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Free-Induction-Decay Shapes in a Dipolar-Coupled Rigid Lattice of Infinite Nuclear Spins

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A molecular-dynamics experiment is used to derive free-induction-decay curves for a rigid cubic lattice with 216 classical spins coupled by magnetic dipole interaction. Free-induction-decay curves are obtained for the magnetic field along the [100], [110], and [111] axes of the lattice. The shapes of these curves are compared to the shapes derived from the expansion theory of Gade and Lowe and to the shapes based on Abragam's trial function. A considerable difference is found between the predictions of the Gade and Lowe theory and our experiments, whereas Abragam's trial functions reproduce the experiments quite well except for long times. Continuous-wave spectra obtained by Fourier transformation of the free-induction-decay curves are also reported as well as their first few moments. The moments are in good agreement with the second and fourth moments given by Van Vleck.

I. INTRODUCTION

The free-induction decay (FID) of the transverse magnetization in a dipolar-coupled rigid lattice is a fundamental problem in magnetic resonance and in the theory of many-body interactions. Until now no theory has been presented which covers the diversity of FID experiments. Lowe and Norberg[1] (LN) first calculated the FID for a system of identical particles with spin $I=\frac{1}{2}$, obtaining a good agreement with the FID experiments on CaF_2 and with the Fourier transform of Bruce's[2] continuous-wave (cw) spectra. The theory is based on an expansion procedure which has been challenged[3,4] since its convergence properties are not known and since it is not unique.

In 1966 Gade and Lowe[5] (GL) generalized the LN theory to arbitrary spin and calculated FID shapes and cw spectra for a simple cubic (sc) lattice, a face-centered-cubic (fcc) lattice, and a body-centered-cubic (bcc) lattice with spin $I=\frac{1}{2}$, 1, $\frac{3}{2}$, and ∞. Their set of equations (13)–(16) (hereafter referred to as GLI) constitute one of the most general expressions for the FID shapes of a single-ingredient-spin system published until now. Since the expression GLI is rather complicated, an approximation is always used in numerical calculations. We shall refer to this approximation [Eq. (17) in Ref. 5] as GLII. For spin $\frac{1}{2}$ GLII is identical to the LN expansion.

Another theoretical approach is that of Ewans and Powles[6] and of Lee, Tse, Goldburg, and Lowe.[7] Both groups consider the exchange term in the truncated dipolar Hamiltonian as a perturbation to the Ising term and use perturbation theory to obtain the FID. This method has been developed further by several investigators.[8-12]

A third method uses expansion theorems to express the FID curve in terms of known functions adjusted to give correct values of the first few moments of the cw spectra. Abragam's[3] trial function is an example of this approach. It gives an agreement with the CaF_2 experiments[3] which is comparable to that of the LN theory.

The error-bound theory, developed by Gordon[13] in 1968, constitutes the most general procedure in this direction. With this theory the error bounds for the line shape may be calculated from the known moments. The usefulness of these approaches is restricted by the difficulties in calculating the moments of order higher than four.[14,15]

Most of the more recent experiments[7,11,16,17] performed on single crystals (LiF, NaF, SrF_2, CsF, NaCl) indicate that the expansion theory of GL is not in all cases adequate, and it is therefore of considerable interest to test the theory by comparison with experiments on additional spin systems, in particular on systems with $I > \frac{1}{2}$ free of any disturbing quadrupole effects.

Molecular-dynamics experiments are currently applied successfully in investigations of interacting systems.[18-20] We have used this technique to generate experimental FID shapes for a simple cubic lattice consisting of 216 classical spins ($I = \infty$) coupled by a truncated dipolar interaction. The obtained FID shapes are compared to the results of the expansion theories which may be extrapolated to the classical limit of $I = \infty$ by letting $I \to \infty$, $\hbar \to 0$ while $I\hbar$ remains finite. Our FID shapes can therefore test the limiting behavior of the theories which is not possible by any real experiment. On the other hand, molecular-dynamics experiments are difficult to perform on systems consisting of N particles with finite spin[21] since a quantum state must be specified by $(2I+1)^N$ complex numbers. In the classical limit ($I \to \infty$), however, this number reduces to $2N$ (i.e., two polar angles for each spin).

