SINGLE-SITE ANISOTROPIC PAIR POTENTIAL BASED ON SPHERICAL HARMONIC FUNCTIONS

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TO MY PARENTS

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ABSTRACT

The main objective of this work is to model a less computationaly demanding flexible single-site anisotropic potential using generalized spherical harmonic functions. These functions have been used to expand the well depth, $\epsilon(r,\omega_1,\omega_2)$, and the range, $\sigma(r,\omega_1,\omega_2)$, parameters of the shifted Lennard-Jones (LJ) potential.

We have compared our potential with site-site LJ and single-site Gay-Berne (GB) potentials. The results are in better agreement with LJ site-site potential for all tested orientations. The computational time is comparable to that of GB potential.

The new potential has the additional advantage of flexibility. By taking additional spherical harmonic terms any desired accuracy can be obtained. The important virtues of this model is that it can also be used to model non-cylindrically symmetric molecules.

INTRODUCTION

Liquid crystal mesophases are formed by many substances which show strong shape anisotropy purpose of understanding Liquid Crystal. The development of simple, accurate pair potentials representative of these molecules is thus of tremendous value in order to evaluate their macroscopic properties[1]. The potential must be mathematically simple, involving only functions which are easy to calculate; it must not be time consuming.

Until recently, an accurate intermolecular potential for any system was just a gleam in the theorist's eye. The structure and dynamics of monatomic solids and liquids were calculated on the basis of simple, convenient, and reasonable pair potentials and often enough the results were surprisingly good; for example molecular dynamics studies have shown that the Lennard-Jones (12-6) (LJ) potential accounts quite adequately for argon. The application of the same model to polyatomic molecules has met with good results compared to that obtained experimentally, but the calculations are time consuming. Some approximate potentials to simulate cylindrically symmetric molecules have been developed such as the Gay-Berne[2] (GB) potential to overcome the time problem. However, it is not clear how to extend such potentials to molecules with non-cylindrical symmetry. Recently much work

has been done using spherical harmonics to represent shape anisotropy in MonteCarlo (MC) simulation of systems such as halogens. Spherical harmonics have proved a flexible and cost effective route to the simulation of such systems[3].

In the first chapter of this work we shall give a brief description of the concept of liquid crystal theories and general ideas about the main types of potential models used in liquid crystals modelling including their merits and flaws. The second chapter is focused on our potential (ZM) describing the general form of the potential, the method to determine S functions, computational details including evaluation of the coefficients of the strength and range parameters, and determination of ZM, LJ and GB potentials as functions of distance r. The third chapter includes results and discussion, while the fourth is the conclusion. At the end of this thesis Appendices contain some tables used in our calculations as well as the general references.

CHAPTER ONE

POTENTIAL MODELS

1.1. INTRODUCTION

A liquid crystal differs in many ways from the isotropic phase from which it is formed but the difference of prime importance is the extent of long-range orientational order while positional order is limited[4]. It is this difference which any molecular theory of liquid crystals should first explain. The development of such theories must begin with the formulation of the intermolecular potential and the identification of those features which are responsible for the transition from the liquid to the liquid crystal phase. The problem of calculating the thermodynamic observables of a system from a given intermolecular potential is complex. There are only two possibilities open: one is to use approximate theories[5,6,7,8,9,10,11,12,13,14,15,16,17] and the other is to resort to computer simulations[18,19,20,21,22,23].

1.2. TYPES OF MODEL POTENTIALS

Accurate theoretical potentials are few for polyatomic molecules[24]. Statistical Mechanical calculations are usually done with model potentials. The intermolecular potentials can

be broadly classified into three categories: Purely Hard Potentials, Purely Attractive Potentials and Mixed Type Potentials. The flow chart is given in fig 1.1.

1.2.1. PURELY HARD POTENTIALS

The simplest type of hard intermolecular potential is

$U_{ij} = \infty$, if i and j overlapped

$= 0$, otherwise. (1.1)

Onsager[25] used this potential to study right circular cylinders capped on both ends with half spheres. In this model the phase transition is induced purely by the density of the particles. As the result it is unsuitable for thermotropic systems where phase transition is induced by both density and temperature.

1.2.2. PURELY ATTRACTIVE POTENTIALS

The simplest type of purely attractive potential is the Lebwohl-Lasher potential defined as

$$U(\beta_{ij}) = -1/2\epsilon_{ij} (3\cos^2\beta_{ij} - 1) = -\epsilon_{ij}P_2(\cos\beta_{ij}) \quad (1.2)$$

where β_{ij} is the angle between the two molecular symmetry axes, $P_2(\cos\beta_{ij})$ is the second order Legendre Polynomial and ϵ_{ij} is the positive interaction strength parameter.

This potential model is extensively studied by mean-field theory[5,7,12] as well as computer simulation [12,18,19,21,22].

The advantages of this model are that its simplicity and ability to predict the spontaneous ordering of the nematic

phase from the isotropic phase. The main flaw of this model is that it is not able to distinguish between side-by-side and head-to-tail configurations.

1.2.3. MIXED TYPE POTENTIALS

1.2.3.1 Atom-Atom Lennard-Jones Potential[24,26,27,28,29]

The repulsion at short range is a consequence of the overlap of the electron clouds and it varies with distance[30].

One of the popular forms of mixed type potentials used to describe the potential between two molecules is the atom-atom potential. Here the intermolecular potential is assumed to be a sum of interatomic potential U_{ab} between the atoms which constitute the two molecules

$$U_{ab} = \Sigma_{ab}\, U_{ab}(r_{ab}) \qquad (1.3)$$

where r_{ab} is the separation between atoms a and b of molecules 1 and 2 respectively. The atomic potentials U_{ab} are usually taken as LJ (12,6) form

$$U_{ab} = 4\epsilon[(\sigma/r_{ab})^{12} - (\sigma/r_{ab})^{6}] \qquad (1.4)$$

where ϵ is the energy scaling parameter, σ is a distance scaling parameter, and r_{ab} is the distance between the two atoms a and b. The atom-atom potential is expected to model the effects of the short-range asymmetric repulsive core and the long-range asymmetric attractive force[31].

It has been demonstrated that the above model effectively reproduces the thermodynamic properties of liquid nitrogen and oxygen over broad ranges of temperature and density, as well

as it leads to accurate calculations of the second virial coefficient over a wide temperature range[32].

In computer simulations of aspherical molecules, a popular and effective representation of the short-range attractive and repulsive interaction consists of designating several sites in each molecule between which L-J 12-6 potentials act[2,33]. For larger molecules it may be necessary to employ many sites, typically one at each atom position, to represent the short-range interactions. So the time to evaluate the potential increases as the square of the number of sites. This leads to a computational inefficiency. This problem can be overcome by modelling site-site potential with a single-site anisotropic potential. The two models are given in fig 1.2.

1.2.3.2. Single-Site Model Potentials

Like the atom-atom models their principal virtue is in providing a reasonable description of the molecular shape. In these models a general expression of the type,

$$U(r) = \epsilon f(r/\sigma) \qquad (1.5)$$

where ϵ and σ are assumed to be dependent of the orientation of molecules, is used for the pair potential.

i) Corner's Single-Site Potential Model[29]

Corner proposed modelling an intermolecular potential $U(r\omega_1\omega_2)$ by a pseudo-atomic form of Eq 1.5.

Thus the LJ form of Eq 1.4. becomes

$$U(r\omega_1\omega_2) = 4\epsilon(\omega_1\omega_2\omega)[\{\sigma(\omega_1\omega_2\omega)/r\}^{12} - \sigma(\omega_1\omega_2\omega)/r\}^6] \quad (1.6)$$

where ω_1, ω_2 and ω denote the orientations of molecule 1, molecule 2, and the intermolecular vector r respectively. Corner considered a molecule with four LJ sites to determine the potential parameters in Eq 1.6. The LJ potential parameters for the site-site interactions are assumed to be ϵ_{ss} and σ_{ss}. By comparing the exact site-site potential with Eq 1.6 he was able to find approximate empirical expressions for $\epsilon(\omega_1\omega_2\omega)$ and $\sigma(\omega_1\omega_2\omega)$. These relations are

$$\epsilon(\omega_1\omega_2\omega) = \epsilon_o[1+(10/9L^{*2}-9/8L^*)(C_1'^2+C_2'^2)+9/8L^*S_1'C_1'S_2'C_2'C' - 1/7L^*S_1'^2S_2'^2C'^2]^2 \quad (1.7)$$

$$\sigma(\omega_1\omega_2\omega) = \sigma_o [1-2/5L^*+7/2L^*(C_1'^2+C_2'^2) + 21L^{*3}C_1'^2C_2'^2 - 14L^{*3}S_1'C_1'S_2'C_2'C' + 16L^{*3}(C_1'^2+C_2'^2)S_1'^2S_2'^2C'^2 - 25L^{*3}S_1'^2S_2'^2C_1'^2C_2'^2C'^2]^{1/3} \quad (1.8)$$

where $S_1' = \mathrm{Sin}\theta_1'$, $C_1' = \mathrm{Cos}\theta_1'$, $C' = \mathrm{Cos}(\varphi_1' - \varphi_2')$ [Fig 1.3], $\epsilon_o = 16\epsilon_{ss}$, $\sigma_o = \sigma_{ss}$ and $L^* = 2^{7/6}(L/\sigma_o)$ with L, the intermolecular distance between the first and fourth sites. The above expressions are valid for $L^* \leq 1.5$, and are not the simplest that could be found from the numerical fitting. Also it is valid only for prolate molecules of restricted anisotropy.

ii) Kihara Core Model[34]

In this model the molecules are considered as convex hard cores (eg. spherocylinders, ellipsoids, etc). The pair

potential $U(r\omega_1\omega_2)$ is assumed to be a function of S where $S = S(r\omega_1\omega_2)$ is the shortest distance between the cores. The potential is given by

$$U(r\omega_1\omega_2) = 4\epsilon_o[\{\sigma_o/S(r\omega_1\omega_2)\}^{12} - \{\sigma_o/S(r\omega_1\omega_2)\}^{6}] \quad (1.9)$$

where ϵ_o and σ_o are angle independent parameters. The difficulty inherent in this model is that the calculation of the distance S is difficult and time consuming and also ϵ is wrongly taken to be independent of orientation.

iii) Gaussian Overlap Models

a) General Gaussian Models[29,35,36,37]

An axially symmetric molecule whose charge density is $\rho(r)$ is assumed to be Gaussian, where

$$\rho(r) \sim \exp[-(x^2+y^2)/\sigma_.^2 - z^2/\sigma_{|}^2] \quad (1.10)$$

Where x, y, and z refer to the principal axes, z being the symmetry axis. The contour surfaces of ρ are ellipsoids of revolution about z axis, with $\sigma_.$ and $\sigma_{|}$ the diameters of the distribution perpendicular and parallel to the symmetry axis, respectively. If two such distributions separated by r and having orientations ω_1 and ω_2 interact, the coulomb energy is rigorously given by

$$J(r\omega_1\omega_2) = \int dr_1 dr_2 \rho_1(r_1)\rho_2(r_2)|r_1-r_2|^{-1} \quad (1.11)$$

It is assumed that at short-range where the distribution overlap slightly, U is approximately proportional to the overlap volume integral of the two distributions

$$J(r\omega_1\omega_2) \sim \int dr_1 \rho_1(r_1)\rho_2|r_1-r|. \quad (1.12)$$

This integral can be evaluated exactly and has a Gaussian form

$$U(r\omega_1\omega_2) = \epsilon(\omega_1\omega_2\omega)\exp[-r^2/\sigma(\omega_1\omega_2\omega)^2] \quad (1.13)$$

where $\epsilon(\omega_1\omega_2\omega)$ and $\sigma(\omega_1\omega_2\omega)$ are angle-dependent strength and range parameters given by

$$\epsilon(\omega_1,\omega_2,\omega) = \epsilon_o[1-\chi^2C_{12}^2]^{-1/2} \quad (1.14)$$

and

$$\sigma(\omega_1,\omega_2,\omega) = \sigma_o[1-\chi^2C_{12}^2]^{1/2}((1-\chi(C_{12}^2+C_2^2)+\chi^2(2C_1C_2C_{12}-C_{12}^2))^{-1/2} \quad (1.15)$$

where

$$\chi = (\sigma_l^2-\sigma_s^2)/(\sigma_l^2+\sigma_s^2) \quad (1.16)$$

is anisotropic parameter, ϵ_o and σ_o are constants and σ_l and σ_s are major and minor axes. If U_1,U_2 and R are unit vectors along ω_1, ω_2 and ω respectively, then the three cosines in Eqs 1.14 and 1.15 are given by:

$C_{12} = U_1.U_2$, $\quad C_1 = R.U_1$ $\quad$ and $\quad C_2=R.U_2$

In this model, $\epsilon(\omega_1,\omega_2,\omega)$ is independent of ω, the intermolecular vector orientation. It is a maximum when the molecules are parallel and a minimum when they are perpendicular. Although it is valid for oblate as well as prolate shapes of arbitrary anisotropy, the r dependence in Eq 1.13 is not realistic. Therefore it is replaced by the more realistic LJ form in Berne-Pechukas and Gay-Berne models.

b) Berne and Pechukas Model[2,35]

Berne and Pechukas have proposed a Gaussian Overlap Model in which the overlap between two ellipsoidal charge distributions is first calculated. They suggested that the

functional forms $\epsilon(\Omega)$ and $\sigma(\Omega)$ may give a reasonable description of the well depth and Van der Waals diameter as functions of orientation so that the potential could be written in the form

$$V(U_1,U_2,R)=4\epsilon(U_1,U_2)\{[\sigma(U_1,U_2,R)/r]^{12}-[\sigma(U_1,U_2,R)/r]^{6}\} \quad (1.17)$$

where

$$\epsilon(U_1,U_2) = \epsilon_0[1-\chi^2(U_1,U_2)^2]^{-1/2} \quad (1.18)$$

and

$$\sigma(U_1,U_2,R)=\sigma_0[1-1/2\chi\{(R.U_1+R.U_2)^2/(1+\chi(U_1.U_2) + (R.U_1-R.U_2)/(1-\chi(U_1.U_2)\}]^{-1/2} \quad (1.19)$$

are called strength and range parameters respectively. The anisotropy parameter χ is determined by the parameters of the ellipsoidal Gaussians as given by Eq 1.16.

This potential has the following unrealistic features

(1) For parallel molecules, the well depth is independent of their orientation with respect to the intermolecular vector, whereas in reality the well depths for these configurations should be significantly different for elongated mesogenic molecules.

(2) The width of the attractive well is found to vary with respect to the intermolecular vector.

c) <u>Tsykalo and Bagment Model</u>[38]

Tsykalo and Bagment proposed that the pair potential should be scaled with a function which depended on the orientation of the particles with respect to their

intermolecular vector. They redefined the strength parameter as

$$\epsilon(U_1,U_2,R) = \epsilon(U_1,U_2)\sigma_o^2/\sigma^2(U_1,U_2,R) \qquad (1.20)$$

However the anisotropy in the strength parameter is then determined by shape anisotropy via χ which may not be appropriate. In addition this class of potential has another unrealistic feature in that the width of attractive well varies with the orientation of the particles with respect to the intermolecular vector although the width should be essentially independent of this orientation.

d) Gay and Berne Model[2,39]

As we have seen before many attempts have been undertaken to model site-site potential with single-site potential.

To rectify the deficiencies of the Berne-Pechukas potential mentioned above Gay and Berne have modified the original Gaussian Overlap Potential in an essentially phenomenological manner. Thus they attempted to obtain a function which gave the best fit to the pair potential for a linear array of four equidistant Lennard-Jones centres with a separation of $2\sigma_o$ between the first and fourth sites [Fig 1.4]. On the basis of such modelling they have proposed a new functional form for the strength parameter, namely

$$\epsilon(U_1,U_2,R) = \epsilon'(U_1,U_2)\epsilon'^{\mu}(U_1,U_2,R) \qquad (1.21)$$

where

$$\epsilon'(U_1,U_2,R) = 1-\chi'/2\{(U_1.R+U_2.R)^2/(1+\chi' U_1.U_2) + (U_1.R-U_2.R)^2/(1-\chi' U_1.U_2)\} \quad (1.22)$$

which is the function of **R** as well as U_1 and U_2.

The new parameter χ' is used to adjust the ratio of side-by-side to end-to-end well depths and reflects the anisotropy,

$$\chi' = \{(\epsilon_1/\epsilon_s)^{1/\mu}-1\}/\{(\epsilon_1/\epsilon_s)^{1/\mu}+1\} \quad (1.23)$$

To overcome the problem associated with the width of the attractive well, the anisotropic range and strength parameters were used in the shifted potential

$$V(U_1,U_2,R)=\epsilon(U_1,U_2,R)\{[r-\sigma(U_1,U_2,R)+1]^{-12}-[r-\sigma(U_1,U_2,R+1]^{6}]\} \quad (1.24)$$

rather than the Lennard-Jones scaled potential of Eq 1.4, where $\sigma(U_1,U_2,R)$ is given by Eq 1.19.

Gay and Berne considered a linear array of four equidistant LJ sites per molecule to determine their potential parameters. The best fit to LJ potential gave μ= 1, ν= 2 and ϵ_e/ϵ_s = 0.2.

The ability of particles interacting via the Gay Bern potential to exhibit liquid-crystalline behaviour has been demonstrated using MD simulation. The system exhibits a nematic phase with long-range orientational order and just short-range translational order[39].

The disadvantages of GB potential are that it is complicated, contains many terms with different parameters, and there is no obvious way to extend it to describe general shape molecules.

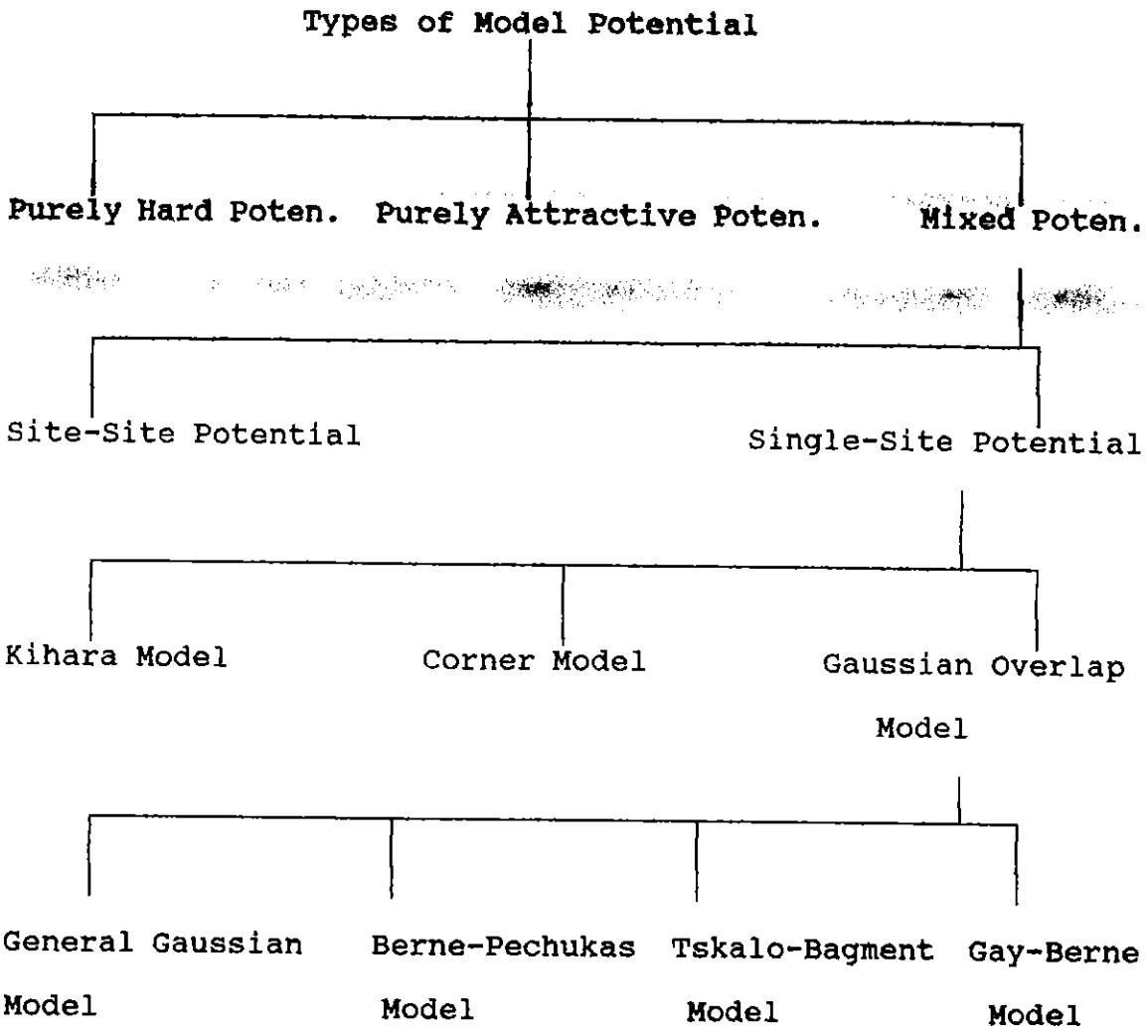
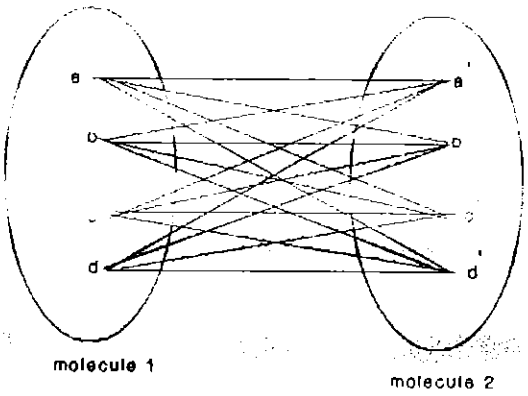


Fig. 1.1. Flow chart showing different types of model potential.

a)



b)

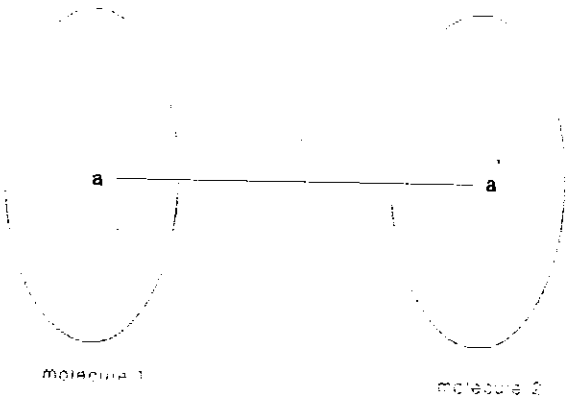


Fig. 1.2. Two types of potential model:
a) site-site and b) single-site

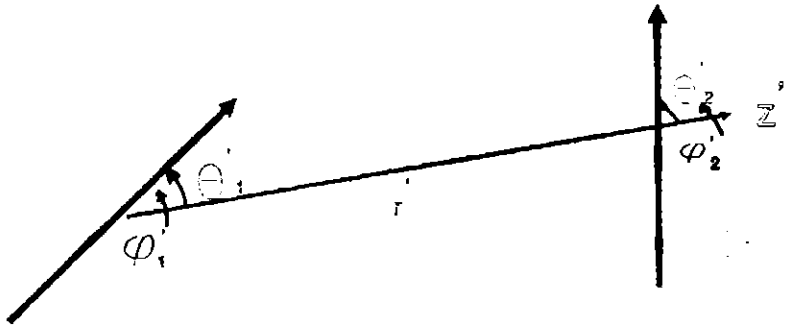


Fig. 1.3. The intermolecular axis coordinate system (the 'r-frame'), with polar Z- axis along r.

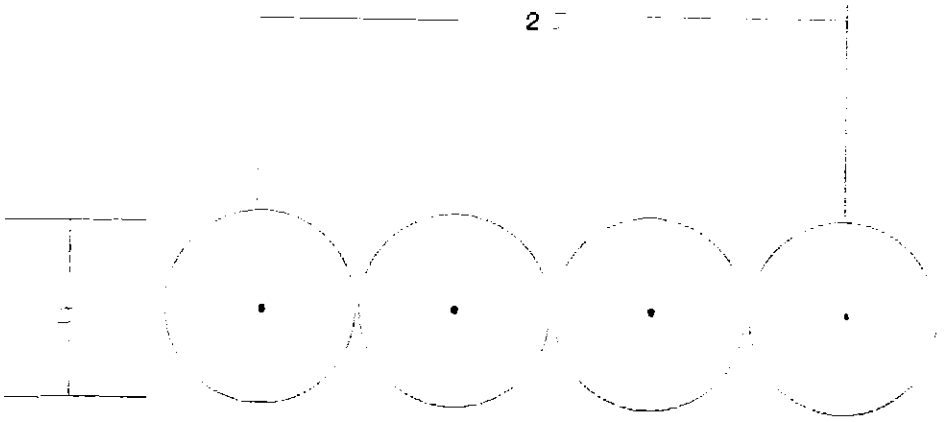


Fig. 1.4. A four-site molecule. LJ potentials with parameter □ act between sites on neighbouring molecules which are located on the axes of the molecules 2□/3 apart

CHAPTER TWO

SINGLE-SITE ANISOTROPIC PAIR POTENTIAL BASED ON SPHERICAL HARMONIC EXPANSION

2.1. INTRODUCTION

Our objective is to develop a general single-site anisotropic pair potential that can be used for efficient computer simulation.

Our model is based on the shifted form of the Lennard-Jones (LJ) potential given by

$$V(\mathbf{r},\Omega_1,\Omega_2)=4\epsilon(\mathbf{r},\Omega_1,\Omega_2)\{[r-\sigma(\mathbf{r},\Omega_1,\Omega_2)+1]^{-12} - [r-\sigma(\mathbf{r},\Omega_1,\Omega_2)+1]^{-6}\} \quad (2.1)$$

Where the first term of the right hand side represents the anisotropic repulsion and the second term represents anisotropic dispersion interaction between molecules 1 and 2. $\mathbf{r}$ is the intermolecular unit vector from molecule 1 to molecule 2 and r is the distance between their centres. $\sigma(\mathbf{r},\Omega_1,\Omega_2)$ is the intermolecular separation for a given molecular and intermolecular vector orientation where the intermolecular potential is zero and $\epsilon(\mathbf{r},\Omega_1,\Omega_2)$ is the potential well depth for the given molecular and intermolecular orientations. The well depth, ϵ, and the range parameters, σ, are determined as reduced units with respect to ϵ_o and σ_o.

Using the Gaussian overlap model Berne and Pechukas and

Gay and Berne proposed particular forms for the functions σ and ϵ. The main advantage of the Gaussian overlap model is that it is a simple physical model. The disadvantages are that it is unlikely to represent the true potential very accurately and that there is no obvious way to improve it, or to modify it to describe non-cylindrically symmetric molecules.

In our model the range parameter which is the separation between molecules at zero intermolecular potential, $\sigma(r,\Omega_1,\Omega_2)$, and the energy scaling parameter which is the potential depth, $\epsilon(r,\Omega_1,\Omega_2)$, for two arbitrary shape molecules are considered to be functions of intermolecular vector orientation r as well as the intermolecular orientations Ω_1 and Ω_2. These parameters are expanded in the flexible generalized spherical harmonics which are functions of the orientations of the two molecules (Ω_1,Ω_2) and that of the intermolecular vector (ω), as

$$\sigma(r,\Omega_1,\Omega_2)= \sum_{\ell_1\ell_2\ell} \sum_{m_1,m_2,m} \sum_{k_1,k_2} \sigma_{\ell_1\ell_2\ell}^{k_1k_2} S_{\ell_1\ell_2\ell}^{k_1k_2}(\Omega) \qquad (2.2)$$

and

$$\epsilon(r,\Omega_1,\Omega_2)= \sum_{\ell_1\ell_2\ell} \sum_{m_1,m_2,m} \sum_{k_1,k_2} \epsilon_{\ell_1\ell_2\ell}^{k_1k_2} S_{\ell_1\ell_2\ell}^{k_1k_2}(\Omega) \qquad (2.3)$$

where $\Omega \equiv (\Omega_1,\Omega_2,\omega)$.

