Low-frequency elastic response of a spherical particle

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A continuum approach is discussed to describe the low-frequency response of a small spherical particle under laser irradiation. This response is interpreted in terms of essentially long-wavelength normal vibrations of a perfectly elastic globe. The eigenfrequencies of spheroidal and torsional oscillations are derived as functions of the particle radius R and multipole degree l. The frequencies of spheroidal vibrations are found to be $\omega_s = \omega_0\,[2(2l+1)(l-1)]^{1/2}$; the eigenmodes of torsional oscillations are given by $\omega_t = \omega_0\,[(2l+3)(l-1)]^{1/2}$, with $\omega_0^2 = \mu/(\rho_0 R^2)$, where μ and ρ_0 are the shear modulus and the bulk density, respectively. The obtained results may be tested on spherical clusters with $R \sim$ 100–200 Å, irradiated by infrared laser light with the wavelength $\lambda \sim 5000$ Å.

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I. INTRODUCTION

The experimental methods of infrared spectroscopy such as the photoabsorption and the low-frequency Raman inelastic scattering afford a unique opportunity to study the matter in the form of fine particles. In particular, these methods allow one to carry out direct measurements of vibration frequencies of nanoparticles (see, for instance, [1], and references therein). The latter problem has attracted much attention in the past [2–5], and nowadays is a subject of renewed interest in the physics of clusters.

Theoretical analysis of the particle response given in Refs. [2–5] has been performed within the well known model of Lamb [6]. This model presumes that (i) a fine particle may be pictured by a perfectly elastic globe with continuous distribution of atoms and (ii) the linear dynamics of oscillations is governed by the Lamé equation (analysis of this model can be found elsewhere [7]):

$$\rho_0\, \ddot{\mathbf{s}} = (\lambda + \mu)\,\mathrm{grad}\,\mathrm{div}\,\mathbf{s} + \mu \Delta\, \mathbf{s}, \tag{1.1}$$

where ρ_0 is the bulk density; $\mathbf{s}(\mathbf{r}, t)$ is the field of displacements; μ is the shear modulus and λ is the squeezing modulus (parameter of isotropic dilatations). According to Lamb, the eigenmodes of an absolutely elastic globe are classified as the spheroidal and torsional ones. The general solution of (1.1) for harmonic in time displacements resulting in the spheroidal modes is represented as a superposition of the potential field and the solenoidal, poloidal field. The torsional vibrations are described by a pure solenoidal, toroidal field of displacements. The full analysis of solenoidal (poloidal and toroidal) vector fields is given in [8]. The above mentioned displacements corresponding to the spheroidal $\mathbf{s}_s$ and torsional $\mathbf{s}_t$ vibrations are written as

$$\mathbf{s}_s = A\,\mathrm{grad}\,\phi + B\,\mathrm{rot}\,\mathrm{rot}\,\mathbf{r}\,\phi, \quad \mathbf{s}_t = C\,\mathrm{rot}\,\mathbf{r}\,\phi, \tag{1.2}$$

with the scalar function ϕ of the form

$$\phi = j_l(kr)\, P_l(\cos\theta)\, \sin \omega t. \tag{1.3}$$

Here A, B, and C are arbitrary constants which are fixed by boundary conditions; $j_l(kr)$ is the spherical Bessel function of the multipole order l, and $P_l(\cos\theta)$ is the Legendre polynomial (hereafter the frame with fixed polar axis is used). The fundamental frequencies of normal vibrations crucially depend on the vector nature of displacements, i.e., whether or not this field is longitudinal, transverse, or their superposition. The second factor affecting the absolute value of eigenfrequencies is related to the choice of boundary conditions (Neumann or Dirichlet, to be exact). These two factors uniquely determine the modes of eigenvibrations and are sources of uncertainties in the predictions of frequencies computed in [2–5] on the basis of the Lamé equation.

