NMR line-shape calculation for a linear dipolar chain*†

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Numerical calculations have been performed to obtain *exact* results for the high-temperature space- and time-dependent transverse spin pair correlation functions $G_r^x(t)$, where r is the spatial separation of the spins, and the free-induction-decay function $F(t)$, for a linear chain of N dipolar coupled spins $(S = \frac{1}{2})$ for $N = 7$, 9, and 11 with periodic boundary conditions. The range Δ of the dipolar interaction was allowed to take separately the values 1, 2, and 3. The moments of the NMR line shape (M_2 through M_{20}) were obtained for the *infinite* chain with $\Delta = 1$ and $\Delta = 2$ and were found to be in excellent agreement with previous reliable moment calculations (M_2 through M_8). By an evaluation of the contribution of the four-particle terms to M_6, it is shown that the Bersohn-Das theorem fails in one dimension. We find that $G_r^x(t)$ for even (odd) values of r are predominantly positive (negative). This gives rise to a damped beat structure for $F(t)$. A study of $F(t)$ for $N = 7$, 9, and 11 shows that our 11-spin solution reproduces that of the *infinite* chain at least up through the fifth node, and is also in good agreement with recent ^{19}F free-induction-decay measurement in fluoroapatite. The nodes of $F(t)$ were found to shift towards shorter values of time as Δ was allowed to increase. Further, for $N = 11$, a comparison of our exact results for $F(t)$ for $\Delta = 3$, with a Gaussian broadening of $F(t)$ for $\Delta = 2$, shows that the effect of the weak interactions is not well approximated by a simple Gaussian broadening for this case.

I. INTRODUCTION

The nuclear-magnetic-resonance (NMR) line-shape calculation for a rigid lattice of dipolar coupled spins remains a formidable problem.[1] Various proposed expansion schemes[2,3] fail at long times because of a lack of a proper perturbation parameter. A number of microscopic theories[4,5] have been advanced, but to get specific results, one is forced to make phenomenological approximations. Recently, various empirical fitting schemes, based on the known moments of the line shape, have been proposed.[6] Although they yield reasonable agreement with experiments, they fail to give any physical insight into the problem. Almost all the theories have been tested against experimental measurements[2,7] in CaF$_2$ where the ^{19}F nuclei form a simple cubic lattice.

Recently, the spin dynamics in one-dimensional systems has received a great deal of attention.[8] The simpler nature of the problem allows one to obtain exact results for the various physical properties of interest. These are very useful in bringing forth the nature of approximations of more general theories. Moreover, a large class of linear-chain materials have been found[9] where the magnetic part of the interaction is dominated by intrachain interactions. For nuclear spins, an example of such a system is the mineral fluoroapatite [Ca$_5$F(PO$_4$)$_3$]. Recently, it has been shown[10,11] that if the external magnetic field is applied along the hexagonal c axis, the ^{19}F resonance can be well explained in terms of the truncated dipolar inter-

action of the F nuclei situated along this axis.

In this paper we shall examine the behavior of a linear chain of N dipolar coupled spins in an applied magnetic field $\vec{B}_0$. The z axis of our coordinate system will be chosen to lie along $\vec{B}_0$ and the chain axis will be at an angle θ to $\vec{B}_0$. For $\vec{B}_0$ much larger than the local fields arising from the dipolar interaction, the NMR line shape is governed by the usual truncated dipolar Hamiltonian[12] given by

$$\mathcal{H}_d = A \sum_{i=1}^{N} \sum_{j=1}^{\Delta} \frac{S_i^x S_{i+j}^x + S_i^y S_{i+j}^y - 2 S_i^z S_{i+j}^z}{j^3} , \tag{1}$$

where Δ is the range of the dipolar interaction and the interaction parameter A is given by