Following the convention of A. J. Stone[30], the generalized spherical harmonic is expressed as

$$S_{\ell_1\ell_2\ell}^{k_1k_2}(\Omega) = \sum C(\ell_1\ell_2\ell;m_1m_2m)D^{\ell_1}_{m_1k_1}(\Omega_1)D^{\ell_2}_{m_2k_2}(\Omega_2)Y_{\ell m}^*(\omega) \qquad (2.4)$$

where

$\Omega_1 \equiv (\alpha_1,\beta_1,\gamma_1)$ describes the orientation of molecule 1 with respect to an arbitrary space-fixed axis system.

$\Omega_2 \equiv (\alpha_2, \beta_2, \gamma_2)$ describes the orientation of molecule 2. $\omega \equiv (\theta, \varphi)$ (polar angles) describes the direction of the intermolecular vector, r, defined from the origin of molecule 1 to the origin of molecule 2. The coordinate system is shown in Fig 2.1. $Y_{\ell m}^*(\theta,\varphi)$ is spherical harmonic and $D^{\ell}_{mk}(\alpha,\beta,\gamma)$ is the Wigner Rotation Matrix or generalized spherical harmonic[40]. Some properties of Wigner Rotation Matrices and related functions are given in Appendix A1.

$C(\ell_1\ell_2\ell;m_1m_2m)$ is a Clebsch-Gordan Coefficient (CGC) given by

$$C(\ell_1\ell_2\ell;m_1m_2m) = \delta_{m,m1+m2}((2\ell+1)(\ell_1+\ell_2-\ell)!(\ell_1-\ell_2+\ell)!(-\ell_1+\ell_2+\ell)!)^{1/2}$$
$$\times((\ell_1+m_1)!(\ell_1-m_1)!(\ell_2+m_2)!(\ell_2-m_2)!(\ell+m)!(\ell-m)!)^{1/2}$$
$$\times\sum_z \frac{(-1)^z}{z!(\ell_1+\ell_2-\ell-z)!(\ell_1-m_1-z)!(\ell_2+m_2-z)!}$$
$$\times(\ell-\ell_2+m_1+z)!(\ell-\ell_1-m_2+z)! \qquad (2.5)$$

Where z runs over those positive integer values such that none of the factorials in the denominator is negative. The last two factorials in Eq 2.5 are in the denominator[20]. Some values of CGC are calculated and listed in Table A2.2. ℓ_1, ℓ_2, and ℓ are ranks of vectors on molecules 1, 2 and that joining their centres respectively.

In the language of vector spaces, the direct product space $\ell_1 \otimes \ell_2$ is invariant under rotations. It is, however, reducible (contains invariant subspaces), and it can be decomposed into a direct sum of irreducible subspaces. The general result is that $\ell_1 \otimes \ell_2$ contains irreducible parts

$\ell=\ell_1+\ell_2,\ \ell_1+\ell_2-1,\ldots\ldots,|\ell_1-\ell_2|$ (which can be written symbolically as $(\ell_1+\ell_2)\oplus(\ell_1+\ell_2-1)\oplus\ldots.$). Also CGC = 0 unless $m = m_1+m_2$ These two results are summarized in the selection rules; $C(\ell_1\ell_2\ell;m_1m_2m) = 0$ unless

1) $$|\ell_1-\ell_2| \le \ell \le |\ell_1+\ell_2| \qquad (2.6)$$

2) $$m = m_1+m_2 \qquad (2.7)$$

For a given rank of ℓ, m has the values of

$$m = +\ell,\ell-1,\ell-2,..,-\ell \qquad (2.8)$$

For integer ℓ and m CGC has the following properties:

$$C(\ell_1\ell_2\ell;m_1m_2m) = (-)^{\ell1+\ell2+\ell} C(\ell_1\ell_2\ell;-m_1,-m_2,-m) \qquad (2.9.a)$$

$$= (-)^{\ell1+\ell2+\ell} C(\ell_1\ell_2\ell;m_2,m_1,m) \qquad (2.9.b)$$

$$= (-)^{\ell1+m1}\left\{\frac{2\ell+1}{2\ell_2+1}\right\}^{1/2} C(\ell_1\ell\ell_2;-m_1,-m,-m_2) \qquad (2.9.c)$$

$$= (-)^{\ell2+m2}\left\{\frac{\ell+1}{2\ell_1+1}\right\}^{1/2} C(\ell\ell_2\ell_1;-m,-m_2,-m_1) \qquad (2.9.d)$$

$$= (-)^{\ell1-m1}\left\{\frac{2\ell+1}{2\ell_2+1}\right\}^{1/2} C(\ell\ell_1\ell_2;m,-m_1,m_2) \qquad (2.9.e)$$

$$= (-)^{\ell2+m2}\left\{\frac{2\ell+1}{2\ell_1+1}\right\}^{1/2} C(\ell_2\ell\ell_1;-m_2,m,m_1). \qquad (2.9.f)$$

For linear molecules all terms with non-zero k_1 or k_2 vanish and $\ell_1+\ell_2+\ell$ must be even; for linear centrosymmetric molecules there is the further restriction that ℓ_1 and ℓ_2 must both be even.

Thus due to the centrosymmetric restriction imposed on our model, the ellipsoidal model, ℓ_1, ℓ_2 and ℓ are even.

If the molecule has a C_n symmetry-axis, such a rotation

must leave the energy unchanged, so terms involving $D_{m,k}$' can only appear if k = 0. In particular, it has been shown that for linear molecules, $k_1 = k_2 = 0$, hence simpler functions can be used. Thus Eqs 2.2 and 2.3 for molecules with cylindrical symmetry reduce to

$$\sigma(r\omega_1\omega_2) = \sum_{\ell_1\ell_2\ell}\sum_{m_1m_2m} \sigma_{\ell_1\ell_2\ell}\, S_{\ell_1\ell_2\ell}(\omega_{12}) \qquad (2.10)$$

and

$$\epsilon(r\omega_1\omega_2) = \sum_{\ell_1\ell_2\ell}\sum_{m_1m_2m} \epsilon_{\ell_1\ell_2\ell}\, S_{\ell_1\ell_2\ell}(\omega_{12}) \qquad (2.11)$$

where

$$S_{\ell_1\ell_2\ell}(\omega_{12}) = \sum_{m_1m_2m} C(\ell_1\ell_2\ell; m_1m_2m)\, Y_{\ell_1m_1}(\omega_1)\, Y_{\ell_2m_2}(\omega_2)\, Y_{\ell m}^*(\omega) \qquad (2.12)$$

$\omega_{12} \equiv (\omega_1, \omega_2, \omega)$, $\omega_1 \equiv (\beta_1, \alpha_1)$ and $\omega_2 \equiv (\beta_2, \alpha_2)$

Some expressions for $Y_{\ell m}(\beta\alpha)$ are determined and given in Appendix A2, Table A2.1.

Here we shall restrict the expansion to the lowest order terms of ℓ_1 and ℓ_2. Thus restricting the values of ℓ_1 and ℓ_2 to be 0 and 2 the resulting values of ℓ can be determined from Eq 2.6 as tabulated below

ℓ_1	0	2	0	2
ℓ_2	0	0	2	2
ℓ	0	2	2	0,2,4

hence the surviving terms of $S_{\ell_1\ell_2\ell}$ are

S_{000}, S_{202}, S_{022}, S_{220}, S_{222} and S_{224}

These functions are determined and summarized in Table 2.1. The details of the determination of these functions is provided in Appendix A3.

Using the various assumptions and approximations given above the expansions of Eqs 2.10 and 2.11 reduce to

$$\sigma(r,\omega_1,\omega_2) = \sigma_{000}\, S_{000}(\omega_{12}) + \sigma_{202}\, S_{202}(\omega_{12}) + \sigma_{022}\, S_{022}(\omega_{12}) + \sigma_{220}\, S_{220}(\omega_{12}) + \sigma_{222}\, S_{222}(\omega_{12}) + \sigma_{224}\, S_{224}(\omega_{12}) \quad (2.13)$$

and

$$\epsilon(r,\omega_1,\omega_2) = \epsilon_{000}\, S_{000}(\omega_{12}) + \epsilon_{202}\, S_{202}(\omega_{12}) + \epsilon_{022}\, S_{022}(\omega_{12}) + \epsilon_{220}\, S_{220}(\omega_{12}) + \epsilon_{222}\, S_{222}(\omega_{12}) + \epsilon_{224}\, S_{224}(\omega_{12}). \quad (2.14)$$

Assuming that both interacting molecules are identical leads to $\sigma_{202} = \sigma_{022}$ and $\epsilon_{202} = \epsilon_{022}$. Thus Eqs 2.13 and 2.14 become

$$\sigma(r,\omega_1,\omega_2) = \sigma_{000}\, S_{000}(\omega_{12}) + \sigma_{022}\, \{S_{202}(\omega_{12}) + S_{022}(\omega_{12})\} + \sigma_{220}\, S_{220}(\omega_{12}) + \sigma_{222}\, S_{222}(\omega_{12}) + \sigma_{224}\, S_{224}(\omega_{12}) \quad (2.15)$$

and

$$\epsilon(r,\omega_1,\omega_2) = \epsilon_{000}\, S_{000}(\omega_{12}) + \epsilon_{022}\, \{S_{202}(\omega_{12}) + S_{022}(\omega_{12})\} + \epsilon_{220}\, S_{220}(\omega_{12}) + \epsilon_{222}\, S_{222}(\omega_{12}) + \epsilon_{224}\, S_{224}(\omega_{12}) \quad (2.16)$$

It is clear, from the last two equations, that there are two sets of expansion coefficients. Five for the range parameter, σ_{000}, σ_{022}, σ_{220}, σ_{222} and σ_{224}, and five for the well depth, ϵ_{000}, ϵ_{022}, ϵ_{220}, ϵ_{222} and ϵ_{224}.

In order to determine these coefficients, the range and strength parameters of our potential are fit to those determined from the LJ site-site potential for carefully chosen crucial configurations. The complete procedure is explained in the computational details.

2.2. COMPUTATIONAL DETAILS

The coefficients in Eqs 2.15 and 2.16 are estimated according to the following procedure. A flow chart is presented in Fig 2.2.

A four-site molecule described in Figure 1.4 is considered for determining the expansion coefficients in our potential. LJ potential with range parameter σ and strength parameter ϵ act between sites on neighbouring molecules which are located on the axes of the molecules $2\sigma/3$ apart is used.

Four crucial configurations are identified for these molecules, viz, side-by-side (S-S), head-to-tail (H-T), Tee (T), and cross (X) configurations. These configurations are given in Fig 2.3. For these four configurations the LJ potentials as functions of intermolecular separations are calculated. The potential well depth, ϵ_{min}, and the intermolecular separations at zero potential, σ_{zero}, are determined for these crucial configurations. These results are used to calculate the strength, ϵ_{sizes}, and range, σ_{sizes}, parameter coefficients of our potential. Minimization routine is used to determine these coefficients[41].

Generally to determine the strength coefficients, for example, the following algorithm is used. Let $\epsilon_i = \epsilon_{min}^i$ be the reduced minimum potential well depth for the i^{th} configuration determined for LJ potential. Where ϵ_{min}^i is the LJ potential well depth of Eq 1.4 for the i^{th} configuration. The corresponding potential well depth, ϵ_i', for our model is

given by Eq 2.16 as

$$\epsilon_i' = \epsilon_{000}S_{000} + \epsilon_{202}\,(S_{202}+S_{022}) + \epsilon_{220}S_{220} + \epsilon_{222}\,S_{222} + \epsilon_{224}\,S_{224}.$$

For the identified crucial k configurations the spherical harmonic functions are determined and substituted in the above equation. An error function is defined as

$$f = \Sigma_{i=1}^{k}(\epsilon_i - \epsilon_i')^2 \qquad (2.17)$$

Where f is a function of the five coefficients

$$f = f(\epsilon_{000},\ \epsilon_{022},\ \epsilon_{220},\ \epsilon_{222},\ \epsilon_{224}).$$

These coefficients are determined as those that can minimize this error function using the conjugate gradient method.

Similarly the range coefficients of Eq 2.15 are determined as follows: the separation at zero potential, $\sigma_i = \sigma_{zero}^i$, is determined for the i^{th} configuration using LJ potential. Where σ_{zero}^i is the reduced LJ range scaling parameter of Eq 1.4 at zero potential for the given i^{th} configuration. The corresponding separation, σ_i', for our single-site potential is expressed as

$$\sigma_i' = \sigma_{000}S_{000} + \sigma_{022}(S_{202}+S_{022}) + \sigma_{220}S_{220} + \sigma_{222}S_{222} + \sigma_{224}S_{224}$$

The spherical harmonic functions are obtained for the identified crucial k configurations. An error function f is given by

$$f = \Sigma_{i=1}^{k}(\sigma_i - \sigma_i')^2 \qquad (2.18)$$

Where f is a function of the five coefficients,

$$f = f(\sigma_{000},\ \sigma_{022},\ \sigma_{220},\ \sigma_{222},\ \sigma_{224}).$$

The coefficients chosen are those which minimize this error function using the conjugate gradient method.

The strength and range parameters determined using LJ site-site potential are given in Table 3.1. The two sets of parameter coefficients determined as those which gave the minimum errors of Eqs 2.17 and 2.18 are listed in Table 3.2.

We have compared our potential with the Site-Site LJ and the Single-Site GB potentials. The results are presented in the following section.

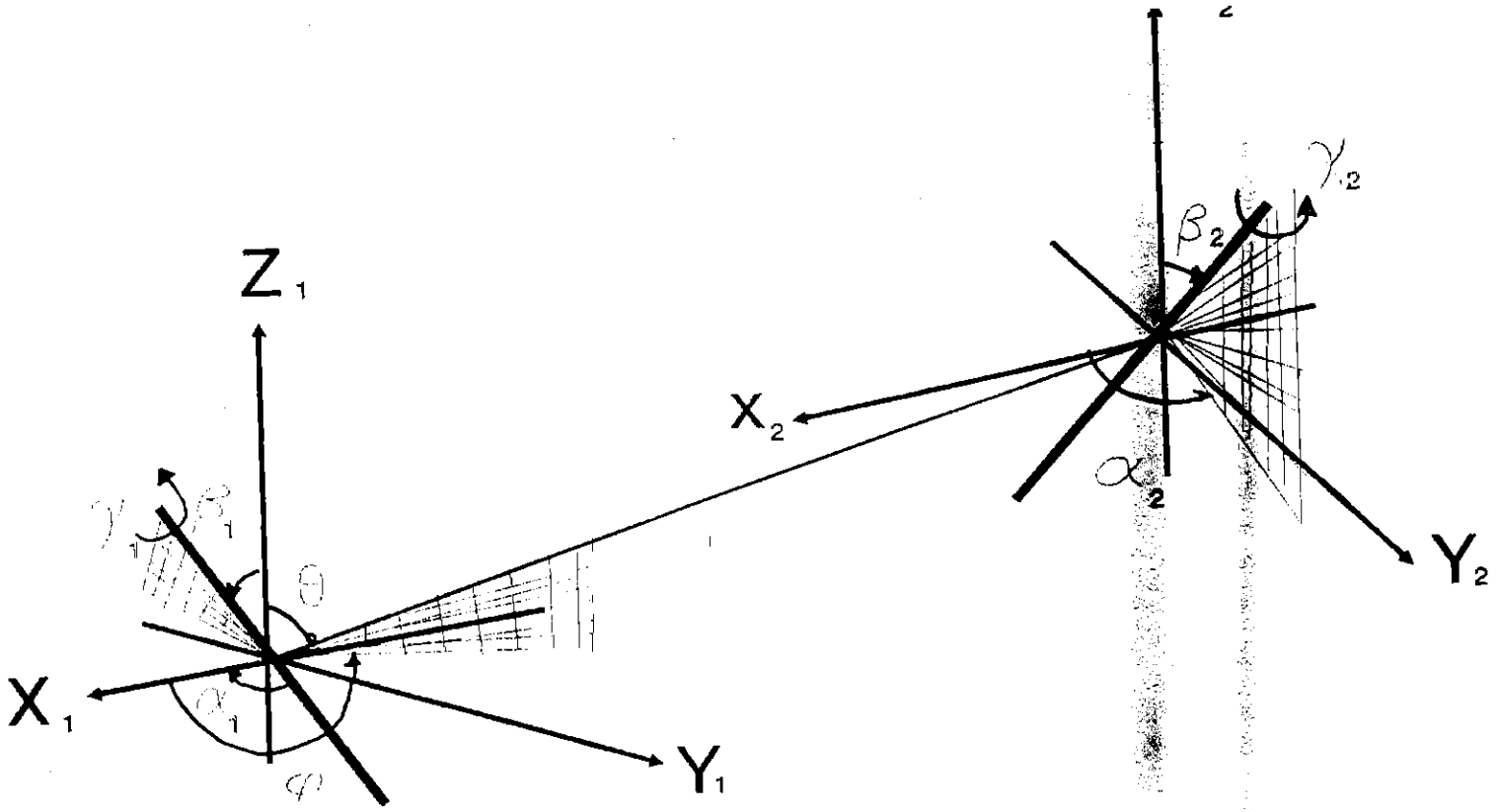


Fig. 2.1. Angular variables used to describe the orientation of two molecules relative to the laboratory frame with the director as Z- axis

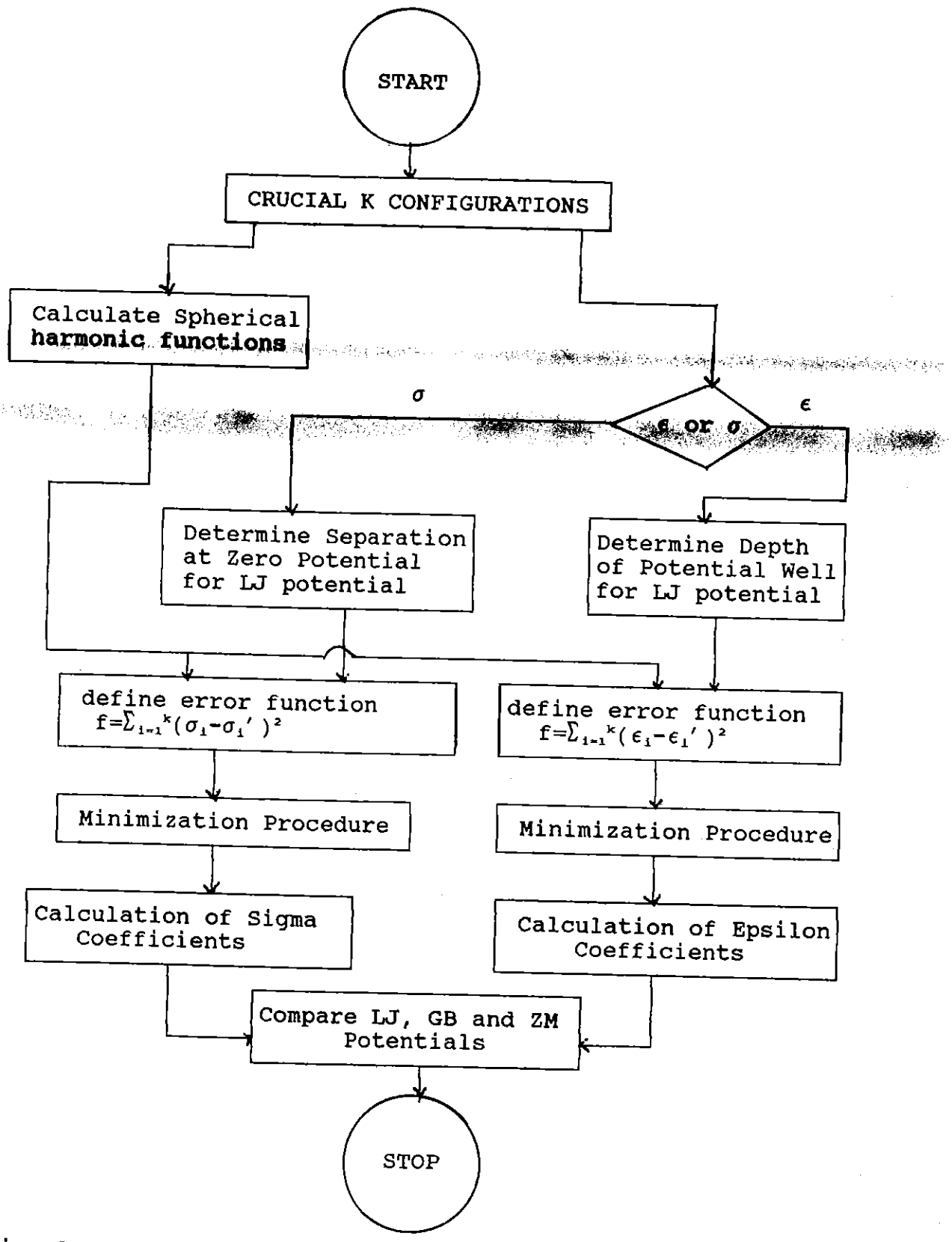


Fig. 2.2. Flow Chart showing computational details.

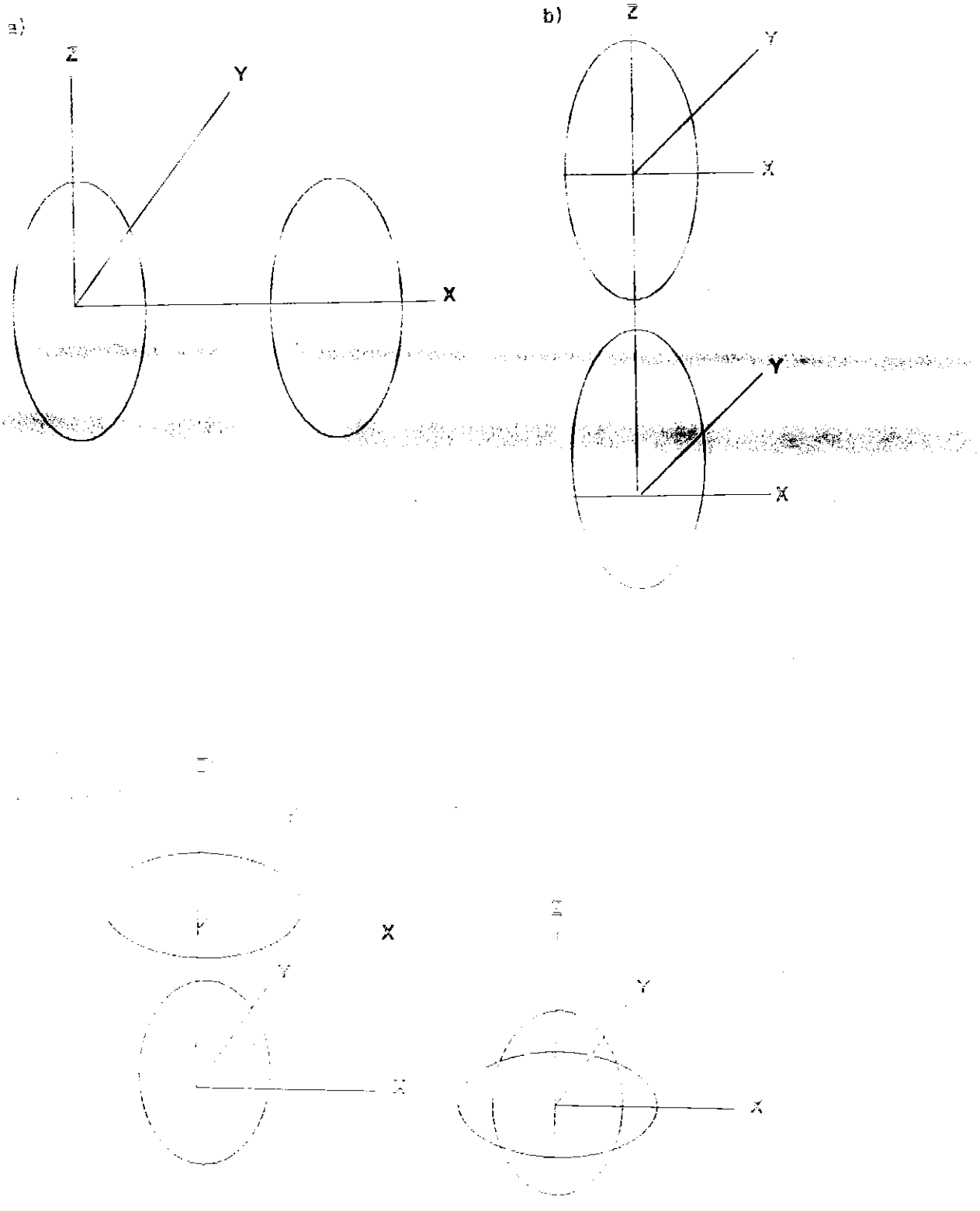


Fig. 2.3. Four crucial configurations; a) side-by-side
b) head-to-tail, c) Tee (T) and d) Cross (X) configurations

Table 2.1 S functions $S_{\ell_1\ell_2\ell}$ for some values of ℓ_1, ℓ_2 and ℓ (further functions are tabulated in Appendix A3).