Our spin system is described by $N(=216)$ unit vectors representing the orientation of the magnetic dipoles. We describe the spins in a coordinate system rotating about the static field with the Larmor frequency. Each spin sees in this system the dipolar field from its 26 nearest neighbors. The time evolution of the orientation of an individual spin is found by solving the classical equations of motion numerically. The FID is evaluated as the time correlation function of the transverse components of the magnetization.[1] The theories in question may be applied to systems with interaction between any number of neighbors. Accordingly, our restriction to the 26 nearest neighbors is immaterial in comparing the theories with our molecular-dynamics experiments.

II. NUMERICAL METHOD

We consider a system of $N(=216)$ nuclear magnetic dipoles $\vec{\mu}_j = (\mu_{jx}, \mu_{jy}, \mu_{jz})$ forming a simple cubic lattice. The dipoles are placed in a static magnetic field $\vec{B}_0$ and they interact by a dipolar coupling. We assume that $|\vec{B}_0|$ is so large that only the terms of the truncated dipolar coupling[14] influence the FID shape significantly. The truncated dipolar coupling V may be written as follows:

$$V = \frac{1}{4} \sum_{j}^{N} \sum_{k(\neq j)}^{N} \frac{(3\mu_{jz}\mu_{kz} - \vec{\mu}_j \cdot \vec{\mu}_k)(1 - 3\cos^2\theta_{jk})}{r_{jk}^3}, \tag{1}$$

where θ_{jk} is the angle between $\vec{B}_0$ and the vector $\vec{r}_{jk}$, connecting the dipoles j and k. We may also write Eq. (1) as

$$V = \frac{1}{2} \sum_{j}^{N} \sum_{k(\neq j)}^{N} \vec{\mu}_j \cdot \vec{H}_{jk}, \tag{2}$$

where $\vec{H}_{jk}$ is the local field at the position of dipole $\vec{\mu}_j$ produced by the kth dipole.

$\vec{H}_{jk}$ is given by

$$\vec{H}_{jk} = -\frac{\frac{1}{2}(\mu_{kx}, \mu_{ky}, -2\mu_{kz})(1 - 3\cos^2\theta_{jk})}{r_{jk}^3}. \tag{3}$$

We describe the time dependence of the spin system using a coordinate system rotating with the Larmor frequency about $\vec{B}_0$. The time evolution of the orientation of a single spin with $I = \infty$ in this coordinate system is governed by the equation of motion

$$\frac{d\vec{\mu}_j(t)}{dt} = \gamma\vec{\mu}_j(t) \times \sum_{k(\neq j)}^{N} \vec{H}_{jk}(t), \tag{4}$$

where γ is the magnetogyric ratio of the nucleus. Equation (4) may be rewritten as

$$\frac{d\vec{e}_j(t)}{dt} = \vec{e}_j(t) \times \sum_{k(\neq j)}^{N} \vec{H}'_{jk}(t), \tag{5}$$

where t now is in units of

$$X = 2r_0^3/3\gamma|\vec{\mu}|, \tag{6}$$

and $\vec{H}'_{jk}$ is given by

$$\vec{H}'_{jk}(t) = \frac{-\frac{1}{3}(e_{kx}, e_{ky}, -2e_{kz})(1 - 3\cos^2\theta_{jk})}{a_{jk}^3}. \tag{7}$$

In Eqs. (5) and (7) $\vec{e}_j$ is a unit vector along $\vec{\mu}_j$ and $r_{jk} = r_0 a_{jk}$, where r_0 is the lattice parameter of the cubic unit cell. The equations of motion, Eq. (5), are solved for a number K of initial states. Each initial state consists of N randomly chosen unit vectors $\vec{e}_j(0)$ $(j = 1, \ldots, N)$ and corresponds to a spin system with a certain dipole energy and a certain magnetization. For large K we obtain in this way a uniform distribution of energies and magnetizations corresponding to the high-temperature approximation. We follow the time evolution of these systems by integrating Eq. (5) numerically using the method of Runge–Kutta.[22] The summation over k in Eq. (5) is restricted to the 26 nearest dipoles, and periodic boundary conditions are applied to ensure that all dipoles have the same number of neighbors. The integration leads to $\vec{e}_j(n\Delta t)$, where Δt is the time increment, which in our computations is equal to 0.025. For each value of n we determine the dipole energy

