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Sixth Moment of Dipolar-Broadened Magnetic-Resonance-Absorption Line Shapes in Crystals

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Several recently developed theories of broad-line NMR presume the knowledge of the first several moments of the line shape. An exact expression for the sixth moment for the purely dipolar-broadened case is presented here. The result indicates that the sixth moment consists of one type of two-particle term, five types of three-particle terms, and nine types of four-particle terms, one of which has a vanishing coefficient. Most of the contribution comes from the four-particle terms.

I. INTRODUCTION

Historically, the first serious attempt at measuring resonance in bulk matter by looking for the resonance of Li^7 in lithium fluoride and for the proton resonance in potassium alum by using a calorimetric method was made by Gorter.[1] The first successful NMR absorption measurements were made independently by two groups: Purcell, Torrey, and Pound[2] and Bloch, Harrison, and Packard.[3] Since then, various broad-line-NMR theories have been suggested by Bloembergen,[4] Lowe and Norberg,[5] Lee, Tse, Goldburg, and Lowe,[6] Evans and Powles,[7] Demco,[8] Gibbs,[9] Fornes, Parker and Memory,[10] Gravely and Memory,[11] and Lado, Memory, and Parker.[12]

The majority of recent developments in broad-line-NMR line-shape theory can be classified into three groups. (i) Since the original work of Lowe and Norberg,[5] who proved that the free-induction decay (FID) was the Fourier transform of the absorption curve, various methods of approximation have been proposed to evaluate directly the time autocorrelation function of the transverse magnetization; among those are methods proposed by Evans and Powles,[7] Lee, Tse, Goldburg, and Lowe,[6] Demco,[8] Gibbs,[9] and Fornes, Parker, and Memory[10]; (ii) expansion theorems have been developed which are based on some parameters related to the crystal structure and a knowledge of the first few moments of the line shape, examples of which are the Gram-Charlier three-moment expansion suggested by Gravely and Memory,[11] and the generalized Neumann expansion obtained by Parker[13]; (iii) various approximate expressions have been proposed for the autocorrelation function or the associated memory function based on the general mathematical techniques employed in many-body problems or nonequilibrium statistical mechanics, such as presented by Lado, Memory, and Parker.[12]

A number of these approaches have in common the fact that they presume the knowledge of at least the first several of the line-shape moments, in terms of which the corresponding function of physical interest can be expressed approximately. However, the calculation of these moments, based on Van Vleck's formula,[14] involves the evaluation of the traces of the square of some complicated quantum-mechanical operators, and there is no general systematic pattern of evaluating the double sums involved in different types of particle-interaction terms. These facts have made difficult the calculation of moments other than the second and fourth, which were given in Van Vleck's original paper.

As for the sixth moment, Glebashev[15] obtained an approximate estimation for the exchange-narrowed case by considering only nearest-neighboring particles in the presence of a large static magnetic field and at high frequency. Also, Bersohn and Das[16] obtained some information about the sixth moment for the case of purely dipolar broadening, from a many-body-analysis approach. The purpose of our paper is to report the exact expression of the sixth-order moment of the magnetic-resonance-absorption curve in crystals.

II. SIXTH MOMENT OF MAGNETIC-RESONANCE LINE SHAPES

The normalized moment of a magnetic-resonance-absorption curve may be defined by the relation

$$\langle (\Delta\nu)^{2n}\rangle = \int_{-\infty}^{\infty} g(\nu-\nu_0)\,(\nu-\nu_0)^{2n}\,d\nu\ , \qquad (2.1)$$

where $g(\nu-\nu_0)$ is the line-shape function with the line centered at the frequency ν_0. The general expression for the $(2n)$th moment originally suggested by Waller[17] and proved by mathematical induction is

$$\langle \nu^{2n}\rangle = \frac{(ih)^{-2n}\,\mathrm{Tr}\{[\mathcal{H},[\mathcal{H}\cdots[\mathcal{H},s_x]\cdots]^2]\}}{\mathrm{Tr}\{S_x^2\}}\ , \qquad (2.2)$$

where $\mathcal{H}$ is the Hamiltonian of the spin system of physical interest, h is Planck's constant, S_x is the

x component of the total spin operator of the system, and there are n $\mathcal{H}$'s in the numerator. Since we are using the departure of the mean-square frequency from the square of the main line, $g^2\mu_B^2H^2/h^2$, as a measure of the mean-square line breadth of the main line, the subsidiary lines whose frequencies are near 0, $2g\mu_B H/h$, and $3g\mu_B H/h$ are not those of interest to us. Consequently, terms in the complete Hamiltonian that give contribution to the lines centering about 0, $2g\mu_B H/h$, and $3g\mu_B H/h$ will be discarded. This omission is not to be regarded as merely a simplification. Retention of these terms, in fact, would be completely erroneous in the computation of various mean powers of the frequencies by the commutator method. In the calculations of the second and fourth moments presented in his original paper, Van Vleck[14] showed that the correct truncated Hamiltonian of spin systems consisting of only one type of magnetic active ingredient was given by

$$\mathcal{H} = Hg\mu_B \sum_j S_{zj} + \sum_{k>j} A_{jk}\vec{S}_j \cdot \vec{S}_k + \sum_{k>j} B_{jk} S_{jk} S_{zk} , \tag{2.3}$$

where H_0 is the constant magnetic field applied in the z direction; μ_B is the Bohr, or the nuclear, magneton as the case may be; g is the corresponding Landé factor;

$$A_{jk} = \tilde{A}_{jk} + g^2\beta^2 r_{jk}^{-3}[\tfrac{3}{2}\cos^2\theta_{jk} - \tfrac{1}{2}] , \quad A_{jk} = A_{kj} ;$$

$$B_{jk} = -3g^2\beta^2 r_{jk}^{-3}[\tfrac{3}{2}\cos^2\theta_{jk} - \tfrac{1}{2}] , \qquad B_{jk} = B_{kj} ;$$

$$\tilde{A}_{jk} = -2z^2 J_{jk} ;$$

J_{jk} is the usual exchange integral; z is the number of electrons not in complete shell in each atom; θ_{jk} is the angle between the vector joining the jth and kth particle relative to the z axis; $S_x = \sum_j S_{xj}$; and r_{jk} is the distance between the jth and the kth particle. It must be pointed out that in the case of NMR absorption, where the exchange effect is negligible, the following relation holds:

$$A_{jk} = -\tfrac{1}{3}B_{jk} . \tag{2.4}$$

Also, he obtained,

$$[\mathcal{H}, S_x] = H_0 g\mu_B i \sum_j S_{yj} + i\sum_{k>j} B_{jk}(S_{yj}S_{zk} + S_{yk}S_{zj}) , \tag{2.5}$$

$$\langle \Delta\nu^2 \rangle = h^{-2}N^{-1}S(S+1)\tfrac{2}{3} \sum_{k>j}^{N} B_{jk}^2 , \tag{2.6}$$

where S is the spin quantum number of an individual atom and N is the total number of atoms in the crystal. Furthermore,

$$[\mathcal{H}, [\mathcal{H}, S_x]] = H_0^2 g^2 \mu_B^2 \sum_j S_{xj} + \sum_{k>j} ([jk] + [kj]) + \sum_{l>k>j} ([jkl] + [klj] + [ljk]) , \tag{2.7}$$

where

$$[jk] = 2H_0 g\mu_B B_{jk}S_{zk} + B_{jk}^2 S_{xj}S_{zk}^2 + A_{jk}B_{jk}(-S_{zj}S_{xk}S_{zk} + S_{yk}S_{yj}S_{xj} - S_{yj}^2 S_{xk} + S_{xj}S_{zk}^2) ,$$
$$[jkl] = 2B_{jk}B_{kl}S_{zj}S_{xk}S_{zl} + (-B_{jl}A_{jk} + B_{kl}A_{jk} - B_{jl}A_{kl} + B_{jk}A_{kl})(S_{zj}S_{xk}S_{zl} - S_{yj}S_{xk}S_{yl}) , \tag{2.8}$$

with

$$A_{jk} = A_{kj} , \quad B_{jk} = B_{kj} ,$$

$$\langle \Delta\nu^4 \rangle = h^{-4}N^{-1} \sum_{j\neq k\neq l} [3B_{jk}^2 B_{jl}^2 + 2A_{jk}^2(B_{jl} - B_{kl})^2 + 2A_{jk}A_{kl}(B_{jl} - B_{jk})(B_{jl} - B_{kl}) + 2A_{jk}B_{jk}(B_{jl} - B_{kl})^2][\tfrac{1}{3}S(S+1)]^2$$
$$+ h^{-4}2N^{-1} \sum_{k>j} \{B_{jk}^4 \tfrac{1}{5}[S^2(S+1)^2 - \tfrac{1}{3}S(S+1)] + 2B_{jk}^3 A_{jk}\tfrac{1}{5}[\tfrac{2}{3}S^2(S+1)^2 - \tfrac{1}{2}S(S+1)] + \tfrac{1}{2}B_{jk}^2 A_{jk}^2[\tfrac{4}{5}S^2(S+1)^2 - \tfrac{3}{5}S(S+1)]\} . \tag{2.9}$$

We observe that by the way the (jkl)'s are defined there exists symmetry between j and l:

$$[jkl] = [lkj] . \tag{2.10}$$

From (2.1) and (2.2), the sixth moment can be written in the form

$$\langle \nu^6 \rangle = \frac{-h^{-6}\,\mathrm{Tr}[\mathcal{H}, [\mathcal{H}, [\mathcal{H}, S_x]]]^2}{\mathrm{Tr}\, S_x^2} \tag{2.11}$$

or the normalized sixth moment

$$\langle \Delta\nu^6 \rangle = \langle \nu^6 \rangle - 15\frac{g^2\mu_B^2H_0^2}{h^2}\langle \Delta\nu^4 \rangle - 15\frac{g^4\mu_B^4H_0^4}{h^4}\langle \Delta\nu^2 \rangle - \frac{g^6\mu_B^6H_0^6}{h^6} . \tag{2.12}$$

