} \int_0^e e^{3ix} \frac{x^3}{\sin^3 x} e^{-\tau v} dv. \quad (4.5)$$

Again, when τ is large, the main contributions to this integral come from a small neighbourhood of $v = 0$. When v is small, the equation $v = e^{x \cot x} \sin(x)/x$ has a solution x with $x \sim \pi$. Hence, replacing $x \cot x$ by $-x/\sin x$, we find that the solution x satisfies

$$\frac{x}{\sin x} = -\ln v + \mathcal{O}[\ln(-\ln v)], \quad \text{as } v \rightarrow 0.$$

Figure 2
Real and imaginary parts of $G(\tau)$.

Since $x \sim \pi$ we have $e^{3ix} \sim -1 + 3i \sin x$. It follows that a first approximation of $G_1^{(2)}(\tau)$ is given by

$$G_1^{(2)}(\tau) \sim \frac{-i\tau}{6} \int_0^\infty (-\ln v)^3 e^{-v\tau} dv - \frac{\pi\tau}{2} \int_0^\infty (-\ln v)^2 e^{-v\tau} dv,$$

giving

$$G_1^{(2)}(\tau) \sim -\frac{\pi}{2} \ln^2 \tau - \frac{i}{6} \ln^3 \tau, \quad \text{as } \tau \rightarrow \infty. \quad (4.6)$$

Collecting these results for $G(\tau) = G_1(\tau) + G_2(\tau) \sim G_1^{(2)}(\tau) + G_2(\tau)$ from (4.4)–(4.6), we finally have

$$G(\tau) \sim \frac{-i}{3} \ln^3 \tau - \frac{\pi}{2} \ln^2 \tau, \quad \text{as } \tau \rightarrow \infty. \quad (4.7)$$

5. Numerical evaluation of $G(\tau)$

When computing $G(\tau)$ for, say $\tau \geq 3$, direct numerical integration of (1.1) is not recommended. Because of the singularity at the origin and the strong oscillations of the integrand, especially when τ is large, numerical quadrature is quite impossible. For instance, earlier experiments with straightforward applications to (1.1) for $\tau > 15$ of library quadrature routines (that claimed to be suitable for oscillating integrands) yielded results of order 10^{10} , while the function $G(\tau)$ is of order $\mathcal{O}(\ln^3 \tau)$ as $\tau \rightarrow \infty$.

In Table 1 we give the real and imaginary parts of $G(\tau)$ for τ -values in the interval $[0, 50]$. Table 2 gives values for very large τ -values. Only for these τ -values the asymptotic result in (4.7) does come close to the numerical values. This is due to the logarithmic scale that shows up in the asymptotic expansion and to the order of magnitude of the neglected terms in (4.7); they are of lower order, but not much smaller than the dominant terms given in (4.7).

We have computed the tables by using the splitting $G(\tau) = G_1^{(1)}(\tau) + G_1^{(2)}(\tau) + G_2(\tau)$ and the integral representations given in (3.2), (3.3) and (3.4). For the numerical computations we have used the NAG subroutines D01AHF and D01AMF.

6. Discussion and physical interpretations

As has been mentioned in Section 1, the function $G(\tau)$ describes the influence of resonant dipole-dipole coupling on the field-field correlation function $C(\tau)$ inside polarizable particles. This correlation function is the

Table 1
Real and imaginary parts of $G(\tau)$

τ	$\Re G(\tau)$	$\Im G(\tau)$	τ	$\Re G(\tau)$	$\Im G(\tau)$
0.00	0.000000E+00	0.000000E+00	10.00	-0.349571E+01	-0.693905E-01
0.50	-0.204561E-01	0.509865E-01	12.00	-0.412071E+01	-0.330307E+00
1.00	-0.108133E+00	0.150845E+00	14.00	-0.469942E+01	-0.605984E+00
1.50	-0.253001E+00	0.248874E+00	16.00	-0.523879E+01	-0.889528E+00
2.00	-0.433038E+00	0.327306E+00	18.00	-0.574441E+01	-0.117678E+01
2.50	-0.632011E+00	0.383101E+00	20.00	-0.622078E+01	-0.146511E+01
3.00	-0.839893E+00	0.418198E+00	22.00	-0.667154E+01	-0.175286E+01
3.50	-0.105081E+01	0.435715E+00	24.00	-0.709967E+01	-0.203893E+01
4.00	-0.126136E+01	0.438666E+00	26.00	-0.750766E+01	-0.232260E+01
4.50	-0.146963E+01	0.429627E+00	28.00	-0.789758E+01	-0.260340E+01
5.00	-0.167453E+01	0.410699E+00	30.00	-0.827119E+01	-0.288104E+01
5.50	-0.187546E+01	0.383576E+00	32.00	-0.863000E+01	-0.315532E+01
6.00	-0.207216E+01	0.349623E+00	34.00	-0.897531E+01	-0.342616E+01
6.50	-0.226452E+01	0.309943E+00	36.00	-0.930823E+01	-0.369349E+01
7.00	-0.245254E+01	0.265435E+00	38.00	-0.962977E+01	-0.395733E+01
7.50	-0.263630E+01	0.216836E+00	40.00	-0.994078E+01	-0.421768E+01
8.00	-0.281591E+01	0.164755E+00	42.00	-0.102420E+02	-0.447459E+01
8.50	-0.299151E+01	0.109698E+00	44.00	-0.105342E+02	-0.472812E+01
9.00	-0.316324E+01	0.520913E-01	46.00	-0.108179E+02	-0.497833E+01
9.50	-0.333126E+01	-0.770639E-02	48.00	-0.110937E+02	-0.522530E+01
10.00	-0.349571E+01	-0.693905E-01	50.00	-0.113620E+02	-0.546909E+01