$$A = \gamma^2 \hbar^2 (3 \cos^2 \theta - 1) / 2a^3 , \tag{2}$$

where a is the distance between neighboring spins. To facilitate comparison with the Heisenberg model, we will often denote $\frac{1}{2} A$ by J. The normalized infinite temperature transverse-spin-correlation functions for spins r units apart are given by[13]

$$G_r^x(t) = 4 \, \text{Tr}\{\exp[i(\mathcal{H}_d/\hbar)t] S_0^x$$
$$\times \exp[-i(\mathcal{H}_d/\hbar)t] S_r^x\}/\text{Tr}(1) , \tag{3}$$

where we have imposed the periodic boundary conditions for the spins. The free-induction-decay (FID) function $F(t)$, which is the Fourier transform of the NMR absorption line shape $G(\omega)$, is related to $G_r^x(t)$ by

$$F(t) = \sum_r G_r^x(t)$$

$$= \text{Tr}\left[\exp\left(i\frac{\mathfrak{IC}_d}{\hbar}t\right)S^x \exp\left(-i\frac{\mathfrak{IC}_d}{\hbar}t\right)S^x \right]\bigg/ \text{Tr}(S^x S^x),$$

$$(4)$$

where S^x is the x component of the total spin.[2] We present here *ab initio* calculations for $G_r^x(t)$ and $F(t)$ for different values of N and Δ, by a method[14] used in our recent studies of the spin dynamics in Heisenberg[13] and[15] XY chains. We also obtain the first 20 frequency moments of $G(\omega)$ which are given by the Taylor series expansion of $F(t)$ around $t = 0$, namely,

$$F(t) = 1 + \sum_{k=1}^{\infty}(-1)^k M_{2k}\frac{t^{2k}}{(2k)!}. \quad (5)$$

We compare our results with the recent experimental measurements of Engelsberg, Lowe, and Carolan[11] (ELC). We also study the validity of the Gaussian-broadening approximation for weak interactions.

Our method of calculation is outlined in Sec. II, the results are presented in Sec. III, and a summary of our work is given in Sec. IV.

II. CALCULATION

The method of calculating $F(t)$ by solving the Hamiltonian for a small number of spins is well known in NMR.[16-18] In one-dimensional systems, the problem is further simplified if one imposes the periodic boundary condition on the spins. If T is the translation operator, then it is easy to show that T, S^z (the z component of the total spin), and $\mathfrak{IC}_d$ form a set of commuting operators. It is therefore possible to obtain numerically all the eigenvectors and the corresponding eigenvalues of $\mathfrak{IC}_d$ in a representation in which T and S^z are diagonal.[11,13] One can also obtain the matrix elements of S^x and S_r^x in this representation, and evaluate the traces in Eqs. (3) and (4), and the moments, in a straightforward manner.[19]

The computer[20] was programmed to evaluate all the relevant matrix elements before and after the diagonalization. The only input were N, Δ, and the states of the Ising basis which are not connected by the translation operator T. Whereas $F(t)$ could be easily calculated once the eigenvalues and eigenvectors are properly stored, because of lack of translation invariance of S_r^x, evaluation of $G_r^x(t)$ was more difficult. As a check on our computations we used the fact that $F(t)$ is independent of time for the isotropic Heisenberg model. But perhaps the best checks are the second and fourth moments which can be calculated by using Van Vleck's results.

III. RESULTS

A. Moments of $F(t)$

The "moment method" of Van Vleck[12] is one of the important theoretical methods of studying NMR line shapes in solids. The conventional starting expression for M_{2k} in Eq. (5) can be obtained from Eq. (4) by expanding the exponential operators, namely,

$$M_{2k} = \text{Tr}([\mathfrak{IC}_d, [\mathfrak{IC}_d, \ldots [\mathfrak{IC}_d, S^x]]\ldots]S^x)/\text{Tr}(S^x S^x).$$

$$(6)$$

There are $2k$ commutators in the above expression. Analytical expressions for M_{2k}, in terms of the interaction parameters of $\mathfrak{IC}_d$, are normally obtained by evaluating the commutations in Eq. (6) and then calculating the trace of the resulting expression in a representation in which S_i^z is diagonal for each i. The labor of calculating M_{2k} increases very rapidly with k, and analytical expressions for only M_2, M_4, M_6, and M_8 have been published[12,21-24] to date.