$$V(n\Delta t)=\frac{1}{2}\sum_{j}^{N}\vec{\mu}_j(n\Delta t)\cdot\sum_{k(\neq j)}\vec{H}_{jk}(n\Delta t) \tag{8}$$

and the components of the magnetization

$$M_q(n\Delta t)=\sum_{j}^{N}\mu_{jq}(n\Delta t), \quad q=x, y, z\ . \tag{9}$$

It follows from the form of the interaction, Eq. (1), that both V and M_z are constants of motion. In most computations both were constant in the first eight digits.

The FID is given by[1]

$$F(t)=\langle M_x M_x(t)\rangle/\langle M_x^2\rangle\ , \tag{10}$$

where the brackets $\langle\ \rangle$ indicate an ensemble average. We approximate the averaging process by

$$\langle M_x M_x(t_n)\rangle\simeq\sum_{m}^{K}\sum_{h}^{W} M_x^m(t_h)M_x^m(t_h+t_n)\ , \tag{11}$$

where $M_x^m(t_h)$ is the value of M_x at time t_h for a system, which has evolved in time from the mth spin configuration and W is the number of time steps used in the averaging process. We have also determined $\langle M_y M_y(t_n)\rangle$ by the same procedure. $\langle M_x M_x(t_n)\rangle$ and $\langle M_y M_y(t_n)\rangle$ [Eq. (11)] are identical if we average over an infinite number of initial states (i.e., $K\rightarrow\infty$), and we have therefore evaluated $F(t)$ as

$$F(t_n)=(\langle M_x M_x(t_n)\rangle+\langle M_y M_y(t_n)\rangle)/(\langle M_x^2\rangle+\langle M_y^2\rangle)\ . \tag{12}$$

The computations were performed for $\vec{B}_0$ parallel to the three directions [100], [110], and [111]. The product KW was in all three cases 69×10^4. However, K and W varied from one direction to the other. K and W were 115 and 6000 for the directions [100] and [111] and they were 230 and 3000 for the direction [110]. It was found that $\langle M_x M_x(t_n)\rangle$ and $\langle M_y M_y(t_n)\rangle$ are slightly more similar in the former case than in the latter. The averaging procedure was checked by evaluating

$$\sum_{m}^{K}\sum_{h}^{W}[M_x^m(t_h)]^2/(KWN|\vec{\mu}|^2)\ . \tag{13}$$

In the thermodynamic limit this quantity is equal to $\frac{1}{3}$. In our computer experiments it has the value 0.333 ± 0.002 for all three directions.

The correlation functions reported here are obtained for $N=216$. However, trial runs with $\vec{B}_0$ along [100] for $N=512$ and $N=64$ indicate that the curves are not very sensitive to the size of the spin system. In particular, both the position and the amplitude of the first minimum of $F(t)$ are independent of N within the estimated statistical error.

The computations were performed on the CDC 6400 computer at the Regional EDP-Center, Aarhus University. Some of the preliminary computations were made on the IBM 360/75 at the Technical University of Denmark.

III. RESULTS AND DISCUSSION

Our experimental FID shapes are shown in Fig. 1 together with corresponding shapes calculated from GLI and GLII. Also shown is Abragam's trial function. The curves based on GLI and GLII were evaluated in the limit $I\rightarrow\infty$, $\hbar\rightarrow 0$, and $I\hbar$ finite. In this calculation only the 26 nearest neighbors to a given spin were considered. The difference between GLI and GLII is that the latter neglects all terms containing odd powers of $(1-3\cos^2\theta_{jk})/r_{jk}^3$.

Abragam's trial function is given by

$$F(t)=e^{-a^2t^2/2}\sin(bt)/(bt)\ . \tag{14}$$

The parameters a and b are expressed in terms of the second and fourth moments, M_2 and M_4, by using the relationship

$$F(t)=\sum_{n=0}^{\infty}(-1)^n(M_{2n}/M_0)t^{2n}/(2n)! \tag{15}$$

and equating the first three terms of the Taylor-series expansion of Eq. (14) with the first three terms of Eq. (15). The second and fourth moments were calculated from the exact theoretical expressions given by Van Vleck in Ref. 14 by going to the limit of spin infinite as described above. Here we also restricted the interaction to the 26 nearest neighbors. The exact moments are given in Table I together with the corresponding moments obtained from our experiments by the procedure described below.