Following Van Vleck's result of (2.7) and (2.8), we carried one step further and obtained the following expression:

$$Z = [\mathcal{H}, [\mathcal{H}, [\mathcal{H}, S_x]]]$$
$$= i\sum_j [j] + i\sum_{k>j} ([jk] + [kj]) + i\sum_{l>k>j} ([jkl] + [klj] + [ljk]) + i\sum_{m>l>k>j} ([jklm] + [klmj] + [lmjk] + [mjkl]) , \tag{2.13}$$

where cyclic permutation of indices is to be understood, with

$$[j]=H_0^3 g^3 \mu_B^3 S_{zj}\ ;$$

$$\begin{aligned}
[jk]={}&3H_0^2 g^2 \mu_B^2 B_{jk} S_{yj} S_{zk}+3H_0 g\mu_B B_{jk}^2 S_{yj}^2 S_{zk}^2+3H_0 g\mu_B A_{jk}B_{jk}(-S_{zj}S_{yk}S_{zk}+S_{xj}S_{yj}S_{xk}-S_{xj}^2 S_{yk}+S_{yj}S_{zk}^2)\\
&+A_{jk}^2 B_{jk}(S_{yj}S_{xk}S_{zk}S_{xk}-S_{xj}S_{xk}S_{zk}\ S_{yk}+S_{xj}S_{yj}S_{xj}S_{zk}+S_{xj}S_{zj}S_{xk}S_{yk}+S_{zj}S_{xj}S_{yk}S_{xk}-S_{zj}S_{yj}S_{xk}^2+S_{yj}S_{zj}S_{zk}^2\\
&-S_{yj}S_{zj}S_{yk}^2-2S_{yj}S_{zj}S_{xk}^2+S_{yj}^3 S_{zk})+(A_{jk}^2 B_{jk}+A_{jk}B_{jk}^2)(-S_{zj}S_{yj}S_{xj}S_{xk}+2S_{xj}S_{yj}S_{zk}S_{xk}-S_{zj}^2 S_{yk}S_{zk}+S_{yj}S_{xj}S_{zk}S_{xk}\\
&-S_{yj}^2 S_{zj}S_{yk}+S_{xj}S_{yj}S_{xk}S_{zk}-S_{xj}^2 S_{zk}S_{yk}+S_{yj}^2 S_{yk}S_{zk}-2S_{xj}^2 S_{yk}S_{zk}-S_{zj}S_{yk}S_{zk}^2+S_{yj}S_{zk}^3)+A_{jk}B_{jk}^2 S_{yj}S_{zk}^3+B_{jk}^3 S_{yj}S_{zk}^3\ ;
\end{aligned}$$

$$\begin{aligned}
[jkl]={}&H_0 g\mu_B[6B_{jk}B_{kl}S_{zj}S_{yk}S_{zl}+(-B_{jl}A_{jk}+B_{kl}A_{jk}-B_{jl}A_{kl}+B_{jk}A_{kl})(3S_{zj}S_{yk}S_{zl}-3S_{xj}S_{yk}S_{xl}-S_{yj}S_{yk}S_{yl})]\\
&-(S_{yj}S_{xk}S_{xl}S_{zl}+S_{zj}S_{xj}S_{xk}S_{yl})[2A_{jl}B_{jk}B_{kl}+A_{jl}(-B_{jl}A_{jk}+B_{kl}A_{jk}-B_{jl}A_{kl}+B_{jk}A_{kl})]-(S_{yj}S_{xj}S_{xk}S_{zl}\\
&+S_{zj}S_{xk}S_{xl}S_{yl})[A_{jl}(-B_{jl}A_{jk}+B_{kl}A_{jk}-B_{jl}A_{kl}+B_{jk}A_{kl})]+(S_{xj}S_{xk}S_{yl}S_{zl}+S_{zj}S_{yj}S_{xk}S_{xl})[2A_{jl}B_{jk}B_{kl}+(2A_{jl}+B_{jl})\\
&\times(-B_{jl}A_{jk}+B_{kl}A_{jk}-B_{jl}A_{kl}+B_{jk}A_{kl})]-S_{yj}S_{yk}S_{zk}S_{yl}[A_{kl}A_{kj}B_{kj}+A_{kj}A_{kl}B_{kl}+(A_{jk}+B_{jk})(-B_{lk}A_{lj}+B_{jk}A_{lj}\\
&-B_{lk}A_{jk}+B_{lj}A_{jk})+(A_{kl}+B_{kl})(-B_{kj}A_{kl}+B_{lj}A_{kl}-B_{kj}A_{lj}+B_{kl}A_{lj})]-S_{zj}S_{yk}S_{zk}S_{zl}\ [A_{kl}A_{jk}B_{jk}+A_{kj}A_{kl}B_{kl}\\
&+B_{kl}A_{jk}B_{jk}+B_{kj}A_{kl}B_{kl}+2A_{jk}B_{jk}B_{lj}+A_{jk}(-B_{lk}A_{lj}+B_{jk}A_{lj}-B_{lk}A_{jk}+B_{lj}A_{jk})+2A_{kl}B_{kl}B_{lj}+A_{kl}(-B_{kj}A_{kl}\\
&+B_{lj}A_{kl}-B_{kj}A_{lj}+B_{kl}A_{lj})]-(S_{xj}S_{zk}S_{yk}S_{xl}+S_{xj}S_{yk}S_{zk}S_{xl})[A_{lk}B_{jk}^2+A_{jk}B_{kj}^2+2(A_{jk}A_{lk}B_{lk}+A_{lk}A_{jk}B_{jk})]\\
&+S_{xj}S_{xk}S_{zk}S_{yl}(A_{lk}B_{jk}^2+A_{lj}A_{lk}B_{lk}-A_{lj}A_{jk}B_{jk}+A_{lk}A_{jk}B_{jk})+S_{yj}S_{xk}S_{zk}S_{xl}(A_{jk}B_{lk}^2+A_{jl}A_{jk}B_{jk}-A_{jl}A_{lk}B_{lk}\\
&+A_{jk}A_{lk}B_{lk})+S_{zj}S_{yk}S_{xk}S_{xl}(A_{jk}A_{kl}B_{kl}-A_{jl}A_{kl}B_{kl}+A_{jl}A_{kj}B_{kj}-B_{lj}A_{lk}B_{lk}+B_{jk}A_{lk}B_{lk})+S_{xj}S_{yk}S_{xk}S_{zl}(A_{lj}A_{kl}B_{kl}\\
&-A_{lj}A_{kj}B_{kj}+A_{lk}A_{kj}B_{kj}-B_{lj}A_{kj}B_{kj}+B_{lk}A_{kj}B_{kj})+S_{xj}S_{zk}S_{xk}S_{yl}[A_{kl}B_{jk}^2+A_{jk}A_{kl}B_{kl}+A_{kl}A_{kj}B_{kj}\\
&+(A_{jk}+B_{jk})(-B_{jl}A_{jk}+B_{kl}A_{jk}-B_{jl}A_{kl}+B_{jk}A_{kl})]\\
&+S_{yj}S_{zk}S_{xk}S_{xl}\ [A_{kj}B_{lk}^2+A_{kl}A_{kj}B_{kj}+A_{kj}A_{lk}B_{lk}+(A_{kl}+B_{kl})(-B_{jl}A_{jk}+B_{kl}A_{jk}-B_{jl}A_{lk}+B_{jk}A_{kl})]\\
&+S_{zj}S_{xk}S_{yk}S_{xl}\ [3A_{kl}B_{kl}B_{jk}+A_{kj}A_{kl}B_{kl}+A_{kl}A_{jk}B_{jk}+A_{kl}(-B_{jl}A_{jk}+B_{kl}A_{jk}-B_{jl}A_{kl}+B_{jk}A_{kl})]\\
&+S_{xj}S_{xk}S_{yk}S_{zl}\ [3A_{jk}B_{jk}B_{kl}+A_{jk}A_{lk}B_{lk}+A_{kl}A_{jk}B_{jk}+A_{jk}(-B_{jl}A_{jk}+B_{kl}A_{jk}-B_{jl}A_{kl}+B_{jk}A_{kl})]\\
&+S_{yj}^2 S_{zk}S_{yl}[A_{jk}A_{jl}B_{jl}-A_{kl}A_{jl}B_{jl}+A_{kl}A_{jk}B_{jk}-B_{kl}A_{jl}B_{jl}+B_{jk}A_{jl}B_{jl}+A_{jk}(-B_{jl}A_{jk}+B_{kl}A_{jk}-B_{jl}A_{kl}+B_{jk}A_{kl})]\\
&+S_{yj}S_{zk}S_{yl}^2\ [A_{jk}A_{lk}B_{lk}-A_{jk}A_{lj}B_{lj}+A_{kl}A_{lj}B_{lj}-B_{jk}A_{jl}B_{jl}+B_{kl}A_{jl}B_{jl}+A_{kl}(-B_{jl}A_{jk}+B_{kl}A_{jk}-B_{jl}A_{kl}+B_{jk}A_{kl})]\\
&-S_{zj}S_{xk}^2 S_{yl}[3A_{kl}B_{kl}B_{jk}+A_{jk}A_{lk}B_{lk}+A_{kl}A_{kj}B_{kj}+(A_{jk}+A_{kl})(-B_{jl}A_{jk}+B_{kl}A_{jk}-B_{jl}A_{kl}+B_{jk}A_{kl})]\\
&-S_{yj}S_{xk}^2 S_{zl}[3A_{jk}B_{jk}B_{kl}+A_{jk}A_{lk}B_{lk}+A_{kl}A_{kj}B_{kj}+(A_{jk}+A_{kl})(-B_{jl}A_{jk}+B_{kl}A_{jk}-B_{jl}A_{kl}+B_{jk}A_{kl})]\\
&+S_{zj}^2 S_{yk}S_{zl}[3B_{kl}B_{jk}^2+3A_{jk}B_{jk}B_{kl}-A_{kl}B_{lj}^2+A_{kl}B_{jk}^2+A_{jk}A_{lj}B_{lj}-A_{kl}A_{lj}B_{lj}+A_{kl}A_{kj}B_{kj}\\
&+(A_{jk}+B_{jk})(-B_{jl}A_{jk}+B_{kl}A_{jk}-B_{jk}A_{kl}+B_{jk}A_{kl})]+S_{zj}S_{yk}S_{zl}^2[3B_{jk}B_{kl}^2+3A_{kl}B_{kl}B_{jk}-A_{jk}B_{jl}^2+A_{jk}B_{lk}^2+A_{jk}A_{kl}B_{kl}\\
&\qquad-A_{jk}A_{jl}B_{jl}+A_{kl}A_{jl}B_{jl}+(A_{kl}+B_{kl})(-B_{jl}A_{jk}+B_{kl}A_{jk}-B_{jl}A_{kl}+B_{jk}A_{kl})]\ ;
\end{aligned}$$