For the one-dimensional problem, an alternative method[13] of calculating M_{2k} for different values of k is to evaluate the traces in a representation in which $\mathfrak{IC}_d$ is diagonal. To examine how the moments obtained from finite chains approach their asymptotic values as $N \to \infty$, we have evaluated the first 20 moments for $N = 9$ and $N = 11$, while Δ is allowed to take separately the values 1 and 2. Our results for $\Delta = 1$ and $\Delta = 2$ are shown in Tables I and II. By

TABLE I. Moments of the NMR line shape for a linear dipolar chain containing N spins. The range of the interaction is denoted by Δ. The interaction constant A is equal to $\gamma^2/\hbar^2(3\cos^2\theta - 1)/2a^3$ with standard notation. This table corresponds to $\Delta = 1$. The theoretical values for M_2 and M_4 are calculated from Van Vleck,[a] while those for M_6 and M_8 are calculated from Jensen and Hansen.[b] These results are shown in last column of the table.

$2n$	$(M_{2n})^{1/2n}/A$			
				Other theoretical values
	$N=9$	$N=11$	$N=\infty$	
2	2.121 32	2.121 32	2.121 32	2.121 32
4	2.410 28	2.410 28	2.410 28	2.410 28
6	2.573 13	2.573 13	2.573 13	2.573 13
8	2.685 90	2.685 90	2.685 90	2.685 90
10	2.774 87	2.774 87	2.774 87	
12	2.852 87	2.852 87	2.852 87	
14	2.928 28	2.928 28	2.928 28	
16	3.008 01	3.008 00	3.008 00	
18	3.098 00	3.097 91	3.0979	
20	3.201 72	3.201 31	3.201	

[a] J. H. Van Vleck, Phys. Rev. **74**, 1168 (1948).
[b] S. J. K. Jensen and E. K. Hansen, Phys. Rev. B **7**, 2910 (1973).

TABLE II. Moments of the NMR line shape for a linear dipolar chain containing N spins. The range of the interaction is denoted by Δ. The interaction constant A is equal to $\gamma^2 \hbar^2 (3 \cos\theta - 1)/2a^3$ with standard notation. This table corresponds to $\Delta = 2$. The theoretical values for M_2 and M_4 are calculated from Van Vleck,[a] while those for M_6 and M_8 are calculated from Jensen and Hansen.[b] These results are shown in the last column of the table.

| $2n$ | $(M_{2n})^{1/2n}/A$ | | | Other theoretical values |
	$N=9$	$N=11$	$N=\infty$	
2	2.137 83	2.137 83	2.137 83	2.137 83
4	2.466 79	2.466 79	2.466 79	2.466 79
6	2.655 17	2.655 17	2.655 17	2.655 17
8	2.786 48	2.786 48	2.786 48	2.786 48
10	2.892 38	2.892 38	2.892 38	
12	2.990 55	2.990 55	2.990 55	
14	3.094 39	3.094 31	3.094 31	
16	3.214 07	3.213 39	3.2134	
18	3.353 18	3.350 40	3.350	
20	3.507 51	3.500 10	3.50	

[a] See Ref. a of Table I.
[b] See Ref. b of Table I.

a diagrammatic analysis, it can be shown[19] that for $\Delta = 1$, the moments M_2 through M_{N-1} must be equal to those of the infinite chain if N is odd. We note from Table I, however, that for this case the first 14 moments for $N=9$ are the same as those for $N=11$, within the accuracy of our calculations.[25] The rather close agreement of a few of the moments for these two values of N enables us to obtain exact results for the moments $M_2 - M_{10}$ and estimate the moments $M_{12} - M_{20}$ for the *infinite* chain. As the range of the interaction increases, we expect our moments for a given value of N to converge less rapidly towards those of the infinite chain. This is verified by a comparison of the moments for $N=9$ and $N=11$ when $\Delta = 2$ as shown in Table II. We note that only the first 12 moments are identical for these two cases. Some of the higher terms for $N=\infty$ are estimated in a manner similar to that for $\Delta = 1$.

