Sixth Moment of the Magnetic-Resonance Line Shape*

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(Received 13 April 1972)

The sixth moment of the magnetic-resonance line shape is presented as the sum of nine lattice sums instead of ten as reported by Das and Bersohn. The sixth moment is evaluated for simple-cubic, bcc, and fcc symmetry and the applied magnetic field along the [100], [110], and [111] directions. The three-index lattice sums are evaluated two different ways: the usual way by summing each index over a sphere of lattice points and secondly by rewriting the three-index sums as a combination of one- and two-index sums. The two-index sums were then evaluated by summing the second index over a sphere centered on the first index. Lattice sums containing odd powers of B_{ij} are not always much smaller than lattice sums containing only even powers, as has been assumed in previous free-induction-decay (FID) calculations. A moment analysis of the Lowe–Norberg and Evans–Powles FID expansions shows that they contain relatively small parts of M_6 and the higher moments, so that they cannot be improved by the addition of complete higher-moment terms.

I. INTRODUCTION

Even since Van Vleck showed how to calculate the moments of the line shape, the moment concept has been one of the more important concepts of the line-shape problem in magnetic resonance.[1] Moments of experimental curves are determined and compared with theoretical values. Theoretical expansions of the free-induction decay are proposed using the known moments in the evaluation of expansion parameters. The second and fourth moments have been calculated for general conditions, but the sixth moment has been calculated only for simple-cubic (sc) symmetry and the applied magnetic field along the [100] direction.[2]

Das and Bersohn have shown that the $n+1$ particle terms in the $2n$th moment (M_{2n}) constitute the major part of the moment so that one need not calculate the entire moment to get a good approximation to its value.[3] So we present here a calculation of the four-particle terms in M_6 for the purpose of using it to improve the fit of theoretical expansions (in particular the Lowe–Norberg expansion which contains complete M_2 and M_4 terms) to the experimental curves.[4,5] Also Parker has given expansion theorems for the free-induction decay (FID) which involve all the moments of the line shape, and the number of terms which can be included depends on the number of moments which are known.[6]

In Sec. II, our method of obtaining the four-particle part of M_6 is presented. Our method is explained rather thoroughly, since we obtain nine lattice sums [Eq. (10)], whereas Das and Bersohn say that there should be ten. In Sec. III, the four-particle part of M_6 and the nine lattice sums are evaluated for sc, bcc, and fcc symmetry and [100], [110], and [111] directions for the applied magnetic field. We do not *a priori* discard the lattice sums containing odd powers of B_{ij}, but do evaluate them and show that they are not necessarily much smaller than the even-power lattice sums.

In Sec. IV, a moment analysis of the Lowe–Norberg and Evans–Powles FID expansions is given and a M_6 correction term is applied.[7]

II. FOUR-PARTICLE PART OF M_6

Since Das and Bersohn's four-particle part of the sixth moment consists of ten lattice sums, while ours consists of nine, it is appropriate to show how our four-particle part was obtained. We proceed as follows.

The sixth moment M_6 of the magnetic-resonance absorption relative to the Larmor frequency is

$$M_6 = -\hbar^{-6}\,\mathrm{Tr}([H,[H,[H,I_x]]])^2/\mathrm{Tr}(I_x^2), \tag{1}$$

where H is the truncated part of the magnetic dipole-dipole Hamiltonian.[8] That is,

$$H = \tfrac{1}{2}\sum_{j\neq k}^{N} B_{jk} I_{jz} I_{kz} - \tfrac{1}{6}\sum_{j\neq k}^{N} B_{jk}\vec{I}_j\cdot\vec{I}_k\,, \tag{2}$$

where

$$B_{jk} = (3\gamma^2\hbar^2/2r_{jk}^2)(1-3\cos^2\theta_{jk}),$$

and all the symbols have their usual meanings.

M_6 consists of four-, three-, and two-particle terms with the four-particle part comprising the major part and fortunately the easiest of the three parts to calculate. The four-particle part of M_6 (hereafter referred to as ${}^4M_6$) was determined as follows.