Here the following designations are used

$$X \equiv \cos\theta, \quad X_1 \equiv \cos\beta_1, \quad \text{and} \quad X_2 \equiv \cos\beta_2$$

$$S_{000} = (1/4\pi)^{3/2} \tag{2.19}$$

$$\begin{aligned} S_{202} = {} & C(202,202)k_{22}^2\, k_{00}P_{22}(X_1)P_{00}(X_2)P_{22}(X)\{2\cos(2\alpha_1-2\varphi)\} \\ & + \\ & C(202,101)k_{21}^2\, k_{00}P_{21}(X_1)P_{00}(X_2)P_{21}(X)\{2\cos(\alpha_1-\varphi)\} \\ & + \\ & C(202;0,0,0)k_{20}^2\, k_{00}P_{20}(X_1)P_{00}(X_2)P_{20}(X) \end{aligned} \tag{2.20}$$

$$\begin{aligned} S_{022} = {} & C(022,022)k_{22}^2\, k_{00}P_{00}(X_1)P_{22}(X_2)P_{22}(X)\{2\cos(2\alpha_2-2\varphi)\} \\ & + \\ & C(022,011)k_{21}^2\, k_{00}P_{00}(X_1)P_{21}(X_2)P_{21}(X)\{2\cos(\alpha_2-\varphi)\} \\ & + \\ & C(022;0,0,0)k_{20}^2\, k_{00}P_{00}(X_1)P_{20}(X_2)P_{20}(X) \end{aligned} \tag{2.21}$$

$$\begin{aligned} S_{220} = {} & C(220,2{-}20)k_{22}^2\, k_{00}P_{22}(X_1)P_{22}(X_2)P_{00}(X)\{2\cos(-2\alpha_2+2\alpha_1)\} \\ & + \\ & C(220,1{-}10)k_{21}^2\, k_{00}P_{21}(X_1)P_{12}(X_2)P_{00}(X)\{2\cos(-\alpha_2+\alpha_1)\} \\ & + \\ & C(220;0,0,0)k_{20}^2\, k_{00}P_{20}(X_1)P_{20}(X_2)P_{00}(X) \end{aligned} \tag{2.22}$$

$$
\begin{aligned}
S_{222} = \; & C(222,202)k_{22}^2\, k_{20}P_{22}(X_1)P_{20}(X_2)P_{22}(X)\{2\cos(2\alpha_1-2\varphi)\} \\
& + \\
& C(222,022)k_{22}^2\, k_{20}P_{20}(X_1)P_{22}(X_2)P_{22}(X)\{2\cos(2\alpha_2-2\varphi)\} \\
& + \\
& C(222,112)k_{21}^2\, k_{22}P_{21}(X_1)P_{12}(X_2)P_{22}(X)\{2\cos(\alpha_2+\alpha_{1-2\varphi})\} \\
& + \\
& C(222,2\text{-}20)k_{22}^2\, k_{20}P_{22}(X_1)P_{22}(X_2)P_{20}(X)\{2\cos(-2\alpha_2+2\alpha_1)\} \\
& + \\
& C(222,1\text{-}10)k_{21}^2\, k_{20}P_{21}(X_1)P_{12}(X_2)P_{20}(X)\{2\cos(-\alpha_2+\alpha_1)\} \\
& + \\
& C(222;0,0,0)k_{20}^3\, P_{20}(X_1)P_{20}(X_2)P_{20}(X) \\
& + \\
& -C(222,2\text{-}11)k_{21}^2\, k_{22}P_{22}(X_1)P_{21}(X_2)P_{21}(X)\{2\cos(2\alpha_1-\alpha_2-\varphi)\} \\
& + \\
& -C(222,-121)k_{21}^2\, k_{22}P_{22}(X_1)P_{22}(X_2)P_{21}(X)\{2\cos(2\alpha_2-\alpha_1-\varphi)\} \\
& + \\
& C(222,101)k_{21}^2\, k_{20}P_{21}(X_1)P_{20}(X_2)P_{21}(X)\{2\cos(\alpha_1-\varphi)\} \\
& + \\
& C(222,011)k_{21}^2 k_{20}P_{20}(X_1)P_{21}(X_2)P_{21}(X)\{2\cos(\alpha_2-\varphi)\} \qquad (2.23)
\end{aligned}
$$

$$
\begin{aligned}
S_{224} = \; & C(224,224)k_{22}^2\, k_{44}P_{22}(X_1)P_{22}(X_2)P_{24}(X)\{2\cos(2\alpha_1+2\alpha_2-4\varphi) \\
& + \\
& C(224,213)k_{22}k_{21}k_{43}P_{22}(X_1)P_{21}(X_2)P_{43}(X)\{2\cos(2\alpha_1+\alpha_2-3\varphi)\} \\
& + \\
& C(224,213)k_{21}k_{22}k_{43}P_{21}(X_1)P_{22}(X_2)P_{43}(X)\{2\cos(\alpha_1+2\alpha_2-3\varphi)\} \\
& + \\
& C(224,202)k_{22}k_{20}k_{42}P_{22}(X_1)P_{20}(X_2)P_{42}(X)\{2\cos(2\alpha_1-2\varphi)\} \\
& + \\
& C(224,022)k_{20}k_{22}k_{42}P_{20}(X_1)P_{22}(X_2)P_{42}(X)\{2\cos(2\alpha_2-2\varphi)\} \\
& + \\
& C(224,112)k_{21}^2\, k_{42}P_{21}(X_1)P_{21}(X_2)P_{42}(X)\{2\cos(\alpha_2+\alpha_1-2\varphi)\} \\
& + \\
& -C(224,2\text{-}11)k_{21}k_{22}k_{41}P_{22}(X_1)P_{21}(X_2)P_{41}(X)\{2\cos(2\alpha_1-\alpha_2-\varphi)\} \\
& + \\
& C(224,101)k_{21}k_{20}k_{41}P_{21}(X_1)P_{20}(X_2)P_{41}(X)\{2\cos(\alpha_1-\varphi)\} \\
& + \\
& -C(224,-121)k_{21}k_{22}k_{41}P_{21}(X_1)P_{22}(X_2)P_{41}(X)\{2\cos(2\alpha_2-\alpha_1-\varphi)\} \\
& + \\
& C(224,011)k_{20}k_{21}k_{41}P_{20}(X_1)P_{21}(X_2)P_{21}(X)\{2\cos(\alpha_2-\varphi)\} \\
& + \\
& C(224,2\text{-}20)k_{22}^2 k_{40}P_{22}(X_1)P_{22}(X_2)P_{40}(X)\{2\cos(-2\alpha_2+2\alpha_1)\} \\
& + \\
& -C(224,1\text{-}10)k_{21}^2\, k_{40}P_{21}(X_1)P_{12}(X_2)P_{40}(X)\{2\cos(-\alpha_2+\alpha_1)\} \\
& + \\
& C(224;0,0,0)k_{20}^2\, k_{40}P_{20}(X_1)P_{20}(X_2)P_{40}(X) \qquad (2.24)
\end{aligned}
$$

CHAPTER THREE

RESULTS AND DISCUSSION

We have performed a series of test calculations on our pair potentials. In all cases we have given comparisons with LJ and GB potentials. The potentials are scaled with respect to the corresponding potential depths for side-by-side configurations.

In all the diagrams series 1, series 2, and series 3 correspond to LJ, GB, and our potential models respectively. For brevity we shall refer to our potential as ZM potential.

Figure 3.1 represents the intermolecular separation dependence of the pair potentials for side-by-side (S.S) configuration. The potential well depth and separation at the well depth are remarkably represented by our pair potential model. The GB potential, on the other hand, although it fits the well depth of LJ potential, but it overestimates the separation at the potential depth. It appears that the GB potential is shifted by some distance indicating that the potential is more repulsive.

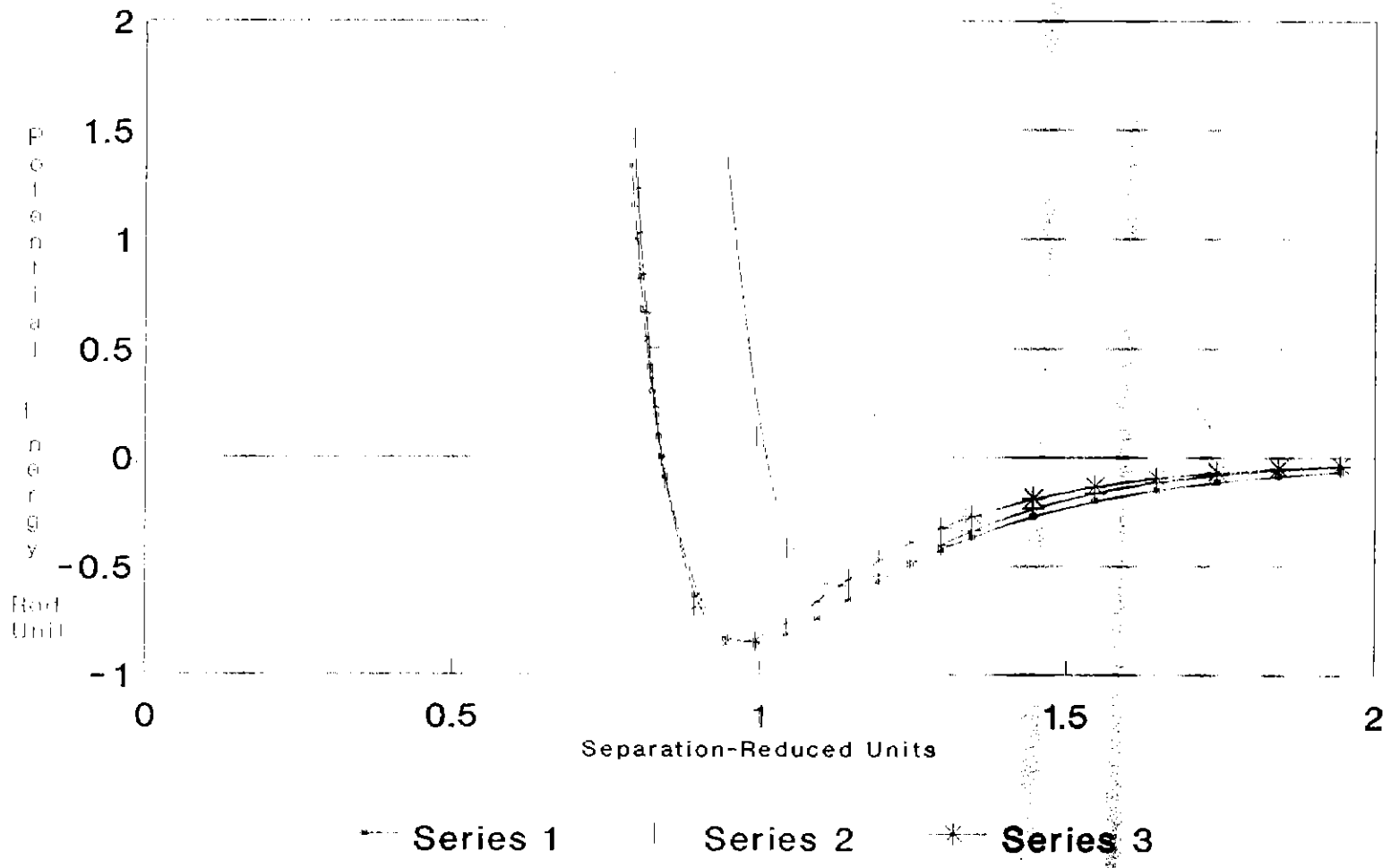


Fig. 3.1. Energies of LJ (series 1), GB (series 2), and ZM (series 3) as functions of separation for side-by-side configuration.

Plotted in Figure 3.2 are head-to-tail (H.T) configuration results for the three potentials. Remarkable agreement between LJ and ZM potentials has been obtained with S.S to H.T ratio of approximately 0.15, i.e. the side-by-side potential well is approximately 6.67 times deeper than the end-to-end. While the repulsive part problem found in the previous configuration (Figure 3.1) for GB potential is partially solved here, the well depth is much deeper than that of LJ, with S.S to H.T ratio of approximately 0.2, i.e. the side-by-side potential well is approximately 5 times deeper than the end-to-end.

Figures 3.3 and 3.4 compare the potentials for Tee (T) and cross (X) configurations, respectively. Our potential model fits that of LJ remarkably. The GB potential shows significant discrepancy both in the potential well and separation at the potential minimum. This dramatic difference between curves of series 1 and series 2 supports the view that the GB model has some qualitative deficiencies probably resulting from the use of only two configurations, namely side-by-side and head-to-tail, to fit all the parameters used in their potential.

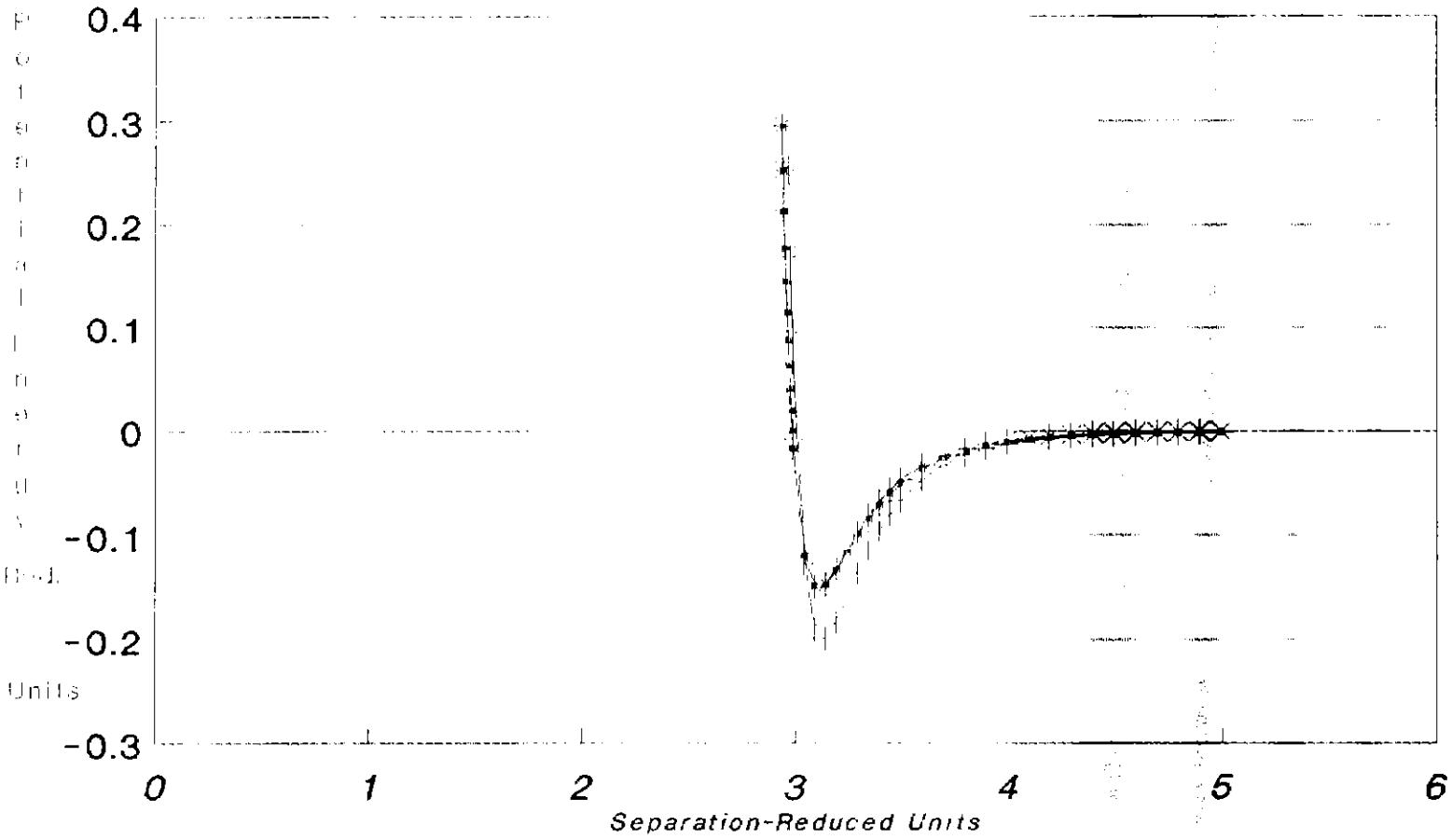


Fig. 3.2. Energies of LJ (series 1), GB (series 2), and ZM (series 3) as functions of separation for head-to-tail configuration.

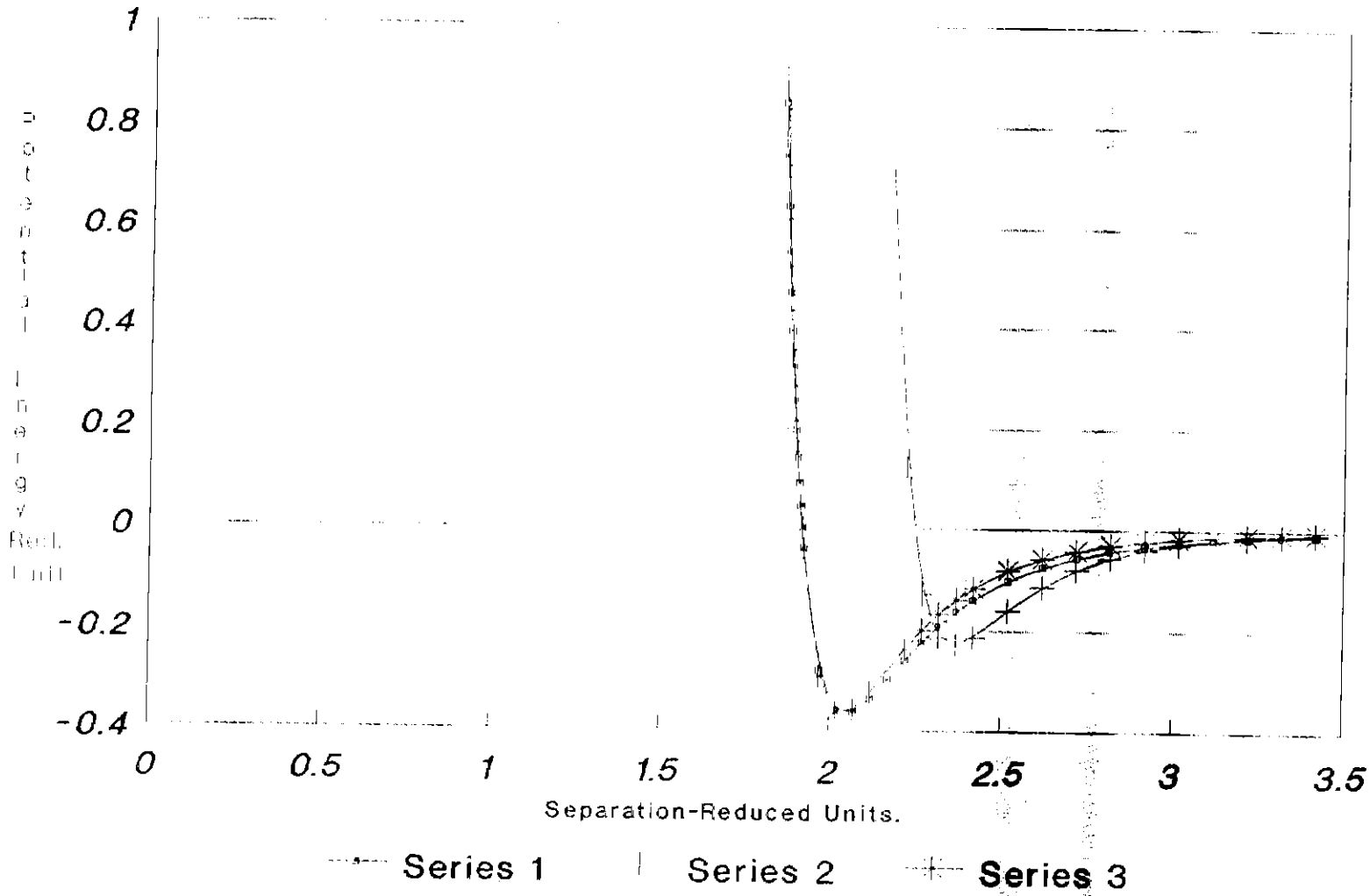


Fig. 3.3. Energies of LJ (series 1), GB (series 2) , and ZM (series 3) as functions of separation for Tee configuration

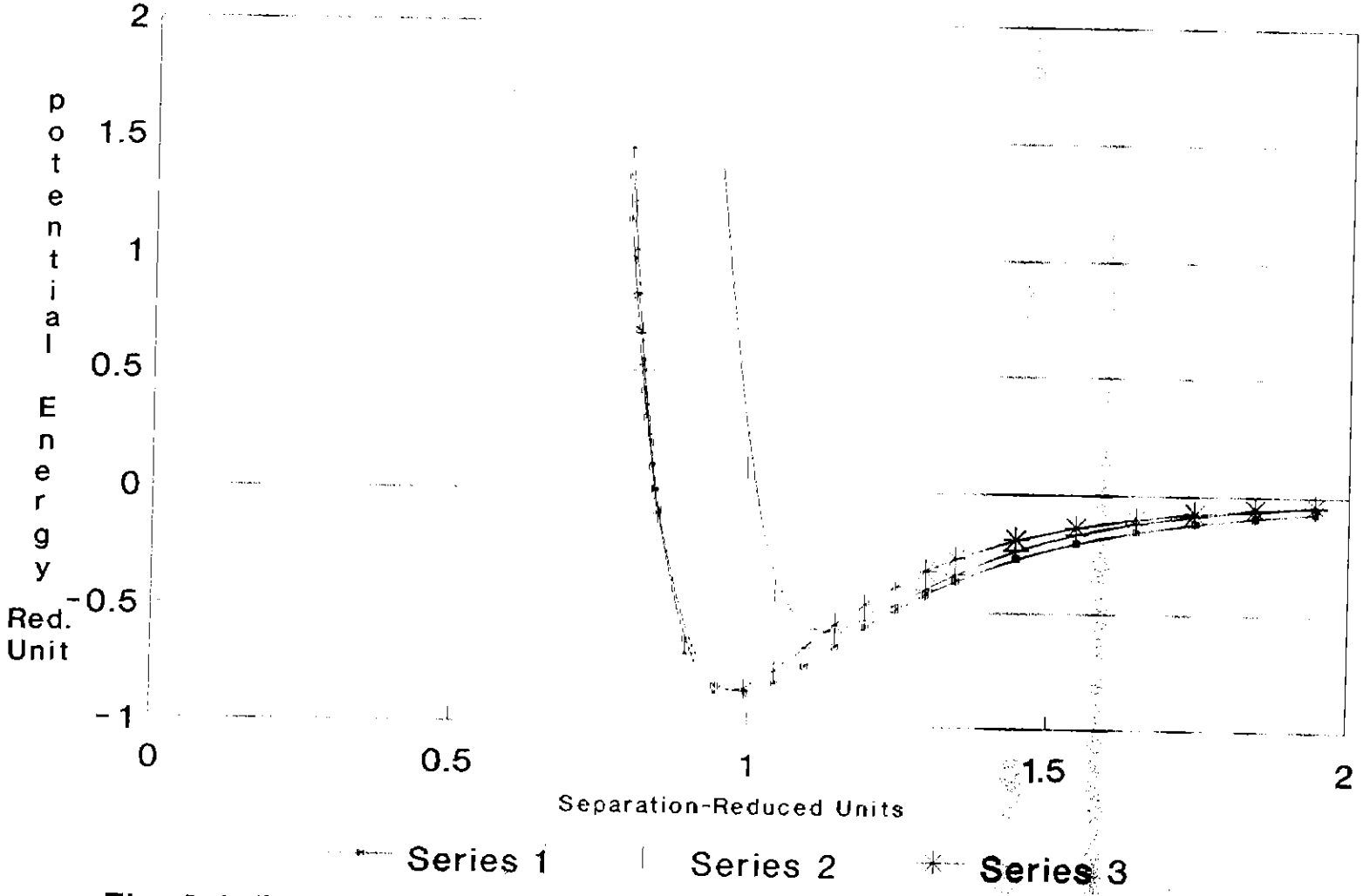


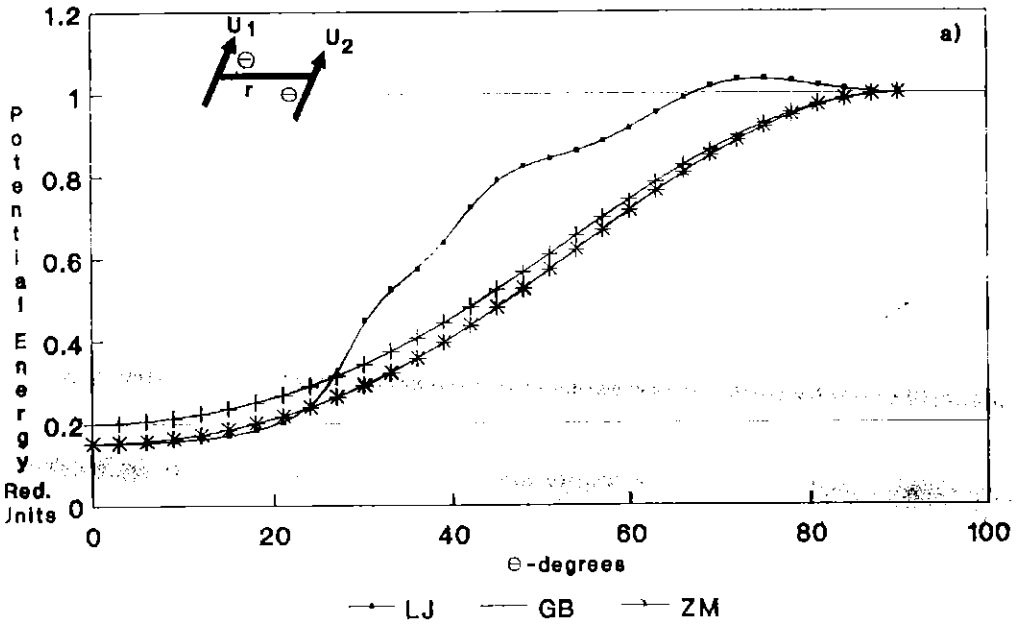
Fig. 3.4. Energies of LJ (series 1), Gb (series 2), and ZM (series 3) as functions of separation for cross configuratin

For further comparison the above three potentials have been tested on five additional cases. These cases include: rotation from side-by-side to head-to-tail maintaining the molecules parallel, rotation from side-by-side to head-to-tail through a Vee configuration, rotation from side-by-side to a Tee configuration, rotation from head-to-tail to a Tee configuration, and the rotation from side-by-side to a crossed configuration. The results of maximum well depth (a) and separation at maximum well depth (b) are given in Figures 3.5, 3.6, 3.7, 3.8 and 3.9 for the above five cases respectively.

On the average the ZM model potential represents the LJ potential better than the GB potential model. The fit can be improved by considering additional critically important configurations and including higher-order spherical harmonic terms in the expansion of strength and range parameters.

The computational time required to evaluate the potentials of 10,000 randomly generated configurations was determined. The results for LJ, GB and ZM are in the ratio 3:1:1 respectively.

It should be mentioned that the ZM potential also shows a high degree of flexibility. Ten-site molecules have been studied. The corresponding strength and range coefficients have been determined. The results compare very well with the corresponding LJ models.



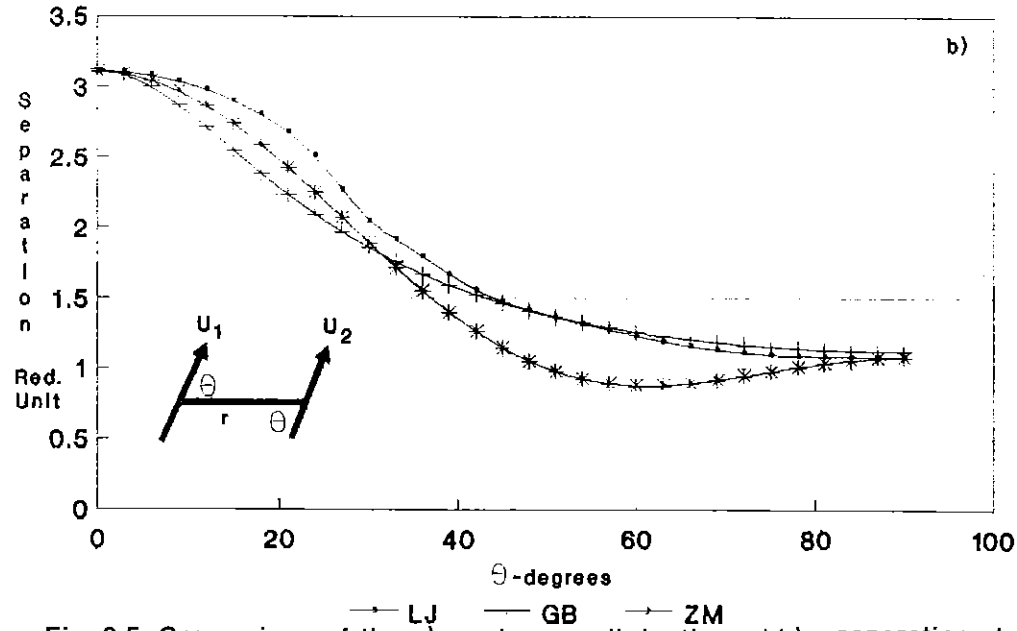
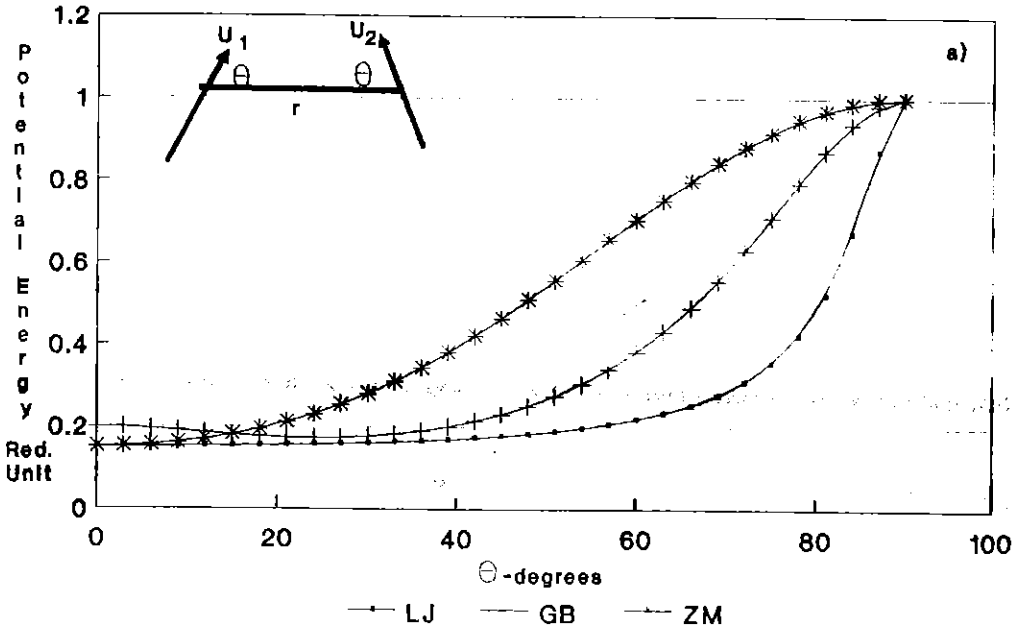


Fig. 3.5. Comparison of the a) maximum well depth, and b) separation at maximum well depth for LJ, GB, and ZM potentials as function of angle for the planar configuration shown above.