For the linear chain, we have also evaluated $M_2 - M_8$ from the various published analytical expressions for these moments[12,21-24] for each given value of N and Δ. In each of the cases investigated, our results for M_2 and M_4 are found to be in excellent agreement with those obtained from Van Vleck.[12] Similarly, our results for M_6 and M_8 agree very well with those obtained from Jensen and Hansen.[24] This comparison is shown in the last columns in Tables I and II. However, our result for M_6 is not in agreement with that obtained either from Cheng and Memory[21] or from Wurzbach *et al.*[23] This

suggests that the expressions for M_6 as given in these two papers are not correct.

It is well known that clusters containing only $2, 3, \ldots, (k+1)$ particles contribute to M_{2k}. By analyzing the many-body diagrams that enter in the calculation of M_{2k}, Bersohn and Das[26] concluded that the dominant contribution comes from the $k+1$ particle clusters. This is generally found to be true in three-dimensional lattices.[24] Recently, the sum of all the four-particle terms in M_6, denoted by $^4M_6$, was determined by Wurzbach *et al.*[22,23] A comparison of their results for $^4M_6$ with our result for M_6 for the infinite chain, shows that $^4M_6$ constitutes only 29.36%, 33.02%, and 33.45% of M_6 as Δ is allowed to take the values 1, 2, and 3, respectively. This clearly indicates that the Bersohn-Das theorem breaks down in one dimension even when the long-range nature of the dipolar interaction is taken into account. This is the reason why our finite chain results converge to the $N=\infty$ limit so rapidly (see Tables I and II).

Based upon the listed comparisons of our results for $M_2 - M_8$ with the now accepted reliable calculations by previous authors, it seems reasonable that the higher moments listed in Tables I and II can provide a good check as to the accuracy of future moment calculations.

B. Transverse-spin-correlation functions

To gain some insight as to how the beat structure appears in the FID, we studied the time dependence of the transverse-spin-correlation functions $G_r^x(t)$ for different values of r. A detailed study of $G_r^x(t)$ for $N=9$ is shown in Fig. 1, where we allow the range Δ to take up the values 1, 2, and 3, separately. For $G_0^x(t)$ our result is essentially identical to that of the infinite chain up through $Jt \sim 3$, and is non-negative in this time domain.[27] The pronounced secondary maxima in $G_0^x(t)$ [see Fig. 1(a)] is similar to those observed in $G_0^z(t)$ for the Heisenberg model,[13] and is typical of one-dimensional spin dynamics. As shown in Fig. 1(b), $G_1^x(t)$ shows a pronounced negative peak around $Jt \sim 0.5$, and other negative minima for larger values of time. Similarly, $G_2^x(t)$ shows a maxima at $Jt \sim 1$ [see Fig. 1(c)]. Considerable structure is also seen in $G_3^x(t)$ and $G_4^x(t)$ as shown in Figs. 1(d) and 1(e), respectively. A striking pattern that emerges from a study of these figures is that $G_r^x(t)$ at even (odd) sites are predominantly positive (negative). We also note that increasing the range of dipolar interaction does not change the basic structure appreciably but shifts the general pattern towards shorter values of time, and also increases the damping somewhat.