The commutator in M_6 consists of four-particle, three-particle, and two-particle terms. That is,

$$[H,[H,[H,I_x]]] = C_4 + C_3 + C_2\,, \tag{3}$$

where C_4, C_3, and C_2 are the four-, three-, and two-particle parts, respectively. For example,

a typical term in C_4 is

$$\sum_{i\neq j\neq k\neq 1} B_{ij}B_{k1}B_{jk}I_{ix}I_{jy}I_{kz}I_{1x}.$$

Terms in C_3 are of the types

$$\sum_{i\neq j\neq k} B_{ij}B_{jk}^2 I_{ix}I_{jx}I_{jy}I_{kz}$$

and

$$\sum_{i\neq j\neq k} B_{ij}^2 B_{jk}I_{ix}^2 I_{jy}I_{kz}.$$

A typical term in C_2 is

$$\sum_{j\neq k} B_{jk}^3 I_{jx}^2 I_{ky}I_{kz}.$$

Substituting Eq. (3) into (1) yields M_6 as the sum of six traces, that is,

$$M_6 = -\hbar^{-6}\left[\sum_{i=2}^{4} \mathrm{Tr}C_i^2 + \sum_{i>j=2}^{4} \mathrm{Tr}(C_iC_j)\right]/\mathrm{Tr}(I_x^2). \quad (4)$$

Only two of the six traces in Eq. (4) contribute to ${}^4M_6$, namely, $\mathrm{Tr}(C_4^2)$ and $\mathrm{Tr}(C_3^2)$. This occurs as follows: Since the operators in C_4 are of the type $I_{ix}I_{jy}I_{kz}I_{1x}$, that is four traceless operators, C_4 contributes only to ${}^4M_6$ and not to the three- and two-particle parts of M_6. Furthermore, the two cross terms in Eq. (4) involving C_4 are zero because at least one of the traceless single-particle operators in C_4 remains unmatched, when C_4 is combined with the three-particle operators of C_3 or the two-particle operators of C_2.

The three-particle term C_3 consists of two types of terms. The first type such as $I_{ix}I_{jx}I_{jy}I_{kz}$ has three traceless single-particle operators. The second type such as $I_{ix}^2 I_{jy}I_{kz}$ has two traceless single-particle operators and a third single-particle operator $(I_{ix})^2$ which is not traceless. Only the latter type of term in C_3 contributes to ${}^4M_6$, and this contribution comes from the $\mathrm{Tr}(C_3^2)$ term in Eq. (4). This occurs as follows: The second type of term in C_3 contributes to M_6 a trace of the form

$$\mathrm{Tr}(I_{ix}^2 I_{jy}I_{kz}\sum_{1\neq m\neq n} I_{1y}^2 I_{ny}I_{mz}).$$

A four-particle term is obtained if $n=j$, $m=k$, and i and 1 remain unmatched. The necessity of having a nonzero trace for one of the single-particle operators in C_3 is seen from this example.

$\mathrm{Tr}(C_3C_2)$ in Eq. (4) is obviously zero because at least one of the traceless single-particle operators in C_3 is unmatched by the two single-particle operators in C_2. Likewise, $\mathrm{Tr}(C_2^2)$, while not zero, cannot contribute to ${}^4M_6$, because at least one of the two single-particle operators in C_2 is traceless and does have to be matched. Therefore, $\mathrm{Tr}(C_2^2)$ can at most contribute to the three-particle and two-particle terms of M_6.

C_4 and the part of C_3 which contributes to ${}^4M_6$ are

$$C_4 = \frac{i}{9}\sum_{i\neq j\neq k\neq 1} I_{ix}I_{jy}I_{kz}I_{1x}[B_{i1}B_{j1}B_{k1} + B_{jk}(B_{ij}B_{j1}+B_{ik}B_{k1}) + 2B_{ij}B_{k1}(B_{jk}-4B_{i1}) - 3B_{ij}B_{ik}(B_{k1}-2B_{j1})]$$
$$+\frac{i}{9}\sum_{i\neq j\neq k\neq 1} I_{iz}I_{jy}I_{ky}I_{1y}[B_{ik}B_{i1}(B_{ij}-B_{jk}) + 2B_{i1}B_{k1}(B_{j1}-B_{jk})] + \frac{i}{9}\sum_{i\neq j\neq k\neq 1} I_{iy}I_{jz}I_{kz}I_{1z}[B_{i1}B_{k1}(B_{jk}+2B_{j1})$$
$$+2B_{ik}B_{i1}(B_{jk}+2B_{ij})], \quad (5)$$