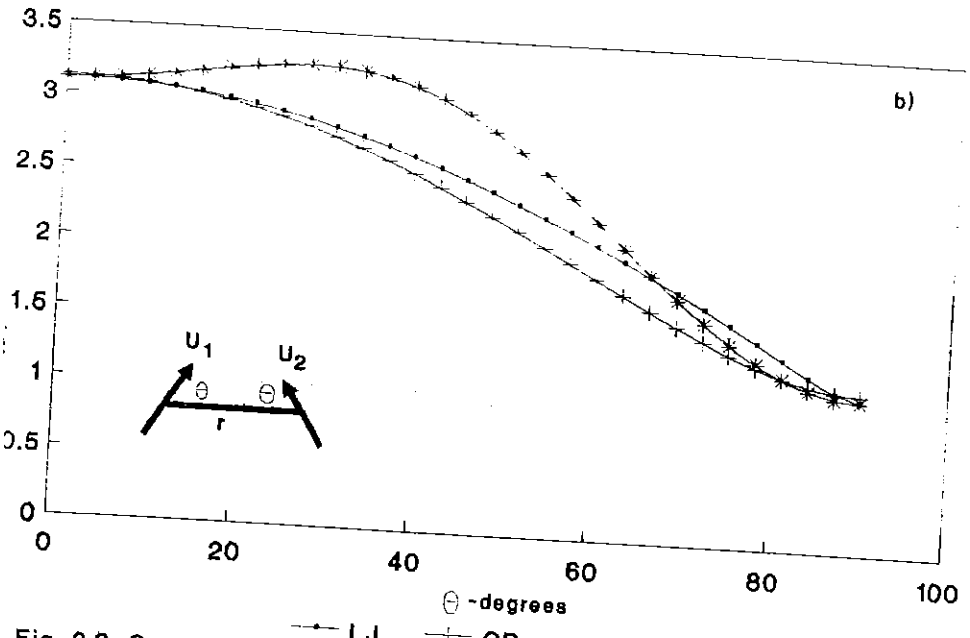
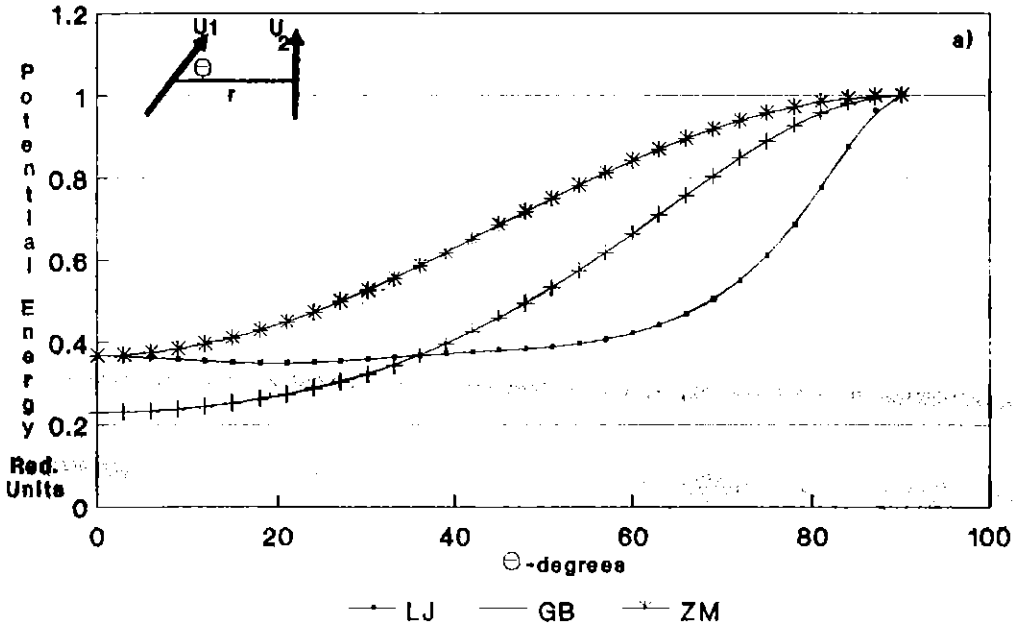


Fig. 3.6. Comparison of the (a) maximum well depth and (b) separation at maximum well depth for LJ, GB and ZM potentials as functions of angle for the planar configuration shown above.



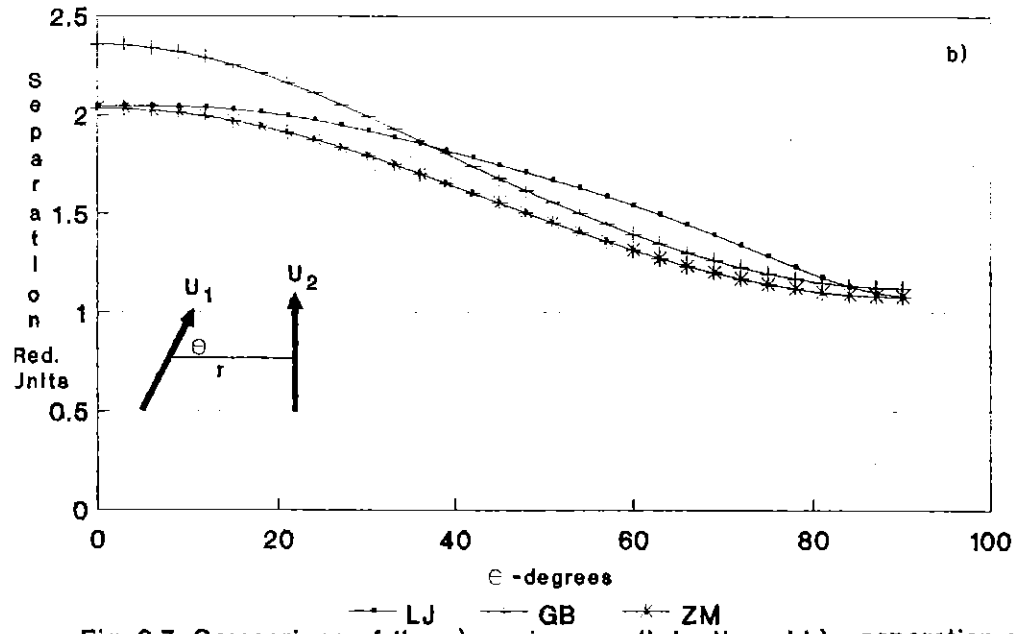
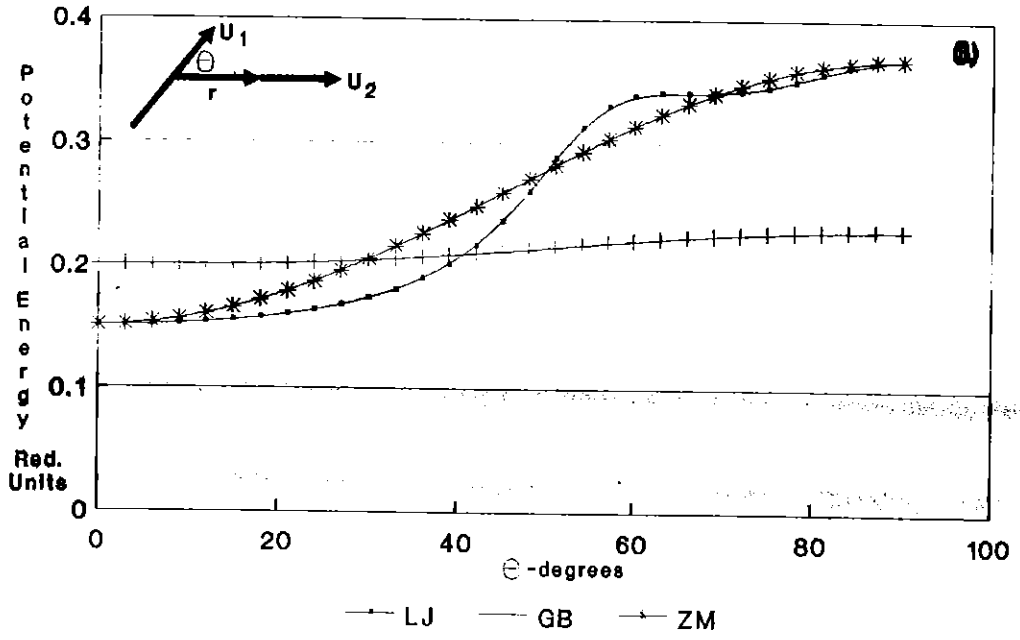


Fig. 3.7. Comparison of the a) maximum well depth and b) separation at maximum well depth for LJ, GB, ZM potentials as functions of angle for the planar configuration shown above.



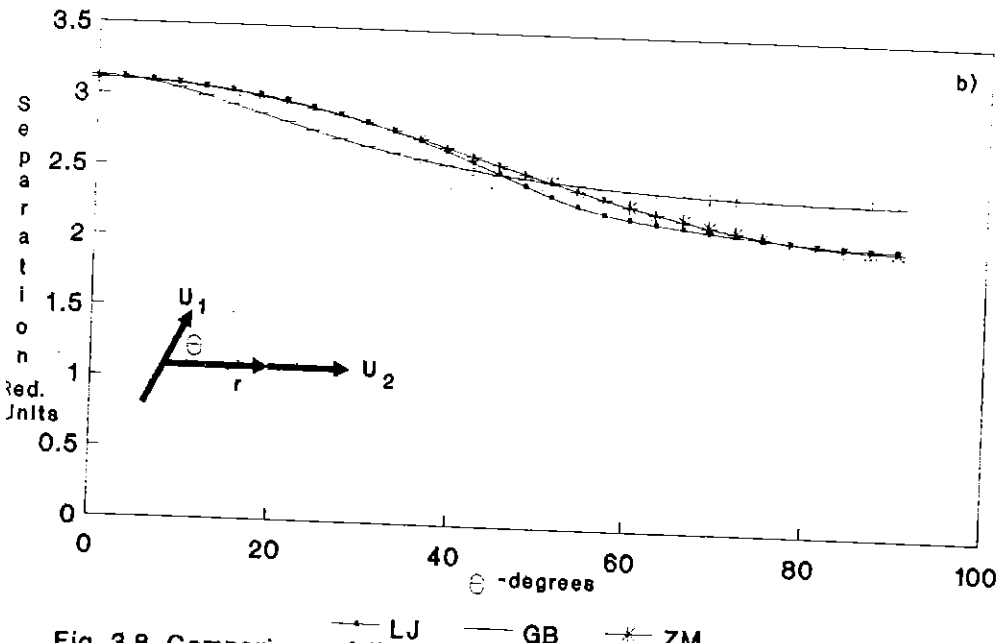
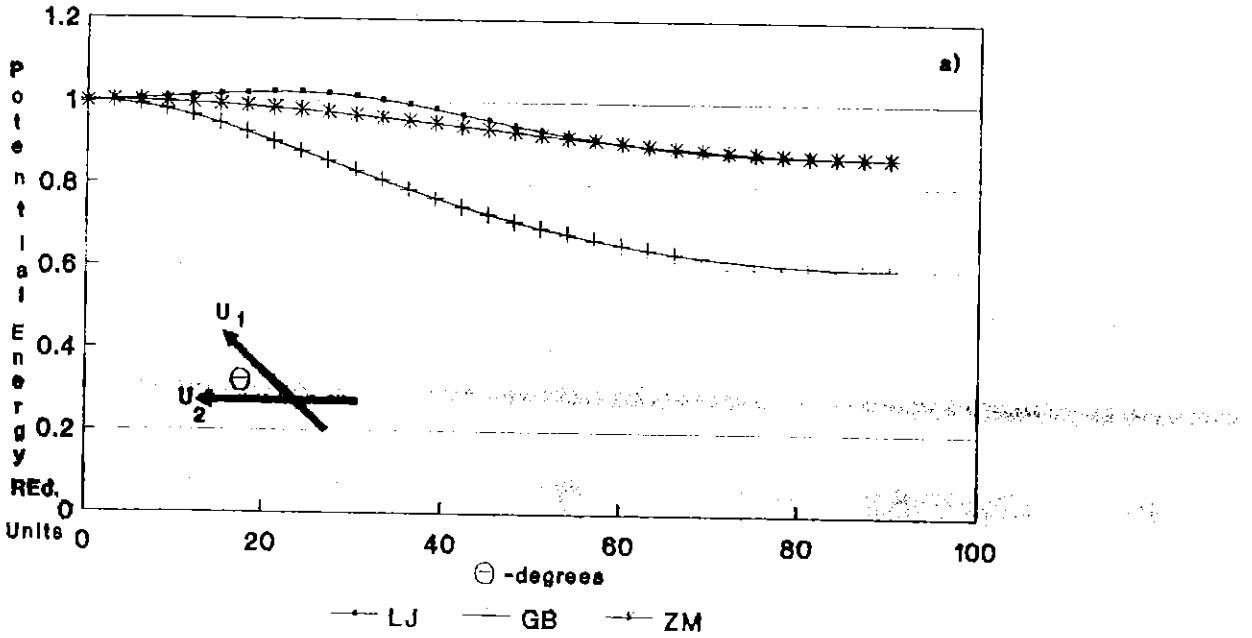


Fig. 3.8. Comparison of the a0 maximum well depth, and b) separation at maximum well depth for LJ, GB, and ZM potentials as function of angle for the planar configuration shown above.



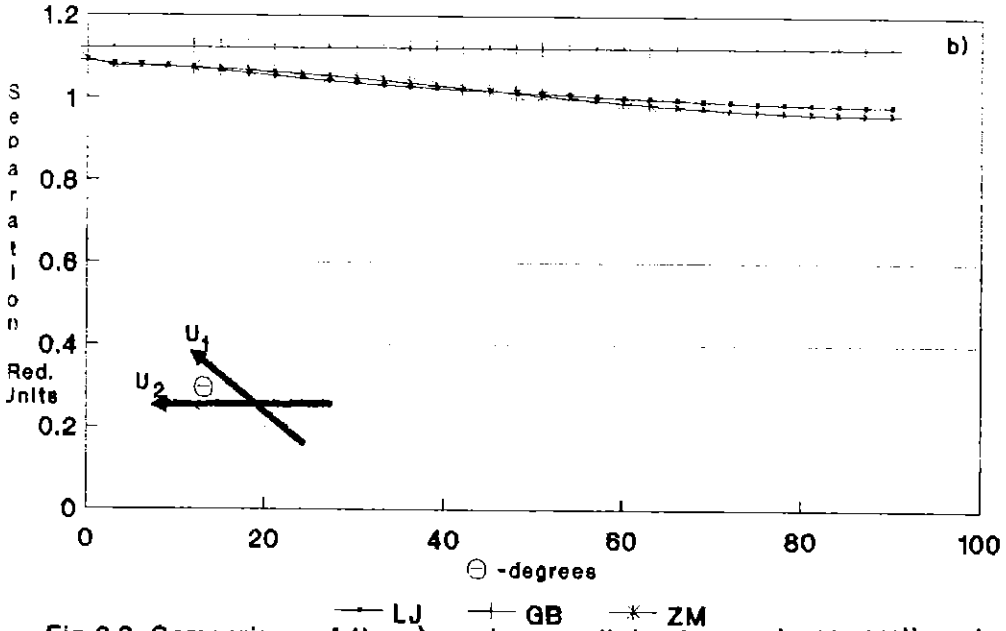


Fig 3.9. Comparison of the a) maximum well depth and b) separation at maximum well depth for LJ, GB, and ZM potentials as function of angle for the planar configuration shown above in which U1 and U2 are per. to r.

In summary, three potentials have been compared according to the number of parameters and their determinations. In the three cases the well depth, ϵ, and the range parameters, σ, are determined as reduced units with respect to ϵ_o and σ_o. LJ potential has only two parameters ϵ and σ. GB potential is characterized by σ_i $\sigma_\perp$, ϵ_i, $\epsilon_\perp$, χ, χ', ν, and μ parameters. ZM potential is determined using σ_{000}, σ_{022}, σ_{220}, σ_{222}, σ_{224} ϵ_{000}, ϵ_{022}, ϵ_{220}, ϵ_{222}, and ϵ_{224} parameters. Our potential contains ten parameters. These parameters are evaluated easily by fitting σ and ϵ of our potential to that of LJ potential. As we have seen in the computational details these parameters are determined by solving a set of linear equations using conjugate gradient minimization methods. The GB potential, on the other hand, contains about eight parameters. In addition to there is no simple clear mathematical procedure to determine these parameters. The simple mathematical procedure used to determine ZM potential parameters makes it more competent and easy to be applied to different configurations and sites.

Table 3.1. The potential well depth and the separation at zero potential calculated using LJ potential for S-S, H-T, T and X configurations.

potential parameters	S-S	H-T	T	X
$\epsilon(r\omega_1\omega_2)$	-8.75852	-1.32091	-3.22390	-7.61438
$\sigma(r\omega_1\omega_2)$	0.95549	2.98290	1.91236	0.83892

Table 3.2. The coefficients of the well depth and the separation at zero potential determined to minimize the corresponding error function.

Strength Coefficients		Range Coefficients	
ϵ_{000}	-.23244592E+03	σ_{000}	.70389342E+02
ϵ_{022}	.23417850E+02	σ_{022}	.61391961E+01
ϵ_{220}	-.12241262E+02	σ_{220}	-.90241715E+01
ϵ_{222}	-.15379991E+00	σ_{222}	.28821168E+01
ϵ_{224}	-.31720260E+01	σ_{224}	.35789736E+01

CHAPTER FOUR

CONCLUSION

We have used generalized spherical harmonic functions to develop a pair potential which is flexible. The flexibility of the potential has been tested using more sites, and can be modelled to accurately fit the LJ potential. It appears promising to model non-cylindrically symmetric molecules.

We have shown that the overall agreement between our potential and LJ is remarkable. Most of the important quantitative features, strength and range parameters of the LJ potentials have been correctly predicted for different types of configurations. In addition, our potential combines the accuracy of LJ potential and the computational speed of the GB potential.

APPENDIX A1

Some properties of Wigner Rotation Matrices and related functions.

Wigner rotation matrix is given by

$$D^{\ell}_{mk}(\alpha,\beta,\gamma)=e^{-im\alpha}\ e^{-ik\gamma}\ d^{\ell}_{mk}(\beta) \qquad (A1.1)$$

where the reduced rotation matrices are given by

$$d^{\ell}_{mk}(\beta)=\Sigma\ (-1)^{t}\ \frac{[(\ell+m)!(l-m)!(\ell-k)!(\ell+k)!]^{1/2}}{(\ell+m-t)!(\ell-k-t)!(\ell+k-m)!}\ \cos(\beta/2)^{x}\ \sin(\beta/2)^{y} \qquad (A1.2)$$

where

$x=2\ell+m-k-2t, \quad y=2t-m+t$

For centerosymmetric molecules k = 0, then

$$D^{\ell}_{m0}(\alpha,\beta,\gamma)=e^{-im\alpha}\ d^{\ell}_{m0}(\beta)\ =(-1)^{m}[4\pi/(2l+1)]^{1/2}\ Y_{lm}(\omega) \qquad (A1.3)$$

$$D^{\ell}_{m0}(\alpha,\beta,\gamma)=(-1)^{m}[(\ell-m)!/(\ell+m)!]^{1/2}\ P_{\ell}(\cos\beta)e^{im\alpha} \qquad (A1.4)$$

$$Y_{\ell m}(\beta\alpha)\ =\ (-1)^{m}\ ((2\ell+1)/[4\pi(\ell+m)!])^{1/2}P_{\ell}^{m}(\cos\beta)e^{im\alpha} \qquad (A1.5)$$

where

$$P_{\ell}^{m}(\cos\beta)\ =\ \frac{1}{2^{\ell}\ell!}\ (1-\cos^{2}\beta)^{|m|/2}\ \frac{d^{\ell+|m|}(\cos^{2}\beta-1)^{\ell}}{d(\cos\beta)^{\ell+|m|}} \qquad (A1.6)$$

is called Legender Polynomial[42].

$$Y^{*}_{\ell,m}\ =\ (-1)^{m}Y_{\ell,-m} \qquad (A1.7)$$

$$Y^{*}_{\ell,-m}\ =\ (-1)^{m}Y_{\ell,m} \qquad (A1.8)$$

APPENDIX A2

This appendix contains some of the important harmonics, Clebsch Gordan Coefficients (CGC) and Legender Polynomial functions that have been used in our potential for the determination of $S_{\ell_1\ell_2\ell}$ functions.

Table(A2.1): Some of the lower-order harmonics

m	$Y_{\ell m}(\theta,\varphi)$
0	$(1/4\pi)^{1/2}$
0	$(3/4\pi)^{1/2}\cos\theta$
±1	$\mp(3/8\pi)^{1/2}\sin\theta\exp(\pm i\varphi)$
0	$(5/4\pi)^{1/2}(1/2)(3\cos^2\theta - 1)$
±1	$\mp(5/4\pi)^{1/2}(3/2)^{1/2}\sin\theta\cos\varphi\exp(\pm i\varphi)$
±2	$(5/4\pi)^{1/2}(3/8)^{1/2}\sin^2\theta\exp(\pm i2\varphi)$
0	$(7/4\pi)^{1/2}(1/2)(5\cos^3\theta - 3\cos\theta)$
±1	$\mp(7/4\pi)^{1/2}(3/16)^{1/2}\sin\theta(5\cos^2\theta - 1)\exp(\pm i\varphi)$
±2	$(7/4\pi)^{1/2}(15/8)^{1/2}\sin^2\theta\cos\theta\exp(\pm i2\varphi)$
±3	$\mp(7/4\pi)^{1/2}(5/16)^{1/2}\sin^3\theta\exp(\pm i3\varphi)$
0	$(9/4\pi)^{1/2}(1/8)(35\cos^4\theta - 30\cos^2\theta+3)$
±1	$\mp(9/4\pi)^{1/2}(1/80)^{1/2}\sin\theta(35\cos^3\theta - 15\cos\theta)\exp(i\pm\varphi)$
±2	$(9/4\pi)^{1/2}(5/32)^{1/2}\sin^2\theta(7\cos^2\theta - 1)\exp(i2\pm\varphi)$
±3	$\mp(9/4\pi)^{1/2}(105/48)^{1/2}\sin^3\theta\cos\theta\exp(\pm i3\varphi)$
±4	$(9/4\pi)^{1/2}(105/384)^{1/2}\sin^4\theta\exp(\pm i4\varphi)$

Table(A2.2): Some of the non-zero Clebsch-Gordan Coefficients (CGC)

ℓ_1	ℓ_2	ℓ	m_1	m_2	m	$C(\ell_1,\ell_2,\ell;m_1,m_2,m)$
0	0	0	0	0	0	+1
0	1	1	0	0	0	+1
0	1	1	0	1	1	+1
0	2	2	0	0	0	+1
0	2	2	0	1	1	+1
0	2	2	0	2	2	+1
1	1	0	1	-1	0	$+1/\sqrt{3}$
1	1	0	0	0	0	$-1/\sqrt{3}$
1	1	1	0	1	1	$-1/\sqrt{2}$
1	1	1	0	-1	-1	$+1/\sqrt{2}$
1	1	1	1	0	1	$+1/\sqrt{2}$
1	1	2	0	0	0	$+2/\sqrt{6}$
1	1	2	0	1	1	$+1/\sqrt{2}$
1	1	2	1	-1	0	$+1/\sqrt{6}$
1	1	2	1	0	1	$+1/\sqrt{2}$
1	1	2	1	1	2	+1
1	2	1	0	0	0	$-2/\sqrt{10}$
1	2	1	0	1	1	$-\sqrt{(3/10)}$
1	2	1	1	-2	-1	$+\sqrt{(3/5)}$
1	2	1	1	-1	0	$+\sqrt{3/10}$
1	2	1	1	0	1	$+1/\sqrt{10}$
1	2	2	0	1	1	$-1/\sqrt{6}$

Table (A2.2): continued

ℓ_1	ℓ_2	ℓ	m_1	m_2	m	$C(\ell_1,\ell_2,\ell;m_1,m_2,m)$
1	2	2	0	2	2	$-2/\sqrt{6}$
1	2	2	1	-2	-1	$+1/\sqrt{3}$
1	2	2	1	-1	0	$+1/\sqrt{2}$
1	2	2	1	0	1	$+1/\sqrt{2}$
1	2	2	1	1	2	$+1/\sqrt{3}$
2	2	0	0	0	0	$+1/\sqrt{5}$
2	2	0	1	-1	0	$-1/\sqrt{5}$
2	2	0	2	-2	0	$+1/\sqrt{5}$
2	2	1	0	1	1	$+\sqrt{(3/10)}$
2	2	1	1	-2	-1	$+1/\sqrt{5}$
2	2	1	1	-1	0	$-1/\sqrt{10}$
2	2	1	1	0	1	$\sqrt{(3/10)}$
2	2	1	2	-2	0	$+2/\sqrt{10}$
2	2	1	2	-1	1	$+1/\sqrt{5}$
2	2	2	0	0	0	$-\sqrt{(2/7)}$
2	2	2	0	1	1	$+1/\sqrt{14}$
2	2	2	0	2	2	$+2/\sqrt{7}$
2	2	2	1	-2	-1	$+\sqrt{(3/7)}$
2	2	2	1	-1	0	$+1/\sqrt{14}$
2	2	2	1	0	1	$-1/\sqrt{14}$
2	2	2	1	1	2	$-\sqrt{(3/7)}$
2	2	2	2	-2	0	$+\sqrt{(2/7)}$

Table (A2.2): continued

ℓ_1	ℓ_2	ℓ	m_1	m_2	m	$C(\ell_1,\ell_2,\ell;m_1,m_2,m)$
2	2	2	2	-1	1	$+\sqrt{(3/7)}$
2	2	2	2	0	2	$+\sqrt{(2/7)}$
2	2	4	0	0	0	$+6/\sqrt{70}$
2	2	4	0	1	1	$+\sqrt{(3/7)}$
2	2	4	0	2	2	$+3/\sqrt{42}$
2	2	4	1	-2	-1	$+1/\sqrt{14}$
2	2	4	1	-1	0	$+4/\sqrt{70}$
2	2	4	1	0	1	$+\sqrt{(3/7)}$
2	2	4	1	1	2	$+2/\sqrt{7}$
2	2	4	1	2	3	$+1/\sqrt{2}$
2	2	4	2	-2	0	$+1/\sqrt{70}$
2	2	4	2	-1	1	$+1/\sqrt{70}$
2	2	4	2	0	2	$+3/\sqrt{42}$
2	2	4	2	1	3	$+1/\sqrt{2}$
2	2	4	2	2	4	+1

Table(A2.2): Continued

ℓ_1	ℓ_2	ℓ	m_1	m_2	m	$C(\ell_1,\ell_2,\ell;m_1,m_2,m)$
2	4	2	2	-4	-2	+4.47214
2	4	2	1	-3	-2	-1.05409
2	4	2	0	2	2	+.34503
2	4	2	1	1	2	-.19920
2	4	2	2	0	2	+.08909
2	4	2	2	-3	-1	+3.16228
2	4	2	1	-2	-1	-1.12687
2	4	2	0	1	1	+.48795
2	4	2	1	0	1	-.35635
2	4	2	2	-1	1	+.19920
2	4	2	2	-2	0	+.34503
2	4	2	1	-1	0	-.48795
2	4	2	0	0	0	+.53452
2	4	4	2	2	4	+.33029
2	4	4	1	3	4	-.61791
2	4	4	0	4	4	+.71351
2	4	4	2	1	3	+.49543
2	4	4	1	2	3	-.58387
2	4	4	0	3	3	+.17838
2	4	4	1	-4	-3	+.61791
2	4	4	2	0	2	+.59216
2	4	4	1	1	2	-.39723