Figure 1 shows that our experimental curves are practically identical to the corresponding theoretical curves for $t\leq M_2^{-1/2}$ (note that GLI, GLII, and

TABLE I. Moments M_{2n} of the cw spectra for infinite nuclear spin in a simple cubic lattice. Each spin interacts with its 26 nearest neighbors. M_{2n} is given in units of $[2r_0^3/3\gamma|\vec{\mu}|]^{-2n}$. A: Theoretical values of M_{2n} (Ref. 3); B: M_{2n} obtained from the molecular-dynamics experiments.

	M_2		M_4		M_6	M_8
Direction of $\vec{B}_0$	A	B	A	B	B	B
[100]	4.25	4.29	38.4	38.9	513	9321
[110]	1.54	1.55	5.33	5.48	28.7	209
[111]	0.632	0.627	0.907	0.887	1.92	5.84

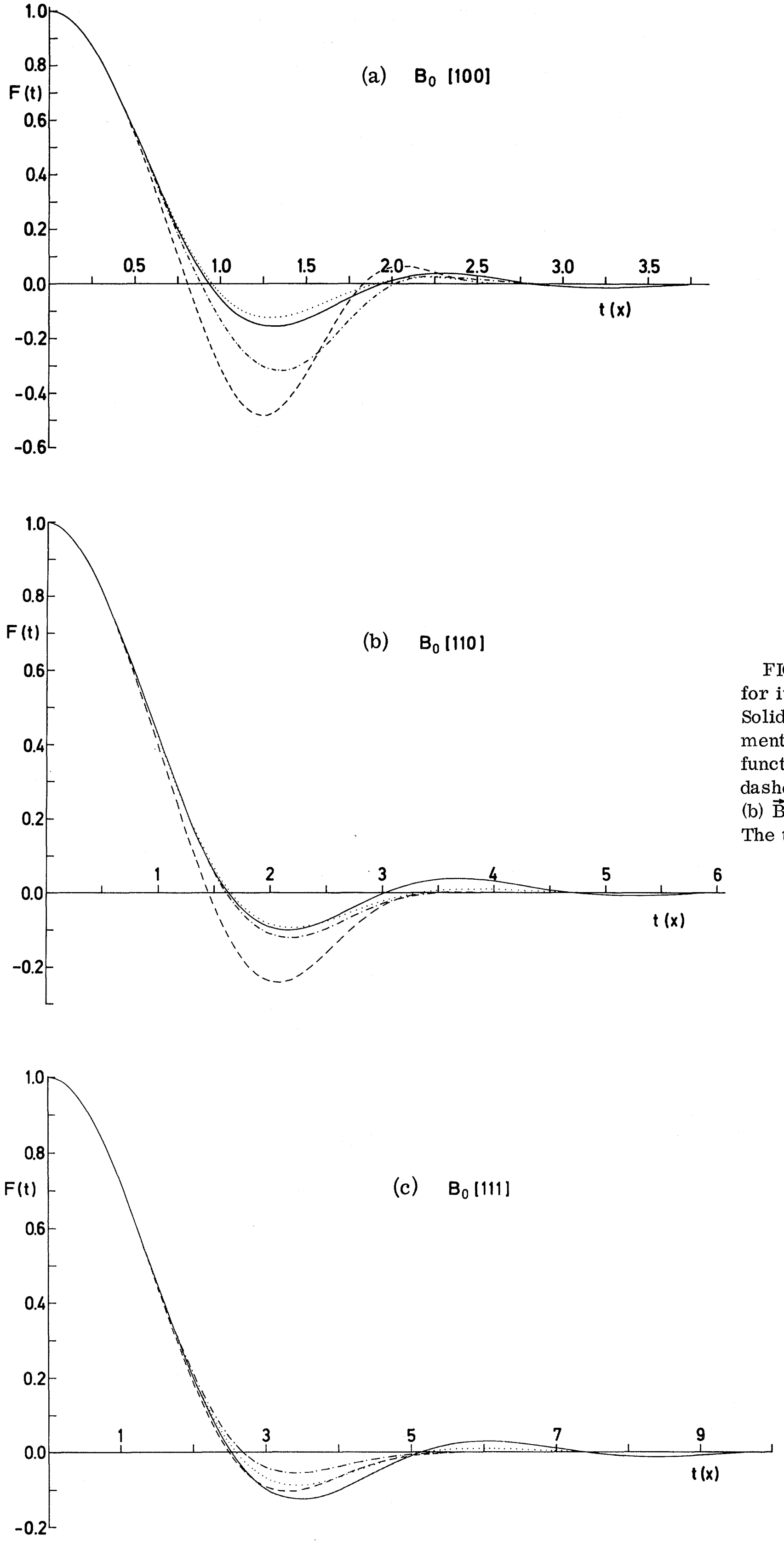

FIG. 1. Free-induction-decay shapes for infinite spin in a simple cubic lattice. Solid line: molecular-dynamics experiment; dotted line: Abragam's trial function; dot-dashed line: GLI; and dashed line: GLII. (a) $\vec{B}_0$ along [100], (b) $\vec{B}_0$ along [110], (c) $\vec{B}_0$ along [111]. The time is in units of $X = 2r_0^3/3\gamma|\vec{\mu}|$.