$$C_3' = \frac{i}{9}\sum_{i\neq j\neq k} I_{ix}^2 I_{jy}I_{kz}[B_{ij}^2(B_{jk}+4B_{ik}) + B_{ik}^2(B_{jk}-2B_{ij}) + B_{ij}B_{ik}B_{jk}] + \frac{i}{9}\sum_{i\neq j\neq k} I_{iy}^2 I_{jy}I_{kz}[2B_{ij}^2 B_{jk} + B_{ik}^2(3B_{jk}-B_{ij})]$$
$$+\frac{i}{9}\sum_{i\neq j\neq k} I_{iz}^2 I_{jy}I_{kz}[2B_{ij}^2(B_{ik}+6B_{jk}) + 2B_{ik}B_{jk}(B_{ij}+B_{ik})]. \quad (6)$$

Therefore, ${}^4M_6$ becomes

$${}^4M_6 = -\hbar^{-6}\{\mathrm{Tr}(C_4)^2 + [\mathrm{Tr}(C_3')^2]_4\}/\mathrm{Tr}(I_x^2), \quad (7)$$

where $[\mathrm{Tr}]_4$ means the four-particle terms in the enclosed trace. Each of the two traces in Eq. (7) involves nine lattice sums, which are designated $S_1, S_2, \ldots, S_9$ and which are listed in the Appendix.

$$\mathrm{Tr}(C_4)^2 = P[123S_1 + 123S_2 + 78S_3 + 27S_4 - 60S_5 + 15S_6 + 243S_7 - 6S_8 - 24S_9], \quad (8)$$

$$[\mathrm{Tr}(C_3')^2]_4 = P[261S_1 + 180S_2 + 270S_3 + 45S_4 - 36S_5 - 18S_6 + 9S_7 + 18S_8], \quad (9)$$

where

$$P = -I^4(I+1)^4(2I+1)^N/(81)^2.$$

Inserting Eqs. (8) and (9) into (7) and using $\mathrm{Tr}I_x^2 = \frac{1}{3}NI(I+1)(2I+1)^N$ yields

$${}^4M_6 = -[I(I+1)/9\hbar^2]^3 N^{-1}[128S_1 + 101S_2 + 116S_3 + 24S_4 - 32S_5 - 1S_6 + 84S_7 + 4S_8 - 8S_9]. \quad (10)$$

III. ${}^4M_6$ FOR CUBIC SYMMETRY

Using Eq. (10), ${}^4M_6$ was evaluated for spins arranged in sc, bcc, and fcc lattices for the three directions of the applied magnetic field [100], [110],

and [111]. The lattice sums in ${}^4M_6$ were evaluated assuming identical spins and using 80, 168, and 200 nearest neighbors for sc, bcc, and fcc, respectively. The values of the lattice sums which were used are listed in Table I in units of x^{-6}, where $x=2d^3/3\gamma^2\hbar[I(I+1)]^{1/2}$ and d is the nearest-neighbor distance for the sc lattice, $2/\sqrt{3}$ times the nearest-neighbor distance for the bcc lattice, and $\sqrt{2}$ times the nearest-neighbor distance for the fcc lattice. The values of ${}^4M_6$ are also listed in Table I in units of x^{-6}.