Table(A2.2): Continued

ℓ_1	ℓ_2	ℓ	m_1	m_2	m	$C(\ell_1,\ell_2,\ell;m_1,m_2,m)$
2	4	4	0	2	2	-.20386
2	4	4	1	-3	-2	+.58387
2	4	4	2	-4	-2	+.33029
2	4	4	2	-1	1	+.62419
2	4	4	1	0	1	-.13957
2	4	4	0	1	1	-.43320
2	4	4	1	-2	-1	+.39723
2	4	4	2	-3	1	+.49543
2	4	4	2	-2	0	+.59216
2	4	4	1	-1	0	+.13957
2	4	4	0	0	0	-.50965
2	4	6	2	4	6	+1.00000
2	4	6	2	3	5	+.81650
2	4	6	1	4	5	+.57735
2	4	6	2	2	4	+.65134
2	4	6	1	3	4	+.69631
2	4	6	0	4	4	+.30151
2	4	6	2	1	3	+.50452
2	4	6	1	2	3	+.71351
2	4	6	0	3	3	+.46710
2	4	6	-1	4	3	+.13484
2	4	6	2	0	2	+.37605

Table(A2.2): Continued

ℓ_1	ℓ_2	ℓ	m_1	m_2	m	$C(\ell_1,\ell_2,\ell;m_1,m_2,m)$
2	4	6	1	1	2	+.67270
2	4	6	0	2	2	+.58258
2	4	6	-1	3	2	+.25426
2	4	6	-2	4	2	+.04495
2	4	6	2	-1	1	+.26591
2	4	6	1	0	1	+.59459
2	4	6	0	1	1	+.65134
2	4	6	-1	2	1	+.37605
2	4	6	-2	3	1	+.10050
2	4	6	2	-2	0	+.17408
2	4	6	1	-1	0	+.49237
2	4	6	0	0	0	+.67420
4	2	2	4	-2	2	+.74536
4	2	2	3	-1	2	-.52705
4	2	2	2	0	2	+.34503
4	2	2	1	1	2	-.39841
4	2	2	0	2	2	+.53452
4	2	2	3	-2	1	+.52705
4	2	2	2	-1	1	-.56344
4	2	2	1	0	1	+.48795
4	2	2	0	1	1	-.71270
4	2	2	1	-2	-1	+1.19523

Table(A2.2): Continued

ℓ_1	ℓ_2	ℓ	m_1	m_2	m	$C(\ell_1,\ell_2,\ell;m_1,m_2,m)$
4	2	2	2	-2	0	+.34503
4	2	2	1	-1	0	-.48795
4	2	4	4	0	4	+.71351
4	2	4	3	1	4	-.61791
4	2	4	2	2	4	+.33029
4	2	4	4	-1	3	+.61791
4	2	4	3	0	3	+.17838
4	2	4	2	1	3	-.58387
4	2	4	1	2	3	+.49543
4	2	4	4	-2	2	+.33029
4	2	4	3	-1	2	+.58387
4	2	4	2	0	2	-.20386
4	2	4	1	1	2	-.39723
4	2	4	0	2	2	+.59216
4	2	4	3	-2	1	+.49543
4	2	4	2	-1	1	+.39723
4	2	4	1	0	1	-.43320
4	2	4	0	1	1	-.13957
4	2	4	1	-2	-1	+.62419
4	2	4	2	-2	0	+.59216
4	2	4	1	-1	0	+.13957
4	2	4	0	0	0	-.50965

Table(A2.2): Continued

ℓ_1	ℓ_2	ℓ	m_1	m_2	m	$C(\ell_1,\ell_2,\ell;m_1,m_2,m)$
4	4	0	4	-4	0	+.33333
4	4	0	3	-3	0	-.33333
4	4	0	2	-2	0	+.33333
4	4	0	1	-1	0	-.66667
4	4	0	0	0	0	+2.00000
0	4	4	0	4	4	+1.00000
0	4	4	0	3	3	+1.00000
0	4	4	0	2	2	+1.00000
0	4	4	0	1	1	+1.00000
0	4	4	0	0	0	+1.00000
4	0	4	4	0	4	+1.00000
4	0	4	3	0	3	+1.00000
4	0	4	2	0	2	+1.00000
4	0	4	1	0	1	+1.00000
4	0	4	0	0	0	+1.00000
4	4	2	4	-2	2	+.24618
4	4	2	3	-1	2	-.36927
4	4	2	2	0	2	+.44137
4	4	2	1	1	2	-.93048
4	4	2	0	2	2	+2.64820
4	4	2	1	-3	-2	-.36927

Table(A2.2): Continued

ℓ_1	ℓ_2	ℓ	m_1	m_2	m	$C(\ell_1,\ell_2,\ell;m_1,m_2,m)$
4	4	2	2	-4	-2	+.24618
4	4	2	4	-3	1	+.46057
4	4	2	3	-2	1	-.43519
4	4	2	2	-1	1	+.29608
4	4	2	1	0	1	-.52016
4	4	2	0	1	1	+1.04031
4	4	2	1	-2	-1	+.29608
4	4	2	2	-3	-1	-.43519
4	4	2	3	-4	-1	+.46057
4	4	2	4	-4	0	+.53182
4	4	2	3	-3	0	-.13295
4	4	2	2	-2	0	-.15195
4	4	2	1	-1	0	+.13295
4	4	2	0	0	0	-.41786
4	4	4	4	0	4	+.31289
4	4	4	3	1	4	-.49473
4	4	4	2	2	4	+.56097
4	4	4	1	3	4	-.98945
4	4	4	0	4	4	+1.87736
4	4	4	4	-1	3	+.49473
4	4	4	3	0	3	-.46934
4	4	4	2	1	3	+.18699

Table(A2.2): Continued

ℓ_1	ℓ_2	ℓ	m_1	m_2	m	$C(\ell_1,\ell_2,\ell;m_1,m_2,m)$
4	4	4	1	2	3	-.18699
4	4	4	0	3	3	-.31289
4	4	4	-1	4	3	+2.96836
4	4	4	4	-2	2	+.56097
4	4	4	3	-1	2	-..18699
4	4	4	2	0	2	-.24584
4	4	4	1	1	2	+.21203
4	4	4	0	2	2	-.62579
4	4	4	-1	3	2	+1.12194
4	4	4	-2	4	2	+3.36581
4	4	4	4	-3	1	+.49473
4	4	4	3	-2	1	+.18699
4	4	4	2	-1	1	-.42405
4	4	4	1	0	1	+.11175
4	4	4	0	1	1	-.22350
4	4	4	-1	2	1	-.21203
4	4	4	-2	3	1	+2.61785
4	4	4	-3	4	1	+2.96836
4	4	4	4	-4	0	+.31289
4	4	4	3	-3	0	+.46934
4	4	4	2	-2	0	-.24584
4	4	4	1	-1	0	-.22350

Table(A2.2): Continued

ℓ_1	ℓ_2	ℓ	m_1	m_2	m	$C(\ell_1,\ell_2,\ell;m_1,m_2,m)$
4	4	4	0	0	0	+.13857
4	4	6	4	2	6	+.51640
4	4	6	3	3	6	-.68313
4	4	6	4	1	5	+.63246
4	4	6	3	2	5	-.31623
4	4	6	4	0	4	+.60302
4	4	6	3	1	4	+.09535
4	4	6	2	2	4	-.50452
4	4	6	4	-1	3	+.49237
4	4	6	3	0	3	-.68313
4	4	6	2	1	3	-.32567
4	4	6	4	-2	2	+.34816
4	4	6	3	-1	2	+.52223
4	4	6	2	0	2	+.64324
4	4	6	1	1	2	-.46057
4	4	6	4	-3	1	+.20597
4	4	6	3	-2	1	+.50602
4	4	6	2	-1	1	+.30896
4	4	6	1	0	1	-.32567
4	4	6	4	-4	0	+.08989
4	4	6	3	-3	0	+.38205
4	4	6	2	-2	0	+.49441

Table(A2.2): Continued

ℓ_1	ℓ_2	ℓ	m_1	m_2	m	$C(\ell_1,\ell_2,\ell;m_1,m_2,m)$
4	4	6	1	-1	0	-.02247
4	4	6	0	0	0	-.44947
4	4	8	4	4	8	+.00254
4	4	8	4	3	7	+.00720
4	4	8	4	2	6	+.01521
4	4	8	3	3	6	-.68313
4	4	8	4	1	5	+.04578
4	4	8	3	2	5	+.09157
4	4	8	4	0	4	+.05097
4	4	8	3	1	4	+.12896
4	4	8	2	2	4	+.17059
4	4	8	4	-1	3	+.02943
4	4	8	3	0	3	+.09307
4	4	8	2	1	3	+.15573
4	4	8	4	-2	2	+.01537
4	4	8	3	-1	2	+.06148
4	4	8	2	0	2	+.12859
4	4	8	1	1	2	+.16266
4	4	8	4	-3	1	+.00687
4	4	8	3	-2	1	+.03637
4	4	8	2	-1	1	+.09623
4	4	8	1	0	1	+.15215

Table(A2.2): Continued

ℓ_1	ℓ_2	ℓ	m_1	m_2	m	$C(\ell_1,\ell_2,\ell;m_1,m_2,m)$
4	4	8	4	-4	0	+.00229
4	4	8	3	-3	0	+.01833
4	4	8	2	-2	0	+.06415
4	4	8	1	-1	0	+.12830
4	4	8	0	0	0	+.16038

Table(A2.3): Some of the lower Legender Polynomial functions

ℓ	m	$P_\ell^m(\cos\theta)$
0	0	1
1	0	$\cos\theta$
1	1	$\sin\theta$
2	0	$1/2\ (3\cos^2\theta - 1)$
2	1	$3\sin\theta\cos\theta$
2	2	$3\sin^2\theta$
3	0	$1/2\ (5\cos^3\theta - 3\cos\theta)$
3	1	$1/2\ \sin\theta(15\cos^2\theta - 3)$
3	2	$15\sin^2\theta\cos\theta$
3	3	$15\sin^3\theta$
4	0	$1/8\ (35\cos^4\theta - 30\cos^2\theta + 3)$
4	1	$1/2\ \sin\theta\ (35\cos^3\theta - 15\cos\theta)$
4	2	$1/2\ \sin^2\theta(105\cos^2\theta - 15)$
4	3	$105\ \sin\theta^3\theta\cos\theta$
4	4	$105\ \sin^4\theta$
6	0	$1/16(231\cos^6\theta - 315\cos^4\theta + 105\cos^2\theta - 5)$
6	1	$1/8\ \sin\theta\ (693\cos^5\theta - 630\cos^3\theta + 105\cos\theta)$
6	2	$1/8\ \sin^2\theta\ (3465\cos^4\theta - 1890\cos^2\theta + 105)$
6	3	$1/4\ \sin^3\theta\ (6930\cos^3\theta - 1890\cos\theta)$
6	4	$1/2\ \sin^4\theta\ (10395\cos^2\theta - 945)$
6	5	$10395\ \sin^5\theta\ \cos\theta$
6	6	$10395\ \sin^6\theta$

Table (A2.3) continued

ℓ	m	$P_\ell^m(\cos\theta)$
8	0	$1/128\ (6435\cos^8\theta - 12012\cos^6\theta + 6930\cos^4\theta - 1260\cos^2\theta + 35)$
8	1	$1/16\ \sin\theta\ (6435\cos^7\theta - 9009\cos^5\theta + 3465\cos^3\theta - 315\cos\theta)$
8	2	$1/16\ \sin^2\theta\ (45045\cos^6 - 45045\cos^4\theta + 10395\cos^2\theta - 315)$
8	3	$1/8\ \sin^3\theta\ (135135\cos^5\theta - 90090\cos^3\theta + 10395\cos\theta)$
8	4	$1/8\ \sin^4\theta\ (675675\cos^4\theta - 270270\cos^2\theta + 10395)$
8	5	$1/2\ \sin^5\theta\ (675675\cos^3\theta - 135135\cos\theta)$
8	6	$1/2\ \sin^6\theta\ (2027025\cos^2\theta - 135135)$
8	7	$2027025\ \sin^7\theta\ \cos\theta$
8	8	$2027025\ \sin^8\theta$

APPENDIX A3

This Appendix explains the calculation details for some lower-order Spherical Harmonic Functions. Here for brevity the Legender Polynomial functions are designated as:

$P_{ij}(\cos\theta) \equiv P_{ij}(X)$

$P_{ij}(\cos\beta_1) \equiv P_{ij}(X1)$

$P_{ij}(\cos\beta_2) \equiv P_{ij}(X2)$

A3.1 DETERMINATION OF S_{000}

$$S_{000} = \Sigma_{000}\, C(000;000)\, Y_{00}(\omega_1)\, Y_{00}(\omega_2)\, Y_{00}^*(\omega)$$
$$= (1/4\pi)^{1/2}\, (1/4\pi)^{1/2}\, (1/4\pi)^{1/2}$$
$$= (1/4\pi)^{3/2} \qquad (A3.1)$$

A3.2 DETERMINATION OF S_{202}:-

$$S_{202} = \Sigma_{m1,0,m}\, C(202;m_1,0,m)\, Y_{2m1}(\omega_1)\, Y_{0m2}(\omega_2)\, Y_{2m}^*(\omega)$$

from Eq 2.8

when $\ell_1 = 2$ the values of $m_1 = -2,-1,0,1,2$

$\ell_2 = 0$ the value of $m_2 = 0$

$\ell = 2$ the values of $m = -2,-1,0,1,2$

The values of orders m_1, m_2 and m that make CGC not equal zero are determined from Eq 2.7 and tabulated below

m_1	-2	-1	0	1	2
m_2	0	0	0	0	0
m	-2	-1	0	1	2

$$S_{202} = \sum_{m1m} C(202;-20-2)\ Y_{2m1}(\omega_1)\ Y_{00}(\omega_2)\ Y_{2m}^*(\omega) \quad (A3.2)$$

$$= Y_{00}(\omega_2)\ \{\ C(202;-2,0,-2)Y_{2,-2}(\omega_1)Y_{2,-2}^*(\omega)$$
$$+ C(202;2,0,2)Y_{2,2}(\omega_1)Y_{2,2}^*(\omega)$$
$$+ C(202;-1,0,-1)Y_{2,-1}(\omega_1)Y_{2,-1}^*(\omega)$$
$$+ C(202;1,0,1)Y_{2,1}(\omega_1)Y_{2,1*}(\omega)$$
$$+ C(202;0,0,0)Y_{2,0}(\omega_1)Y_{2,0}^*(\omega)\ \} \quad (A3.3)$$

For $m = \pm 2$

$$a = C(202;2,0,2)\{Y_{2,2}(\omega_1)Y_{00}(\omega_2)(\omega_1)Y_{2,2}^*(\omega) + Y_{2,-2}(\omega_1)Y_{00}(\omega_2)(\omega_1)Y_{2,-2}^*(\omega)\} \quad (A3.4)$$

$$= C(202,202)\begin{vmatrix} k_{22}P_{22}(X_1)\exp(i2\alpha_1) & k_{22}P_{22}(X_1)\exp(-i2\alpha_1) \\ k_{00}P_{00}(X_2) & + \quad k_{00}P_{00}(X_2) \\ k_{22}P_{22}(X)\exp(-i2\varphi) & k_{22}P_{22}(X)\exp(i2\varphi) \end{vmatrix} \quad (A3.5)$$

we have

$$\exp(im\alpha) = \cos(m\alpha) + i\sin(m\alpha) \quad (A3.6.a)$$

$$\exp(-im\alpha) = \cos(m\alpha) - i\sin(m\alpha) \quad (A3.6.b)$$

$$\exp(im\alpha) + \exp(-im\alpha) = 2\cos(m\alpha) \quad (A3.6.c)$$

substituting Eq A3.6.c into Eq A3.5 we get

$$a = C(202,202)k_{22}^2\ k_{00}P_{22}(X_1)P_{00}(X_2)P_{22}(X)\{2\cos(2\alpha_1-2\varphi)\} \quad (A3.7)$$

For $m = \pm 1$

$$b = C(202;1,0,1)\{Y_{2,1}(\omega_1)Y_{00}(\omega_2)(\omega_1)Y_{2,1}^*(\omega) + Y_{2,-1}(\omega_1)Y_{00}(\omega_2)(\omega_1)Y_{2,-1}^*(\omega)\} \quad (A3.8)$$

$$= C(202,101)\begin{vmatrix} k_{21}P_{21}(X_1)\exp(i\alpha_1) & k_{21}P_{21}(X_1)\exp(-i\alpha_1) \\ k_{00}P_{00}(X_2) & + \quad k_{00}P_{00}(X_2) \\ k_{21}P_{21}(X)\exp(-i\varphi) & k_{21}P_{21}(X)\exp(i\varphi) \end{vmatrix} \quad (A3.9)$$

Eq A3.6.c into Eq A3.9 gives

$$b = C(202,101)k_{21}^2\, k_{00}P_{21}(X_1)P_{00}(X_2)P_{21}(X)\{2\cos(\alpha_1-\varphi)\} \quad (A3.10)$$

For m = 0

$$c = C(202;0,0,0)Y_{2,0}(\omega_1)Y_{00}(\omega_2)Y_{2,0}^*(\omega) \quad (A3.11)$$

$$= C(202;0,0,0)k_{20}^3\, P_{20}(X_1)P_{00}(X_2)P_{20}(X) \quad (A3.12)$$

Then S_{202} will be obtained by substituting Eqs A3.7, A3.10 and A3.12 into Eq A3.2.

$$S_{202} = C(202,202)k_{22}^2\, k_{00}P_{22}(X_1)P_{00}(X_2)P_{22}(X)\{2\cos(2\alpha_1-2\varphi)\} + C(202,101)k_{21}^2\, k_{00}P_{21}(X_1)P_{00}(X_2)P_{21}(X)\{2\cos(\alpha_1-\varphi)\} + C(202;0,0,0)k_{20}^2\, k_{00}P_{20}(X_1)P_{00}(X_2)P_{20}(X)$$

where (A3.13)

$$k_{\ell m} = (-1)^m\, \{(2\ell+1)/[4\pi(\ell+m)!]\}^{1/2} \quad (A3.14)$$

and P_ℓ^m is determined from Eq A1.6 and some values are tabulated in table (A2.3).

A3.3. DETERMINATION OF S_{022}:-

$S_{022} = \sum_{o,m2,m} C(022;0,m_2,m)\; Y_{0m1}(\omega_1)\; Y_{2m2}(\omega_2)\; Y_{2m}^*(\omega)$

from Eq 2.8

when $\ell_1 = 0$ the values of $m_1 = 0$

$\ell_2 = 2$ the value of $m_2 = -2,-1,0,1,2$

$\ell = 2$ the values of $m = -2,-1,0,1,2$

The values of orders m_1, m_2 and m that make CGC not equal zero re determined from Eq 2.7 and tabulated below

m_1	0	0	0	0	0
m_2	-2	-1	0	1	2
m	-2	-1	0	1	2

$$S_{022} = \Sigma_{m2,m}\, C(022;0,m_2,m)\; Y_{00}(\omega_1)\; Y_{2m2}(\omega_2)\; Y_{2m}^{*}(\omega) \quad \text{(A3.15)}$$

$$= Y_{00}(\omega_1)\,\{\, C(022;0,-2,-2)Y_{2,-2}(\omega_2)Y_{2,-2}^{*}(\omega) + C(022;0,2,2)Y_{2,2}(\omega_2)Y_{2,2}^{*}(\omega) + C(022;0,-1,-1)Y_{2,-1}(\omega_2)Y_{2,-1}^{*}(\omega) + C(022;0,1,1)Y_{2,1}(\omega_2)Y_{2,1}^{*}(\omega) + C(022;0,0,0)Y_{2,0}(\omega_2)Y_{2,0}^{*}(\omega)\,\} \quad \text{(A3.16)}$$

For $m = \pm 2$

$$a = C(022;022)\{Y_{00}(\omega_1)Y_{22}(\omega_2)(\omega_1)Y_{2,2}^{*}(\omega) + Y_{00}(\omega_1)Y_{2-2}(\omega_2)(\omega_1)Y_{2,-2}^{*}(\omega)\} \quad \text{(A3.17)}$$

$$= C(022,022)\begin{vmatrix} k_{00}P_{00}(X_1) & k_{00}P_{00}(X_1) \\ k_{22}P_{22}(X_2)\exp(i2\alpha_2) & + k_{22}P_{22}(X_2)\exp(-i2\alpha_2) \\ k_{22}P_{22}(X)\exp(-i2\varphi) & k_{22}P_{22}(X)\exp(i2\varphi) \end{vmatrix} \quad \text{(A3.18)}$$

substituting Eq A3.6.c into Eq A3.18 we get

$$a = C(022,022)k_{22}^{2}\, k_{00}P_{00}(X_1)P_{22}(X_2)P_{22}(X)\{2\cos(2\alpha_2-2\varphi)\} \quad \text{(A3.19)}$$

For $m = \pm 1$

$$b = C(022;011)\{Y_{0,0}(\omega_1)Y_{21}(\omega_2)(\omega_1)Y_{2,1}^{*}(\omega) + Y_{0,0}(\omega_1)Y_{2-1}(\omega_2)(\omega_1)Y_{2,-1}^{*}(\omega)\} \quad \text{(A3.20)}$$

$$= C(022,011)\begin{vmatrix} k_{00}P_{00}(X_1) & k_{00}P_{00}(X_1) \\ k_{21}P_{21}(X_2)\exp(i\alpha_2) & + k_{21}P_{21}(X_2)\exp(-i\alpha_2) \\ k_{21}P_{21}(X)\exp(-i\varphi) & k_{21}P_{21}(X)\exp(i\varphi) \end{vmatrix} \quad \text{(A3.21)}$$

Eq A3.6.c into Eq A3.21 gives

$$b = C(022,011)k_{21}^{2}\, k_{00}P_{00}(X_1)P_{21}(X_2)P_{21}(X)\{2\cos(\alpha_2-\varphi)\} \quad \text{(A3.22)}$$

For $m = 0$

$$c = C(022;000)Y_{00}(\omega_1)Y_{20}(\omega_2)Y_{2,0}^{*}(\omega) \quad \text{(A3.23)}$$

$$= C(022;0,0,0)k_{20}^{2}k_{00}P_{00}(X_1)P_{20}(X_2)P_{20}(X) \quad \text{(A3.24)}$$

Substituting Eqs A3.19, A3.22 and A3.24 into Eq A3.15 S_{022} can be calculated as below

$$S_{022} = C(022,022)k_{22}^2 k_{00} P_{00}(X_1)P_{22}(X_2)P_{22}(X)\{2\cos(2\alpha_2-2\varphi)\}$$
$$+$$
$$C(022,011)k_{21}^2 k_{00} P_{00}(X_1)P_{21}(X_2)P_{21}(X)\{2\cos(\alpha_2-\varphi)\}$$
$$+$$
$$C(022;0,0,0)k_{20}^2\ k_{00} P_{00}(X_1)P_{20}(X_2)P_{20}(X) \qquad \text{(A3.25)}$$

A3.4 DETERMINATION OF S_{220}:-

$S_{220} = \sum_{m1,m2,o} C(220;m_2,m_1,0)\ Y_{2m1}(\omega_1)\ Y_{2m2}(\omega_2)\ Y_{om}^*(\omega)$ from Eq 2.8

when $\ell_1 = 2$ the values of $m_1 = -2,-1,0,1,2$

$\ell_2 = 2$ the value of $m_2 = -2,-1,0,1,2$

$\ell = 0$ the values of $m = 0$

The values of orders m_1, m_2 and m that make CGC not equal zero are determined from Eq 2.7 and tabulated below

m_1	-2	-1	0	1	2
m_2	2	1	0	-1	-2
m	0	0	0	0	0

$$S_{220} = \sum_{m1,m2} C(220;m_1,m_2,0)\ Y_{2,m1}(\omega_1)\ Y_{2m2}(\omega_2)\ Y_{oo}^*(\omega) \qquad \text{(A3.26)}$$

$$= Y_{oo}^*(\omega)\ (\ C(220;-2,2,0)Y_{2,-2}(\omega_1)Y_{2,2}(\omega_2)$$
$$+\ C(220;2,-2,0)Y_{2,2}(\omega_1)Y_{2,-2}(\omega_2)$$
$$+\ C(220;-1,1,0)Y_{2,-1}(\omega_1)Y_{2,1}(\omega_2)$$
$$+\ C(220;1,-1,0)Y_{2,1}(\omega_1)Y_{2,-1}(\omega_2)$$
$$+\ C(220;0,0,0)Y_{2,0}(\omega_1)Y_{2,0}(\omega_2)\ ) \qquad \text{(A3.27)}$$

For $m = \pm 2$

$$a = C(220;2-20)\{Y_{22}(\omega_1)Y_{2-2}(\omega_2)(\omega_1)Y_{oo}^*(\omega)$$
$$+$$
$$Y_{2-2}(\omega_1)Y_{22}(\omega_2)(\omega_1)Y_{oo}^*(\omega)\} \qquad \text{(A3.28)}$$

$$= C(220,2\text{-}20)\begin{vmatrix} k_{22}P_{22}(X_1)\exp(i2\alpha_1) & k_{22}P_{22}(X_1)\exp(-i2\alpha_1) \\ k_{22}P_{22}(X_2)\exp(-i2\alpha_2) & + \; k_{22}P_{22}(X_2)\exp(i2\alpha_2) \\ k_{00}P_{00}(X) & k_{00}P_{00}(X) \end{vmatrix} \quad (A3.29)$$

substituting Eq A3.6.c into Eq A3.29 we get

$$a = C(220,2\text{-}20)k_{22}^2 k_{00}P_{22}(X_1)P_{22}(X_2)P_{00}(X)\{2\cos(-2\alpha_2+2\alpha_1)\} \quad (A3.30)$$

For m = ± 1

$$b = C(220;1\text{-}10)\{Y_{21}(\omega_1)Y_{2-1}(\omega_2)(\omega_1)Y_{00}^*(\omega) + Y_{21}(\omega_1)Y_{2-1}(\omega_2)(\omega_1)Y_{00}^*(\omega)\} \quad (A3.31)$$

$$= C(220,1\text{-}10)\begin{vmatrix} k_{21}P_{21}(X_1)\exp(i\alpha_1) & k_{21}P_{21}(X_1)\exp(-i\alpha_1) \\ k_{21}P_{21}(X_2)\exp(-i\alpha_2) & + \; k_{21}P_{21}(X_2)\exp(i\alpha_2) \\ k_{00}P_{00}(X) & k_{00}P_{00}(X) \end{vmatrix} \quad (A3.32)$$

substituting Eq A3.6.c into Eq A3.32 we get

$$b = C(220,1\text{-}10)k_{21}^2 k_{00}P_{21}(X_1)P_{12}(X_2)P_{00}(X)\{2\cos(-\alpha_2+\alpha_1)\} \quad (A3.33)$$

For m = 0

$$c = C(022;000)Y_{20}(\omega_1)Y_{20}(\omega_2)Y_{00}^*(\omega) \quad (A3.34)$$

$$= C(220;0,0,0)k_{20}^2 k_{00}P_{20}(X_1)P_{20}(X_2)P_{00}(X) \quad (A3.35)$$

Substituting Eqs A3.30, A3.33 and A3.35 into Eq A3.26

S_{220} can be calculated

$$S_{220} = C(220,2\text{-}20)k_{22}^2 k_{00}P_{22}(X_1)P_{22}(X_2)P_{00}(X)\{2\cos(-2\alpha_2+2\alpha_1)\} + C(220,1\text{-}10)k_{21}^2 k_{00}P_{21}(X_1)P_{12}(X_2)P_{00}(X)\{2\cos(-\alpha_2+\alpha_1)\} + C(220;0,0,0)k_{20}^2 k_{00}P_{20}(X_1)P_{20}(X_2)P_{00}(X) \quad (A3.36)$$