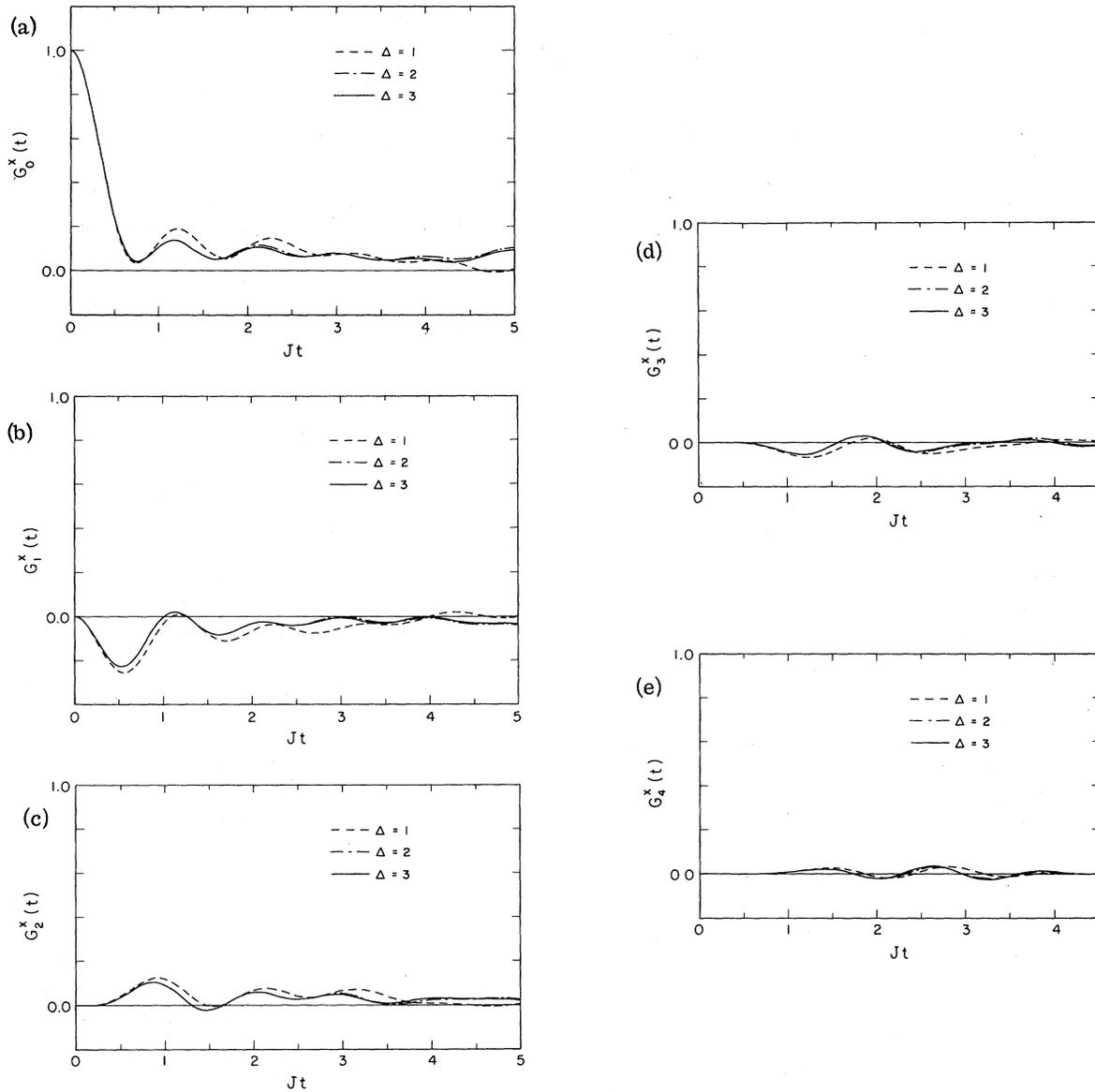The FID function $F(t)$ for this case is shown in Fig. 2. We note that at very short times, the main

