EXACT ENERGY OF INTERACTION BETWEEN NONPOLAR MOLECULES REPRESENTED AS DIPOLES

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Abstract

A new non-perturbative method is developed for deriving the energy of interaction between molecules represented as polarizable dipoles. The method is applied to obtain the exact energy of interaction for two anisotropic electric dipoles and for three dipoles in the isotropic case. The results of London and of Casimir and Polder are special limiting cases. Deviations of the London energy from the exact result are investigated. A new feature which emerges is that the potential energy becomes repulsive at very small distances of separation.

I. INTRODUCTION

The first calculations of the interaction energy $V$ between two nonpolar molecules or atoms as a function of separation $r$ were made by London (1942), who found $V \sim r^{-6}$. Subsequently Casimir and Polder (1948) showed that for very large $r$ this result breaks down. For $r \gg c/\nu_0$, where $\nu_0$ is the principal absorption frequency of the interacting atoms, $V \sim r^{-7}$. Their expression has since been rederived by a number of authors using quantum mechanical perturbation theory (see e.g. Craig and Power 1969, where references to earlier work are given). Our purpose here is to draw attention to a different procedure which can be used to calculate interaction energies between two or more atoms represented as polarizable dipoles. The method is extremely simple, is not based on perturbation theory, and gives the energy exactly for all distances of separation of the dipoles. The application of this technique to the two-dipole problem was reported briefly earlier (Mitchell, Ninham, and Richmond 1971). It was first developed by Mitchell and Ninham (1972) to calculate interaction energies between macroscopic bodies (i.e. those with a well-defined electromagnetic susceptibility) in the non-retarded limit. To illustrate the method here we solve two specific problems. The first deals with the exact interaction energy of two dipoles with anisotropic polarizabilities. From the general formula the results of London (1942), Casimir and Polder (1948), and Craig and Power (1969) emerge as special limiting cases. Secondly, we solve the corresponding three-body problem exactly. Here, to avoid unnecessarily cumbersome algebra, we restrict the analysis to isotropic dipoles. We would hope that this last problem has some interest simply because it is an exact solution of a three-body problem.

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II. Interaction Energy for Two Anisotropic Dipoles

(a) General Formulation

Consider first two dipoles and choose a cartesian coordinate system with origin at dipole 1. Suppose that the positive $z$ axis is in the direction joining the origin to dipole 2. The principal axes of the polarizability tensors of the dipoles have arbitrary orientation with respect to the space coordinates. The interaction energy arises due to the mutually induced polarization which we must first calculate as follows.

Suppose that the instantaneous dipole moment of dipole 1 is

$$P(1, t) = P \exp(-i\omega t).$$

The vector and scalar potentials $A$ and $\phi$ generated at an arbitrary point distant $r$ from the dipole are

$$A(r, t) = -\frac{i\omega}{c} P \exp(-i\omega t) \frac{\exp(i\omega r/c)}{r}$$

and

$$\phi(r, t) = \frac{P \cdot r}{r^2} \left(1 - \frac{i\omega}{c}\right) \exp(-i\omega t) \exp(i\omega r/c).$$

The associated electric field is

$$E(r, t) = -c^{-1} \frac{\partial A}{\partial t} - \nabla \phi.$$ 

Consequently we have $E(r, t) = E \exp(-i\omega t)$ where

$$E = P \left(\frac{\omega^2}{c^2} + \frac{i\omega}{cr} - \frac{1}{r^2}\right) \exp(i\omega r/c)$$

$$- (P \cdot r) \frac{r}{r^3} \left(\frac{\omega^2}{c^2} + \frac{3i\omega}{cr} - \frac{3}{r^2}\right) \exp(i\omega r/c).$$

Hence if $|r|$ is the distance between the dipoles, and $r$ is in the direction of the $z$ axis, the components of $E(1)$ generated by dipole 1 at the position of dipole 2 are

$$E_x(1) = f(r) P_x(1), \quad E_y(1) = f(r) P_y(1), \quad E_z(1) = h(r) P_z(1),$$

where

$$f(r) = \frac{\exp(i\omega r/c)}{r} \left(\frac{\omega^2}{c^2} + \frac{i\omega}{cr} - \frac{1}{r^2}\right), \quad h(r) = \frac{2\exp(i\omega r/c)}{r} \left(\frac{1}{r^2} - \frac{i\omega}{cr}\right).$$