$$\begin{aligned}
[jklm]={}&S_{xj}S_{yk}S_{xl}S_{zm}[-2A_{jk}B_{kl}B_{lm}+2A_{jk}B_{jl}B_{lm}-2A_{kl}B_{mj}B_{jk}\\
&+2A_{kl}B_{mj}B_{jl}+(B_{lm}+A_{lm})(-B_{lk}A_{lj}+B_{jk}A_{lj}-B_{lk}A_{jk}+B_{lj}A_{jk})\\
&+(B_{jm}+A_{jm})(-B_{kj}A_{kl}+B_{lj}A_{kl}+B_{kj}A_{lj}+B_{kl}A_{lj})+A_{kl}(-B_{ml}A_{mj}+B_{jl}A_{mj}-B_{ml}A_{jl}+B_{mj}A_{jl})\\
&+A_{jk}(-B_{jm}A_{jl}+B_{lm}A_{jl}-B_{jm}A_{lm}+B_{jl}A_{lm})-(A_{jk}+A_{jm})(-B_{km}A_{kl}+B_{lm}A_{kl}-B_{km}A_{lm}+B_{kl}A_{lm})\\
&-(A_{kl}+A_{lm})(-B_{mk}A_{mj}+B_{jk}A_{mj}-B_{mk}A_{jk}+B_{mj}A_{jk})]+S_{yj}S_{xk}S_{xl}S_{zm}[-2A_{jl}B_{jk}B_{km}+2A_{jk}B_{kl}B_{lm}-2A_{jk}B_{jl}B_{lm}\\
&+2A_{jl}B_{mk}B_{kl}+(B_{lm}+A_{lm})(-B_{jl}A_{jk}+B_{kl}A_{jk}-B_{jl}A_{kl}+B_{jk}A_{kl})+(B_{km}+A_{km})(-B_{kj}A_{kl}+B_{lj}A_{kl}-B_{kj}A_{lj}\\
&+B_{kl}A_{lj})+A_{jk}(-B_{km}A_{kl}+B_{lm}A_{kl}-B_{km}A_{lm}+B_{kl}A_{lm})+A_{jl}(-B_{ml}A_{mk}+B_{kl}A_{mk}-B_{ml}A_{kl}+B_{mk}A_{kl})\\
&-(A_{lm}+A_{jl})(-B_{jm}A_{jk}+B_{km}A_{jk}-B_{jm}A_{km}+B_{jk}A_{km})-(A_{km}+A_{jk})(-B_{jm}A_{jl}+B_{lm}A_{jl}-B_{jm}A_{lm}+B_{jl}A_{lm})]\\
&+S_{zj}S_{xk}S_{xl}S_{ym}[-2A_{lm}B_{jk}B_{km}+2A_{lm}B_{jk}B_{kl}-2A_{km}B_{jl}B_{lm}+2A_{km}B_{kl}B_{lj}\\
&+(B_{jk}+A_{jk})(-B_{km}A_{kl}+B_{lm}A_{kl}-B_{km}A_{lm}+B_{kl}A_{lm})+(B_{jl}+A_{jl})(-B_{ml}A_{mk}+B_{kl}A_{mk}-B_{ml}A_{kl}+B_{mk}A_{kl})\\
&+A_{lm}(-B_{jl}A_{jk}+B_{kl}A_{jk}-B_{jl}A_{kl}+B_{jk}A_{kl})+A_{km}(-B_{jk}A_{kl}+B_{jl}A_{kl}-B_{kj}A_{lj}+B_{kl}A_{lj})
\end{aligned}$$

$$
\begin{aligned}
&-(A_{lm}+A_{jl})(-B_{jm}A_{jk}+B_{km}A_{jk}-B_{jm}A_{km}+B_{jk}A_{km})-(A_{km}+A_{jk})(-B_{jm}A_{jl}+B_{lm}A_{jl}-B_{jm}A_{lm}+B_{jl}A_{lm})] \\
&+S_{yj}S_{zk}S_{zl}S_{zm}[6B_{mj}B_{jk}B_{jl}-A_{jm}B_{lm}B_{mk}+A_{jm}B_{lj}B_{jk}-A_{jk}B_{mk}B_{kl}+A_{jk}B_{mj}B_{jl}-A_{jl}B_{kl}B_{lm}+A_{jl}B_{mj}B_{jk} \\
&+(B_{jm}+A_{jm})(-B_{lk}A_{lj}+B_{jk}A_{lj}-B_{lk}A_{jk}+B_{lj}A_{jk})+(B_{jl}+A_{jl})(-B_{mk}A_{mj}+B_{jk}A_{mj}-B_{mk}A_{jk}+B_{mj}A_{jk}) \\
&+(B_{jk}+A_{jk})(-B_{ml}A_{mj}+B_{jl}A_{mj}-B_{ml}A_{jl}+B_{mj}A_{jl})-A_{jl}(-B_{km}A_{kl}+B_{lm}A_{kl}-B_{km}A_{lm}+B_{kl}A_{lm}) \\
&-A_{jm}(-B_{lk}A_{lm}+B_{mk}A_{lm}-B_{lk}A_{mk}+B_{lm}A_{mk})-A_{jk}(-B_{ml}A_{mk}+B_{kl}A_{mk}-B_{ml}A_{kl}+B_{mk}A_{kl})] \\
&+S_{zj}S_{yk}S_{yl}S_{ym}[-(B_{jk}+A_{jk})(-B_{ml}A_{mk}+B_{kl}A_{mk}-B_{ml}A_{kl}+B_{mk}A_{kl})-(B_{jm}+A_{jm})(-B_{lk}A_{lm}+B_{mk}A_{lm} \\
&-B_{lk}A_{mk}+B_{lm}A_{mk})-(B_{jl}+A_{jl})(-B_{km}A_{kl}+B_{lm}A_{kl}-B_{km}A_{lm}+B_{kl}A_{lm})+A_{jm}(-B_{lk}A_{lj}+B_{jk}A_{lj} \\
&-B_{lk}A_{jk}+B_{lj}A_{jk})+A_{jl}(-B_{mk}A_{mj}+B_{jk}A_{mj}-B_{mk}A_{jk}+B_{mj}A_{jk})+A_{jk}(-B_{ml}A_{mj}+B_{jl}A_{mj}-B_{ml}A_{jl}+B_{mj}A_{jl})] \;.
\end{aligned}
$$

With the aids of (i) the usual quantum-mechanical-operator commutation relations such as $S_{xj}S_{yk}-S_{yk}S_{xj} = i\delta_{jk}S_{zj}$; (ii) simple trace equations such as $\mathrm{Tr}S_{zj}S_{zl}=0$ when $j\neq l$; (iii) from the manner the cyclic terms are defined, relations such as

$$
\begin{gathered}
\mathrm{Tr}([j][jk])=\mathrm{Tr}([j][jk])=\mathrm{Tr}([j][jkl])=\mathrm{Tr}([j][jklm])=0 \;, \\
\mathrm{Tr}([jk][jklm])=0 \;, \quad \mathrm{Tr}([jkl][jklm])=0 \;, \quad \mathrm{Tr}([jklm][klmj])=0 \;;
\end{gathered}
$$

(iv) the cyclic property of the trace, (v) the commutativity of multiplication of scalar functions; and (vi) flipping indices, we obtained, from (2.13),

$$-\mathrm{Tr}Z^2=R+S+T+U+V+W \;, \tag{2.14}$$

where

$$R=\mathrm{Tr}\{\sum_j [j]^2+2\sum_{k>j}([j][jk]+[kj][k])+2\sum_{k>j}([jk]^2+[jk][kj])\} \;, \tag{2.15}$$

$$S=\mathrm{Tr}\sum_{l\neq j\neq k}([jk][jl]+[kj][lj]+[jk][lj]+[kj][jl]) \;, \tag{2.16}$$

$$
\begin{aligned}
T=\mathrm{Tr}\;2\sum_{l>k>j}&([jkl][kl]+[jkl][jl]+[jkl][jk]+[klj][kl]+[klj][jl]+[klj][jk]+[ljk][kl]+[ljk][jl]+[ljk][jk] \\
&+[jkl][lk]+[jkl][lj]+[jkl][kj]+[klj][lk]+[klj][lj]+[klj][kj]+[ljk][lk]+[ljk][lj]+[ljk][kj]) \;,
\end{aligned} \tag{2.17}
$$

$$U=\mathrm{Tr}\sum_{l>k>j}([jkl]^2+2[jkl][klj]+[klj]^2+2[jkl][ljk]+[ljk]^2+2[klj][ljk]) \;, \tag{2.18}$$