FIG. 1. Effect of increasing the range of interaction Δ on the normalized infinite temperature spin-pair-correlation functions $G_r^x(t)$, where r is the spatial separation of the spins, for a linear dipolar chain containing nine spins. We have used periodic boundary conditions on the spins. For details, see text.

contribution to $F(t)$ comes from $G_0^x(t)$ and $G_1^x(t)$. Around the first minimum of $F(t)$, the contribution from $G_1^x(t)$ is most important. As $G_2^x(t)$ shows a positive peak, the combined effect of $G_0^x(t)$, $G_1^x(t)$, and $G_2^x(t)$ results in a positive peak in $F(t)$. For larger values of time the far away cross-correlation functions become more and more important and we obtain a damped beat structure for $F(t)$. We however, would like to point out that our present result for $F(t)$ for $N = 9$ (see Fig. 2) reproduces that of the infinite chain in the short time region only ($Jt \sim 2$). The absence of alternate positive maxima

and negative minima in the long time tail of $F(t)$ is believed to be a manifestation of the small size of our system. The behavior of the infinite chain is further discussed in Sec. III C.

C. Comparison with experiments

The interaction of the fluorine nuclei in fluoro-apatite can be well described by the Hamiltonian given in Eq. (1), as indicated by earlier experiments by Van der Lugt et al.[10] and recent FID measurements by Engelsberg et al.[11] The latter authors calculated $F(t)$ for $N = 5$, and $\Delta = 2$ and
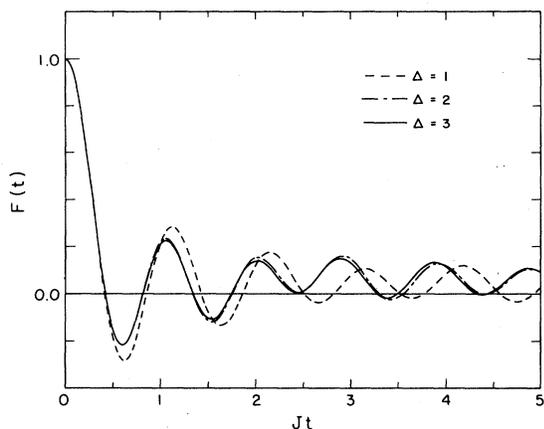
FIG. 2. Effect of increasing the range of interaction Δ on the FID function $F(t)$ for a linear dipolar chain containing nine spins.

approximated the effect of off chain spins by a simple Gaussian broadening. They found good agreement with experiment. Here we evaluate $F(t)$ for larger number of spins and longer range of interaction.

To obtain the behavior of the infinite chain, we study the effect of increasing the length of the chain on $F(t)$. Our results for $F(t)$ for $N = 7$, 9, 11 and $\Delta = 2$, is shown in Fig. 3. If τ_N is the time domain up to which $F(t)$ for a given value of N is expected to reproduce that for the infinite chain, a study of Fig. 3 shows that $\tau_7 \sim 120$ μsec, and $\tau_9 \sim 200$ μsec. We therefore expect that $\tau_{11} \sim 300$ μsec, which includes the first five nodes of $F(t)$. We note that the beat structure is less damped

compared to that of CaF_2. This arises because of the smaller number of neighbors interacting with a given spin in one dimension.