3.5. DETERMINATION OF S_{222}:-

$$S_{222} = \Sigma_{m1,m2,m}\, C(222;m_1,m_2,m)\; Y_{2m1}(\omega_1)\; Y_{2m2}(\omega_2)\; Y_{2m}^*(\omega)$$

from Eq 2.8

when $\ell_1 = 2$ the values of $m_1 = -2,-1,0,1,2$

$\ell_2 = 2$ the value of $m_2 = -2,-1,0,1,2$

$\ell = 2$ the values of $m = -2,-1,0,1,2$

The values of orders m_1, m_2 and m that make CGC not equal zero are determined from Eq 2.7 and tabulated below

m	-2			-1				0					1				2		
m_1	-2	-1	0	-2	-1	0	1	2	1	0	-1	-2	2	1	0	-1	2	1	0
m_2	0	-1	-2	1	0	-1	-2	-2	-1	0	1	2	-1	0	1	2	0	1	2

For $m = \pm 2$

$$X = \sum_{m1,m2,2} C(222;m1,m2,2)\, Y_{2m1}(\omega_1)\, Y_{2m2}(\omega_2)\, Y_{22}^*(\omega) + \sum_{m1,m2,-2} C(222;m1,m2,-2)\, Y_{2m1}(\omega_1)\, Y_{2m2}(\omega_2)\, Y_{2-2}^*(\omega) \quad \text{(A3.37)}$$

$$a = C(222;2,0,2)\{Y_{2,2}(\omega_1)Y_{2,0}(\omega_2)Y_{22}^*(\omega)+Y_{2,-2}(\omega_1)Y_{2,0}(\omega_2)Y_{2-2}^*(\omega)\}$$

$$b = C(222;0,2,2)\{Y_{2,0}(\omega_1)Y_{2,2}(\omega_2)Y_{22}^*(\omega)+Y_{2,0}(\omega_1)Y_{2,-2}(\omega_2)Y_{2-2}^*(\omega)\}$$

$$c = C(222;1,1,2)\{Y_{2,1}(\omega_1)Y_{2,1}(\omega_2)Y_{22}^*(\omega)+Y_{2,-1}(\omega_1)Y_{2,-1}(\omega_2)Y_{2-2}^*(\omega)\}$$

$$a = C(222,202)\begin{vmatrix} k_{22}P_{22}(X_1)\exp(i2\alpha_1) & k_{22}P_{22}(X_1)\exp(-i2\alpha_1) \\ k_{20}P_{20}(X_2) & + \quad k_{20}P_{20}(X_2) \\ k_{22}P_{22}(X)\exp(-i2\varphi) & k_{22}P_{22}(X)\exp(i2\varphi) \end{vmatrix} \quad \text{(A3.38)}$$

substituting Eq A3.6.c into Eq A3.38 we get

$$a = C(222,202)k_{22}^2\, k_{20}P_{22}(X_1)P_{20}(X_2)P_{22}(X)\{2\cos(2\alpha_1-2\varphi)\} \quad \text{(A3.39)}$$

$$= C(222,022)\begin{vmatrix} k_{20}P_{20}(X_1) & k_{20}P_{20}(X_1) \\ k_{22}P_{22}(X_2)\exp(i2\alpha_2) & + k_{22}P_{22}(X_2)\exp(-i2\alpha_2) \\ k_{22}P_{22}(X)\exp(-i2\varphi) & k_{22}P_{22}(X)\exp(i2\varphi) \end{vmatrix} \quad \text{(A3.40)}$$

ubstituting Eq A3.6.c into Eq A3.40 we get

$$b = C(222,022)k_{22}^2\, k_{20}P_{20}(X_1)P_{22}(X_2)P_{22}(X)\{2\cos(2\alpha_2-2\varphi)\} \quad (A3.41)$$

$$c = C(222,112)\begin{vmatrix} k_{21}P_{21}(X_1)\exp(i\alpha_1) & k_{21}P_{21}(X_1)\exp(-i\alpha_1) \\ k_{21}P_{21}(X_2)\exp(i\alpha_2) & + \; k_{21}P_{21}(X_2)\exp(-i\alpha_2) \\ k_{22}P_{22}(X)\exp(-i2\varphi) & k_{22}P_{22}(X)\exp(i2\varphi) \end{vmatrix} \quad (A3.42)$$

substituting Eq A3.6.c into Eq A3.42 we get

$$c = C(222,112)k_{21}^2\, k_{22}P_{21}(X_1)P_{21}(X_2)P_{22}(X)\{2\cos(\alpha_2+\alpha_1-2\varphi)\} \quad (A3.43)$$

For m = 0

$$y = \sum_{m1,m2,0} C(222;m1,m2,0)\; Y_{2m1}(\omega_1)\; Y_{2m2}(\omega_2)\; Y_{20}^*(\omega) \quad (A3.44)$$

$$\begin{aligned} a &= C(222;2,-2,0)\{Y_{2,2}(\omega_1)Y_{2,-2}(\omega_2)Y_{20}^*(\omega) \\ &\qquad + \\ &\qquad Y_{2,-2}(\omega_1)Y_{2,2}(\omega_2)Y_{20}^*(\omega)\;\} \\ b &= C(222;1,-1,0)\{Y_{2,1}(\omega_1)Y_{2,-1}(\omega_2)Y_{20}^*(\omega) \\ &\qquad + \\ &\qquad Y_{2,-1}(\omega_1)Y_{2,1}(\omega_2)Y_{20}^*(\omega)\} \\ c &= C(222;0,0,0)Y_{2,0}(\omega_1)Y_{2,0}(\omega_2)Y_{20}^*(\omega) \end{aligned} \quad (A3.45)$$

$$= C(222,2\text{-}20)\begin{vmatrix} k_{22}P_{22}(X_1)\exp(i2\alpha_1) & k_{22}P_{22}(X_1)\exp(-i2\alpha_1) \\ k_{22}P_{22}(X_2)\exp(-i2\alpha_2) & + \; k_{22}P_{22}(X_2)\exp(i2\alpha_2) \\ k_{20}P_{20}(X) & k_{20}P_{20}(X) \end{vmatrix} \quad (A3.46)$$

substituting Eq A3.6.c into Eq A3.46 we get

$$a = C(222,2\text{-}20)k_{22}^2\, k_{20}P_{22}(X_1)P_{22}(X_2)P_{20}(X)\{2\cos(-2\alpha_2+2\alpha_1)\} \quad (A3.47)$$

$$= C(222,1\text{-}10)\begin{vmatrix} k_{21}P_{21}(X_1)\exp(i\alpha_1) & k_{21}P_{21}(X_1)\exp(-i\alpha_1) \\ -k_{21}P_{21}(X_2)\exp(-i\alpha_2) & + \; k_{21}P_{21}(X_2)\exp(i\alpha_2) \\ k_{20}P_{20}(X) & k_{20}P_{20}(X) \end{vmatrix} \quad (A3.48)$$

substituting Eq A3.6.c into Eq A3.48 we get

$$b = -C(222,1\text{-}10)k_{21}^2\, k_{20}P_{21}(X_1)P_{12}(X_2)P_{20}(X)\{2\cos(-\alpha_2+\alpha_1)\} \quad (A3.49)$$

$$c = C(222;0,0,0)k_{20}^3\, P_{20}(X_1)P_{20}(X_2)P_{20}(X) \quad (A3.50)$$

For $m = \pm 1$

$$X = \sum_{m_1,m_2,1} C(222;m_1,m_2,1)\, Y_{2m_1}(\omega_1)\, Y_{2m_2}(\omega_2)\, Y_{2,1}^*(\omega) + \sum_{m_1,m_2,-1} C(222;m_1,m_2,-1)\, Y_{2m_1}(\omega_1)\, Y_{2m_2}(\omega_2)\, Y_{2,-1}^*(\omega) \quad \text{(A3.51)}$$

$$a = C(222;2,-1,1)\{Y_{2,2}(\omega_1)Y_{2,-1}(\omega_2)Y_{2,1}^*(\omega) + Y_{2,-2}(\omega_1)Y_{2,1}(\omega_2)Y_{2,-1}^*(\omega)\}$$

$$b = C(222;1,0,1)\{Y_{2,1}(\omega_1)Y_{2,0}(\omega_2)Y_{2,1}^*(\omega) + Y_{2,-1}(\omega_1)Y_{2,0}(\omega_2)Y_{2,-1}^*(\omega)$$

$$c = C(222;0,1,1)\{Y_{2,0}(\omega_1)Y_{2,1}(\omega_2)Y_{2,1}^*(\omega) + Y_{2,0}(\omega_1)Y_{2,-1}(\omega_2)Y_{2,-1}^*(\omega)\}$$

$$d = C(222;-1,2,1\{Y_{2,-1}(\omega_1)Y_{2,2}(\omega_2)Y_{2,1}^*(\omega) + Y_{2,1}(\omega_1)Y_{2,-2}(\omega_2)Y_{2,-1}^*(\omega)\} \quad \text{(A3.52)}$$

$$a = C(222,2\text{-}11)\begin{vmatrix} k_{22}P_{22}(X_1)\exp(i2\alpha_1) & k_{22}P_{22}(X_1)\exp(-i2\alpha_1) \\ k_{21}P_{21}(X_2)\exp(-i\alpha_2) & + \; -k_{21}P_{21}(X_2)\exp(i\alpha_2) \\ -k_{21}P_{21}(X)\exp(-i\varphi) & k_{21}P_{21}(X)\exp(i\varphi) \end{vmatrix} \quad \text{(A3.53)}$$

Eq A3.6.c into Eq A3.53 gives

$$a = -C(222,2\text{-}11)k_{21}^2 k_{22}P_{22}(X_1)P_{21}(X_2)P_{21}(X)\{2\cos(2\alpha_1-\alpha_2-\varphi)\} \quad \text{(A3.54)}$$

$$b = C(222,101)\begin{vmatrix} k_{21}P_{21}(X_1)\exp(i\alpha_1) & k_{21}P_{21}(X_1)\exp(-i\alpha_1) \\ k_{20}P_{20}(X_2) & + \quad k_{20}P_{20}(X_2) \\ k_{21}P_{21}(X)\exp(-i\varphi) & k_{21}P_{21}(X)\exp(i\varphi) \end{vmatrix} \quad \text{(A3.55)}$$

Eq A3.6.c into Eq A3.55 gives

$$b = C(222,101)k_{21}^2\, k_{20}P_{21}(X_1)P_{20}(X_2)P_{21}(X)\{2\cos(\alpha_1-\varphi)\} \quad \text{(A3.56)}$$

$$= C(222,011)\begin{vmatrix} k_{20}P_{20}(X_1) & k_{20}P_{20}(X_1) \\ k_{21}P_{21}(X_2)\exp(i\alpha_2) & + \; k_{21}P_{21}(X_2)\exp(-i\alpha_2) \\ k_{21}P_{21}(X)\exp(-i\varphi) & k_{21}P_{21}(X)\exp(i\varphi) \end{vmatrix} \quad \text{(A3.57)}$$

Eq A3.6 c into Eq A3.57 gives

$$c = C(222,011)k_{21}^2\, k_{20}P_{20}(X_1)P_{21}(X_2)P_{21}(X)\{2\cos(\alpha_2-\varphi)\} \quad \text{(A3.58)}$$

$$d = C(222,-121)\begin{vmatrix} k_{21}P_{21}(X_1)\exp(-i\alpha_1) & k_{21}P_{21}(X_1)\exp(i\alpha_1) \\ k_{22}P_{22}(X_2)\exp(2i\alpha_2) & -k_{22}P_{22}(X_2)\exp(-2i\alpha_2) \\ -k_{21}P_{21}(X)\exp(-i\varphi) & k_{21}P_{21}(X)\exp(i\varphi) \end{vmatrix} \quad \text{(A3.59)}$$

Eq A3.6.c into Eq A3.59 gives

$$d = -C(222,-121)k_{21}^2 k_{22}P_{21}(X_1)P_{22}(X_2)P_{21}(X)\{2\cos(2\alpha_2-\alpha_1-\varphi)\} \quad \text{(A3.60)}$$

Substitution of Eqs A3.39, A3.41, A3.43, A3.47, A3.49, A3.50, A3.54, A3.56, A3.58 and A3.60 into A3.39 gives.

$$\begin{aligned} S_{222} = {} & C(222,202)k_{22}^2\, k_{20}P_{22}(X_1)P_{20}(X_2)P_{22}(X)\{2\cos(2\alpha_1-2\varphi)\} \\ & + C(222,022)k_{22}^2\, k_{20}P_{20}(X_1)P_{22}(X_2)P_{22}(X)\{2\cos(2\alpha_2-2\varphi)\} \\ & + C(222,112)k_{21}^2\, k_{22}P_{21}(X_1)P_{12}(X_2)P_{22}(X)\{2\cos(\alpha_2+\alpha_{1-2\varphi})\} \\ & + \\ & C(222,2-20)k_{22}^2\, k_{20}P_{22}(X_1)P_{22}(X_2)P_{20}(X)\{2\cos(-2\alpha_2+2\alpha_1)\} \\ & + C(222,1-10)k_{21}^2\, k_{20}P_{21}(X_1)P_{12}(X_2)P_{20}(X)\{2\cos(-\alpha_2+\alpha_1)\} \\ & + C(222;0,0,0)k_{20}^3\, P_{20}(X_1)P_{20}(X_2)P_{20}(X) \\ & + \\ & - C(222,2-11)k_{21}^2\, k_{22}P_{22}(X_1)P_{21}(X_2)P_{21}(X)\{2\cos(2\alpha_1-\alpha_2-\varphi)\} \\ & - C(222,-121)k_{21}^2\, k_{22}P_{21}(X_1)P_{22}(X_2)P_{21}(X)\{2\cos(2\alpha_2-\alpha_1-\varphi)\} \\ & + C(222,101)k_{21}^2\, k_{20}P_{21}(X_1)P_{20}(X_2)P_{21}(X)\{2\cos(\alpha_1-\varphi)\} \\ & + C(222,011)k_{21}^2\, k_{20}P_{20}(X_1)P_{21}(X_2)P_{21}(X)\{2\cos(\alpha_2-\varphi)\} \end{aligned} \quad \text{(A3.61)}$$

A3.6. DETERMINATION OF S_{224}:-

$$S_{224} = \sum_{m1,m2,m} C(224;m_1,m_2,m)\ Y_{2m1}(\omega_1)\ Y_{2m2}(\omega_2)\ Y_{4m}^*(\omega) \quad \text{(A3.62.a)}$$

from Eq 2.8

when $\ell_1 = 2$ the values of $m_1 = -2,-1,0,1,2$

$\ell_2 = 2$ the value of $m_2 = -2,-1,0,1,2$

$\ell = 4$ the values of $m = -4,-3,-2,-1,0,1,2,3,4$

The values of orders m_1,m_2 and m that make CGC not equal zero are determined from Eq 2.7 and tabulated below

-4	-3	-2	-1	0	1	2	3	4
-2	-2 -1	-2 -1 0	-2 -1 0 1	-2 -1 0 1 2	2 1 0 -1	2 1 0	1 2	2
-2	-1 -2	0 -1 -2	1 0 -1 -2	2 1 0 -1 -2	-1 0 1 2	0 1 2	2 1	2

For $m = \pm 4$

$$x = \sum_{m1,m2,4} C(224;m1,m2,4)\ Y_{2m1}(\omega_1)\ Y_{2m2}(\omega_2)\ Y_{44}^*(\omega) + \sum_{m1,m2,-4} C(224;m1,m2,-4)\ Y_{2m1}(\omega_1)\ Y_{2m2}(\omega_2)\ Y_{4-4}^*(\omega) \quad \text{(A3.62.b)}$$

$$a = C(224;2,2,4)\{Y_{2,2}(\omega_1)Y_{2,2}(\omega_2)Y_{44}^*(\omega) + Y_{2,-2}(\omega_1)Y_{2,-2}(\omega_2)Y_{4-4}^*(\omega)\}$$

$$= C(224,224)\begin{vmatrix} k_{22}P_{22}(X_1)\exp(i2\alpha_1) & k_{22}P_{22}(X_1)\exp(-i2\alpha_1) \\ k_{22}P_{22}(X_2)\exp(i2\alpha_2) & + \ k_{22}P_{22}(X_2)\exp(-i2\alpha_2) \\ k_{44}P_{44}(X)\exp(-i4\varphi) & k_{44}P_{44}(X)\exp(i4\varphi)) \end{vmatrix} \quad \text{(A3.63)}$$

substituting Eq A3.6.c into Eq A3.63 we get

$$= C(224,224)k_{22}^2\, k_{44}P_{22}(X_1)P_{22}(X_2)P_{24}(X)\{2\cos(2\alpha_1+2\alpha_2-4\varphi)\} \quad \text{(A3.64)}$$

For $m = \pm 3$

$$= \sum_{m1,m2,3} C(224;m1,m2,3)\ Y_{2m1}(\omega_1)\ Y_{2m2}(\omega_2)\ Y_{43}^*(\omega) + \sum_{m1,m2,-3} C(224;m1,m2,-3)\ Y_{2m1}(\omega_1)\ Y_{2m2}(\omega_2)\ Y_{4-3}(\omega)$$

$$a = C(224;2,1,3)\{Y_{2,2}(\omega_1)Y_{2,1}(\omega_2)Y_{43}^*(\omega) + Y_{2,-2}(\omega_1)Y_{2,-1}(\omega_2)Y_{4-3}^*(\omega)\}$$

$$b = C(224;1,2,3)\{Y_{2,1}(\omega_1)Y_{2,2}(\omega_2)Y_{43}^*(\omega) + Y_{2,1}(\omega_1)Y_{2,2}(\omega_2)Y_{43}^*(\omega)\}$$

(A3.65)

$$a = C(224,213)\begin{vmatrix} k_{22}P_{22}(X_1)\exp(i2\alpha_1) & k_{22}P_{22}(X_1)\exp(-i2\alpha_1) \\ k_{21}P_{21}(X_2)\exp(i\alpha_2) & + \quad k_{21}P_{21}(X_2)\exp(-i\alpha_2) \\ k_{43}P_{43}(X)\exp(-i3\varphi) & k_{43}P_{43}(X)\exp(i3\varphi) \end{vmatrix} \quad \textbf{(A3.66)}$$

Eq A3.6.c into Eq A3.65 gives

$$a = C(224,213)k_{22}k_{21}k_{43}P_{22}(X_1)P_{21}(X_2)P_{43}(X)\{2\cos(2\alpha_1+\alpha_2-3\varphi)\} \quad (A3.67)$$

$$b = C(224,123)\begin{vmatrix} k_{21}P_{21}(X_1)\exp(i\alpha_1) & k_{21}P_{21}(X_1)\exp(-i\alpha_1) \\ k_{22}P_{22}(X_2)\exp(2i\alpha_2) & + \quad k_{22}P_{22}(X_2)\exp(-i2\alpha_2) \\ k_{43}P_{43}(X)\exp(-i3\varphi) & k_{43}P_{43}(X)\exp(i3\varphi) \end{vmatrix} \quad (A3.68)$$

Eq A3.6.c into Eq A3.68 gives

$$b = C(224,213)k_{21}k_{22}k_{43}P_{21}(X_1)P_{22}(X_2)P_{43}(X)\{2\cos(\alpha_1+2\alpha_2-3\varphi)\} \quad (A3.69)$$

For $m = \pm 2$

$$= \sum_{m1,m2,2} C(224;m1,m2,2)\ Y_{2m1}(\omega_1)\ Y_{2m2}(\omega_2)\ Y_{42}^*(\omega) + \sum_{m1,m2,-2} C(224;m1,m2,-2)\ Y_{2m1}(\omega_1)\ Y_{2m2}(\omega_2)\ Y_{4-2}^*(\omega) \quad (A3.70)$$

$$a = C(224;2,0,2)\{Y_{2,2}(\omega_1)Y_{2,0}(\omega_2)Y_{42}^*(\omega) + Y_{2,-2}(\omega_1)Y_{2,0}(\omega_2)Y_{4-2}^*(\omega)\}$$

$$b = C(224;0,2,2)\{Y_{2,0}(\omega_1)Y_{2,2}(\omega_2)Y_{42}^*(\omega) + Y_{2,0}(\omega_1)Y_{2,-2}(\omega_2)Y_{4-2}^*(\omega)\}$$

$$c = C(224;1,1,2)\{Y_{2,1}(\omega_1)Y_{2,1}(\omega_2)Y_{42}^*(\omega) + Y_{2,-1}(\omega_1)Y_{2,-1}(\omega_2)Y_{4-2}^*(\omega)\} \quad (A3.71)$$

$$a = C(224,202)\begin{vmatrix} k_{22}P_{22}(X_1)\exp(i2\alpha_1) & k_{22}P_{22}(X_1)\exp(-i2\alpha_1) \\ k_{20}P_{20}(X_2) & + \quad k_{20}P_{20}(X_2) \\ k_{42}P_{42}(X)\exp(-i2\varphi) & k_{42}P_{42}(X)\exp(i2\varphi) \end{vmatrix} \quad (A3.72)$$

substituting Eq A3.6.c into Eq A3.72 we get

$$a = C(224,202)k_{22}k_{20}k_{42}P_{22}(X_1)P_{20}(X_2)P_{42}(X)\{2\cos(2\alpha_1-2\varphi)\} \quad (A3.73)$$

$$b = C(224,022)\begin{vmatrix} k_{20}P_{20}(X_1) & k_{20}P_{20}(X_1) \\ k_{22}P_{22}(X_2)\exp(i2\alpha_2) & + k_{22}P_{22}(X_2)\exp(-i2\alpha_2) \\ k_{24}P_{24}(X)\exp(-i2\varphi) & k_{42}P_{42}(X)\exp(i2\varphi) \end{vmatrix} \quad (A3.74)$$

substituting Eq A3.6.c into Eq A3.74 we get

$$b = C(224,022)k_{20}k_{22}k_{42}P_{20}(X_1)P_{22}(X_2)P_{42}(X)\{2\cos(2\alpha_2-2\varphi)\} \quad (A3.75)$$

$$c = C(224,112)\begin{vmatrix} k_{21}P_{21}(X_1)\exp(i\alpha_1) & k_{21}P_{21}(X_1)\exp(-i\alpha_1) \\ k_{21}P_{21}(X_2)\exp(i\alpha_2) & + k_{21}P_{21}(X_2)\exp(-i\alpha_2) \\ k_{42}P_{42}(X)\exp(-i2\varphi) & k_{42}P_{42}(X)\exp(i2\varphi) \end{vmatrix} \quad (A3.76)$$

substituting Eq A3.6.c into Eq A3.76 we get

$$= C(224,112)k_{21}^2\, k_{42}P_{21}(X_1)P_{21}(X_2)P_{42}(X)\{2\cos(\alpha_2+\alpha_1-2\varphi)\} \quad (A3.77)$$

For $m = \pm 1$

$$= \sum_{m1,m2,1} C(224;m_1,m_2,1)Y_{2m1}(\omega_1)\, Y_{2m2}(\omega_2)\, Y_{4,1}^*(\omega) + \sum_{m1,m2,-1} C(224;m_1,m_2,-1)Y_{2m1}(\omega_1)\, Y_{2m2}(\omega_2)\, Y_{4,-1}^*(\omega) \quad (A3.78)$$

$$a = C(224;2,-1,1)\{Y_{2,2}(\omega_1)Y_{2,-1}(\omega_2)Y_{4,1}^*(\omega) + Y_{2,-2}(\omega_1)Y_{2,1}(\omega_2)Y_{4,-1}^*(\omega)\}$$

$$b = C(224;1,0,1)\{Y_{2,1}(\omega_1)Y_{2,0}(\omega_2)Y_{4,1}^*(\omega) + Y_{2,-1}(\omega_1)Y_{2,0}(\omega_2)Y_{4,-1}^*(\omega)\}$$

$$c = C(224;-1,2,1)\{Y_{2,-1}(\omega_1)Y_{2,2}(\omega_2)Y_{4,1}^*(\omega) + Y_{2,1}(\omega_1)Y_{2,-2}(\omega_2)Y_{4,-1}^*(\omega)\}$$

$$d = C(224;0,1,1)\{Y_{2,0}(\omega_1)Y_{2,1}(\omega_2)Y_{4,1}^*(\omega) + Y_{2,0}(\omega_1)Y_{2,-1}(\omega_2)Y_{4,-1}^*(\omega)\} \quad (A3.79)$$

$$a = C(224,2\text{-}11)\begin{vmatrix} k_{22}P_{22}(X_1)\exp(i2\alpha_1) & k_{22}P_{22}(X_1)\exp(-i2\alpha_1) \\ k_{21}P_{21}(X_2)\exp(-i\alpha_2) & + \; -k_{21}P_{21}(X_2)\exp(i\alpha_2) \\ -k_{41}P_{41}(X)\exp(-i\varphi) & k_{41}P_{41}(X)\exp(i\varphi) \end{vmatrix} \quad \text{(A3.80)}$$

Eq A3.6.c into Eq A3.80 gives

$$a = -C(224,2\text{-}11)k_{21}k_{22}k_{41}P_{22}(X_1)P_{21}(X_2)P_{41}(X)\{2\cos(2\alpha_1-\alpha_2-\varphi)\} \quad \text{(A3.81)}$$

$$b = C(224,101)\begin{vmatrix} k_{21}P_{21}(X_1)\exp(i\alpha_1) & k_{21}P_{21}(X_1)\exp(-i\alpha_1) \\ k_{20}P_{20}(X_2) & + \quad k_{20}P_{20}(X_2) \\ k_{41}P_{41}(X)\exp(-i\varphi) & k_{41}P_{41}(X)\exp(i\varphi) \end{vmatrix} \quad \text{(A3.82)}$$

Eq A3.6.c into Eq A3.82 gives

$$b = C(224,101)k_{21}k_{20}k_{41}P_{21}(X_1)P_{20}(X_2)P_{41}(X)\{2\cos(\alpha_1-\varphi)\} \quad \text{(A3.83)}$$

$$c = C(224,\text{-}121)\begin{vmatrix} k_{21}P_{21}(X_1)\exp(-i\alpha_1) & -k_{21}P_{21}(X_1)\exp(i\alpha_1) \\ k_{22}P_{22}(X_2)\exp(2i\alpha_2) & + \; k_{22}P_{22}(X_2)\exp(-2i\alpha_2) \\ -k_{41}P_{41}(X)\exp(-i\varphi) & k_{41}P_{41}(X)\exp(i\varphi) \end{vmatrix} \quad \text{(A3.84)}$$

Eq A3.6.c into Eq A3.84 gives

$$c = -C(222,\text{-}121)k_{21}k_{22}k_{41}P_{21}(X_1)P_{22}(X_2)P_{41}(X)\{2\cos(2\alpha_2-\alpha_1-\varphi)\} \quad \text{(A3.85)}$$

$$d = C(224,011)\begin{vmatrix} k_{20}P_{20}(X_1) & k_{20}P_{20}(X_1) \\ k_{21}P_{21}(X_2)\exp(i\alpha_2) & + \; k_{21}P_{21}(X_2)\exp(-i\alpha_2) \\ k_{41}P_{41}(X)\exp(-i\varphi) & k_{41}P_{41}(X)\exp(i\varphi) \end{vmatrix} \quad \text{(A3.86)}$$

Eq A3.6.c into Eq A3.86 gives

$$d = C(224,011)k_{20}k_{21}k_{41}P_{20}(X_1)P_{21}(X_2)P_{21}(X)\{2\cos(\alpha_2-\varphi)\} \quad \text{(A3.87)}$$