$$
\begin{aligned}
V=2\mathrm{Tr}\sum_{m>l>k>j}&([jkl][klm]+[klm][jlm]+[jkl][jkm]+[jkm][jlm]+[klm][jkm]+[jlm][jkl]+[jkl][lmk] \\
&+[klm][lmj]+[jkl][kmj]+[jkm][lmj]+[klm][kmj]+[jlm][klj]+[jkl][mkl]+[klm][mjl]+[jkl][mjk] \\
&+[jkm][mjl]+[klm][mjk]+[jlm][ljk]+[klj][klm]+[lmk][jlm]+[klj][jkm]+[kmj][jlm]+[lmk][jkm] \\
&+[lmj][jkl]+[klj][lmk]+[lmk][lmj]+[klj][kmj]+[kmj][lmj]+[lmk][kmj]+[lmj][klj]+[klj][mkl] \\
&+[lmk][mjl]+[klj][mjk]+[kmj][mjl]+[lmk][mjk]+[lmj][ljk]+[ljk][klm]+[mkl][jlm]+[ljk][jkm] \\
&+[mjk][jlm]+[mkl][jkm]+[mjl][jkl]+[ljk][lmk]+[mkl][lmj]+[ljk][kmj]+[mjk][lmj]+[mkl][kmj] \\
&+[mjl][klj]+[ljk][mkl]+[mkl][mjl]+[ljk][mjk]+[mjk][mjl]+[mkl][mjk]+[mjl][ljk]) \;,
\end{aligned} \tag{2.19}
$$

$$W=\mathrm{Tr}\sum_{m>l>k>j}([jklm]^2+[klmj]^2+[lmjk]^2+[mjkl]^2) \;. \tag{2.20}$$

III. EVALUATION OF THE SIXTH MOMENT

A. Evaluation of the Two-Particle Interaction Terms

To compute the two-particle interaction terms, we have to evaluate (2.15). The trace equations to be used in the calculation are listed in Table I. Some of their derivations will be given in the Appendix. Since there are only two indices involved, the calculation is straightforward but extremely cumbersome. Therefore, we omit the tedious details, but just present our results. With the help of Table I and the familiar commutation relations of the spin operators, we finally obtain, by collecting all the nonvanishing product terms as given in (2.15),

$$\mathrm{Tr}[j]^2 = NH_0^6 g^6 \mu_B^6 \tfrac{1}{3} S(S+1)(2S+1)^N \ , \qquad (3.1)$$

$$2\mathrm{Tr}\sum_{k>j}([kj][k]+[j][jk]) = \tfrac{4}{3} H_0^4 g^4 \mu_B^6 \tfrac{1}{3} S^2(S+1)^2 \times (2S+1)^N \sum_{k>j} B_{jk}^2 \ , \qquad (3.2)$$

$$\begin{aligned}
&\mathrm{Tr}\sum_{k>j}[jk]^2+[jk][kj] \\
&= H_0^4 g^4 \mu_B^4 (2S+1)^N S^2(S+1)^2 \sum_{k>j} B_{jk}^2 + H_0^2 g^2 \mu_B^2 (2S+1)^N S^2(S+1)^2 [2S(S+1)-\tfrac{3}{2}] \sum_{k>j} A_{jk}^2 B_{jk}^2 \\
&\quad + H_0^2 g^2 \mu_B^2 (2S+1)^N S^2(S+1)^2 [\tfrac{4}{3} S(S+1)-1] \sum_{k>j} A_{jk} B_{jk}^3 + H_0^2 g^2 \mu_B^2 (2S+1)^N S^2(S+1)^2 [S(S+1)-\tfrac{1}{3}] \sum_{k>j} B_{jk}^4 \\
&\quad + (2S+1)^N S^2(S+1)^2 \tfrac{1}{21} [S^2(S+1)^2 - S(S+1)+\tfrac{1}{3}] \sum_{k>j} B_{jk}^6 + (2S+1)^N S^2(S+1)^2 \tfrac{1}{21} [\tfrac{76}{25} S^2(S+1)^2 - \tfrac{347}{75} S(S+1) + \tfrac{44}{25}] \\
&\quad \times \sum_{k>j} A_{jk} B_{jk}^5 + (2S+1)^N S^2(S+1)^2 \tfrac{1}{21} [\tfrac{270}{25} S^2(S+1)^2 - \tfrac{2355}{150} S(S+1) + \tfrac{855}{150}] \sum_{k>j} A_{jk}^2 B_{jk}^4 + (2S+1)^N S^2(S+1)^2 \tfrac{1}{21} [\tfrac{520}{25} S^2(S+1)^2 \\
&\qquad - \tfrac{2190}{75} S(S+1) + \tfrac{765}{75}] \sum_{k>j} A_{jk}^3 B_{jk}^3 + (2S+1)^N S^2(S+1)^2 \tfrac{1}{21} [\tfrac{448}{25} S^2(S+1)^2 - \tfrac{3612}{150} S(S+1) + \tfrac{1197}{150}] \sum_{k>j} A_{jk}^4 B_{jk}^2 \ , \qquad (3.3)
\end{aligned}$$

and

$$(2.15) = (3.1) + (3.2) + 2(3.3) \ . \qquad (3.4)$$

After dividing (3.4) by the equation

$$h^6 \,\mathrm{Tr}\, S_x^2 = \tfrac{1}{3} NS(S+1)(2S+1)^N h^6 \qquad (3.5)$$

the two-particle interaction terms for the second and fourth moment in (2.12) will be cancelled out. If we let G denote the two-particle interaction terms of the sixth moment, then

$$\begin{aligned}
G = 2N^{-1}h^{-6} \sum_{k>j} & B_{jk}^6 \tfrac{1}{7} [S^3(S+1)^3 - S^2(S+1)^2 + \tfrac{1}{3} S(S+1)] + B_{jk}^5 A_{jk} \tfrac{1}{7} [\tfrac{76}{25} S^3(S+1)^3 - \tfrac{347}{75} S^2(S+1)^2 + \tfrac{44}{25} S(S+1)] \\
& + B_{jk}^4 A_{jk}^2 \tfrac{1}{7} [\tfrac{270}{25} S^3(S+1)^3 - \tfrac{2355}{150} S^2(S+1)^2 + \tfrac{855}{150} S(S+1)] + B_{jk}^3 A_{jk}^3 \tfrac{1}{7} [\tfrac{520}{25} S^3(S+1)^3 - \tfrac{2190}{75} S^2(S+1)^2 + \tfrac{765}{75} S(S+1)] \\
& + B_{jk}^2 A_{jk}^4 \tfrac{1}{7} [\tfrac{448}{25} S^3(S+1)^3 - \tfrac{3612}{150} S^2(S+1)^2 + \tfrac{1197}{150} S(S+1)] \ . \qquad (3.6)
\end{aligned}$$

As pointed out by Van Vleck,[15] a necessary criterion to check the correctness of the algebra is that when $S=\frac{1}{2}$ and the B's are assumed to be equal to a constant, independent of their subscripts, all the terms involving the A's will vanish and should not appear in the moment of any order. To check the correctness of our algebra for the two-particle terms of the sixth moment, we find, indeed, all the terms involving the A's will vanish when $S=\frac{1}{2}$, satisfying the necessary criterion suggested by Van Vleck. For the case of purely dipolar broadening, we set $A=-\frac{1}{3}B$, and (3.6) reduces to

$$G_p = N^{-1}h^{-6} \tfrac{1}{42525} (7746\lambda^3 - 5079\lambda^2 + 1224\lambda) \sum_{k>j} B_{jk}^6 \ , \qquad (3.7)$$

where $\lambda \equiv S(S+1)$ and G_p denotes the two-particle interaction terms of the sixth moment for the purely dipolar case.

B. Evaluation of the Three-Particle Interaction Terms

The three-particle interaction terms come from (2.16)–(2.18). Since (2.16) has already been in the form $\sum_{l\neq j\neq k}$ notation, the calculation is straightforward. We obtain.

$$\begin{aligned}
(2.16) = \sum_{l\neq j\neq k} & [H_0^2 g^2 \mu_B^2 \tfrac{9}{27} (2S+1)^N \lambda^3 B_{jk}^2 B_{jl}^2 + (2S+1)^N \lambda^3 \tfrac{1}{45} (\tfrac{1}{3}\lambda - \tfrac{1}{4})(36 A_{jk}^2 B_{jk} A_{jl}^2 B_{jl} + 18 A_{jk}^2 B_{jk} A_{jl} B_{jl}^2 \\
& + 18 A_{jl}^2 B_{jl} A_{jk} B_{jk}^2 - 9 A_{jk} B_{jk}^2 A_{jl} B_{jl}^2)] \ . \qquad (3.8)
\end{aligned}$$

Equation (2.17) can be evaluated in two ways: (i) either by making use of the symmetric property between j and l in $[jkl]$, and flipping indices, or (ii) by actually picking out the nonvanishing terms and redefining terms, then finally flipping indices. Both approaches lead to the same following result:

$$(2.17)=2\sum_{l\neq j\neq k}\mathrm{Tr}[jkl][jk]+[jkl][kl]+[jkl][lj]\ . \tag{3.9}$$