Table 2

Real and imaginary parts of $G(\tau)$ for large values of τ . $\Delta\Re G(\tau)$ and $\Delta\Im G(\tau)$ are the relative errors with respect to the asymptotic estimate given in (4.7)

τ	$\Re G(\tau)$	$\Im G(\tau)$	$\Delta\Re G(\tau)$	$\Delta\Im G(\tau)$
1.0E+10	-0.6319E+3	-0.2843E+4	0.24E-0	0.30E-0
1.0E+20	-0.2819E+4	-0.2613E+5	0.15E-0	0.20E-0
1.0E+30	-0.6630E+4	-0.9335E+5	0.11E-0	0.15E-0
1.0E+40	-0.1208E+5	-0.2284E+6	0.93E-1	0.12E-0
1.0E+50	-0.1918E+5	-0.4555E+6	0.79E-1	0.10E-0
1.0E+60	-0.2792E+5	-0.7987E+6	0.69E-1	0.91E-1
1.0E+70	-0.3833E+5	-0.1282E+7	0.61E-1	0.83E-1
1.0E+80	-0.5038E+5	-0.1930E+7	0.55E-1	0.74E-1
1.0E+90	-0.6409E+5	-0.2767E+7	0.49E-1	0.67E-1
1.0E+100	-0.7946E+5	-0.3817E+7	0.45E-1	0.61E-1

Fourier transform of the total ‘potential energy’ in the medium [1] containing the particles as a function of frequency. It is given by

$$C(\tau) = e^{-\tau} \Re[e^{i\omega_0 \tau} (1 + \varrho G(\tau))],$$

where ω_0 is the eigenfrequency of the dipoles and ϱ is a dimensionless quantity proportional to the number density of the dipoles. The parameter τ has been scaled with the inverse linewidth of the radiation resonance of the dipole. For a single dipole $C(\tau)$ decreases exponentially with τ , suggest-

ing that this inverse linewidth is a sort of 'dwell time' of the light in the particle. In general we might associate a 'dwell time' with the decay properties of $C(\tau)$.

The function $G(\tau)$ describes the influence of recurrent scattering between two dipoles on this correlation function, the various orders of recurrency being obtained separately by expanding the integrand of Eq. (1.1) formally into a Taylor series. The first order of recurrency is in fact at the base of the $1/r^6$ Van-der-Waals interaction between two polarizable particles separated by a distance r . At larger times, higher orders of recurrent scattering take over.

From the analysis of the present paper we can draw various conclusions:

- The field-field correlation function achieves an out-phase component, reflected by the imaginary part of $G(\tau)$. Physically this happens because the line profile is no longer symmetric with respect to the resonance.
- At large times, the correlation function $C(\tau)$ is completely determined by high orders of recurrent scattering, and mainly out-phase.
- The exponential decay is not replaced by an asymptotic algebraic decay. Instead it takes the form $\exp(-\tau) \ln^3 \tau$. This means that the original inverse linewidth is still a characteristic time scale, although the real dwell time seems to be increased.

The last point raises the interesting (but difficult) question what happens to correlation function and dwell time if recurrent scattering between more than 2 scatterers is taken into account.

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Abstract

We consider an integral that recently showed up in the calculation of the time-dependent field-field correlation function of the electric field inside polarizable (dielectric) particles. We derive new integral representations on which numerical algorithms can be based and which give information on the asymptotic behaviour for large values of a time parameter. We interpret the results of the paper in terms of the physical problem.

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