A comparison of our numerical result for $F(t)$, with $N = 11$ and $\Delta = 3$, with experimental measurements of Engelsberg et al.[11] is shown in Fig. 4. We note that within the time domain of interest (< 300 μsec) our result is a very good approximation to that of the infinite chain. As we see from Fig. 4, the nodes and the positions of the peaks of our theoretical result for $F(t)$ are in very good agreement with the experimental result $\phi(t)$, but $\phi(t)$ damps faster than $F(t)$. An examination of the peak amplitudes of $\phi(t)$ and $F(t)$ shows that one needs a rather slowly decaying damping factor $h(t)$ so that $h(t)F(t)$ will agree fairly well with $\phi(t)$. As shown by previous authors,[10,11] about 3% of the total M_2 comes from the small F – F interchain interaction and the small fluorine-phosphorus interaction. For $t < 120$ μsec, $0.03M_2 t^2 < \frac{1}{2}$, and these off chain interactions cannot explain the observed damping completely. The crystals on which measurements were made had $T_1 \sim 0.1$ sec at 32 MHz, indicating the presence of a large amount of paramagnetic impurities. This was further evidenced by later FID measurements on the same crystals. It was found that the FID at 4.2 K was much more highly damped than at room temperatures, probably due to increase in correlation time of the paramagnetic centers. Futher study of the damping function should be carried out by FID measurements on purer crystals.

D. Gaussian approximation

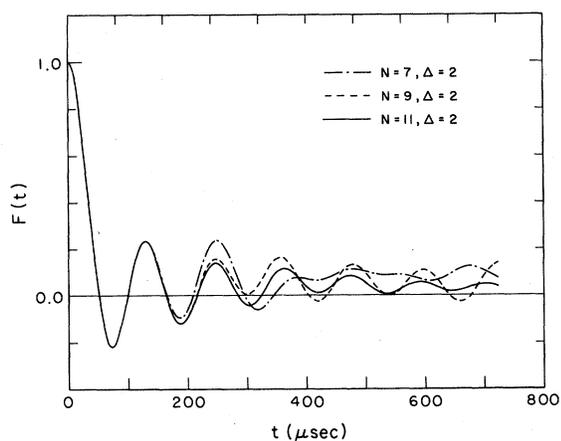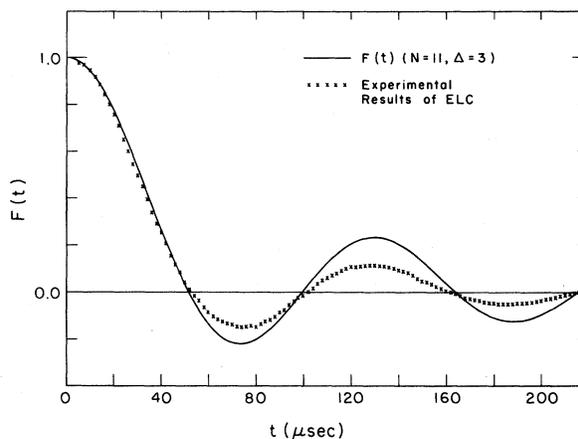Let us write the Hamiltonian describing the spin-spin interaction in the form

FIG. 3. Theoretical FID function $F(t)$ for ^{19}F resonance which arises from the F-F intrachain interaction in fluoroapatite for different values of N, the number of spins on the chain, where the dipolar interaction is allowed to extend up through second-nearest neighbors ($\Delta = 2$).

FIG. 4. Comparison of the present calculation of $F(t)$ for $N = 11$ and $\Delta = 3$ for the ^{19}F resonance in fluoroapatite which arises from the F-F intrachain interaction with experimental measurements of Engelsberg, Lowe, and Carolan (Ref. 11).

$$\mathcal{H} = \mathcal{H}_0 + \mathcal{V} , \tag{7}$$

where $\mathcal{H}_0$ is the Hamiltonian for the unperturbed system, and $\mathcal{V}$ is a small perturbation. Let $F_0(t)$ be the FID corresponding to $\mathcal{H}_0$. In this case the customary procedure in NMR[11,16,17] is to assume that $F(t)$, which corresponds to $\mathcal{H}$, can be well approximated by $F_1(t)$ given by

$$F_1(t) = h(t) F_0(t) , \tag{8}$$

where $h(t)$ is a smooth decaying function of time. Very often $h(t)$ is chosen to be a Gaussian, namely,

$$h(t) = e^{-\sigma^2 t^2/2} , \tag{9}$$

where σ is chosen so as to preserve the second moment of $F(t)$. Although there is little physical justification for this procedure, it is convenient, and in some cases, it does explain observed line shapes fairly well.[16,17] Since we do not make any *ad hoc* approximation in the evaluation of the FID function, we are able to test the validity of this procedure.