The difficulty of evaluating (2.18) lies in the calculation of the cross-product terms. Though we still have symmetry between j and l in $[jkl]$ from the way we construct the $[jkl]$, ours would somewhat be different from Van Vleck's. In our use, symmetry comes from a pair of successive terms inside $[jkl]$, rather than just from a single term. This means

$$\mathrm{Tr}[jkl][klj]\neq\mathrm{Tr}[lkj][klj]\ .$$

Whereas in Van Vleck's case, he had

$$\mathrm{Tr}[jkl][klj]=\mathrm{Tr}[lkj][klj]\ .$$

Therefore, we have to pick out the actual nonvanishing terms and then redefine them. Again, the process is cumbersome, and we just present the result. Combining (3.8), (3.9), and (2.18) together and subtracting the three-particle terms of the fourth moment in (2.12), the three-particle interaction terms of the sixth moment are denoted by D, where after dividing out by $\mathrm{Tr}S_x^2$ with $\lambda\equiv S(S+1)$,

$$\begin{aligned}
D=N^{-1}h^{-6}\sum_{l\neq j\neq k}&\{B_{kl}^4B_{jk}^2(\lambda^3-\tfrac{1}{3}\lambda^2)+[A_{jk}B_{jk}B_{kl}^4\tfrac{18}{15}(\lambda^3-\tfrac{1}{3}\lambda^2)-A_{jk}B_{jk}B_{jl}B_{kl}^3\tfrac{12}{15}(\lambda^3-\tfrac{1}{3}\lambda^2)+A_{jk}B_{jk}^3B_{kl}^2\tfrac{1}{45}(96\lambda^3-47\lambda^2)\\
&-A_{jk}B_{jk}B_{jl}^2B_{lk}^2\tfrac{6}{15}(\lambda^3-\tfrac{1}{3}\lambda^2)-A_{jk}B_{jk}^3B_{jl}B_{kl}\tfrac{2}{45}(18\lambda^3-\lambda^2)]+[A_{jk}^2B_{kl}^4\tfrac{1}{45}(59\lambda^3-28\lambda^2)-A_{jk}^2B_{jl}B_{lk}^3\tfrac{1}{45}(86\lambda^3-37\lambda^2)\\
&+A_{jk}^2B_{jl}^2B_{kl}^2\tfrac{1}{45}(27\lambda^3-9\lambda^2)-A_{jk}^2B_{jk}^2B_{jl}B_{kl}\tfrac{1}{45}(78\lambda^3-21\lambda^2)+A_{jk}^2B_{jk}^2B_{lk}^2\tfrac{1}{45}(168\lambda^3-\tfrac{354}{4}\lambda^2)]\\
&+[A_{lk}B_{lk}A_{jk}B_{jk}^3\tfrac{1}{45}(118\lambda^3-61\lambda^2)-A_{kl}B_{kl}B_{jk}A_{jk}B_{jk}^2\tfrac{1}{45}(10\lambda^3-\tfrac{30}{4}\lambda^2)-A_{lk}B_{lj}A_{jk}B_{jk}^2\tfrac{1}{45}(46\lambda^3-7\lambda^2)\\
&+A_{jk}B_{jk}^2A_{kl}B_{lj}^2\tfrac{1}{45}(38\lambda^3-6\lambda^2)-A_{jk}B_{jk}A_{lk}B_{lk}B_{jl}^2\tfrac{1}{45}(90\lambda^3-45\lambda^2)-A_{jk}B_{jk}A_{kl}B_{lj}^3\tfrac{1}{45}(28\lambda^3-16\lambda^2)\\
&+A_{kl}A_{jk}B_{jl}^4\tfrac{1}{45}(18\lambda^3-\tfrac{34}{4}\lambda^2)]+[A_{jk}^2B_{jk}A_{kl}B_{kl}^2\tfrac{1}{45}(150\lambda^3-90\lambda^2)-A_{jk}^2B_{jl}A_{kl}B_{kl}^2\tfrac{1}{45}(172\lambda^3-\tfrac{426}{4}\lambda^2)\\
&+A_{jk}^2B_{jk}^2A_{kl}B_{kl}\tfrac{1}{45}(188\lambda^3-121\lambda^2)-A_{jk}^2B_{jk}B_{jl}A_{kl}B_{kl}\tfrac{1}{45}(226\lambda^3-\tfrac{458}{4}\lambda^2)+A_{jk}^2A_{kl}B_{kl}B_{lj}^2\tfrac{1}{45}(84\lambda^3-\tfrac{142}{4}\lambda^2)\\
&-A_{jk}^2B_{jk}^2A_{kl}B_{lj}\tfrac{1}{45}(80\lambda^3-40\lambda^2)+A_{jk}^2B_{jk}A_{kl}B_{lj}^2\tfrac{1}{45}(76\lambda^3-\tfrac{98}{4}\lambda^2)-A_{jk}^2A_{kl}B_{lj}^3\tfrac{1}{45}(24\lambda^3-\tfrac{42}{4}\lambda^2)\\
&+A_{jk}^2A_{kl}B_{kl}^3\tfrac{1}{45}(114\lambda^2-83\lambda^3)]+[-A_{jk}B_{jk}A_{kl}B_{kl}A_{jl}B_{jl}\tfrac{1}{45}(4\lambda^3-\tfrac{22}{4}\lambda^2)-A_{jk}B_{jk}A_{kl}A_{jl}B_{jl}^2\tfrac{1}{45}(32\lambda^3-24\lambda^2)\\
&+A_{jk}A_{kl}A_{jl}B_{jl}^3\tfrac{1}{45}(30\lambda^3-25\lambda^2)]+A_{jk}^3B_{jk}B_{jl}(B_{jl}-B_{kl})\tfrac{1}{45}(88\lambda^3-36\lambda^2)+A_{jk}^4B_{lj}(B_{lj}-B_{lk})\tfrac{1}{45}(40\lambda^3-10\lambda^2)\\
&\times[+A_{jk}^3B_{jk}A_{jl}(B_{jl}-B_{kl})\tfrac{1}{45}(88\lambda^3-46\lambda^2)+A_{jk}^3A_{jl}B_{kl}^2\tfrac{1}{45}(60\lambda^3-15\lambda^2)-A_{jk}^3A_{jl}B_{jl}B_{kl}\tfrac{1}{45}(80\lambda^3-20\lambda^2)\\
&+A_{jk}^3A_{jl}B_{jl}^2\tfrac{1}{45}(20\lambda^3-5\lambda^2)][+A_{jk}^2B_{jk}A_{kl}^2B_{kl}\tfrac{1}{45}(130\lambda^3-85\lambda^2)-A_{jk}^2B_{jk}A_{kl}^2B_{jl}\tfrac{1}{45}(244\lambda^3-148\lambda^2)\\
&+A_{jk}^2B_{jl}^2A_{kl}^2\tfrac{1}{45}(56\lambda^3-\tfrac{98}{4}\lambda^2)+A_{jk}^2B_{jk}^2A_{kl}^2\tfrac{1}{45}(172\lambda^3-124\lambda^2)][+A_{jl}A_{lk}A_{jk}^2B_{jk}^2\tfrac{1}{45}(44\lambda^3-38\lambda^2)\\
&-A_{jl}B_{jl}^2A_{lk}A_{jk}^2\tfrac{1}{45}(44\lambda^3-23\lambda^2)+A_{jl}B_{jl}A_{lk}B_{lk}A_{jk}^2\tfrac{1}{45}(4\lambda^3+2\lambda^2)-A_{jl}A_{lk}B_{lk}A_{jk}^2B_{jk}\tfrac{1}{45}(16\lambda^3-22\lambda^2)]\ .
\end{aligned}\tag{3.10}$$

To check the correctness of the algebra in (3.10), we find when $S=\frac{1}{2}$ and the B's are assumed a constant value, independent of the subscript, all the terms involving the A's will vanish. For the purely dipolar case, by setting $A=-\frac{1}{3}B$, (3.10) reduces to

$$\begin{aligned}
D_p=N^{-1}h^{-6}\tfrac{1}{7290}\sum_{l\neq j\neq k}&[(3660\lambda^3-1035\lambda^2)B_{jk}^4B_{kl}^2+(696\lambda^3-27\lambda^2)B_{jk}^4B_{jl}B_{kl}+(2016\lambda^3-657\lambda^2)B_{jk}^3B_{jl}B_{kl}^2\\
&-(600\lambda^3-360\lambda^2)B_{jk}^3B_{kl}^3-(522\lambda^3-459\lambda^2)B_{jk}^2B_{jl}^2B_{kl}^2]\ .
\end{aligned}\tag{3.11}$$

C. Evaluation of the Four-Particle Terms

The contribution of the four-particle terms to the sixth moment comes from (2.19) and (2.20). The calculations are actually less formidable than they may seem. In evaluating (2.19), we observe the following properties: (i) only the last six terms in the three-indices cyclic terms, in the order of their appearance in the cyclic-permutation bracket terms as defined in (2.13), 18 to 23, will give nonvanishing contributions. (ii) as 18 and 19, 20 and 21, 22 and 23 are of the same scalar function forms, there would be, basically, six types of scalar functions; (iii) each term like $\mathrm{Tr}[jkl][klm]$ would give rise to two nonvanishing products; (iv) all the trace values are equal in magnitude, namely $\pm\frac{1}{81}\lambda^4(2S+1)^N$, where $\lambda\equiv S(S+1)$; (v) symmetries between the first and third indices inside the three-indices cyclic term, e.g., $[jkl]=[lkj]$.

Then, by picking out all the nonvanishing products, factorizing out the common trace value, rearranging terms, and then flipping indices, we obtained the following result:

$$(2.19)=2(2S+1)^N\frac{\lambda^4}{81}\sum_{l\neq j\neq m\neq k}\{\tfrac{1}{2}(20\text{ of}[jlm])(20\text{ of}[jkm])+\tfrac{1}{2}(23\text{ of}[jkm])(23\text{ of}[jkl])+\tfrac{1}{2}(19\text{ of}[jkm])(19\text{ of }[jkl])\cdot$$
$$+(23\text{ of }[klm])(18\text{ of }[jkl])-(18\text{ of }[jkl])(21\text{ of }[lmk])-(22\text{ of }[jkl])(20\text{ of }[lmk])\}\,. \qquad (3.12)$$

To evaluate (2.20), we notice the following properties.

(a) There are only five terms in each of the four four-index permutation brackets. All the traces of the cross product of terms in each of the permutation brackets vanish. If we label the terms in $[jklm]$ by names of $\phi 1$, $\theta 1$, $\gamma 1$, $\alpha 1$, and $\beta 1$, respectively, in the successive order they appear in the bracket, use the numeral 2 to denote terms from $[klmj]$, 3 from $[lmjk]$, and 4 from $[mjkl]$. We observe that all the trace values of these terms are equal to $\frac{1}{81}\lambda^4(2S+1)^N$. After bringing out all the trace values, we have again to deal with scalar functions. Without further confusion, we call these functions according to the previous notations.