In this paper, we formulate a continuum approach to the problem of normal elastic vibrations of a macroscopically small spherical particle. Instead of the Lamé equation we use the closed chain of equations of the first order for such variables as the density, mean velocity, and strain tensor. In the modern theory of continuum this approach is known as "13-moment approximation" [9]. In Sec. II, it is shown that new equations for linear vibrations of an elastic solid may be reduced to the Lamé equation (1.1), and, thereby, all results of the Lamb model are recovered. The method under consideration turns out to be especially effective in the calculations of *low-frequency* eigenmodes corresponding to the *long-wavelength* elastic vibrations of an elastic globe. This just is the case when the procedure of the frequency calculations (from the boundary conditions placed on the Lamb solutions) ceases to be valid. Therefore one of our goals is to supplement the investigations performed in Refs. [2–5]. The long-wavelength vibrations of a homogeneous elastic globe are studied in Sec. III, where the low-frequency spectra of spheroidal and torsional modes are derived in analytic form as functions of particle radius, shear modulus, density, and multipole degree. The discussion of the results obtained is contained in Sec. IV. The details of calculations are presented in Appendix.

II. GOVERNING EQUATIONS OF AN ELASTIC CONTINUUM

The continuum approach is expected to be an effective tool in the theoretical analysis of spectroscopic data when it is known in advance that the wavelengths of absorbed or inelastically scattered photons are much larger than the lattice spacing in the particle interior. The underlying idea of a macroscopic description of the particle response is the following. When a macroscopically small particle is irradiated by intense long-wavelength laser light, a large number of atoms are set in collective vibrations; the center of mass of the particle stays at rest. Owing to the fact that the length of electromagnetic wave is much larger than the crystal lattice spacing (of the same order or larger than the particle size), the coherent vibration of atoms is supposed to be modeled by vibrations of a continunm. Having accepted the model of an elastic solid for particle material, the observable response may be interpreted in terms of resonant scattering of the photons on the phonons of long-wavelength elastic vibrations.

In this section, we show that the consequent mathematical treatment of elastic vibrations may be given within the framework of equations of the 13-moment approximation of the continuum theory. The terminology (widely used in plasma physics) originates from the fact that the density ρ, the three components of mean velocity V_i, and nine components of the strain tensor P_{ij} are defined as, respectively, zero, first, and second velocity moments of distribution function of atoms. Equations for these variables are obtained from the collisionless kinetic equation by taking successive velocity moments of the latter [9]. A similar method has recently been considered in nuclear physics where it has been found that giant nuclear resonances may be interpreted as a manifestation of the quantum elasticity of nuclear matter (see [11–13], and references therein).

The governing equations of the method have the form

$$\frac{d\rho}{dt} + \rho\frac{\partial V_k}{\partial x_k} = 0, \tag{2.1}$$

$$\rho\frac{dV_i}{dt} + \frac{\partial P_{ik}}{\partial x_k} + \rho\frac{\partial U}{\partial x_i} = 0, \tag{2.2}$$

$$\frac{dP_{ij}}{dt} + P_{ik}\frac{\partial V_j}{\partial x_k} + P_{jk}\frac{\partial V_i}{\partial x_k} + P_{ij}\frac{\partial V_k}{\partial x_k} = 0. \tag{2.3}$$

Equation (2.1) is the equation of continuity. Equation (2.2) is the Euler equation (only when $P_{ij} = \delta_{ij}P$, to be exact). The internal potential energy of intermolecular forces is denoted by U. Equation (2.3) describes the evolution of internal stresses in the material.

Let us consider the problem of normal oscillations. The linearized equations (2.1)–(2.3) are represented as

$$\frac{\partial\delta\rho}{\partial t} + \rho_0\frac{\partial\delta V_k}{\partial x_k} = 0, \tag{2.4}$$

$$\rho_0\frac{\partial\delta V_i}{\partial t} + \frac{\partial\delta P_{ij}}{\partial x_j} + \rho_0\frac{\partial\delta U}{\partial x_i} = 0, \tag{2.5}$$

$$\frac{\partial\delta P_{ij}}{\partial t} + P_0\left(\frac{\partial\delta V_i}{\partial x_j} + \frac{\partial\delta V_j}{\partial x_i} + \delta_{ij}\frac{\partial\delta V_k}{\partial x_k}\right) = 0. \tag{2.6}$$

By ρ_0 and P_0 we denoted the equilibrium density and pressure which are presumed to be uniform and isotropic. By definition, the fluctuations in the velocity δV_i are related to the field of displacements s_i by means of

$$\delta V_i(\mathbf{r},t) = \frac{\partial s_i(\mathbf{r},t)}{\partial t}. \tag{2.7}$$