We first note, from Fig. 2, that increasing Δ from 1 to 2, causes a pronounced phase shift of $F(t)$ towards smaller values of time, rather than increase the damping, except when $Jt < 1$. This clearly indicates that *no* smooth decaying functional form for $h(t)$, when multiplied to $F_0(t)$ corresponding to $\Delta = 1$, will be able to reproduce $F(t)$ corresponding to $\Delta = 2$. To investigate this point further, we consider the 11 spin chain, and Δ is allowed to take the values 2 and 3, separately. For this case we have

$$\mathcal{H} = \mathcal{H}_d(11,3), \quad \mathcal{H}_0 = \mathcal{H}_d(11,2) , \tag{10}$$

where $\mathcal{H}_d(N, \Delta)$ is the dipolar Hamiltonian as given in Eq. (1) for given values of N and Δ. The corresponding FID functions are denoted by $F(t; N, \Delta)$, for $N = 11$ and $\Delta = 3$, and $\Delta = 2$. If we now assume that the third-nearest-neighbor interaction produces identical Gaussian broadening for each of the multiplets arising from $\mathcal{H}_d(11,2)$, then $F_1(t)$ is given by

$$F_1(t) = e^{-\sigma^2 t^2/2} F(t; 11, 2) , \tag{11}$$

where σ is chosen so that $F_1(t)$ and $F(t; 11, 3)$ have the same second moment. A comparison of $F_1(t)$ with the exact $F(t; 11, 3)$ is shown in Fig. 5. We find that except for a very small improvement at short times, $F_1(t)$ is not a good approximation for $F(t; 11, 3)$. We, therefore, conclude that, at least in one dimension, the effect of weak interactions is
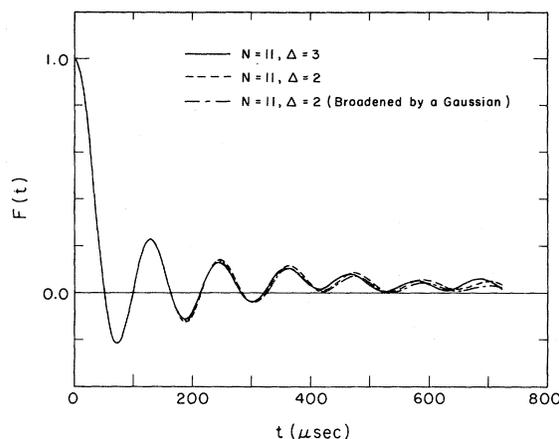

FIG. 5. Test of Gaussian broadening approximation in NMR. See text for details.

more complicated than a simple Gaussian broadening of the unperturbed spectrum.

IV. SUMMARY

We have calculated that first 20 moments of the NMR line shape for the one-dimensional problem, and found good agreement with previous reliable calculations for the first eight moments. The higher moments are expected to provide useful test on the accuracy of future moment calculations. For the first time, the transverse two-time spin-correlation functions have been studied in detail for the truncated dipolar interaction. We find, for example, that the first minima in the FID is dominated by the nearest-neighbor cross-correlation functions. By considering chains of increasing length, we show that the FID function $F(t)$ for $N = 11$ is identical to that for the infinite chain up through at least the fifth node. Our result for $F(t)$ is found to be in good agreement with recent ^{19}F FID measurement in fluoroapatite. As we allow the range of the dipolar interaction to increase, the general features of $F(t)$ do not alter appreciably, but the nodes are found to shift towards shorter values of time. Consequently, the standard Gaussian broadening approximation for the weak interactions in one-dimensional systems is inadequate.

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