Lattice Sums

It should be noted that care must be taken in evaluating the four-index lattice sums of ${}^4M_6$.[9] It is by no means obvious that if each index runs over the lattice points in a finite sphere a good approximation to the true value of the sum will result. Therefore, S_1 and S_2 were evaluated two different ways for the sc [100] case. First, S_1 and S_2 were computer calculated by summing the indices over a sphere of 81 lattice points or spins. Secondly, S_1 and S_2 were calculated by simplifying the sums by reducing the number of indices to one, if possible. This was completely possible with S_1 and almost so with S_2 as shown in the following equations:

$$S_1=N[(\sum_j B_{ij}^2)^3-3(\sum_j B_{ij}^2)(\sum_j B_{ij}^4)+2\sum_j B_{ij}^6], \tag{11}$$

$$S_2=N[(\sum_j B_{ij}^2)^3-2(\sum_j B_{ij}^2)(\sum_j B_{ij}^4)+2\sum_j B_{ij}^6 -\sum_{i\neq j} B_{0j}^2 B_{0i}^2 B_{ij}^2], \tag{12}$$

where N is the total number of spins.

The values obtained for S_1 and S_2 by the two different methods are shown in Table II and they agree within a few percent.

The other seven lattice sums of ${}^4M_6$ ($S_3, S_4, \ldots, S_9$) contain odd powers of B. In previous lineshape calculations for cubic symmetry, these lattice sums were neglected because they were thought to be small relative to lattice sums such as S_1 and S_2 which contain only even powers of B. This was checked by evaluating S_3, $S_4, \ldots, S_9$ and, as may be seen from their values listed in Table I, all of them are not small relative to S_1 and S_2. That is, the double B^2 sums S_3, S_4, S_5, and S_6, in general, are an order of magnitude larger than the single B^2 sums S_7, S_8, and S_9 and are less than an order of magnitude smaller than S_1 and S_2. So that in evaluating ${}^4M_6$ from Eq. (10), all lattice sums were used although neglecting S_7, S_8, and S_9 would introduce an error only on the order of 1%.

Since the terms in the sums containing odd powers of B alternate in sign, it was thought that the way in which the range of the summation in-

TABLE I. ${}^4M_6$ and lattice constants in units of x^{-6} for sc, bcc, and fcc lattices and B_0 along [100], [110], and [111].

	${}^4M_6$	S_1	S_2	S_3	S_4	S_5	S_6	S_7	S_8	S_9
sc										
[100]	402	1128	1415	147.1	−104.1	220.3	123.7	40.5	−2.2	−1.4
[110]	27.1	79.8	82.3	14.1	−1.07	10.6	14.5	3.17	0.89	0.94
[111]	2.44	8.13	6.43	0.76	0.086	0.314	0.528	0.469	−0.017	−0.178
bcc										
[100]	6.16×10^2	1.77×10^3	2.06×10^3	2.39×10^2	-7.65×10^1	2.68×10^2	2.26×10^2	6.83×10^1	−2.10	−4.33
[110]	4.13×10^3	1.20×10^4	1.26×10^4	2.15×10^3	2.36×10^2	1.36×10^3	2.22×10^3	8.90×10^2	1.52×10^2	-4.01×10^2
[111]	3.73×10^3	1.03×10^4	1.36×10^4	1.12×10^3	-4.75×10^2	2.02×10^3	1.05×10^3	1.53×10^2	-4.8×10^1	-2.8×10^1
fcc										
[100]	8.34×10^4	2.41×10^5	2.53×10^5	5.66×10^4	1.35×10^4	3.16×10^4	6.10×10^4	2.27×10^4	5.18×10^3	-6.30×10^3
[110]	1.50×10^5	4.09×10^5	5.41×10^5	5.12×10^4	-2.81×10^4	7.88×10^4	5.19×10^4	9.10×10^3	-1.30×10^3	-4.23×10^2
[111]	3.28×10^5	9.81×10^5	10.18×10^5	1.45×10^5	2.07×10^4	9.68×10^4	1.49×10^5	7.67×10^4	7.19×10^3	-4.01×10^4

TABLE II. Values of the lattice sums for the sc [100] case in units of x^{-6}. The values in the first row were obtained by summing over a sphere of 80 nearest neighbors. The values in the second row were obtained by using Eqs. (11)–(13) and summing the second index over a sphere centered on first index.