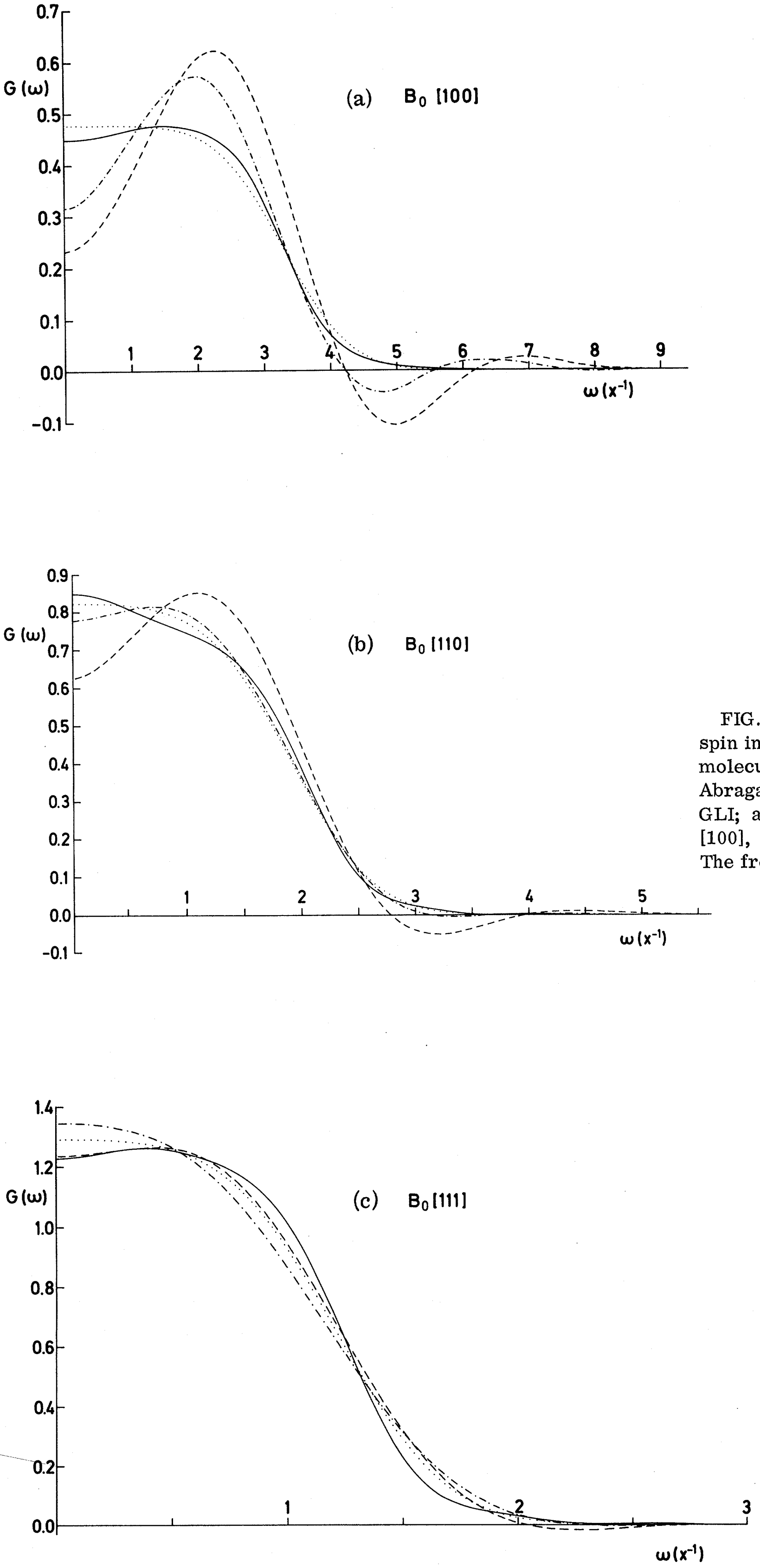

FIG. 2. cw spectra for infinite nuclear spin in a simple cubic lattice. Solid line: molecular-dynamics experiment; dotted line: Abragam's trial function, dot-dashed line; GLI; and dashed line GLII. (a) $\vec{B}_0$ along [100], (b) $\vec{B}_0$ along [110], (c) $\vec{B}_0$ along [111]. The frequency is in units of $X^{-1} = [2r_0^3/3\gamma|\vec{\mu}|]^{-1}$.

Abragam's trial function all have the same M_2). For larger t, however, the curves start to differ. The deviation is pronounced in the vicinity of the first minimum, in particular for $\vec{B}_0$ along the [100] direction [Fig. 1(a)]. Here the minimum value of our FID curve and the one predicted from GLII differ by a factor of 3. In contrast, Abragam's curves give a much better agreement with our experiment in this region. For still larger t values they are closer to our experimental curves than GLI and GLII although they have too small an amplitude.

We have Fourier transformed all the FID shapes to obtain the corresponding cw spectra [23]

$$G(\omega)=\int_0^\infty F(t)\cos(\omega t)\,dt \qquad (16)$$

shown in Fig. 2.

It is seen from this figure that the expansion theory of GL results in spectra having regions of negative absorption, whereas no negative absorption appears in the other spectra. The negative absorption region is most pronounced for GLII.

The moments of our experimental absorption curves given in Table I are calculated as

$$M_{2n}=\int_0^R \omega^{2n}G(\omega)\,d\omega\Big/\int_0^R G(\omega)\,d\omega\,, \qquad (17)$$