(b) The functions $\phi 1$, $\theta 1$, and $\gamma 1$ are symmetric with respect to the indices j and l; functions $\alpha 1$ and $\beta 1$ are symmetric among indices k, l, and m. Consequently, the squares of the functions have the same kinds of the symmetries as the functions themselves.

(c) Furthermore, if we interchange j and k, $\phi 1$ and $\theta 1$ will interchange; if we have $j\to l$, $k\to m$, $l\to k$, $m\to j$, then $\phi 1\to\gamma 1$. This means $\phi 1^2$, $\theta 1^2$, and $\gamma 1^2$ will have the same functional form; whereas $\alpha 1^2$ and $\beta 1^2$ will have different functional forms.

Then by flipping indices bringing out the common trace value, we obtained

$$(2.20)=(2S+1)^N\tfrac{1}{81}\lambda^4\sum_{l\neq j\neq k\neq m}(\tfrac{1}{2}\phi 1^2+\tfrac{1}{6}\alpha 1^2+\tfrac{1}{6}\beta 1^2) \qquad (3.13)$$

Therefore, the total number of all the four-particle interaction terms of the sixth moment is given by X_T, where

$$X_T=(3.12)+(3.13)\,. \qquad (3.14)$$

After carrying out the multiplication and regrouping terms, and dividing out by $h^6\frac{1}{3}\lambda(2S+1)^N$, the contribution of the above equation (3.14) to the sixth moment is seen to be S_T where

$$\begin{aligned}S_T=N^{-1}h^{-6}\tfrac{1}{27}\lambda^3\sum_{m\neq l\neq k\neq j}(&15B_{jk}^2B_{kl}^2B_{km}^2-6A_{jk}B_{jk}B_{km}B_{kl}B_{jl}B_{jm}-6A_{jk}B_{jk}B_{jm}^2B_{kl}^2+48A_{jk}B_{jk}B_{jl}^2B_{jm}^2\\
&-36A_{jk}B_{jk}B_{km}B_{jm}B_{kl}^2-A_{jk}^2B_{jl}^2B_{km}^2+30A_{jk}^2B_{km}^2B_{lm}^2+24A_{jk}^2B_{km}^2B_{kl}^2-26A_{jk}^2B_{km}^2B_{jl}B_{kl}+3A_{jk}^2B_{jl}B_{jm}B_{kl}B_{km}\\
&-30A_{jk}^2B_{lm}^2B_{km}B_{jm}+20A_{jk}B_{jk}A_{jl}B_{jl}B_{lm}B_{km}-50A_{jk}B_{jk}A_{jl}B_{jl}B_{jm}B_{lm}+6A_{jk}B_{jk}A_{jl}B_{jl}B_{lm}^2+11A_{jk}B_{lm}A_{jl}B_{km}B_{kl}^2\\
&+16A_{jk}B_{jk}A_{jl}A_{kl}B_{jm}B_{lm}-6A_{jk}B_{jk}A_{jl}B_{km}B_{lm}B_{kl}-18A_{jk}B_{kl}A_{jl}B_{jl}B_{lm}^2-6A_{jl}B_{kl}A_{jk}B_{jk}B_{lm}^2\\
&-10A_{jk}B_{jk}A_{jl}B_{km}B_{jm}B_{kl}-6A_{jk}A_{jl}B_{lm}B_{jm}B_{kl}^2+49A_{jk}B_{jk}A_{jl}B_{jl}B_{jm}^2-26A_{jk}B_{kl}A_{jl}B_{jl}B_{jm}^2+18A_{jk}A_{jl}B_{kl}^2B_{lm}^2\\
&+2A_{jk}A_{jl}B_{jm}^2B_{kl}^2+12A_{jk}A_{lm}A_{jl}B_{km}B_{jm}^2-4A_{jk}A_{lm}A_{jl}B_{kl}B_{jm}^2-4A_{jk}A_{lm}A_{jl}B_{jl}B_{km}^2-12A_{jk}A_{lm}A_{jl}B_{kl}B_{km}^2\\
&+18A_{jk}A_{jl}A_{lm}B_{lm}B_{km}^2+12A_{jk}A_{lm}A_{jl}B_{km}B_{kl}B_{jm}-6A_{jk}B_{lm}A_{jm}A_{jl}B_{kl}^2+4A_{jk}A_{jl}A_{jm}B_{jm}B_{kl}^2-10A_{jk}A_{jl}A_{kl}B_{kl}B_{jm}^2\\
&+6A_{jk}A_{jl}A_{kl}B_{kl}B_{km}^2+26A_{jk}A_{jl}A_{lm}B_{lm}B_{kl}^2-44A_{jk}A_{jl}A_{lm}B_{lm}B_{km}B_{kl}+16A_{jk}B_{km}A_{jl}B_{kl}A_{jm}B_{jm}\\
&+26A_{jk}B_{kl}A_{jl}B_{jm}A_{lm}B_{lm}+18A_{jk}B_{jk}A_{kl}B_{lm}A_{jl}B_{jm}-4A_{jk}A_{lm}A_{jl}B_{jl}B_{jm}B_{k}-18A_{jk}B_{jk}A_{jl}B_{km}A_{lm}B_{kl}\\
&+6A_{jl}B_{jl}A_{jk}B_{lm}A_{jm}B_{kl}-14A_{jl}B_{jl}A_{jk}B_{lm}A_{kl}B_{jm}+18A_{jk}B_{km}A_{jl}B_{jl}A_{lm}B_{lm}-26A_{jl}B_{jl}A_{lm}B_{lm}A_{jk}B_{kl}\\
&+16A_{jl}B_{km}A_{jk}B_{jk}A_{lm}B_{lm}-34A_{jl}B_{kl}A_{jk}B_{jk}A_{jm}B_{jm}+16A_{jl}B_{jl}A_{lm}B_{lm}A_{jk}B_{jk}-32A_{jl}B_{kl}A_{lm}B_{lm}A_{jk}B_{jk}\\
&+14A_{jk}B_{jk}A_{jl}B_{jl}A_{jm}B_{jm}+36A_{jk}^2B_{kl}^2A_{lm}B_{lm}-36A_{jk}^2B_{jm}B_{km}A_{lm}B_{lm}+36A_{jk}^2B_{jl}B_{km}A_{lm}B_{lm}-36A_{jk}^2B_{jl}B_{jm}A_{lm}B_{lm}\\
&-46A_{jk}^2A_{jl}B_{jl}B_{jm}B_{lm}+18A_{jk}^2B_{lm}B_{km}A_{jl}B_{jl}+26A_{jk}^2A_{jl}B_{lm}B_{jm}B_{kl}+2A_{jk}^2B_{lm}B_{kl}A_{jl}B_{km}+8A_{jk}^2A_{jl}B_{jm}B_{kl}B_{km}\\
&-6A_{jk}^2B_{km}^2A_{jl}B_{kl}-14A_{jk}^2B_{jm}^2A_{jl}B_{kl}-36A_{jk}^2B_{km}B_{jm}A_{jl}B_{jl}+38A_{jk}^2B_{jm}^2A_{jl}B_{jl}+10A_{jk}^2B_{km}^2A_{jl}B_{jl}-16A_{jk}^2B_{lm}^2A_{jl}B_{kl}\\
&+16A_{jk}^2B_{lm}^2A_{jl}B_{jl}+8A_{jk}^2A_{jl}^2B_{lm}^2+20A_{jl}^2A_{jk}^2B_{jm}^2-40A_{jk}^2A_{jl}^2B_{jm}B_{km}+12A_{jk}^2A_{jl}^2B_{lm}B_{km}+24A_{jk}^2A_{lm}^2B_{jm}B_{kl}+24A_{jk}^2A_{lm}^2B_{kl}^2\\
&-48A_{jk}^2A_{lm}^2B_{kl}B_{km}+8A_{km}^2A_{jl}A_{jk}B_{lm}^2-8A_{km}^2A_{jk}B_{lm}A_{jl}B_{jm}+32A_{km}^2A_{jk}A_{jl}B_{jm}B_{kl}+40A_{km}^2A_{jk}A_{jl}B_{kl}^2\\
&-40A_{km}^2A_{jk}B_{jk}A_{jl}B_{kl}-48A_{km}^2A_{jk}B_{lm}A_{jl}B_{kl}+24A_{km}^2A_{jk}B_{jk}A_{jl}B_{jl}-24A_{km}^2A_{jl}B_{jl}A_{jk}B_{jm}-24A_{km}^2A_{jl}B_{jl}A_{jk}B_{kl}\\
&+24A_{km}^2A_{jl}B_{jl}A_{jk}B_{lm}+16A_{km}^2A_{jl}B_{lm}A_{jk}B_{jk}-4A_{jk}^2B_{lm}^2A_{jl}A_{km}-4A_{jk}^2A_{km}B_{km}A_{jl}B_{kl}-4A_{jk}^2A_{km}B_{jm}A_{jl}B_{kl}\end{aligned}$$