This field polarizes the second dipole whose polarization will be given by

$$P(2) = \alpha(2) E(1),$$

where $\alpha(2)$ is its polarizability tensor. This induced polarization in turn produces the electric field at the first dipole responsible for its own polarization, so that corresponding to equation (7) we have

$$P(1) = \alpha(1) E(2).$$
INTERACTION ENERGY BETWEEN DIPOLES

It follows from equations (5), (7), and (8) that

\[ P(2) = X(2) P(1), \quad P(1) = X(1) P(2), \]

(9)

where the matrices \( X \) are defined by

\[ X = \begin{pmatrix} \alpha_{11} f & \alpha_{12} f & \alpha_{13} h \\ \alpha_{21} f & \alpha_{22} f & \alpha_{23} h \\ \alpha_{31} f & \alpha_{32} f & \alpha_{33} h \end{pmatrix} \]

(10)

and the \( \alpha_{jk} \) represent the matrix elements of the polarizability tensors \( \alpha(1) \) or \( \alpha(2) \) respectively. Together equations (9) provide a condition for consistency on allowed modes which is

\[ P(1) = Y P(1), \quad Y = X(1) X(2). \]

(11)

The frequencies of the allowed modes are therefore given by a "dispersion" relation

\[ D(\omega) = \det(1 - Y) = 0. \]

(12)

From equations (10) and (11) the matrix \( Y \) has components

\[ y_{11} = \{\alpha_{11}(1) \alpha_{11}(2) + \alpha_{12}(1) \alpha_{21}(2)\} f^2 + \alpha_{13}(1) \alpha_{31}(2) f h, \]

(13a)
\[ y_{22} = \{\alpha_{21}(1) \alpha_{12}(2) + \alpha_{22}(1) \alpha_{22}(2)\} f^2 + \alpha_{23}(1) \alpha_{32}(2) f h, \]

(13b)
\[ y_{33} = \{\alpha_{31}(1) \alpha_{13}(2) + \alpha_{32}(1) \alpha_{23}(2)\} f h + \alpha_{33}(1) \alpha_{33}(2) h^2, \]

(13c)
\[ y_{12} = \{\alpha_{11}(1) \alpha_{12}(2) + \alpha_{12}(1) \alpha_{21}(2)\} f^2 + \alpha_{13}(1) \alpha_{32}(2) f h, \]

(13d)
\[ y_{13} = \{\alpha_{11}(1) \alpha_{13}(2) + \alpha_{12}(1) \alpha_{23}(2)\} f h + \alpha_{13}(1) \alpha_{33}(2) h^2, \]

(13e)
\[ y_{23} = \{\alpha_{21}(1) \alpha_{13}(2) + \alpha_{22}(1) \alpha_{23}(2)\} f h + \alpha_{23}(1) \alpha_{33}(2) h^2. \]

(13f)

The elements \( y_{21}, y_{31}, \) and \( y_{32} \) follow from (13d), (13e), and (13f) by interchanging dipole indices 1 and 2, e.g. \( y_{21} = y_{12}(1 \leftrightarrow 2) \).

The energy of interaction can now be written down immediately as

\[ V(r) = (\hbar/2\pi) \int_0^\infty d\xi \ln D(i\xi). \]

(14)

A derivation and discussion of this expression has been given by Ninham, Parsegian, and Weiss (1970) and Richmond and Ninham (1971).