Inserting (2.7) into (2.4) we obtain

$$\delta\rho = -\rho_0\frac{\partial s_k(\mathbf{r},t)}{\partial x_k}. \tag{2.8}$$

Substitution of (2.7) into (2.6) yields

$$\delta P_{ij} = -P_0\left(\frac{\partial s_i}{\partial x_j} + \frac{\partial s_j}{\partial x_i} + \delta_{ij}\frac{\partial s_k}{\partial x_k}\right). \tag{2.9}$$

The potential energy is the functional of the density $U = U(\rho)$. Thereby, the fluctuations in U may be represented as

$$\delta U = \left(\frac{\partial U}{\partial\rho}\right)_{\rho_0}\delta\rho = -\rho_0\left(\frac{\partial U}{\partial\rho}\right)_{\rho_0}\operatorname{div}\mathbf{s}. \tag{2.10}$$

Substitution of (2.7), (2.9), and (2.10) into (2.5) leads to

$$\rho_0\ddot{\mathbf{s}} = \left[2P_0 + \rho_0^2\left(\frac{\partial U}{\partial\rho}\right)_{\rho_0}\right]\operatorname{grad}\operatorname{div}\mathbf{s} + P_0\Delta\,\mathbf{s}. \tag{2.11}$$

Defining the Lamé coefficients μ, the shear modulus, and λ, the squeezing modulus, as

$$\mu = P_0, \qquad \lambda = P_0 + \rho_0^2\left(\frac{\partial U}{\partial\rho}\right)_{\rho_0}, \tag{2.12}$$

Eq. (2.11) takes the form

$$\rho_0\ddot{\mathbf{s}} = (\lambda+\mu)\operatorname{grad}\operatorname{div}\mathbf{s} + \mu\Delta\,\mathbf{s},$$

and we see that this is exactly the Lamé equation (1.1). Therefore all the results obtained within the Lamb model can be recovered on the basis of Eqs. (2.1)–(2.3). Moreover, the method based on these equations, as will be shown below, is very effective in the analysis of the low-frequency response which corresponds to excitation of long-wavelength elastic vibrations. Meantime, the standard method for frequency estimation (based on the boundary conditions placed on the Lamb solutions of the Lamé equation) becomes questionable.

To illustrate the latter statement, let us consider solely longitudinal vibrations, i.e., when the field $\mathbf{s}(\mathbf{r},t)$ is irrotational (rot $\mathbf{s}$=0). Then, Eq. (1.1) transforms into the wave equation (see, for details, [10])

$$c_l^{-2}\,\ddot{\mathbf{s}} - \Delta\,\mathbf{s} = 0, \tag{2.13}$$

where $c_l = [(\lambda+2\mu)/\rho_0]^{1/2}$ is the speed of longitudinal sound wave. For the standing waves Eq. (2.13) is replaced by the Helmholtz equation

$$\Delta\mathbf{s} + k^2\mathbf{s} = 0, \qquad \text{with} \qquad k^2 = \frac{\omega^2}{c_l^2}. \tag{2.14}$$

Here k is the wave number and ω stands for the frequency of oscillations. The solution of Eq. (2.14), regular in origin, is well known

$$\mathbf{s} = \operatorname{grad}\phi, \quad \text{with } \phi = j_l(kr)\,P_l(\cos\theta)\,\sin\omega t. \tag{2.15}$$

The spectrum of spheroidal modes may be computed by using either the Dirichlet or Neumann boundary conditions. The Dirichlet condition reads

$$\phi\,|_{r=R} = 0, \quad \text{or the same } j_l(kr)|_{r=R} = 0. \tag{2.16}$$

The Neumann boundary condition requires

$$\left.\frac{\partial \phi}{\partial r}\right|_{r=R} = 0, \quad \text{or the same } \left.\frac{\partial j_l(kr)}{\partial r}\right|_{r=R} = 0. \tag{2.17}$$

The eigenfrequencies are computed from the equation

$$k_{ln}\, R = \frac{\omega_{ln}}{c_l}\, R = z_{ln}, \qquad \rightarrow \qquad \omega_{ln} = \frac{c_l}{R} z_{ln}. \tag{2.18}$$

Here index n labels the number of a root at a fixed multipole degree l. It is clearly seen that the Dirichlet and Neumann boundary conditions yield different values of z_{ln} and, hence, lead to the different predictions of eigenfrequencies ω_{ln}. Analogous analysis can be carried out for the general solutions [Eqs. (1.2), (1.3)].