$$-4A_{jk}^2A_{km}B_{lm}A_{jl}B_{jl}+4A_{jk}^2A_{km}B_{km}A_{jl}B_{jl}+12A_{jk}^2A_{km}B_{lm}A_{jl}B_{kl}+16A_{kl}^2A_{jk}B_{lm}A_{jl}B_{jm}-8A_{kl}^2A_{jk}B_{lm}A_{jl}B_{km}$$
$$-8A_{kl}^2B_{jm}^2A_{jk}A_{jl}+4A_{jm}^2B_{kl}^2A_{jl}A_{jk}+22A_{jm}^2A_{jl}B_{jl}A_{jk}B_{jk}+12A_{jm}^2A_{jl}B_{km}A_{jk}B_{kl}+6A_{jm}^2A_{jl}B_{km}A_{jk}B_{lm}$$
$$-24A_{jm}^2A_{jk}B_{jk}A_{jl}B_{lm}-20A_{jm}^2A_{jl}B_{jl}A_{jk}B_{kl}-8A_{jk}A_{lm}A_{jl}A_{kl}B_{jm}^2+4A_{lm}B_{lm}A_{kl}B_{jm}A_{jl}A_{jk}$$
$$+8A_{jk}B_{jk}A_{jl}B_{km}A_{kl}A_{lm}-12A_{kl}B_{kl}A_{jk}A_{lm}A_{jl}B_{km}+8A_{jl}B_{jl}A_{jk}A_{lm}A_{kl}B_{km}+4A_{jl}B_{km}A_{jk}A_{lm}A_{kl}B_{jm}$$
$$-8A_{jk}B_{jk}A_{lm}B_{lm}A_{jl}A_{kl}+4A_{lm}B_{lm}A_{kl}B_{kl}A_{jl}-52A_{jk}B_{jm}A_{km}A_{lm}A_{jl}B_{jl}+22A_{km}A_{lm}A_{jl}B_{jl}A_{jk}B_{jk}$$
$$+18A_{jk}A_{lm}A_{jl}A_{km}B_{kl}^2+8A_{jk}A_{lm}A_{jl}A_{km}B_{jm}B_{kl}+4A_{jk}B_{jk}A_{lm}B_{lm}A_{jl}A_{km})\ . \qquad (3.15)$$

To check the correctness of the algebra, all terms involving the A's will vanish when the B's are assumed a constant value independent of the subscripts. For the purely dipolar case, Eq. (3.15) is reduced to S_p, where

$$S_p=N^{-1}\hbar^{-6}\frac{\lambda^3}{2187}\sum_{l\neq m\neq j\neq k}(462B_{jk}^2B_{jm}^2B_{jl}^2+303B_{jk}^2B_{kl}^2B_{lm}^2+360B_{jk}^2B_{kl}^2B_{jm}B_{km}-36B_{jk}^2B_{lm}^2B_{jm}B_{km}$$
$$-96B_{jk}^2B_{lm}^2B_{jl}B_{km}+72B_{jl}^2B_{lm}^2B_{jk}B_{km}+174B_{jk}^2B_{jl}B_{km}B_{jm}B_{kl}-24B_{jk}B_{lm}B_{jl}B_{km}B_{jm}B_{kl})\ . \qquad (3.16)$$

At last, the sixth moment as given by (2.12) is finally reduced to

$$\langle\Delta\nu^6\rangle=(3.6)+(3.10)+(3.15)\ . \qquad (3.17)$$

The normalized sixth moment for the purely dipolar case is then given by

$$\langle\Delta\nu^6\rangle=(3.7)+(3.11)+(3.16)=N^{-1}\hbar^{-6}\tfrac{1}{42525}(7746\lambda^3-5079\lambda^2+1224\lambda)\sum_{k>j}B_{jk}^6+N^{-1}\hbar^{-6}\tfrac{1}{7290}\sum_{l\neq j\neq k}[(3660\lambda^3$$
$$-1035\lambda^2)B_{jk}^4B_{kl}^2+(696\lambda^3-27\lambda^2)B_{jk}^4B_{jl}B_{kl}+(2016\lambda^3-657\lambda^2)B_{jk}^3B_{kl}^2B_{jl}-(600\lambda^3-360\lambda^2)B_{jk}^3B_{kl}^3$$
$$-(522\lambda^3-459\lambda^2)B_{jk}^2B_{jl}^2B_{kl}^2]+N^{-1}\hbar^{-6}\frac{\lambda^3}{2187}\sum_{l\neq m\neq j\neq k}(462B_{jk}^2B_{jm}^2B_{jl}^2+303B_{jk}^2B_{kl}^2B_{lm}^2+360B_{jk}^2B_{kl}^2B_{jm}B_{km}$$
$$-36B_{jk}^2B_{lm}^2B_{jm}B_{km}-96B_{jk}^2B_{lm}^2B_{jl}B_{km}+72B_{jl}^2B_{lm}^2B_{jk}B_{km}+174B_{jk}^2B_{jl}B_{km}B_{jm}B_{kl}-24B_{jk}B_{lm}B_{jl}B_{km}B_{jm}B_{kl})$$
$$(3.18)$$

IV. CONCLUSION

It is found that in the case when exchange interaction is present the sixth moment consists of five types of two-particle terms, 47 of three-particle terms, and 115 four-particle terms. Two types of four-particle terms, $A_{jl}B_{jl}B_{jm}B_{km}A_{jk}A_{lm}$ and $A_{km}^2B_{lm}^2A_{jk}A_{jm}$, do not appear in the final result, as they happen to have zero coefficient. As the trace value of the four-particle terms is much greater than the two- and three-particle terms, it is only natural that most of the contribution to the sixth moment should come from the four-particle terms. By looking at the sixth moment for the purely dipolar case, we observe that there are one two-particle term, five three-particle terms, and nine four-particle terms, even though the type of term $B_{jk}^2B_{jl}B_{lm}B_{jm}B_{kl}$ has zero coefficient in the final result.

APPENDIX: EVALUATION OF SOME TRACES

A. Trace Value of Odd-Power Spin Operators Product

We first want to evaluate $\mathrm{Tr}\,S_{xj}^3S_{yj}S_{zj}$. We define the ladder operators S_{j+} and S_{j-} where

$$S_{j+}=S_{jx}+iS_{jy}\ , \qquad (A1)$$

$$S_{j-}=S_{jx}-iS_{jy}\ . \qquad (A2)$$

From (A1) and (A2), we have

$$S_{jx}=\tfrac{1}{2}(S_{j+}+S_{j-})\ , \qquad (A3)$$

$$S_{jy}=(S_{j+}-S_{j-})/2i\ . \qquad (A4)$$

It is well known from elementary algebra[18–20] that

$$\sum_{-S}^{S}m^4=\tfrac{1}{5}(2S+1)[S^2(S+1)^2-\tfrac{1}{3}S(S+1)]\ , \qquad (A5)$$

$$\sum_{-S}^{S}m^n=0\ ;$$

and

$$\sum_{-S}^{S}m^2=\tfrac{1}{3}S(S+1)(2S+1)\ , \qquad (A6)$$

where m is either an integer or half an intger, and n stands for an odd integer. Then from elementary quantum mechanics,[21] we have

$$\mathrm{Tr}\,S_{j-}^2S_{j+}^2S_{zj}=\sum_{m=-S}^{m=S}\{[S(S+1)-m(m+1)]$$

$$\times [S(S+1)-(m+1)(m+2)]m\} . \quad (A7)$$

By means of (A5) and (A6), (A7) is reduced to

$$\mathrm{Tr}\, S_{j-}^2 S_{j+}^2 S_{zj} = -S(S+1)(2S+1)[\tfrac{8}{15} S(S+1) - \tfrac{2}{5}] . \quad (A8)$$

In the same fashion, we have

$$\mathrm{Tr}\, S_{j+}^2 S_{j-}^2 S_{zj} = -\mathrm{Tr}\, S_{j-}^2 S_{j+}^2 S_{zj} = (2S+1)S(S+1)[\tfrac{8}{15} S(S+1) - \tfrac{2}{5}] . \quad (A9)$$

Also, by the same reasoning, we have,

$$\mathrm{Tr}\, S_{j+} S_{j-} S_{zj}^2 = \sum_{-S}^{S} [S(S+1) - m(m-1)] m^2 = (2S+1)S(S+1)[\tfrac{2}{15} S(S+1) + \tfrac{1}{15}] . \quad (A10)$$

Likewise, we have,

$$\mathrm{Tr}\, S_{j-} S_{j+} S_{zj}^2 = \mathrm{Tr}\, S_{j+} S_{j-} S_{zj}^2 = (2S+1)S(S+1)[\tfrac{2}{15} S(S+1) + \tfrac{1}{15}] . \quad (A11)$$

Making use of the cyclic property of a trace and the following commutation relation we find

$$[S_{zj}, S_{j+}] = S_{j+} , \quad [S_{zj}, S_{j-}] = -S_{j-} , \quad [S_{j+}, S_{j-}] = 2S_{zj} . \quad (A12)$$

We have, from (A1) and (A2)

$$\mathrm{Tr}\, S_{xj}^3 S_{yj} S_{zj} = \mathrm{Tr}\left(\frac{S_{j+}+S_{j-}}{2}\right)^3 \left(\frac{S_{j+}-S_{j-}}{2i}\right) S_{zj} = \frac{1}{16i}\, \mathrm{Tr}(S_{j-}^2 S_{j+}^2 S_{zj} - S_{j+}^2 S_{j-}^2 S_{zj} - 2S_{j+} S_{j-} S_{zj}^2 - 2S_{j-} S_{j+} S_{zj}^2) , \quad (A13)$$

where properties of the ladder operators in taking the trace value such as $\mathrm{Tr}\, S_{j-}^4 = 0$ have been utilized. Then by (A9) and (A11), we obtain

$$\mathrm{Tr}\, S_{xj}^3 S_{yj} S_{zj} = \frac{-1}{10i} [S(S+1) - \tfrac{1}{3}](2S+1) . \quad (A14)$$

Following similar procedure, we have

$$\mathrm{Tr}\, S_{xj}^3 S_{yj} S_{zj} = \mathrm{Tr}\, S_{xj} S_{yj}^3 S_{zj} = \mathrm{Tr}\, S_{xj} S_{yj} S_{zj}^3 = \frac{-1}{10i} [S(S+1) - \tfrac{1}{3}](2S+1) . \quad (A15)$$

Alternatively, we can get (A15) from (A14) by recalling that the trace value is invariant under orthogonal transformation, in our case, rotation of axes.