(b) Reduction to Known Results

The integrand of the general expression (14) simplifies considerably in the limit of large \( r \), that is, in the London (1942) or Casimir and Polder (1948) limits. This follows from the observation that \( f(r) \) and \( h(r) \) defined in equations (6) are \( O(r^{-3}) \). Therefore the matrix elements \( y_{jk} \) of \( Y \) are \( O(r^{-6}) \) and we have

\[ D(\omega) = \det(1 - Y) = \left(1 - \sum_j y_{jj} + O(r^{-12})\right). \]

(15)
To terms of lowest order in $r^{-1}$ the dispersion relation is then

$$D(\omega) = 1 - q(\omega) + O(r^{-12}),$$

(16)

where

$$q(\omega) = \left[ x_{11}(1)x_{11}(2) + 2x_{22}(1)x_{22}(2) + 2x_{12}(1)x_{12}(2) \right] f^2$$

$$+ \left[ x_{13}(1)x_{13}(2) + x_{23}(1)x_{23}(2) \right] f + x_{33}(1)x_{33}(2) h^2.$$ 

This result has been obtained using equations (13) and the symmetry of $a$, namely $a_{ij} = a_{ji}$.

(c) Retarded Potential for Anisotropic Dipoles

Previous results now follow immediately. For large $r$ we can expand the logarithm in (14) and keep only the leading term in (16). Thus

$$V(r) \sim -(\hbar/2\pi) \int_0^\infty d\xi \; q(i\xi).$$

(17)

It is convenient to introduce the dimensionless variable $x = \xi r/c$. From equations (6) and (15)–(17) we then obtain

$$V(r) \sim -\frac{\hbar c}{2\pi r^7} \int_0^\infty dx \; \left[ x_{11}(1)x_{11}(2) + 2x_{22}(1)x_{22}(2) + 2x_{12}(1)x_{12}(2) \right] (1+x+x^2)^2$$

$$- 2\left[ x_{13}(1)x_{13}(2) + x_{23}(1)x_{23}(2) \right] (1+x)(1+x+x^2)$$

$$+ 4x_{33}(1)x_{33}(2)(1+x^2) \exp(-2x).$$

(18)

Note that the components of the polarizability tensor are evaluated at imaginary frequencies, that is, $a = a(i\xi) = a(\omega_0 c/r)$. In the limit $r \gg c/\omega_0$, where $\omega_0$ is a characteristic frequency for absorption of electromagnetic radiation by the dipoles, the components $a_{jk}(i\xi)$ in approximation (18) can be replaced by their zero frequency static values $a_{0jk}$. The remaining integrals over $x$ can now be evaluated giving

$$V(r) = -\frac{\hbar c}{8\pi r^7} \left( 13x_{11}(1)x_{11}(2) + 2x_{22}(1)x_{22}(2) + 20x_{33}(1)x_{33}(2) \right.$$

$$+ 26x_{12}(1)x_{12}(2) - 30\left[ x_{23}(1)x_{23}(2) + x_{13}(1)x_{13}(2) \right] \left) \right).$$

(19)

The $a_{jk}$ here refer to static values. This expression is identical with one deduced by Craig and Power (1969) using perturbation theory. For isotropic polarizabilities $a_{ij} = a\delta_{ij}$, equation (19) reduces to the result of Casimir and Polder (1948), namely

$$V(r) = -\frac{3}{4} \frac{\hbar c}{\pi r^7} a(1)a(2).$$

(20)

(d) Non-retarded Potential

In the opposite limiting case $r \ll c/\xi_0$ the interaction potential becomes non-retarded. We consider only a simple form for the polarizability, namely

$$a_{jk}(i\xi) = (a_0)_{jk}(1 + \xi^2/\omega_0^2).$$

(21)
If we specialize further to the case where the polarizabilities for the dipoles are identical, we can expand the integrand in (18) in powers of $x$ and keep only the leading terms. This yields

$$V(r) \sim -\frac{\hbar \omega_0}{8r^6} \left( x_{11}(1) x_{11}(2) + x_{22}(1) x_{22}(2) + 4x_{33}(1) x_{33}(2) ight. 
\left. + 2x_{12}(1) x_{12}(2) - 2(x_{13}(1) x_{13}(2) + x_{23}(1) x_{23}(2)) \right) \rho_0. \quad (22)$$