Particular emphasis should be placed on the case of low-frequency response related to excitation in the particle volume of the long-wavelength vibrations, i.e., when $\lambda \to \infty$ and wave number $k = (2\pi/\lambda) \to 0$. In this limit, Eq. (2.14) is replaced by the vector Laplace equation

$$\Delta \mathbf{s} = 0. \tag{2.19}$$

The solution of Eq. (2.19) corresponding to the long-wavelength longitudinal vibrations is given by

$$\mathbf{s} = \text{grad}\phi, \quad \text{with } \phi = r^l\, P_l(\cos\,\theta)\, \sin\omega t, \tag{2.20}$$

that is, the radial dependence is given by the function r^l instead of the Bessel function $j_l(kr)$. As a result, the displacement field $\mathbf{s}$ happens to be independent of the wave number k. In view of this, the boundary conditions do not help in the finding of vibrational frequencies ω. In the next section a method is presented which allows one to calculate the eigenmodes of long-wavelength elastic oscillations.

III. LONG-WAVELENGTH ELASTIC VIBRATIONS OF A SPHERICAL PARTICLE

The low-frequency particle response is induced by the weak fields. Therefore it is natural to believe that external perturbations do not lead to the fluctuations in the density. Therefore the particle material may be considered as an incompressible elastic continuum. In this case equations of linear dynamics are written as

$$\frac{\partial \delta V_k}{\partial x_k} = 0, \tag{3.1}$$

$$\rho_0 \frac{\partial \delta V_i}{\partial t} + \frac{\partial \delta P_{ij}}{\partial x_j} = 0, \tag{3.2}$$

$$\frac{\partial \delta P_{ij}}{\partial t} + \mu \left(\frac{\partial \delta V_i}{\partial x_j} + \frac{\partial \delta V_j}{\partial x_i} \right) = 0, \tag{3.3}$$

where μ is the shear modulus. Combining Eqs. (3.1)–(3.3) we come to the standard equation for transverse waves,

$$\rho_0\, \ddot{\mathbf{V}} - \mu\, \Delta \delta\, \mathbf{V} = 0, \tag{3.4}$$

which for standing waves is reduced to the Helmholtz equation

$$\Delta \delta \mathbf{V} + k^2 \delta \mathbf{V} = 0, \qquad \text{div}\, \delta \mathbf{V} = 0. \tag{3.5}$$

Here $k = \omega/c_t$ and $c_t = \sqrt{\mu/\rho_0}$ is the speed of transverse waves. Next, it is convenient to represent the deviations of velocity in the form

$$\delta \mathbf{V}(\mathbf{r}, t) = \boldsymbol{\xi}(\mathbf{r}) \dot{\alpha}(t), \tag{3.6}$$

where $\boldsymbol{\xi}(\mathbf{r})$ is the field of instantaneous displacements, and $\alpha(t)$ is the harmonic in time amplitude of vibrations.

In the long-wavelength limit ($k \to 0$), Eq. (3.5) is transformed into the vector Laplace equation which [accounting for substitution (3.6)] takes the form

$$\Delta\, \boldsymbol{\xi} = 0, \qquad \text{div}\, \boldsymbol{\xi} = 0. \tag{3.7}$$

Equation (3.7) has two independent solutions: poloidal and toroidal corresponding to spheroidal and torsional vibrations, respectively.

Substitution of (3.6) into (3.3) allows one to represent the fluctuations in the strain tensor as

$$\delta P_{ij} = -\mu \left(\frac{\partial \xi_i}{\partial x_j} + \frac{\partial \xi_j}{\partial x_i} \right) \alpha. \tag{3.8}$$

It is worth noticing that $Sp\, \delta P_{ij} = \delta P_{ii} = 0$; this is due to the condition of incompressibility (3.7).