For m = 0

$$= \Sigma_{m1,m2,o}\, C(224;m1,m2,0)\; Y_{2m1}(\omega_1)\; Y_{2m2}(\omega_2)\; Y_{40}^{*}(\omega) \quad \text{(A3.88)}$$

$$a = C(224;2,-2,0)\{Y_{2,2}(\omega_1)Y_{2,-2}(\omega_2)Y_{40}^{*}(\omega) + Y_{2,-2}(\omega_1)Y_{2,2}(\omega_2)Y_{40}^{*}(\omega)\}$$

$$b = C(224;1,-1,0)\{Y_{2,1}(\omega_1)Y_{2,-1}(\omega_2)Y_{40}^*(\omega) + Y_{2,-1}(\omega_1)Y_{2,1}(\omega_2)Y_{40}^*(\omega)\}$$

$$c = C(224;0,0,0)Y_{2,0}(\omega_1)Y_{2,0}(\omega_2)Y_{40}^*(\omega) \qquad \text{(A3.89)}$$

$$a = C(224,2\text{-}20)\begin{vmatrix} k_{22}P_{22}(X_1)\exp(i2\alpha_1) & k_{22}P_{22}(X_1)\exp(-i2\alpha_1) \\ k_{22}P_{22}(X_2)\exp(-i2\alpha_2) & + \; k_{22}P_{22}(X_2)\exp(i2\alpha_2) \\ k_{40}P_{40}(X) & k_{40}P_{40}(X) \end{vmatrix} \qquad \text{(A3.90)}$$

substituting Eq A3.6.c into Eq A3.90 we get

$$a = C(224,2\text{-}20)k_{22}^2 k_{40}P_{22}(X_1)P_{22}(X_2)P_{40}(X)\{2\cos(-2\alpha_2+2\alpha_1)\} \qquad \text{(A3.91)}$$

$$b = C(224,1\text{-}10)\begin{vmatrix} k_{21}P_{21}(X_1)\exp(i\alpha_1) & k_{21}P_{21}(X_1)\exp(-i\alpha_1) \\ -k_{21}P_{21}(X_2)\exp(-i\alpha_2) & + \; k_{21}P_{21}(X_2)\exp(i\alpha_2) \\ k_{40}P_{40}(X) & k_{40}P_{40}(X) \end{vmatrix} \qquad \text{(A3.92)}$$

substituting Eq A3.6.c into Eq A3.92 we get

$$b = -C(224,1\text{-}10)k_{21}^2\, k_{40}P_{21}(X_1)P_{12}(X_2)P_{40}(X)\{2\cos(-\alpha_2+\alpha_1)\} \qquad \text{(A3.93)}$$

$$c = C(224;0,0,0)k_{20}^2\, k_{40}P_{20}(X_1)P_{20}(X_2)P_{40}(X) \qquad \text{(A3.94)}$$

Substitution of Eqs A3.67, A3.69, A3.73, A3.75, A3.77, A3.81, A3.83, A3.85, A3.87, A3.91, A3.93, A3.94 into A3.62.a gives β_{224}.

$$
\begin{aligned}
S_{224} = {} & C(224,224)k_{22}^{2}\, k_{44}P_{22}(X_1)P_{22}(X_2)P_{24}(X)\{2\cos(2\alpha_1+2\alpha_2-4\varphi)\} \\
& + \\
& C(224,213)k_{22}k_{21}k_{43}P_{22}(X_1)P_{21}(X_2)P_{43}(X)\{2\cos(2\alpha_1+\alpha_2-3\varphi)\} \\
& + \\
& C(224,213)k_{21}k_{22}k_{43}P_{21}(X_1)P_{22}(X_2)P_{43}(X)\{2\cos(\alpha_1+2\alpha_2-3\varphi)\} \\
& + \\
& C(224,202)k_{22}k_{20}k_{42}P_{22}(X_1)P_{20}(X_2)P_{42}(X)\{2\cos(2\alpha_1-2\varphi)\} \\
& + \\
& C(224,022)k_{20}k_{22}k_{42}P_{20}(X_1)P_{22}(X_2)P_{42}(X)\{2\cos(2\alpha_2-2\varphi)\} \\
& + \\
& C(224,112)k_{21}^{2}\, k_{42}P_{21}(X_1)P_{21}(X_2)P_{42}(X)\{2\cos(\alpha_2+\alpha_1-2\varphi)\} \\
& + \\
- {} & C(224,2\text{-}11)k_{21}k_{22}k_{41}P_{22}(X_1)P_{21}(X_2)P_{41}(X)\{2\cos(2\alpha_1-\alpha_2-\varphi)\} \\
& + \\
& C(224,101)k_{21}k_{20}k_{41}P_{21}(X_1)P_{20}(X_2)P_{41}(X)\{2\cos(\alpha_1-\varphi)\} \\
& + \\
- {} & C(222,\text{-}121)k_{21}k_{22}k_{41}P_{21}(X_1)P_{22}(X_2)P_{41}(X)\{2\cos(2\alpha_2-\alpha_1-\varphi)\} \\
& + \\
& C(224,011)k_{20}k_{21}k_{41}P_{20}(X_1)P_{21}(X_2)P_{21}(X)\{2\cos(\alpha_2-\varphi)\} \\
& + \\
& C(224,2\text{-}20)k_{22}^{2}k_{40}P_{22}(X_1)P_{22}(X_2)P_{40}(X)\{2\cos(-2\alpha_2+2\alpha_1)\} \\
& + \\
- {} & C(224,1\text{-}10)k_{21}^{2}\, k_{40}P_{21}(X_1)P_{12}(X_2)P_{40}(X)\{2\cos(-\alpha_2+\alpha_1)\} \\
& + \\
& C(224;0,0,0)k_{20}^{2}\, k_{40}P_{20}(X_1)P_{20}(X_2)P_{40}(X)
\end{aligned}
$$

(A3.95)

Some of the Higher Order Spherical Harmonics calculated following the same above procedure.

$$
\begin{aligned}
S_{422} = \; & C(422;4,-2,2)k_{44}k_{22}^{2}P_{44}(X_1)P_{22}(X_2)P_{22}(X)\{2\cos(4\alpha_1-2\alpha_2-2\varphi)\} \\
& - C(422;3,-1,2)k_{43}k_{21}k_{22}P_{43}(X_1)P_{21}(X_2)P_{22}(X)\{2\cos(3\alpha_1-\alpha_2-2\varphi)\} \\
& + C(422;2,0,2)k_{42}k_{20}k_{22}P_{42}(X_1)P_{20}(X_2)P_{22}(X)\{2\cos(2\alpha_1-2\varphi)\} \\
& + C(422;1,1,2)k_{41}k_{21}k_{22}P_{41}(X_1)P_{21}(X_2)P_{22}(X)\{2\cos(\alpha_1+\alpha_2-2\varphi)\} \\
& + C(422;0,2,2)k_{40}k_{22}k_{22}P_{40}(X_1)P_{22}(X_2)P_{22}(X)\{2\cos(2\alpha_2-2\varphi)\} \\
& + C(422;3,-2,1)k_{43}k_{22}k_{21}P_{43}(X_1)P_{22}(X_2)P_{21}(X)\{2\cos(3\alpha_1-2\alpha_2-\varphi)\} \\
& - C(422;2,-1,1)k_{42}k_{21}k_{21}P_{42}(X_1)P_{21}(X_2)P_{21}(X)\{2\cos(2\alpha_1-\alpha_2-\varphi)\} \\
& + C(422;1,0,1)k_{41}k_{20}k_{21}P_{41}(X_1)P_{20}(X_2)P_{21}(X)\{2\cos(\alpha_1-\varphi)\} \\
& + C(422;0,1,1)k_{40}k_{21}k_{21}P_{40}(X_1)P_{21}(X_2)P_{21}(X)\{2\cos(\alpha_2-\varphi)\} \\
& - C(422;-1,2,1)k_{41}k_{22}k_{21}P_{41}(X_1)P_{22}(X_2)P_{21}(X)\{2\cos(-\alpha_1+2\alpha_2-\varphi)\} \\
& + C(422;2,-2,0)k_{42}k_{22}k_{20}P_{42}(X_1)P_{22}(X_2)P_{20}(X)\{2\cos(2\alpha_1-2\alpha_2)\} \\
& - C(422;1,-1,0)k_{41}k_{21}k_{20}P_{41}(X_1)P_{21}(X_2)P_{20}(X)\{2\cos(\alpha_1-\alpha_2)\} \\
& + C(422;0,0,0)k_{40}k_{20}k_{20}P_{40}(X_1)P_{20}(X_2)P_{20}(X)
\end{aligned}
\tag{A3.96}
$$

$$
\begin{aligned}
S_{424} = \; & C(424;4,0,4)k_{44}k_{20}k_{44}P_{44}(X_1)P_{20}(X_2)P_{44}(X)\{2\cos(4\alpha_1-4\varphi)\} \\
& + C(424;3,1,4)k_{43}k_{21}k_{44}P_{43}(X_1)P_{21}(X_2)P_{44}(X)\{2\cos(3\alpha_1+\alpha_2-4\varphi)\} \\
& + C(424;2,2,4)k_{42}k_{22}k_{44}P_{42}(X_1)P_{22}(X_2)P_{44}(X)\{2\cos(2\alpha_1+2\alpha_2-4\varphi)\} \\
& - C(424;4,-1,3)k_{44}k_{21}k_{43}P_{44}(X_1)P_{21}(X_2)P_{43}(X)\{2\cos(4\alpha_1-\alpha_2-3\varphi)\} \\
& + C(424;3,0,3)k_{43}k_{20}k_{43}P_{43}(X_1)P_{20}(X_2)P_{43}(X)\{2\cos(3\alpha_1-3\varphi)\} \\
& + C(424;2,1,3)k_{42}k_{21}k_{43}P_{42}(X_1)P_{21}(X_2)P_{43}(X)\{2\cos(2\alpha_1+\alpha_2-3\varphi)\} \\
& + C(424;1,2,3)k_{41}k_{22}k_{43}P_{41}(X_1)P_{22}(X_2)P_{43}(X)\{2\cos(\alpha_1+2\alpha_2-3\varphi)\} \\
& + C(424;4,-2,2)k_{44}k_{22}k_{42}P_{44}(X_1)P_{22}(X_2)P_{42}(X)\{2\cos(4\alpha_1-2\alpha_2-2\varphi)\} \\
& - C(424;3,-1,2)k_{43}k_{21}k_{42}P_{43}(X_1)P_{21}(X_2)P_{42}(X)\{2\cos(3\alpha_1-\alpha_2-2\varphi)\} \\
& + C(424;2,0,2)k_{42}k_{20}k_{42}P_{42}(X_1)P_{20}(X_2)P_{42}(X)\{2\cos(2\alpha_1-2\varphi)\}
\end{aligned}
$$

$$
\begin{aligned}
&+ C(424;1,1,2)k_{41}k_{21}k_{42}P_{41}(X_1)P_{21}(X_2)P_{42}(X)\{2\cos(\alpha_1+\alpha_2-2\varphi)\} \\
&+ C(424;0,2,2)k_{40}k_{22}k_{42}P_{40}(X_1)P_{22}(X_2)P_{42}(X)\{2\cos(2\alpha_2-2\varphi)\} \\
&+ C(424;3,-2,1)k_{43}k_{22}k_{41}P_{43}(X_1)P_{22}(X_2)P_{41}(X)\{2\cos(3\alpha_1-2\alpha_2-\varphi)\} \\
&- C(424;2,-1,1)k_{42}k_{21}k_{41}P_{42}(X_1)P_{21}(X_2)P_{41}(X)\{2\cos(2\alpha_1-\alpha_2-\varphi)\} \\
&+ C(424;1,0,1)k_{41}k_{20}k_{41}P_{41}(X_1)P_{20}(X_2)P_{41}(X)\{2\cos(\alpha_1-\varphi)\} \\
&+ C(424;0,1,1)k_{40}k_{21}k_{41}P_{40}(X_1)P_{21}(X_2)P_{41}(X)\{2\cos(\alpha_2-\varphi)\} \\
&- C(424;-1,2,1)k_{41}k_{22}k_{41}P_{41}(X_1)P_{22}(X_2)P_{41}(X)\{2\cos(-\alpha_1+2\alpha_2-\varphi)\} \\
&+ C(424;2,-2,0)k_{42}k_{22}k_{40}P_{42}(X_1)P_{22}(X_2)P_{40}(X)\{2\cos(2\alpha_1-2\alpha_2)\} \\
&- C(424;1,-1,0)k_{41}k_{21}k_{40}P_{41}(X_1)P_{21}(X_2)P_{40}(X)\{2\cos(\alpha_1-\alpha_2)\} \\
&+ C(424;0,0,0)k_{40}k_{20}k_{40}P_{40}(X_1)P_{20}(X_2)P_{40}(X)
\end{aligned}
\qquad (A3.97)
$$

$$
\begin{aligned}
S_{426} = \; & C(426;4,2,6)k_{44}k_{22}k_{66}P_{44}(X_1)P_{22}(X_2)P_{66}(X)\{2\cos(4\alpha_1+2\alpha_2-6\varphi)\} \\
&+ C(426;4,1,5)k_{44}k_{21}k_{65}P_{44}(X_1)P_{21}(X_2)P_{65}(X)\{2\cos(4\alpha_1+1\alpha_2-5\varphi)\} \\
&+ C(426;3,2,5)k_{43}k_{22}k_{65}P_{43}(X_1)P_{22}(X_2)P_{65}(X)\{2\cos(3\alpha_1+2\alpha_2-5\varphi)\} \\
&+ C(426;4,0,4)k_{44}k_{20}k_{64}P_{44}(X_1)P_{20}(X_2)P_{64}(X)\{2\cos(4\alpha_1-4\varphi)\} \\
&+ C(426;3,1,4)k_{43}k_{21}k_{64}P_{43}(X_1)P_{21}(X_2)P_{64}(X)\{2\cos(3\alpha_1+\alpha_2-4\varphi)\} \\
&+ C(426;2,2,4)k_{42}k_{22}k_{64}P_{42}(X_1)P_{22}(X_2)P_{64}(X)\{2\cos(2\alpha_1+2\alpha_2-4\varphi)\} \\
&- C(426;4,-1,3)k_{44}k_{21}k_{63}P_{44}(X_1)P_{21}(X_2)P_{63}(X)\{2\cos(4\alpha_1-\alpha_2-3\varphi)\} \\
&+ C(426;3,0,3)k_{43}k_{20}k_{63}P_{43}(X_1)P_{20}(X_2)P_{63}(X)\{2\cos(3\alpha_1-3\varphi)\} \\
&+ C(426;2,1,3)k_{42}k_{21}k_{63}P_{42}(X_1)P_{21}(X_2)P_{63}(X)\{2\cos(2\alpha_1+\alpha_2-3\varphi)\} \\
&+ C(426;1,2,3)k_{41}k_{22}k_{63}P_{41}(X_1)P_{22}(X_2)P_{63}(X)\{2\cos(\alpha_1+2\alpha_2-3\varphi)\} \\
&+ C(426;4,-2,2)k_{44}k_{22}k_{62}P_{44}(X_1)P_{22}(X_2)P_{62}(X)\{2\cos(4\alpha_1-2\alpha_2-2\varphi)\} \\
&- C(426;3,-1,2)k_{43}k_{21}k_{62}P_{43}(X_1)P_{21}(X_2)P_{62}(X)\{2\cos(3\alpha_1-\alpha_2-2\varphi)\} \\
&+ C(426;2,0,2)k_{42}k_{20}k_{62}P_{42}(X_1)P_{20}(X_2)P_{62}(X)\{2\cos(2\alpha_1-2\varphi)\} \\
&+ C(426;1,1,2)k_{41}k_{21}k_{62}P_{41}(X_1)P_{21}(X_2)P_{62}(X)\{2\cos(\alpha_1+\alpha_2-2\varphi)\} \\
&+ C(426;0,2,2)k_{40}k_{22}k_{62}P_{40}(X_1)P_{22}(X_2)P_{62}(X)\{2\cos(2\alpha_2-2\varphi)\}
\end{aligned}
$$

$$+ C(426;3,-2,1)k_{43}k_{22}k_{61}P_{43}(X_1)P_{22}(X_2)P_{61}(X)\{2\cos(3\alpha_1-2\alpha_2-\varphi)\}$$
$$+ C(426;2,-1,1)k_{42}k_{21}k_{61}P_{42}(X_1)P_{21}(X_2)P_{61}(X)\{2\cos(2\alpha_1-\alpha_2-\varphi)\}$$
$$+ C(426;1,0,1)k_{41}k_{20}k_{61}P_{41}(X_1)P_{20}(X_2)P_{61}(X)\{2\cos(\alpha_1-\varphi)\}$$
$$+ C(426;0,1,1)k_{40}k_{21}k_{61}P_{40}(X_1)P_{21}(X_2)P_{61}(X)\{2\cos(\alpha_2-\varphi)\}$$
$$+ C(426;-1,2,1)k_{41}k_{22}k_{61}P_{41}(X_1)P_{22}(X_2)P_{61}(X)\{2\cos(-\alpha_1+2\alpha_2-\varphi)\}$$
$$+ C(426;2,-2,0)k_{42}k_{22}k_{60}P_{42}(X_1)P_{22}(X_2)P_{60}(X)\{2\cos(2\alpha_1-2\alpha_2)\}$$
$$+ C(426;1,-1,0)k_{41}k_{21}k_{60}P_{41}(X_1)P_{21}(X_2)P_{60}(X)\{2\cos(\alpha_1-\alpha_2)\}$$
$$+ C(426;0,0,0)k_{40}k_{20}k_{60}P_{40}(X_1)P_{20}(X_2)P_{60}(X) \qquad \text{(A3.98)}$$

$$S_{242} = C(242;2,0,2)k_{22}k_{40}k_{22}P_{22}(X_1)P_{40}(X_2)P_{22}(X)\{2\cos(2\alpha_1-2\varphi)\}$$
$$+ C(242;1,1,2)k_{21}k_{41}k_{22}P_{21}(X_1)P_{41}(X_2)P_{22}(X)\{2\cos(\alpha_1+\alpha_2-2\varphi)\}$$
$$+ C(242;0,2,2)k_{20}k_{42}k_{22}P_{20}(X_1)P_{42}(X_2)P_{22}(X)\{2\cos(2\alpha_2-2\varphi)\}$$
$$- C(242;-1,3,2)k_{21}k_{43}k_{22}P_{21}(X_1)P_{43}(X_2)P_{22}(X)\{2\cos(3\alpha_2-\alpha_1-2\varphi)\}$$
$$+ C(242;-2,4,2)k_{22}k_{44}k_{22}P_{22}(X_1)P_{44}(X_2)P_{22}(X)\{2\cos(4\alpha_2-2\alpha_1-2\varphi)\}$$
$$- C(242;2,-1,1)k_{22}k_{41}k_{21}P_{22}(X_1)P_{41}(X_2)P_{21}(X)\{2\cos(2\alpha_1-\alpha_2-\varphi)\}$$
$$+ C(242;1,0,1)k_{21}k_{40}k_{21}P_{21}(X_1)P_{40}(X_2)P_{21}(X)\{2\cos(\alpha_1-\varphi)\}$$
$$+ C(242;0,1,1)k_{20}k_{41}k_{21}P_{20}(X_1)P_{41}(X_2)P_{21}(X)\{2\cos(\alpha_2-\varphi)\}$$
$$- C(242;-1,2,1)k_{21}k_{42}k_{21}P_{21}(X_1)P_{42}(X_2)P_{21}(X)\{2\cos(2\alpha_2-\alpha_1-\varphi)\}$$
$$+ C(242;-2,3,1)k_{22}k_{43}k_{21}P_{22}(X_1)P_{43}(X_2)P_{21}(X)\{2\cos(3\alpha_2-2\alpha_1-\varphi)\}$$
$$+ C(242;2,-2,0)k_{22}k_{42}k_{20}P_{22}(X_1)P_{42}(X_2)P_{20}(X)\{2\cos(2\alpha_1-2\alpha_2)\}$$
$$- C(242;1,-1,0)k_{21}k_{41}k_{20}P_{21}(X_1)P_{41}(X_2)P_{20}(X)\{2\cos(\alpha_1-\alpha_2)\}$$
$$+ C(242;0,0,0)k_{20}k_{40}k_{20}P_{20}(X_1)P_{40}(X_2)P_{20}(X) \qquad \text{(A3.99)}$$

$$_4 = C(244;2,2,4)k_{22}k_{42}k_{44}P_{22}(X_1)P_{42}(X_2)P_{44}(X)\{2\cos(2\alpha_1+2\alpha_2-4\varphi)\}$$
$$+ C(244;1,3,4)k_{21}k_{43}k_{44}P_{21}(X_1)P_{43}(X_2)P_{44}(X)\{2\cos(\alpha_1+3\alpha_2-4\varphi)\}$$
$$+ C(244;0,4,4)k_{20}k_{44}k_{44}P_{20}(X_1)P_{44}(X_2)P_{44}(X)\{2\cos(4\alpha_2-4\varphi)\}$$

$$
\begin{aligned}
&+ C(244;2,1,3)k_{22}k_{41}k_{43}P_{22}(X_1)P_{41}(X_2)P_{43}(X)\{2\cos(2\alpha_1+\alpha_2-3\varphi)\} \\
&+ C(244;1,2,3)k_{21}k_{42}k_{43}P_{21}(X_1)P_{42}(X_2)P_{43}(X)\{2\cos(\alpha_1+2\alpha_2-3\varphi)\} \\
&+ C(244;0,3,3)k_{20}k_{43}k_{43}P_{20}(X_1)P_{43}(X_2)P_{43}(X)\{2\cos(3\alpha_2-3\varphi)\} \\
&- C(244;-1,4,3)k_{21}k_{44}k_{43}P_{21}(X_1)P_{44}(X_2)P_{43}(X)\{2\cos(-\alpha_1+4\alpha_2-3\varphi)\} \\
&+ C(244;2,0,2)k_{22}k_{40}k_{42}P_{22}(X_1)P_{40}(X_2)P_{42}(X)\{2\cos(2\alpha_1-2\varphi)\} \\
&+ C(244;1,1,2)k_{21}k_{41}k_{42}P_{21}(X_1)P_{41}(X_2)P_{42}(X)\{2\cos(\alpha_1+\alpha_2-2\varphi)\} \\
&+ C(244;0,2,2)k_{20}k_{42}k_{42}P_{20}(X_1)P_{42}(X_2)P_{42}(X)\{2\cos(2\alpha_2-2\varphi)\} \\
&- C(244;-1,3,2)k_{21}k_{43}k_{42}P_{21}(X_1)P_{43}(X_2)P_{42}(X)\{2\cos(3\alpha_2-\alpha_1-2\varphi)\} \\
&+ C(244;-2,4,2)k_{22}k_{44}k_{42}P_{22}(X_1)P_{44}(X_2)P_{42}(X)\{2\cos(4\alpha_2-2\alpha_1-2\varphi)\} \\
&- C(244;2,-1,1)k_{22}k_{41}k_{41}P_{22}(X_1)P_{41}(X_2)P_{41}(X)\{2\cos(2\alpha_1-\alpha_2-\varphi)\} \\
&+ C(244;1,0,1)k_{21}k_{40}k_{41}P_{21}(X_1)P_{40}(X_2)P_{41}(X)\{2\cos(\alpha_1-\varphi)\} \\
&+ C(244;0,1,1)k_{20}k_{41}k_{41}P_{20}(X_1)P_{41}(X_2)P_{41}(X)\{2\cos(\alpha_2-\varphi)\} \\
&- C(244;-1,2,1)k_{21}k_{42}k_{41}P_{21}(X_1)P_{42}(X_2)P_{41}(X)\{2\cos(2\alpha_2-\alpha_1-\varphi)\} \\
&+ C(244;-2,3,1)k_{22}k_{43}k_{41}P_{22}(X_1)P_{43}(X_2)P_{41}(X)\{2\cos(3\alpha_2-2\alpha_1-\varphi)\} \\
&+ C(244;2,-2,0)k_{22}k_{42}k_{40}P_{22}(X_1)P_{42}(X_2)P_{40}(X)\{2\cos(2\alpha_1-2\alpha_2)\} \\
&- C(244;1,-1,0)k_{21}k_{41}k_{40}P_{21}(X_1)P_{41}(X_2)P_{40}(X)\{2\cos(\alpha_1-\alpha_2)\} \\
&+ C(244;0,0,0)k_{20}k_{40}k_{40}P_{20}(X_1)P_{40}(X_2)P_{40}(X)
\end{aligned}
\qquad (A3.100)
$$

$$
\begin{aligned}
I_{246} = \; & C(246;2,4,6)k_{22}k_{44}k_{66}P_{22}(X_1)P_{44}(X_2)P_{66}(X)\{2\cos(2\alpha_1+4\alpha_2-6\varphi)\} \\
&+ C(246;2,3,5)k_{22}k_{43}k_{65}P_{22}(X_1)P_{43}(X_2)P_{65}(X)\{2\cos(2\alpha_1+3\alpha_2-5\varphi)\} \\
&+ C(246;1,4,5)k_{21}k_{44}k_{65}P_{21}(X_1)P_{44}(X_2)P_{65}(X)\{2\cos(\alpha_1+4\alpha_2-5\varphi)\} \\
&+ C(246;2,2,4)k_{22}k_{42}k_{64}P_{22}(X_1)P_{42}(X_2)P_{64}(X)\{2\cos(2\alpha_1+2\alpha_2-4\varphi)\} \\
&+ C(246;1,3,4)k_{21}k_{43}k_{64}P_{21}(X_1)P_{43}(X_2)P_{64}(X)\{2\cos(\alpha_1+3\alpha_2-4\varphi)\} \\
&+ C(246;0,4,4)k_{20}k_{44}k_{64}P_{20}(X_1)P_{44}(X_2)P_{64}(X)\{2\cos(4\alpha_2-4\varphi)\} \\
&+ C(246;2,1,3)k_{22}k_{41}k_{63}P_{22}(X_1)P_{41}(X_2)P_{63}(X)\{2\cos(2\alpha_1+\alpha_2-3\varphi)\} \\
&+ C(246;1,2,3)k_{21}k_{42}k_{63}P_{21}(X_1)P_{42}(X_2)P_{63}(X)\{2\cos(\alpha_1+2\alpha_2-3\varphi)\}
\end{aligned}
$$

$$+ C(246;0,3,3)k_{20}k_{43}k_{63}P_{20}(X_1)P_{43}(X_2)P_{63}(X)\{2\cos(3\alpha_2-3\varphi)\}$$
$$+ C(246;-1,4,3)k_{21}k_{44}k_{63}P_{21}(X_1)P_{44}(X_2)P_{63}(X)\{2\cos(-\alpha_1+4\alpha_2-3\varphi)\}$$
$$+ C(246;2,0,2)k_{22}k_{40}k_{62}P_{22}(X_1)P_{40}(X_2)P_{62}(X)\{2\cos(2\alpha_1-2\varphi)\}$$
$$+ C(246;1,1,2)k_{21}k_{41}k_{62}P_{21}(X_1)P_{41}(X_2)P_{62}(X)\{2\cos(\alpha_1+\alpha_2-2\varphi)\}$$
$$+ C(246;0,2,2)k_{20}k_{42}k_{62}P_{20}(X_1)P_{42}(X_2)P_{62}(X)\{2\cos(2\alpha_2-2\varphi)\}$$
$$+ C(246;-1,3,2)k_{21}k_{43}k_{62}P_{21}(X_1)P_{43}(X_2)P_{62}(X)\{2\cos(3\alpha_2-\alpha_1-2\varphi)\}$$
$$+ C(246;-2,4,2)k_{22}k_{44}k_{62}P_{22}(X_1)P_{44}(X_2)P_{62}(X)\{2\cos(4\alpha_2-2\alpha_1-2\varphi)\}$$
$$+ C(246;2,-1,1)k_{22}k_{41}k_{61}P_{22}(X_1)P_{41}(X_2)P_{61}(X)\{2\cos(2\alpha_1-\alpha_2-\varphi)\}$$
$$+ C(246;1,0,1)k_{21}k_{40}k_{61}P_{21}(X_1)P_{40}(X_2)P_{61}(X)\{2\cos(\alpha_1-\varphi)\}$$
$$+ C(246;0,1,1)k_{20}k_{41}k_{61}P_{20}(X_1)P_{41}(X_2)P_{61}(X)\{2\cos(\alpha_2-\varphi)\}$$
$$+ C(246;-1,2,1)k_{21}k_{42}k_{61}P_{21}(X_1)P_{42}(X_2)P_{61}(X)\{2\cos(2\alpha_2-\alpha_1-\varphi)\}$$
$$+ C(246;-2,3,1)k_{22}k_{43}k_{61}P_{22}(X_1)P_{43}(X_2)P_{61}(X)\{2\cos(3\alpha_2-2\alpha_1-\varphi)\}$$
$$+ C(246;2,-2,0)k_{22}k_{42}k_{60}P_{22}(X_1)P_{42}(X_2)P_{60}(X)\{2\cos(2\alpha_1-2\alpha_2)\}$$
$$+ C(246;1,-1,0)k_{21}k_{41}k_{60}P_{21}(X_1)P_{41}(X_2)P_{60}(X)\{2\cos(\alpha_1-\alpha_2)\}$$
$$+ C(246;0,0,0)k_{20}k_{40}k_{60}P_{20}(X_1)P_{40}(X_2)P_{60}(X) \qquad (A3.101)$$