S_1	S_2	S_3	S_4	S_5	S_6
1128	1415	147.1	−104.1	220.3	123.7
1132	1461	148.1	−122.5	220.1	132.2

dices is chosen can significantly affect the values of the sums. Therefore, the sums S_3, S_4, S_5, and S_6 were computer calculated two different ways for the sc lattice and [100]. That is, each sum was computer calculated by summing the three indices over a sphere containing 81 lattice points (Row 1 in Table II). Secondly, each three-index sum was also evaluated by reducing it to a combination of one-index and two-index sums as seen in Eq. (13). The one-index sums

$$S_3 = N[(\sum_j B_{ij}^2)(\sum_{j\neq i} B_{j0}^2 B_{ji} B_{0i}) - \sum_{j\neq i} B_{j0}^4 B_{ji} B_{0i} - \sum_{j\neq i} B_{j0}^2 B_{i0}^3 B_{ji}],$$

$$S_4 = N[\sum_i (\sum_j B_{0j}^2 B_{ij})^2 - (\sum_j B_{0j}^4)(\sum_j B_{0j}^2) + \sum_j B_{0j}^6], \tag{13}$$

$$S_5 = N[\sum_i (\sum_j B_{j0}^2 B_{ji})(\sum_k B_{ki}^2 B_{k0}) - (\sum_j B_{0j}^3)^2 + \sum_j B_{0j}^6],$$

$$S_6 = N[(\sum_j B_{0j}^2)(\sum_{j\neq i} B_{ij}^2 B_{i0} B_{j0}) - 2\sum_{j\neq i} B_{i0}^3 B_{0j} B_{ij}^2]$$

were then evaluated the usual way by summing over a sphere of 80 nearest neighbors. The two-index sums like

$$\sum_{j\neq k} B_{ij}^2 B_{ik} B_{kj}$$

were evaluated by summing one index over a sphere of 80 nearest neighbors and then summing the second index over a sphere of 80 nearest neighbors centered on the first index. So for the sum

$$\sum_{j\neq k} B_{ij}^2 B_{ik} B_{kj},$$

j was summed over a sphere of nearest neighbors centered at i and k was summed over a sphere centered on j. The results are listed in row 2 in Table II, and it can be seen that S_4 and S_6 are significantly different but that S_1, S_2, S_3, and S_5 differ by 3% or less. This result, however, does depend on the number of nearest neighbors summed over, so that in the limit of an infinite crystal the two ways of evaluating the sums become identical, but for small numbers of nearest neighbors they do differ significantly.

IV. M_6 IN THEORETICAL EXPANSIONS OF FREE-INDUCTION DECAY

One of our purposes in calculating ${}^4M_6$ was to use it to improve the fit of existing theoretical expansions to the experimental free-induction decays (FID). Our rationale was that since some of the existing expansions contain complete second- and fourth-moment terms, the addition of a nearly complete sixth moment would improve the fit for intermediate values of t. Das and Bersohn have argued that ${}^4M_6$ comprises the major part of M_6 and for the one case where a theoretical M_6 is known, namely, Glebashev's for sc and [100], our ${}^4M_6$ is 94% of M_6.[2,3] So our procedure was to extract from the FID expansions the t^6 part and to add in $-{}^4M_6 t^6/(6!)$. This was done for the Lowe–Norberg (LN) and Evans–Powles (EP) expansions and the t^6 parts were evaluated for sc symmetry and [100].[5,7] These expansions surprisingly contain a relatively small part of M_6 ranging from 52% for LN to only 4% for EP. Therefore, the correction terms range from about one-half of $[-{}^4M_6 t^6/(6!)]$ for LN to essentially all of it for EP and are significant well before the first node in the FID is reached, increasing rapidly thereafter to values larger than one. For example, the correction term has the value of -0.15 for the LN expansion at the first node ($t \simeq 0.95x$) and the value -0.5 at $t = 1.1x$. The correction for the EP expansion at the same values of t are approximately -0.3 and -1.0 (0.07 and -0.33 if M_4 as well as M_6 is completed). Therefore, the fit of these expansions to the experimental fits cannot be improved by the addition of the complete sixth moment and a few higher moments. This analysis shows that these FID expansions are not close to being moment expansions in that they contain relatively small parts of the moments beyond M_4.

During this analysis the sum of the first four terms in the moment expansion $(1 - M_2 t^2/2! + M_4 t^4/4! - {}^4M_6 t^6/6!)$ was evaluated and found to give a very close fit to the experimental FID out to the first node.