The fundamental frequencies of long-wavelength vibrations may be computed by use of the Rayleigh "method of normal coordinates." The procedure is the following. The scalar multiplication of (3.2) by the velocity departure δV_i and integration over the particle volume yields the equation of energy balance

$$\frac{1}{2} \int \rho_0 \frac{\partial \delta V^2}{\partial t} d\tau + \int \delta V_i \frac{\partial \delta P_{ij}}{\partial x_j} d\tau = 0. \tag{3.9}$$

Substitution of (3.6) and (3.8) into (3.9) leads to the standard equation of harmonic oscillations

$$M \ddot{\alpha} + K \alpha = 0, \tag{3.10}$$

where parameters of inertia M and stiffness K are given by

$$M = \int \rho_0 \xi_i \xi_i\, d\tau, \quad K = \frac{1}{2} \int \mu \left(\frac{\partial \xi_i}{\partial x_j} + \frac{\partial \xi_j}{\partial x_i} \right)^2 d\tau. \tag{3.11}$$

So, in order to compute the frequency $\omega = \sqrt{K/M}$ one only needs to know the distribution field of instantaneous displacements $\boldsymbol{\xi}$. The density ρ_0 and the shear modulus μ entering (3.11) are input parameters of the method.

A. Spheroidal eigenmodes

The poloidal solution of Eq. (3.7) has the form

$$\boldsymbol{\xi}_p(\mathbf{r}) = N_p \text{rot}\, \text{rot}\, \mathbf{r}\, r^l\, P_l(\cos\theta) = N_p(l+1) \nabla\, r^l\, P_l(\cos\theta). \tag{3.12}$$

This field of displacements corresponds to the spheroidal modes and describes the harmonic distortions of the particle surface. The inertia and stiffness computed with field (3.12) are given by

$$M = 3\,m\,N_p^2 \frac{l}{(2l+1)} R^{2l-2}, \;\; K = 6\,m\; N_p^2 c_t^2\, l(l-1)\, R^{2l-4}, \tag{3.13}$$

where m is the mass and R is the radius of a particle. The eigenfrequencies of spheroidal long-wavelength vibrations ω_s are found to be

$$\omega_s = \omega_0\,[2(2l+1)(l-1)]^{1/2}, \tag{3.14}$$

where ω_0 stands for the basic frequency of transverse elastic vibrations

$$\omega_0^2 = \left(\frac{c_t}{R}\right)^2 = \frac{\mu}{\rho_0 R^2}. \tag{3.15}$$

It is worthwhile to notice that surface oscillations characterize the response of liquid drop. If the particle is modeled by the drop of inviscid liquid, the macroscopic response of the particle is described by the modes of solely spheroidal vibrations.

B. Torsional eigenmodes

The toroidal solution of Eq. (3.7) is given by

$$\boldsymbol{\xi}_t(\mathbf{r}) = N_t \mathrm{rot}\,\mathbf{r}\, r^l P_l(\cos\theta). \tag{3.16}$$

This field describes the torsional (essentially rotational) oscillations which are solely due to elastic properties of the particle continuum (in the drop of inviscid liquid these vibrations cannot be excited). The inertia and stiffness of torsional vibrations read

$$M = 3\,m\;N_t^2 \frac{l(l+1)}{(2l+1)(2l+3)} R^{2l},$$
$$K = 3\,m\;N_t^2\, c_t^2 \frac{l(l^2-1)}{(2l+1)}\, R^{2l-2}. \tag{3.17}$$

The eigenmodes of torsional long-wavelength vibrations are given by

$$\omega_t = \omega_0\,[(2l+3)(l-1)]^{1/2}. \tag{3.18}$$

It is worthwhile to notice that arbitrary constants do not enter the final expressions for the frequencies. In other words, the problem of boundary conditions does not appear at all and, therefore, one may set $N_p = N_t = 1$. It is interesting to note that fluctuations in velocity, Eq. (3.7), with field of instantaneous displacements (3.16) may be represented as

$$\delta\mathbf{V} = [\mathbf{r}\times\boldsymbol{\omega}], \tag{3.19}$$

where $\boldsymbol{\omega} = -\nabla r^l P_l(\cos\theta)\,\dot{\alpha}$ is the angular velocity of the local rotational vibrations; α is an azimuthal angle of rotation around polar axis z. For example, the dipole case corresponds to the rigid rotation of the particle. The quadrupole torsional mode, recently discussed in [14], is associated with excitation of motions when the north and south semispheres oscillate in the opposite directions.