$$\beta_{044} = C(044;0,4,4)k_{00}k_{44}k_{44}P_{00}(X_1)P_{44}(X_2)P_{44}(X)\{2\cos(4\alpha_2-4\varphi)\}$$
$$+ C(044;0,3,3)k_{00}k_{43}k_{43}P_{00}(X_1)P_{43}(X_2)P_{43}(X)\{2\cos(3\alpha_2-3\varphi)\}$$
$$+ C(044;0,2,2)k_{00}k_{42}k_{42}P_{00}(X_1)P_{42}(X_2)P_{42}(X)\{2\cos(2\alpha_2-2\varphi)\}$$
$$+ C(044;0,1,1)k_{00}k_{41}k_{41}P_{00}(X_1)P_{41}(X_2)P_{41}(X)\{2\cos(\alpha_2-\varphi)\}$$
$$+ C(044;0,0,0)k_{00}k_{40}k_{40}P_{00}(X_1)P_{40}(X_2)P_{40}(X) \qquad (A3.102)$$

$$_{04} = C(404;4,0,4)k_{44}k_{00}k_{44}P_{44}(X_1)P_{00}(X_2)P_{44}(X)\{2\cos(4\alpha_1-4\varphi)\}$$
$$+ C(404;3,0,3)k_{43}k_{00}k_{43}P_{43}(X_1)P_{00}(X_2)P_{43}(X)\{2\cos(3\alpha_1-3\varphi)\}$$
$$+ C(404;2,0,2)k_{42}k_{00}k_{42}P_{42}(X_1)P_{00}(X_2)P_{42}(X)\{2\cos(2\alpha_1-2\varphi)\}$$
$$+ C(404;1,0,1)k_{41}k_{00}k_{41}P_{41}(X_1)P_{00}(X_2)P_{41}(X)\{2\cos(\alpha_1-\varphi)\}$$
$$+ C(404;0,0,0)k_{40}k_{00}k_{40}P_{40}(X_1)P_{00}(X_2)P_{40}(X) \qquad (A3.103)$$

$$
\begin{aligned}
S_{440} = {} & C(440;-4,4,0)k_{44}k_{44}k_{00}P_{44}(X_1)P_{44}(X_2)P_{00}(X)\{2\cos(4\alpha_2-4\alpha_1)\} \\
& + \\
& - C(440;-3,3,0)k_{43}k_{43}k_{00}P_{43}(X_1)P_{43}(X_2)P_{00}(X)\{2\cos(3\alpha_2-3\alpha_1)\} \\
& + \\
& C(440;-2,2,0)k_{42}k_{42}k_{00}P_{42}(X_1)P_{42}(X_2)P_{00}(X)\{2\cos(2\alpha_2-2\alpha_1)\} \\
& + \\
& - C(440;-1,1,0)k_{41}k_{41}k_{00}P_{41}(X_1)P_{41}(X_2)P_{00}(X)\{2\cos(\alpha_2-\alpha_1)\} \\
& + \\
& C(440;0,0,0)k_{40}k_{40}k_{00}P_{40}(X_1)P_{40}(X_2)P_{00}(X)
\end{aligned}
\qquad (A3.104)
$$

$$
\begin{aligned}
S_{444} = {} & C(444;4,0,4)k_{44}k_{40}k_{44}P_{44}(X_1)P_{40}(X_2)P_{44}(X)\{2\cos(4\alpha_1-4\varphi)\} \\
& + \\
& C(444;3,1,4)k_{43}k_{41}k_{44}P_{43}(X_1)P_{41}(X_2)P_{44}(X)\{2\cos(3\alpha_1+\alpha_2-4\varphi)\} \\
& + \\
& C(444;2,2,4)k_{42}k_{42}k_{44}P_{42}(X_1)P_{42}(X_2)P_{44}(X)\{2\cos(2\alpha_1+2\alpha_2-4\varphi)\} \\
& + \\
& C(444;1,3,4)k_{41}k_{43}k_{44}P_{41}(X_1)P_{43}(X_2)P_{44}(X)\{2\cos(\alpha_1+3\alpha_2-4\varphi)\} \\
& + \\
& C(444;0,4,4)k_{40}k_{44}k_{44}P_{40}(X_1)P_{44}(X_2)P_{44}(X)\{2\cos(4\alpha_2-4\varphi)\} \\
& + \\
& - C(444;4,-1,3)k_{44}k_{41}k_{43}P_{44}(X_1)P_{41}(X_2)P_{43}(X)\{2\cos(4\alpha_1-\alpha_2-3\varphi)\} \\
& + \\
& C(444;3,0,3)k_{43}k_{40}k_{43}P_{43}(X_1)P_{40}(X_2)P_{43}(X)\{2\cos(3\alpha_1-3\varphi)\} \\
& + \\
& C(444;2,1,3)k_{42}k_{41}k_{43}P_{42}(X_1)P_{41}(X_2)P_{43}(X)\{2\cos(2\alpha_1+\alpha_2-3\varphi)\} \\
& + \\
& C(444;1,2,3)k_{41}k_{42}k_{43}P_{41}(X_1)P_{42}(X_2)P_{43}(X)\{2\cos(\alpha_1+2\alpha_2-3\varphi)\} \\
& + \\
& C(444;0,3,3)k_{40}k_{43}k_{43}P_{40}(X_1)P_{43}(X_2)P_{43}(X)\{2\cos(3\alpha_2-3\varphi)\} \\
& + \\
& - C(444;-1,4,3)k_{41}k_{44}k_{43}P_{41}(X_1)P_{44}(X_2)P_{43}(X)\{2\cos(4\alpha_2-\alpha_1-3\varphi)\} \\
& + \\
& C(444;4,-2,2)k_{44}k_{42}k_{42}P_{44}(X_1)P_{42}(X_2)P_{42}(X)\{2\cos(4\alpha_1-2\alpha_2-2\varphi)\} \\
& + \\
& - C(444;3,-1,2)k_{43}k_{41}k_{42}P_{43}(X_1)P_{41}(X_2)P_{42}(X)\{2\cos(3\alpha_1-\alpha_2-2\varphi)\} \\
& + \\
& C(444;2,0,2)k_{42}k_{40}k_{42}P_{42}(X_1)P_{40}(X_2)P_{42}(X)\{2\cos(2\alpha_1-2\varphi)\} \\
& + \\
& C(444;1,1,2)k_{41}k_{41}k_{42}P_{41}(X_1)P_{41}(X_2)P_{42}(X)\{2\cos(\alpha_1+\alpha_2-2\varphi)\} \\
& + \\
& C(444;0,2,2)k_{40}k_{42}k_{42}P_{40}(X_1)P_{42}(X_2)P_{42}(X)\{2\cos(2\alpha_2-2\varphi)\} \\
& + \\
& - C(444;-1,3,2)k_{41}k_{43}k_{42}P_{41}(X_1)P_{43}(X_2)P_{42}(X)\{2\cos(3\alpha_2-\alpha_1-2\varphi)\} \\
& + \\
& C(444;-2,4,2)k_{42}k_{44}k_{42}P_{42}(X_1)P_{44}(X_2)P_{42}(X)\{2\cos(4\alpha_2-2\alpha_1-2\varphi)\} \\
& + \\
& - C(444;4,-3,1)k_{44}k_{43}k_{41}P_{44}(X_1)P_{43}(X_2)P_{41}(X)\{2\cos(4\alpha_1-3\alpha_2-\varphi)\} \\
& + \\
& C(444;3,-2,1)k_{43}k_{42}k_{41}P_{43}(X_1)P_{42}(X_2)P_{41}(X)\{2\cos(3\alpha_1-2\alpha_2-\varphi)\} \\
& +
\end{aligned}
$$

$$
\begin{aligned}
&- C(444;2,-1,1)k_{42}k_{41}k_{41}P_{42}(X_1)P_{41}(X_2)P_{41}(X)\{2\cos(2\alpha_1-\alpha_2-\varphi)\}\\
&+\\
&C(444;1,0,1)k_{41}k_{40}k_{41}P_{41}(X_1)P_{40}(X_2)P_{41}(X)\{2\cos(\alpha_1-\varphi)\}\\
&+\\
&C(444;0,1,1)k_{40}k_{41}k_{41}P_{40}(X_1)P_{41}(X_2)P_{41}(X)\{2\cos(\alpha_2-\varphi)\}\\
&+\\
&- C(444;-1,2,1)k_{41}k_{42}k_{41}P_{41}(X_1)P_{42}(X_2)P_{41}(X)\{2\cos(2\alpha_2-\alpha_1-\varphi)\}\\
&+\\
&C(444;-2,3,1)k_{42}k_{43}k_{41}P_{42}(X_1)P_{43}(X_2)P_{41}(X)\{2\cos(3\alpha_2-2\alpha_1-\varphi)\}\\
&+\\
&-C(444;-3,4,1)k_{43}k_{44}k_{41}P_{43}(X_1)P_{44}(X_2)P_{41}(X)\{2\cos(4\alpha_2-3\alpha_1-\varphi)\}\\
&+\\
&C(444;-4,4,0)k_{44}k_{44}k_{40}P_{44}(X_1)P_{44}(X_2)P_{40}(X)\{2\cos(4\alpha_2-4\alpha_1)\}\\
&+\\
&- C(444;-3,3,0)k_{43}k_{43}k_{40}P_{43}(X_1)P_{43}(X_2)P_{40}(X)\{2\cos(3\alpha_2-3\alpha_1)\}\\
&+\\
&C(444;-2,2,0)k_{42}k_{42}k_{40}P_{42}(X_1)P_{42}(X_2)P_{40}(X)\{2\cos(2\alpha_2-2\alpha_1)\}\\
&+\\
&- C(444;-1,1,0)k_{41}k_{41}k_{40}P_{41}(X_1)P_{41}(X_2)P_{40}(X)\{2\cos(\alpha_2-\alpha_1)\}\\
&+\\
&C(444;0,0,0)k_{40}k_{40}k_{40}P_{40}(X_1)P_{40}(X_2)P_{40}(X)
\end{aligned}
\qquad (A3.105)
$$

$$
\begin{aligned}
S_{446} = \;& C(446;4,2,6)k_{44}k_{42}k_{66}P_{44}(X_1)P_{42}(X_2)P_{66}(X)\{2\cos(4\alpha_1+2\alpha_2-6\varphi)\}\\
&+\\
&C(446;3,3,6)k_{43}k_{43}k_{66}P_{43}(X_1)P_{43}(X_2)P_{66}(X)\{2\cos(3\alpha_1+3\alpha_2-6\varphi)\}\\
&+\\
&C(446;2,4,6)k_{42}k_{44}k_{66}P_{42}(X_1)P_{44}(X_2)P_{66}(X)\{2\cos(2\alpha_1+4\alpha_2-6\varphi)\}\\
&+\\
&C(446;4,1,5)k_{44}k_{41}k_{65}P_{44}(X_1)P_{41}(X_2)P_{65}(X)\{2\cos(4\alpha_1+\alpha_2-5\varphi)\}\\
&+\\
&C(446;3,2,5)k_{43}k_{42}k_{65}P_{43}(X_1)P_{42}(X_2)P_{65}(X)\{2\cos(3\alpha_1+2\alpha_2-5\varphi)\}\\
&+\\
&C(446;2,3,5)k_{42}k_{43}k_{65}P_{42}(X_1)P_{43}(X_2)P_{65}(X)\{2\cos(2\alpha_1+3\alpha_2-5\varphi)\}\\
&+\\
&C(446;1,4,5)k_{41}k_{44}k_{65}P_{41}(X_1)P_{44}(X_2)P_{65}(X)\{2\cos(\alpha_1+4\alpha_2-5\varphi)\}\\
&+\\
&C(446;4,0,4)k_{44}k_{40}k_{64}P_{44}(X_1)P_{40}(X_2)P_{64}(X)\{2\cos(4\alpha_1-4\varphi)\}\\
&+\\
&C(446;3,1,4)k_{43}k_{41}k_{64}P_{43}(X_1)P_{41}(X_2)P_{64}(X)\{2\cos(3\alpha_1+\alpha_2-4\varphi)\}\\
&+\\
&C(446;2,2,4)k_{42}k_{42}k_{64}P_{42}(X_1)P_{42}(X_2)P_{64}(X)\{2\cos(2\alpha_1+2\alpha_2-4\varphi)\}\\
&+\\
&C(446;1,3,4)k_{41}k_{43}k_{64}P_{41}(X_1)P_{43}(X_2)P_{64}(X)\{2\cos(\alpha_1+3\alpha_2-4\varphi)\}\\
&+\\
&C(446;0,4,4)k_{40}k_{44}k_{64}P_{40}(X_1)P_{44}(X_2)P_{64}(X)\{2\cos(4\alpha_2-4\varphi)\}\\
&+\\
&-C(446;4,-1,3)k_{44}k_{41}k_{63}P_{44}(X_1)P_{41}(X_2)P_{63}(X)\{2\cos(4\alpha_1-\alpha_2-3\varphi)\}\\
&+\\
&C(446;3,0,3)k_{43}k_{40}k_{63}P_{43}(X_1)P_{40}(X_2)P_{63}(X)\{2\cos(3\alpha_1-3\varphi)\}\\
&+
\end{aligned}
$$

$$
\begin{aligned}
&C(446;2,1,3)k_{42}k_{41}k_{63}P_{42}(X_1)P_{41}(X_2)P_{63}(X)\{2\cos(2\alpha_1+\alpha_2-3\varphi)\}\\
&+\\
&C(446;1,2,3)k_{41}k_{42}k_{63}P_{41}(X_1)P_{42}(X_2)P_{63}(X)\{2\cos(\alpha_1+2\alpha_2-3\varphi)\}\\
&+\\
&C(446;0,3,3)k_{40}k_{43}k_{63}P_{40}(X_1)P_{43}(X_2)P_{63}(X)\{2\cos(3\alpha_2-3\varphi)\}\\
&+\\
&-C(446;-1,4,3)k_{41}k_{44}k_{63}P_{41}(X_1)P_{44}(X_2)P_{63}(X)\{2\cos(4\alpha_2-\alpha_1-3\varphi)\}\\
&+\\
&C(446;4,-2,2)k_{44}k_{42}k_{62}P_{44}(X_1)P_{42}(X_2)P_{62}(X)\{2\cos(4\alpha_1-2\alpha_2-2\varphi)\}\\
&+\\
&-C(446;3,-1,2)k_{43}k_{41}k_{62}P_{43}(X_1)P_{41}(X_2)P_{62}(X)\{2\cos(3\alpha_1-\alpha_2-2\varphi)\}\\
&+\\
&C(446;2,0,2)k_{42}k_{40}k_{62}P_{42}(X_1)P_{40}(X_2)P_{62}(X)\{2\cos(2\alpha_1-2\varphi)\}\\
&+\\
&C(446;1,1,2)k_{41}k_{41}k_{62}P_{41}(X_1)P_{41}(X_2)P_{62}(X)\{2\cos(\alpha_1+\alpha_2-2\varphi)\}\\
&+\\
&C(446;0,2,2)k_{40}k_{42}k_{62}P_{40}(X_1)P_{42}(X_2)P_{62}(X)\{2\cos(2\alpha_2-2\varphi)\}\\
&+\\
&-C(446;-1,3,2)k_{41}k_{43}k_{62}P_{41}(X_1)P_{43}(X_2)P_{62}(X)\{2\cos(3\alpha_2-\alpha_1-2\varphi)\}\\
&+\\
&C(446;-2,4,2)k_{42}k_{44}k_{62}P_{42}(X_1)P_{44}(X_2)P_{62}(X)\{2\cos(4\alpha_2-2\alpha_1-2\varphi)\}\\
&+\\
&-C(446;4,-3,1)k_{44}k_{43}k_{61}P_{44}(X_1)P_{43}(X_2)P_{61}(X)\{2\cos(4\alpha_1-3\alpha_2-\varphi)\}\\
&+\\
&C(446;3,-2,1)k_{43}k_{42}k_{61}P_{43}(X_1)P_{42}(X_2)P_{61}(X)\{2\cos(3\alpha_1-2\alpha_2-\varphi)\}\\
&+\\
&-C(446;2,-1,1)k_{42}k_{41}k_{61}P_{42}(X_1)P_{41}(X_2)P_{61}(X)\{2\cos(2\alpha_1-\alpha_2-\varphi)\}\\
&+\\
&C(446;1,0,1)k_{41}k_{40}k_{61}P_{41}(X_1)P_{40}(X_2)P_{61}(X)\{2\cos(\alpha_1-\varphi)\}\\
&+\\
&C(446;0,1,1)k_{40}k_{41}k_{61}P_{40}(X_1)P_{41}(X_2)P_{61}(X)\{2\cos(\alpha_2-\varphi)\}\\
&+\\
&-C(446;-1,2,1)k_{41}k_{42}k_{61}P_{41}(X_1)P_{42}(X_2)P_{61}(X)\{2\cos(2\alpha_2-\alpha_1-\varphi)\}\\
&+\\
&C(446;-2,3,1)k_{42}k_{43}k_{61}P_{42}(X_1)P_{43}(X_2)P_{61}(X)\{2\cos(3\alpha_2-2\alpha_1-\varphi)\}\\
&+\\
&-C(446;-3,4,1)k_{43}k_{44}k_{61}P_{43}(X_1)P_{44}(X_2)P_{61}(X)\{2\cos(4\alpha_2-3\alpha_1-\varphi)\}\\
&+\\
&C(446;-4,4,0)k_{44}k_{44}k_{60}P_{44}(X_1)P_{44}(X_2)P_{60}(X)\{2\cos(4\alpha_2-4\alpha_1)\}\\
&+\\
&-C(446;-3,3,0)k_{43}k_{43}k_{60}P_{43}(X_1)P_{43}(X_2)P_{60}(X)\{2\cos(3\alpha_2-3\alpha_1)\}\\
&+\\
&C(446;-2,2,0)k_{42}k_{42}k_{60}P_{42}(X_1)P_{42}(X_2)P_{60}(X)\{2\cos(2\alpha_2-2\alpha_1)\}\\
&+\\
&-C(446;-1,1,0)k_{41}k_{41}k_{60}P_{41}(X_1)P_{41}(X_2)P_{60}(X)\{2\cos(\alpha_2-\alpha_1)\}\\
&+\\
&C(446;0,0,0)k_{40}k_{40}k_{60}P_{40}(X_1)P_{40}(X_2)P_{60}(X)
\end{aligned}
\tag{A3.106}
$$

$$
\begin{aligned}
S_{448} = {} & C(448;4,4,8)k_{44}k_{44}k_{88}P_{44}(X_1)P_{44}(X_2)P_{88}(X)\{2\cos(4\alpha_2+4\alpha_1-8\varphi)\} \\
& + C(448;4,3,7)k_{44}k_{43}k_{87}P_{44}(X_1)P_{43}(X_2)P_{87}(X)\{2\cos(4\alpha_2+3\alpha_1-7\varphi)\} \\
& + C(448;3,4,7)k_{43}k_{44}k_{87}P_{43}(X_1)P_{44}(X_2)P_{87}(X)\{2\cos(3\alpha_2+4\alpha_1-7\varphi)\} \\
& + C(448;4,2,6)k_{44}k_{42}k_{86}P_{44}(X_1)P_{42}(X_2)P_{86}(X)\{2\cos(4\alpha_1+2\alpha_2-6\varphi)\} \\
& + C(448;3,3,6)k_{43}k_{43}k_{86}P_{43}(X_1)P_{43}(X_2)P_{86}(X)\{2\cos(3\alpha_1+3\alpha_2-6\varphi)\} \\
& + C(448;2,4,6)k_{42}k_{44}k_{86}P_{42}(X_1)P_{44}(X_2)P_{86}(X)\{2\cos(2\alpha_1+4\alpha_2-6\varphi)\} \\
& + C(448;4,1,5)k_{44}k_{41}k_{85}P_{44}(X_1)P_{41}(X_2)P_{85}(X)\{2\cos(4\alpha_1+\alpha_2-5\varphi)\} \\
& + C(448;3,2,5)k_{43}k_{42}k_{85}P_{43}(X_1)P_{42}(X_2)P_{85}(X)\{2\cos(3\alpha_1+2\alpha_2-5\varphi)\} \\
& + C(448;2,3,5)k_{42}k_{43}k_{85}P_{42}(X_1)P_{43}(X_2)P_{85}(X)\{2\cos(2\alpha_1+3\alpha_2-5\varphi)\} \\
& + C(448;1,4,5)k_{41}k_{44}k_{85}P_{41}(X_1)P_{44}(X_2)P_{85}(X)\{2\cos(\alpha_1+4\alpha_2-5\varphi)\} \\
& + C(448;4,0,4)k_{44}k_{40}k_{84}P_{44}(X_1)P_{40}(X_2)P_{84}(X)\{2\cos(4\alpha_1-4\varphi)\} \\
& + C(448;3,1,4)k_{43}k_{41}k_{84}P_{43}(X_1)P_{41}(X_2)P_{84}(X)\{2\cos(3\alpha_1+\alpha_2-4\varphi)\} \\
& + C(448;2,2,4)k_{42}k_{42}k_{84}P_{42}(X_1)P_{42}(X_2)P_{84}(X)\{2\cos(2\alpha_1+2\alpha_2-4\varphi)\} \\
& + C(448;1,3,4)k_{41}k_{43}k_{84}P_{41}(X_1)P_{43}(X_2)P_{84}(X)\{2\cos(\alpha_1+3\alpha_2-4\varphi)\} \\
& + C(448;0,4,4)k_{40}k_{44}k_{84}P_{40}(X_1)P_{44}(X_2)P_{84}(X)\{2\cos(4\alpha_2-4\varphi)\} \\
& - C(448;4,-1,3)k_{44}k_{41}k_{83}P_{44}(X_1)P_{41}(X_2)P_{83}(X)\{2\cos(4\alpha_1-\alpha_2-3\varphi)\} \\
& + C(448;3,0,3)k_{43}k_{40}k_{83}P_{43}(X_1)P_{40}(X_2)P_{83}(X)\{2\cos(3\alpha_1-3\varphi)\} \\
& + C(448;2,1,3)k_{42}k_{41}k_{83}P_{42}(X_1)P_{41}(X_2)P_{83}(X)\{2\cos(2\alpha_1+\alpha_2-3\varphi)\} \\
& + C(448;1,2,3)k_{41}k_{42}k_{83}P_{41}(X_1)P_{42}(X_2)P_{83}(X)\{2\cos(\alpha_1+2\alpha_2-3\varphi)\} \\
& + C(448;0,3,3)k_{40}k_{43}k_{83}P_{40}(X_1)P_{43}(X_2)P_{83}(X)\{2\cos(3\alpha_2-3\varphi)\} \\
& - C(448;-1,4,3)k_{41}k_{44}k_{83}P_{41}(X_1)P_{44}(X_2)P_{83}(X)\{2\cos(4\alpha_2-\alpha_1-3\varphi)\} \\
& + C(448;4,-2,2)k_{44}k_{42}k_{82}P_{44}(X_1)P_{42}(X_2)P_{82}(X)\{2\cos(4\alpha_1-2\alpha_2-2\varphi)\} \\
& - C(448;3,-1,2)k_{43}k_{41}k_{82}P_{43}(X_1)P_{41}(X_2)P_{82}(X)\{2\cos(3\alpha_1-\alpha_2-2\varphi)\} \\
& + C(448;2,0,2)k_{42}k_{40}k_{82}P_{42}(X_1)P_{40}(X_2)P_{82}(X)\{2\cos(2\alpha_1-2\varphi)\} \\
& + C(448;1,1,2)k_{41}k_{41}k_{82}P_{41}(X_1)P_{41}(X_2)P_{82}(X)\{2\cos(\alpha_1+\alpha_2-2\varphi)\} \\
& + C(448;0,2,2)k_{40}k_{42}k_{82}P_{40}(X_1)P_{42}(X_2)P_{82}(X)\{2\cos(2\alpha_2-2\varphi)\} \\
& +
\end{aligned}
$$

$$
\begin{aligned}
&- C(448;-1,3,2)k_{41}k_{43}k_{82}P_{41}(X_1)P_{43}(X_2)P_{82}(X)\{2\cos(3\alpha_2-\alpha_1-2\varphi)\}\\
&+\\
&\quad C(448;-2,4,2)k_{42}k_{44}k_{82}P_{42}(X_1)P_{44}(X_2)P_{82}(X)\{2\cos(4\alpha_2-2\alpha_1-2\varphi)\}\\
&+\\
&- C(448;4,-3,1)k_{44}k_{43}k_{81}P_{44}(X_1)P_{43}(X_2)P_{81}(X)\{2\cos(4\alpha_1-3\alpha_2-\varphi)\}\\
&+\\
&\quad C(448;3,-2,1)k_{43}k_{42}k_{81}P_{43}(X_1)P_{42}(X_2)P_{81}(X)\{2\cos(3\alpha_1-2\alpha_2-\varphi)\}\\
&+\\
&- C(448;2,-1,1)k_{42}k_{41}k_{81}P_{42}(X_1)P_{41}(X_2)P_{81}(X)\{2\cos(2\alpha_1-\alpha_2-\varphi)\}\\
&+\\
&\quad C(448;1,0,1)k_{41}k_{40}k_{81}P_{41}(X_1)P_{40}(X_2)P_{81}(X)\{2\cos(\alpha_1-\varphi)\}\\
&+\\
&\quad C(448;0,1,1)k_{40}k_{41}k_{81}P_{40}(X_1)P_{41}(X_2)P_{81}(X)\{2\cos(\alpha_2-\varphi)\}\\
&+\\
&- C(448;-1,2,1)k_{41}k_{42}k_{81}P_{41}(X_1)P_{42}(X_2)P_{81}(X)\{2\cos(2\alpha_2-\alpha_1-\varphi)\}\\
&+\\
&\quad C(448;-2,3,1)k_{42}k_{43}k_{81}P_{42}(X_1)P_{43}(X_2)P_{81}(X)\{2\cos(3\alpha_2-2\alpha_1-\varphi)\}\\
&+\\
&- C(448;-3,4,1)k_{43}k_{44}k_{81}P_{43}(X_1)P_{44}(X_2)P_{81}(X)\{2\cos(4\alpha_2-3\alpha_1-\varphi)\}\\
&+\\
&\quad C(448;-4,4,0)k_{44}k_{44}k_{80}P_{44}(X_1)P_{44}(X_2)P_{80}(X)\{2\cos(4\alpha_2-4\alpha_1)\}\\
&+\\
&- C(448;-3,3,0)k_{43}k_{43}k_{80}P_{43}(X_1)P_{43}(X_2)P_{80}(X)\{2\cos(3\alpha_2-3\alpha_1)\}\\
&+\\
&\quad C(448;-2,2,0)k_{42}k_{42}k_{80}P_{42}(X_1)P_{42}(X_2)P_{80}(X)\{2\cos(2\alpha_2-2\alpha_1)\}\\
&+\\
&- C(448;-1,1,0)k_{41}k_{41}k_{80}P_{41}(X_1)P_{41}(X_2)P_{80}(X)\{2\cos(\alpha_2-\alpha_1)\}\\
&+\\
&\quad C(448;0,0,0)k_{40}k_{40}k_{80}P_{40}(X_1)P_{40}(X_2)P_{80}(X) \qquad (A3.107)
\end{aligned}
$$

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