APPENDIX: NINE LATTICE SUMS

$$S_1 = \sum_{i\neq j\neq k\neq 1} B_{jk}^2 B_{kl}^2 B_{ik}^2,$$

$$S_2 = \sum_{i\neq j\neq k\neq 1} B_{jk}^2 B_{kl}^2 B_{ij}^2,$$

$$S_3 = \sum_{i\neq j\neq k\neq 1} B_{jk}^2 B_{ik}^2 B_{jl} B_{kl},$$

$$S_4 = \sum_{i\neq j\neq k\neq 1} B_{jk}^2 B_{ik}^2 B_{jl} B_{il},$$

$$S_5 = \sum_{i\neq j\neq k\neq 1} B_{jk}^2 B_{il}^2 B_{ik} B_{jl},$$

$$S_6 = \sum_{i\neq j\neq k\neq 1} B_{jk}^2 B_{i1}^2 B_{ik} B_{k1} ,$$

$$S_7 = \sum_{i\neq j\neq k\neq 1} B_{jk}^2 B_{ij} B_{j1} B_{ik} B_{k1} ,$$

$$S_8 = \sum_{i\neq j\neq k\neq 1} B_{jk}^2 B_{ij} B_{j1} B_{ik} B_{i1} ,$$

$$S_9 = \sum_{i\neq j\neq k\neq 1} B_{jk} B_{j1} B_{k1} B_{ij} B_{ik} B_{i1} .$$

*Work supported by NSF-Institutional Grant for Science No. GU-3820.

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[9]Since we are assuming identical spins, the lattice sums are actually three-index sums since only three of the indices have to be summed over.

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Effect of the Anharmonic Interactions on the Spin-Lattice Relaxation of Paramagnetic Ions in a MgO Crystal

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(Received 15 June 1971)

The effect of the anharmonic interaction on the spin-lattice relaxation rates has been studied theoretically for Cr^{3+} and Ni^{2+} ions in a MgO crystal. The modification of the relaxation rates due to the anharmonic interaction has been calculated using the Debye model of phonons and an effective-point-charge model for the ligands. The enhancement of the relaxation rate is shown to depend on the paramagnetic ion under consideration. $1/T_1$ is enhanced by an order of magnitude for the case of Cr^{3+} ion, whereas there is hardly any change for Ni^{2+} ion. This is principally due to the fact that the ratio of spin-phonon-coupling parameters for the direct process to those for the Raman process is smaller in the case of $MgO:Ni^{2+}$ than in the case of $MgO:Cr^{3+}$.

I. INTRODUCTION

The relaxation rates of paramagnetic ions in crystals have so far been calculated on the assumption that the lattice motions are harmonic. The dependence of the relaxation rates on temperature, calculated on the basis of a harmonic lattice and Debye model of phonons, agrees fairly well with that obtained experimentally for a number of paramagnetic ions in crystals.[1] But there are some cases where the strength of the spin-phonon coupling is weak and the relaxation rates are consequently measurable up to high temperatures.[2,3] In these cases the relaxation rates were found to deviate considerably from those predicted by the Debye model of phonons. Recently the Raman relaxation rates for Ni^{2+} and Cr^{3+} ions in MgO have been calculated theoretically[4] taking into account the experimental dispersion relations for the phonons in MgO,[5] but assuming the lattice motions to be harmonic and unmodified due to the substitution of the paramagnetic ions. The agreement between these theoretically calculated values of $1/T_1$ and those obtained experimentally is good for the Ni^{2+} ion, but for $MgO:Cr^{3+}$ the theoretical results are smaller than the experimental ones by a factor of 5 even at low temperatures. The object of the present work is to show that the discrepancies between the theoretical and experimental results for $MgO:Cr^{3+}$ can be accounted for to a fairly good extent by taking the effect of the anharmonic interaction into account.

Van Kranendonk and Walker[6] have recently pointed out the importance of the anharmonic interaction on the nuclear-quadrupole relaxation rates for alkali halide crystals. The anharmonic interaction in conjunction with the direct process of relaxation causes the relaxation rates to depend on tempera-