For isotropic polarizabilities, equation (22) reduces to the London (1942) potential

$$V(r) = -\frac{\hbar \omega_0}{4r^6} x_0^2. \quad (23)$$

### III. Validity of London Potential

From the preceding analysis it is clear that both the London (1942) and Casimir and Polder (1948) potentials are very simple and special asymptotic expressions for the general potential energy of interaction given by equation (14). While it is clear that the Casimir potential is correct for distances $r \gg c/\omega_0$ and the London potential in the opposite limit $r \ll c/\omega_0$, the actual potential in the intermediate region which joins their separate regimes of validity has never been investigated. We now study this problem, and consider for simplicity the special case of isotropic polarizabilities $\alpha_{jk} = \alpha \delta_{jk}$. Again suppose that both dipoles are identical with absorption frequency $\omega_0$.

The ratio of the actual potential (14) to the London potential (23) is plotted in Figure 1 for values of $x_0$ and $\omega_0$ characteristic of helium, namely $x_0 \approx 0.21 \text{ Å}^3$ and $\omega_0 = 3.7 \times 10^{16} \text{ rad s}^{-1}$. We remark on several features of the curve.

1. Over the range 2–30 Å, where the dipole approximation should be an excellent one in modellings the interaction between two atoms, the London potential is an extremely good approximation. Retardation becomes significant at much smaller distances than one might have expected.
2. At smaller separations $r \lesssim 2$ Å, deviations from the London potential are quite substantial. However, in this regime the dipole model for atoms is certainly inappropriate.
At very small distances the potential becomes repulsive! To understand the origin of this result we re-examine the small-distance limit in more detail. In the non-retarded limit \((c \to \infty)\) equations (5) take the simple form

\[
E_x(1) = -\frac{P_x(1)}{r^3}, \quad E_y(1) = -\frac{P_y(1)}{r^3}, \quad E_z(1) = \frac{2P_z(1)}{r^3}.
\]  

(24)

Repeating the analysis after equations (5), we then have corresponding to (9)

\[
P_x(2) = -\frac{\alpha(2)P_x(1)}{r^3}, \quad P_y(2) = -\frac{\alpha(2)P_y(1)}{r^3}, \quad P_z(2) = \frac{2\alpha(2)P_z(1)}{r^3},
\]  

(25)

where again we consider only isotropic polarizabilities, \((\alpha(1))_{jk} = \alpha(1) \delta_{jk}\). The dispersion relations corresponding to equations (12) now become

\[
1 - \frac{\alpha(2)\alpha(1)}{r^6} = 0, \quad 1 - \frac{\alpha(2)\alpha(1)}{r^6} = 0, \quad 1 - \frac{4\alpha(2)\alpha(1)}{r^6} = 0.
\]  

(26)

**Table 1**

**Interaction Energy of Two Dipoles as Function of Separation**

<table>
<thead>
<tr>
<th>Separation (r) (Å)</th>
<th>Energy (V(r)) (erg)</th>
<th>Separation (r) (Å)</th>
<th>Energy (V(r)) (erg)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1.6</td>
<td>-7.72 \times 10^{-14}</td>
<td>0.5</td>
<td>-1.25 \times 10^{-11}</td>
</tr>
<tr>
<td>1.4</td>
<td>-1.72 \times 10^{-13}</td>
<td>0.4</td>
<td>1.73 \times 10^{-11}</td>
</tr>
<tr>
<td>1.2</td>
<td>-4.39 \times 10^{-13}</td>
<td>0.3</td>
<td>7.70 \times 10^{-11}</td>
</tr>
<tr>
<td>1.0</td>
<td>-1.35 \times 10^{-12}</td>
<td>0.2</td>
<td>2.29 \times 10^{-10}</td>
</tr>
<tr>
<td>0.8</td>
<td>-6.17 \times 10^{-12}</td>
<td>0.1</td>
<td>8.50 \times 10^{-10}</td>
</tr>
<tr>
<td>0.6</td>
<td>-2.23 \times 10^{-11}</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