B. Trace Value of Even-Power Spin Operators Product

The derivation of $\mathrm{Tr}\, S_{zj}^6$ is straightforward from the fact that

$$\sum_{-S}^{S} m^6 = \tfrac{1}{7} S(S+1)(2S+1)[S^2(S+1)^2 - S(S+1) + \tfrac{1}{3}] . \quad (A16)$$

Next we want to evaluate $\mathrm{Tr}\, S_{xj}^4 S_{yj}^2$. We first notice that

$$\mathrm{Tr}\, S_{j+} S_{j-} S_{zj} = \sum_{-S}^{S} [(S+1)S - m(m-1)] m = \tfrac{1}{3} S(S+1)(2S+1) . \quad (A17)$$

Then by definition of the ladder operators and all previous properties, after some tedious calculation, we obtain

$$\mathrm{Tr}\, S_{xj}^4 S_{yj}^2 = \tfrac{-1}{64}\, \mathrm{Tr}(-4S_{j+}^3 S_{j-}^3 - 12 S_{j+}^2 S_{j-}^2 S_{zj} - 12 S_{j+} S_{j-} S_{zj} + 16 S_{j+} S_{j-} S_{zj}^2 + 6 S_{j-}^2 S_{j+}^2 S_{zj}) . \quad (A18)$$

Then from the definition of the ladder operation, we have

$$\mathrm{Tr}\, S_{j+}^3 S_{j-}^3 = \sum_{m=-S}^{m=S} \{[S(S+1) - m(m-1)][S(S+1) - (m-1)(m-2)][S(S+1) - (m-2)(m-3)]\}$$
$$= (2S+1)S(S+1)[S^2(S+1)^2 \tfrac{16}{35} - S(S+1)\tfrac{44}{35} + \tfrac{24}{35}] , \quad (A19)$$

where (A5), (A6), and (A16) have been used in the simplification. Then by (A9), (A10), (A17), and (A19), (A18) can be simplified to

$$\mathrm{Tr}\, S_{xj}^4 S_{yj}^2 = \tfrac{1}{7}(2S+1)S(S+1) \times [\tfrac{1}{5} S^2(S+1)^2 + \tfrac{4}{15} S(S+1) - \tfrac{1}{6}] . \quad (A20)$$

Either following the same procedure or by the invariance of the trace value under rotation of axes, we have

$$\mathrm{Tr}\, S_{xj}^4 S_{zj}^2 = \mathrm{Tr}\, S_{xj}^4 S_{yj}^2 = \mathrm{Tr}\, S_{yj}^4 S_{xj}^2 = \mathrm{Tr}\, S_{yj}^4 S_{zj}^2 = \mathrm{Tr}\, S_{zj}^4 S_{yj}^2 = \mathrm{Tr}\, S_{zj}^4 S_{xj}^2 = \tfrac{1}{7}(2S+1)S(S+1)[\tfrac{1}{5} S^2(S+1)^2 + \tfrac{4}{15} S(S+1) - \tfrac{1}{6}] . \quad (A21)$$

C. Trace Value of Homogeneous Product of Spin Operators

Finally, we want to derive $\mathrm{Tr} S_{xj}^2 S_{yj}^2 S_{zj}^2$. We first notice that

$$\mathrm{Tr} S_{j-}^2 S_{j+}^2 S_{zj}^2 = \sum_{-S}^{S} \{[S(S+1) - m(m+1)][S(S+1) - (m+1)(m+2)] m^2\}$$
$$= (2S+1)S(S+1)[S^2(S+1)^2 \tfrac{8}{105} + S(S+1)\tfrac{34}{105} - \tfrac{8}{21}] . \quad (A22)$$

TABLE I. Trace values of the one-particle spin operators.

$\mathrm{Tr}\, S_{zj} = \mathrm{Tr}\, S_{xj} = \mathrm{Tr}\, S_{yj} = 0;$

$\mathrm{Tr}\, S_{xj}^3 = \mathrm{Tr}\, S_{xj}^2 S_{yj} = \mathrm{Tr}\, S_{yj}^2 S_{zj} = 0;$

$\mathrm{Tr}\, S_{xj} S_{xk} = 0,\ j \neq k$ (etc.);

$\mathrm{Tr}\, S_{xj} S_{yj} S_{zj}^2 = \mathrm{Tr}\, S_{xj}^3 S_{yj} = \mathrm{Tr}\, S_{xj} S_{yj}^3 = 0$ (etc.).

$\mathrm{Tr}\, S_{xj}^2 = \mathrm{Tr}\, S_{yj}^2 = \mathrm{Tr}\, S_{zj}^2 = \frac{1}{3} S(S+1)(2S+1)^N;$

$\mathrm{Tr}\, S_{xj} S_{yj} S_{zj} = \frac{1}{6} i S(S+1)(2S+1)^N.$

$\mathrm{Tr} S_{zj}^4 = \mathrm{Tr}\, S_{xj}^4 = \mathrm{Tr}\, S_{zj}^4 = \frac{1}{5}\,[S(S+1) - \frac{1}{3}] S(S+1)(2S+1)^N.$

$\mathrm{Tr}\, S_{xj}^2 S_{yj}^2 = \mathrm{Tr}\, S_{yj}^2 S_{zj}^2 = \mathrm{Tr}\, S_{xj}^2 S_{zj}^2$
$= \frac{1}{5} [\frac{1}{3} S(S+1) + \frac{1}{6}]\, S(S+1)(2S+1)^N.$

$\mathrm{Tr}\, S_{xj}^2 S_{yj}^2 S_{zj}^2 = \mathrm{Tr}\, S_{yj}^2 S_{xj}^2 S_{zj}^2$
$= \frac{1}{7} [\frac{1}{15} S^2 (S+1)^2 - \frac{3}{10} S(S+1) + \frac{1}{3}]\, S(S+1)(2S+1)^N.$

(This is derived directly, *not* by the cyclic-permutation property of the trace.)

$\mathrm{Tr}\, S_{xj}^4 S_{zj}^2 = \mathrm{Tr}\, S_{xj}^4 S_{yj}^2 = \mathrm{Tr}\, S_{yj}^4 S_{xj}^2 = \mathrm{Tr}\, S_{yj}^4 S_{zj}^2 = \mathrm{Tr}\, S_{yj}^2 S_{zj}^4$
$= \mathrm{Tr}\, S_{xj}^2 S_{zj}^4 = \frac{1}{7} [\frac{1}{5} S^2 (S+1)^2 + \frac{4}{15} S(S+1) - \frac{1}{6}]\, S(S+1)(2S+1)^N.$

$\mathrm{Tr}\, S_{zj}^6 = \mathrm{Tr}\, S_{yj}^6 = \mathrm{Tr}\, S_{xj}^6 = \frac{1}{7} [S^2 (S+1)^2 - S(S+1) + \frac{1}{3}]\, S(S+1)(2S+1)$

$\mathrm{Tr}\, S_{xj}^3 S_{yj} S_{zj} = \mathrm{Tr}\, S_{xj} S_{yj}^3 S_{zj} = \mathrm{Tr}\, S_{xj} S_{yj} S_{zj}^3$
$= -(1/10i)\, [S(S+1) - \frac{1}{3}] S(S+1)(2S+1)^N.$

Similarly, it is found that

$$\mathrm{Tr} S_{j-}^2 S_{j+}^2 S_{zj}^2 = \mathrm{Tr} S_{j+}^2 S_{j-}^2 S_{zj}^2 \,. \qquad \text{(A23)}$$

Then expressed in terms of ladder operators, we have

$$\mathrm{Tr} S_{xj}^2 S_{yj}^2 S_{zj}^2 = -\tfrac{1}{16}\, \mathrm{Tr}(12 S_{zj}^4 - S_{j+}^2 S_{j-}^2 S_{zj}^2 - S_{j-}^2 S_{j+}^2 S_{zj}^2 - 8 S_{j+} S_{j-} S_{zj}^2)\,. \qquad \text{(A24)}$$

From (A11), (A23), and (A22), (A24) is found to be

$$\mathrm{Tr} S_{xj}^2 S_{yj}^2 S_{zj}^2 = \tfrac{1}{7} (2S+1) S(S+1) [\tfrac{1}{15} S^2 (S+1)^2 - \tfrac{3}{10} S(S+1) + \tfrac{1}{3}]\,. \qquad \text{(A25)}$$

Likewise, we have

$$\mathrm{Tr} S_{xj}^2 S_{yj}^2 S_{zj}^2 = \mathrm{Tr} S_{yj}^2 S_{xj}^2 S_{zj}^2 = \tfrac{1}{7} (2S+1) S(S+1) \times [\tfrac{1}{15} S^2 (S+1)^2 - \tfrac{3}{10} S(S+1) + \tfrac{1}{3}]\,. \qquad \text{(A26)}$$

It is interesting to notice that in (A26) the permutation is not cyclic and yet both trace values are equal. To check the correctness of our algebra, we observe that when $S = \frac{1}{2}$, (A16), (A21), and (A26) are all reduced to the same value, because in this special case, we have $\mathrm{Tr} S_{zj}^2 = \mathrm{Tr} S_{xj}^2 = \mathrm{Tr} S_{yj}^2 = \frac{1}{4}$; a unique value. In all our previous derivation, we assume that our quantum system consists of a single particle which can have $(2S+1)$ quantum numbers. Physically, this means we have a density matrix of $(2S+1) \times (2S+1)$ elements. In a system containing N particles, we must use a statistical ensemble described by a $(2S+1)^N \times (2S+1)^N$ density matrix. For a system of N identical particles, the quantum wave function is a direct product containing N eigenstates each. This means that instead of $(2S+1)$ eigenvalues for a single particle at a time, we can have $(2S+1)^N$ by evaluating our eigenvalues of N particles simultaneously for a direct product. This implies we must replace $(2S+1)$ by $(2S+1)^N$ in all our previously derived equations. Hence all the trace equations listed in Table I.

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