IV. SUMMARY

Equations (3.14) and (3.18) are the basic finding of the present paper. It follows from (3.14) and (3.18) that the monopole excitations ($l = 0$) of both kinds are excluded. This is due to inertness of the particle material (the fluctuation in density is considered to be negligible). The dipole field ($l = 1$) of poloidal displacements contributes only to the parameter of inertia M, whereas the restoring force (the parameter of rigidity K) of vibrations is canceled [see Eq. (3.13)]. As was mentioned above, the dipole case corresponds to the translation of the particle center of mass. An analogous effect takes place for the torsional dipole excitation. However, in this case we deal with the rotation of a particle as a whole. For torsional vibrations the parameter M is interpreted to be the moment of inertia J. In the case of rigid rotation ($l = 1$) we obtain the well known formula for moment of inertia of the sphere: $J = (2/5)MR^2$. Thus we have obtained that the lowest multipole degree of the spheroidal and torsional modes is quadrupole. It is worthwhile to emphasize that the frequencies of torsional vibrations are systematically lower than those for spheroidal ones. This follows from the ratio

$$\frac{\omega_t}{\omega_s} = \left[\frac{(2l+3)}{2(2l+1)}\right]^{1/2} < 1, \qquad l \geq 2. \tag{4.1}$$

As was noted above, the approach considered may be applied to analyze data when it is known in advance that the wavelengths of photons are much larger compared to the particle size. This requirement is compatible with the conditions of measurements reported in [1], where the particles with radii of the order of 50–200 Å were irradiated by the light of a 4-W argon laser with wavelengths $\lambda \sim 5000$ Å. Therefore the measurable spectra may be interpreted on the basis of the theory presented. As a conclusion we note that the model of long-wavelength vibrations of a perfectly elastic globe is interesting as such and one may hope that the results obtained here can find other physical applications.

APPENDIX A

The links between the Cartesian and spherical components of the strain tensor used in calculations of stiffness are given by

$$\frac{\partial\xi_1}{\partial x_1} = \frac{\partial\xi_r}{\partial r}, \;\; \frac{\partial\xi_2}{\partial x_2} = \frac{1}{r}\frac{\partial\xi_\theta}{\partial\theta} + \frac{\xi_r}{r},$$

$$\frac{\partial\xi_3}{\partial x_3} = \frac{1}{r\sin\theta}\frac{\partial\xi_\phi}{\partial\phi} + \frac{\xi_r}{r} + \frac{\xi_\theta \cot\theta}{r},$$

$$\frac{\partial\xi_1}{\partial x_2} = \frac{1}{r}\frac{\partial\xi_r}{\partial\theta} - \frac{\xi_\theta}{r}, \;\; \frac{\partial\xi_2}{\partial x_1} = \frac{\partial\xi_\theta}{\partial r},$$

$$\frac{\partial\xi_1}{\partial x_3} = \frac{1}{r\sin\theta}\frac{\partial\xi_r}{\partial\phi} - \frac{\xi_\phi}{r},$$

$$\frac{\partial \xi_3}{\partial x_1} = \frac{\partial \xi_\phi}{\partial r}, \quad \frac{\partial \xi_2}{\partial x_3} = \frac{1}{r\sin\theta}\frac{\partial \xi_\theta}{\partial \phi} - \frac{\xi_\phi}{r}\cot\theta,$$

$$\frac{\partial \xi_3}{\partial x_2} = \frac{1}{r}\frac{\partial \xi_\phi}{\partial \theta},$$

where ξ_i are the components of the instantaneous displacements.

The calculations of integrals are much easier to perform if one uses the substitution $\mu = \cos\theta$. The following integrals turn out to be useful:

$$\int_{-1}^{+1} P_l^2(\mu)\, d\mu = \frac{2}{(2l+1)},$$

$$\int_{-1}^{+1} (1-\mu^2)\left(\frac{dP_l(\mu)}{d\mu}\right)^2 d\mu = \frac{2l(l+1)}{(2l+1)},$$

$$\int_{-1}^{+1} \mu P_l(\mu)\frac{dP_l(\mu)}{d\mu}\, d\mu = \frac{2l}{(2l+1)},$$

$$\int_{-1}^{+1} \left(\mu\frac{dP_l(\mu)}{d\mu}\right)^2 d\mu = \frac{l(l+1)(2l-1)}{(2l+1)}.$$

Two last integrals have been calculated on the basis of recurrent relations between Legendre polynomials.

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