For identical isotropic dipoles with polarizability given by equation (21), the allowed frequencies are

\[
\omega = \omega_0(1 \pm \alpha_0/r^3)^{1/2}, \quad \omega = \omega_0(1 \pm 2\alpha_0/r^3)^{1/2}.
\]  

(27)

The first two roots each occur twice, and only real frequencies contribute to the interaction energy. If all roots are real, that is, \(2\alpha_0/r^3 > 1\), the energy of interaction is

\[
V(r) = \frac{1}{2} \sum \{\hbar \omega - \hbar \omega_0 (r \to \infty)\} = \frac{\hbar}{2} \omega_0 (2(1 + \alpha_0/r^3)^{1/2} + (1 + 2\alpha_0/r^3)^{1/2} + (1 - 2\alpha_0/r^3)^{1/2} - 6).
\]  

(28)

This expression goes over to the London result for \(2\alpha_0/r^3 \ll 1\). On the other hand, in the range \(2\alpha_0/r^3 > 1\) the mode \(\omega = \omega_0(1 - 2\alpha_0/r^3)^{1/2}\) becomes purely imaginary and cannot contribute to the (real) dispersion energy. We therefore omit this term. At still smaller distances \(\alpha_0/r^3 > 1\) the mode \(\omega = \omega_0(1 - \alpha_0/r^3)^{1/2}\) also becomes imaginary and must similarly be omitted. Consequently, for \(r \ll \alpha_0^{1/2}\) the energy has the form

\[
V(r) = \frac{\hbar}{2} \omega_0 (2 + \sqrt{2})\alpha_0^{1/2}/r^{3/2} - 6 + O(r^{3/2})\).  
\]  

(29)
The freezing out of some modes at distances of the order of the dimensions of the dipoles which leads to a repulsive energy instead of an attractive energy is an entirely natural result. The phenomenon is analogous to a ferroelectric phase transition in solids (Kanzig 1957; Mahan 1965) and reminiscent of the change in state of two hydrogen atoms interacting to form a molecule. For completeness, numerical results for \( V(r) \) in the short-distance regime, calculated using the procedure outlined here, are given in Table 1.

**IV. Three-body Forces**

We now illustrate the further application of the method to the corresponding three-body problem. Suppose that the three dipoles form the triangle in Figure 2, and choose coordinate axes as shown. To avoid cumbersome algebra we restrict solution to the isotropic case. If the instantaneous dipole moments are

\[ \mathbf{P}(A, t) = \mathbf{P}(A) \exp(-i\omega t) \]

with similar definitions for dipoles B and C, then the instantaneous field at A generated by B and C is

\[ \mathbf{E}(A, t) = \mathbf{E}(A) \exp(-i\omega t), \]

where the components of \( \mathbf{E}(A) \) are

\[
\begin{align*}
E_x(A) &= P_x(B)h(r_3) + P_x(C)f(r_2) - \{P_x(C)\cos \theta_1 + P_y(C)\sin \theta_1\}\cos \theta_1 g(r_2), \quad (30a) \\
E_y(A) &= P_y(B)f(r_3) + P_y(C)f(r_2) - \{P_x(C)\cos \theta_1 + P_y(C)\sin \theta_1\}\sin \theta_1 g(r_2), \quad (30b) \\
E_z(A) &= P_z(B)f(r_3) + P_z(C)f(r_2). \quad (30c)
\end{align*}
\]

Here \( f \) and \( h \) are defined by equations (6) and \( g = f - h \). The induced polarization of dipole A is then given by

\[
\begin{align*}
P_x(A) &= \alpha(A)E_x(A), \\
P_y(A) &= \alpha(A)E_y(A), \\
P_z(A) &= \alpha(A)E_z(A). \quad (31)
\end{align*}
\]

Similarly the fields at B and C are given by

\[
\begin{align*}
E_x(B) &= P_x(A)h(r_3) + P_x(C)f(r_1) - \{P_x(C)\cos \theta_2 + P_y(C)\sin \theta_2\}\cos \theta_2 g(r_1), \quad (32a) \\
E_y(B) &= P_y(A)f(r_3) + P_y(C)f(r_1) + \{P_x(C)\cos \theta_2 + P_y(C)\sin \theta_2\}\sin \theta_2 g(r_1), \quad (32b) \\
E_z(B) &= P_z(A)f(r_3) + P_z(C)f(r_1); \quad (32c)
\end{align*}
\]
\[ E_z(C) = P_x(A) f(r_2) + P_x(B) f(r_1) - \{P_x(A) \cos \theta_1 + P_y(A) \sin \theta_1\} \cos \theta_2 g(r_1), \]
\[ E_y(C) = P_y(A) f(r_2) + P_y(B) f(r_1) - \{P_x(A) \cos \theta_1 + P_y(A) \sin \theta_1\} \sin \theta_2 g(r_1), \]
\[ E_z(C) = P_x(A) f(r_2) + P_x(B) f(r_1). \]

Again, corresponding to equations (31) we have

\[ P(B) = \alpha(B) E(B), \quad P(C) = \alpha(C) E(C). \]

Consider first the \( z \) components of the polarization. Substituting for \( E_z(A), E_z(B), \) and \( E_z(C) \) in equations (31) and (34), we have

\[ P_z(A) = \alpha(A) \{P_z(B) f(r_3) + P_z(C) f(r_2)\}, \]
\[ P_z(B) = \alpha(B) \{P_z(A) f(r_3) + P_z(C) f(r_1)\}, \]
\[ P_z(C) = \alpha(C) \{P_z(A) f(r_2) + P_z(B) f(r_1)\}. \]

These equations lead to one dispersion relation

\[ D_z(\omega) = \det \begin{pmatrix} 1 & -\alpha(A) f(r_3) & -\alpha(A) f(r_2) \\ -\alpha(B) f(r_3) & 1 & -\alpha(B) f(r_1) \\ -\alpha(C) f(r_2) & -\alpha(C) f(r_1) & 1 \end{pmatrix} = 0 \]

or

\[ D_z(\omega) = 1 - \{\alpha(A) \alpha(B) f^2(r_3) + \alpha(B) \alpha(C) f^2(r_1) + \alpha(A) \alpha(C) f^2(r_2) + 2 \alpha(A) \alpha(B) \alpha(C) f(r_1) f(r_2) f(r_3)\} = 0. \]

The corresponding equations linking \( x \) and \( y \) components of the polarization are coupled. We have, after a little algebra,

\[ \begin{bmatrix} P_x(A) \\ P_y(A) \\ P_x(B) \\ P_y(B) \\ P_x(C) \\ P_y(C) \end{bmatrix} = \mathbf{M} \begin{bmatrix} P_x(A) \\ P_y(A) \end{bmatrix}, \]

with \( \mathbf{M} \) now a \( 6 \times 6 \) matrix, and corresponding to equation (36) we have the further dispersion relation

\[ D_{x,y}(\omega) = \det(1 - \mathbf{M}) = 0, \]

where the matrix \( (1 - \mathbf{M}) \) is given by the array

\[ (1 - \mathbf{M}) = \begin{pmatrix} S & T \\ U & V \end{pmatrix}. \]
with

\[
S = \begin{pmatrix}
1 & 0 & -\alpha(A) h(r_3) \\
0 & 1 & 0 \\
-\alpha(B) h(r_3) & 0 & 1
\end{pmatrix},
\]

\[
T = \begin{pmatrix}
0 & -\alpha(A) \{f(r_2) - \cos^2 \theta_1 g(r_2)\} & \alpha(A) \cos \theta_1 \sin \theta_1 g(r_2) \\
-\alpha(A) f(r_3) & \alpha(A) \cos \theta_1 \sin \theta_1 g(r_2) & \alpha(A) \{ -f(r_2) + \sin^2 \theta_1 g(r_2)\} \\
0 & -\alpha(B) \{f(r_1) - \cos^2 \theta_2 g(r_1)\} & -\alpha(B) \sin \theta_2 \cos \theta_2 g(r_1)
\end{pmatrix},
\]

\[
U = \begin{pmatrix}
0 & -\alpha(B) f(r_3) & 0 \\
-\alpha(C) \{f(r_2) - \cos^2 \theta_1 g(r_2)\} & \alpha(C) \sin \theta_1 \cos \theta_1 g(r_2) & -\alpha(C) \{f(r_1) - \cos^2 \theta_2 g(r_1)\} \\
\alpha(C) \cos \theta_1 \sin \theta_1 g(r_2) & -\alpha(C) \{f(r_2) - \sin^2 \theta_1 g(r_2)\} & -\alpha(C) \sin \theta_2 \cos \theta_2 g(r_1)
\end{pmatrix},
\]

\[
V = \begin{pmatrix}
1 & -\alpha(B) \cos \theta_2 \sin \theta_2 g(r_1) & -\alpha(B) \{f(r_1) - \sin^2 \theta_2 g(r_1)\} \\
-\alpha(C) \cos \theta_2 \sin \theta_2 g(r_1) & 1 & 0 \\
-\alpha(C) \{f(r_1) - \sin^2 \theta_2 g(r_1)\} & 0 & 1
\end{pmatrix}.
\]

Corresponding to equation (14) we now have the complete energy of interaction as

\[
V(r) = \left(\frac{\hbar}{2\pi}\right) \int_0^\infty \text{d} \xi \ln \{D_z(\xi) + D_{x, y}(\xi)\}. \tag{40}
\]

To recover known results, we proceed as before and expand the logarithms and determinants to terms of order \(r^{-9}\). Thus

\[
\ln D_z(\xi) \sim -\alpha(A) \alpha(B) f^2(r_3) -\alpha(A) \alpha(C) f^2(r_1) -\alpha(A) \alpha(C) f^2(r_2) -2\alpha(A) \alpha(B) \alpha(C) f(r_1) f(r_2) f(r_3) + O(r^{-12}) \tag{41}
\]

and

\[
\ln D_{x, y}(\xi) \sim -\alpha(A) \alpha(B) \{f^2(r_3) + \hbar^2(r_3)\} -\alpha(A) \alpha(C) \{f^2(r_1) + \hbar^2(r_1)\} -\alpha(A) \alpha(C) \{f^2(r_2) + \hbar^2(r_2)\} -2\alpha(A) \alpha(B) \alpha(C) \{2f_1 f_2 f_3 -f_1 f_2 g_3 -f_3 f_1 g_2 -f_2 f_3 g_1 +g_1 g_2 f_2 \cos^2 \theta_2 +g_1 g_2 f_3 \cos^2 \theta_3 +g_2 g_3 f_1 \cos^2 \theta_1 +g_1 g_2 g_3 \cos \theta_1 \cos \theta_2 \cos \theta_3\}. \tag{42}
\]

Substitution of the approximations (41) and (42) into the exact result (40) gives the energy as a sum of two-body interactions and a "three-body" potential. We use the relations

\[
\cos \theta_1 \cos \theta_2 \cos \theta_3 = \frac{1}{4} \{1 - \cos^2 \theta_1 - \cos^2 \theta_2 - \cos^2 \theta_3\} \tag{43a}
\]

and

\[
\cos^2 \theta_1 + \cos^2 \theta_2 + \cos^2 \theta_3 = 1 - 2 \cos \theta_1 \cos \theta_2 \cos \theta_3 \tag{43b}
\]

to carry out necessary algebraic manipulation. In the non-retarded limit, this three-body potential coincides with the well-known result given by Bell (1970)

\[
V_3(r_1, r_2, r_3) \sim -9\hbar \omega_0 (1 + 3 \cos \theta_1 \cos \theta_2 \cos \theta_3) / 16(r_1 r_2 r_3)^3.
\]
The important point, however, is that the whole energy of interaction, including retardation, is given exactly by the general expression (40). Extension of the method to include higher order multipole interactions is immediate. However, it can be shown that extension to include, for example, quadrupole interactions by perturbation theory is inconsistent, as the finite extent of the quadrupole must be taken into account. Such investigations are deferred to a later paper.

V. References