where R is a cutoff frequency which has been introduced to suppress a small damped oscillation of $G(\omega)$ around zero. R is chosen as the first zero point of $G(\omega)$. The oscillation arises from our replacement of the upper limit in the Fourier transformation [Eq. (16)] with a finite time equal to the fourth zero point of $F(t)$. The maximum amplitude of the oscillation is 10^{-3}.

Exact theoretical expressions are known for the second and fourth moments only.[3] In fact, Glebashev[15] has calculated the sixth moment for $I=\infty$ with $\vec{B}_0$ along the [100] direction. He obtained the value $M_6=665$. However, this number cannot be compared to our value $M_6=513$ since Glebashev did not specify the number of spin-spin interactions in his calculation. In view of the good agreement between our experimental second and fourth moments and the corresponding theoretical ones it is desirable to obtain a general expression for the moments of order higher than four for comparison with our experimental moments.

Such work is in progress in this laboratory and preliminary results indicate that the values of M_6 given in Table I deviate less than 5% from the exact values, whereas the values of M_8 given in Table I deviate 15% at most from the exact values. From this we conclude that the number of spins considered here suffices to ensure the reliability of the FID shapes obtained.

IV. CONCLUSION

We have simulated the FID shapes of a single-ingredient spin system with $I=\infty$ by a molecular-dynamics experiment. The experiment is set up to investigate the FID line shapes in a system which evolves in time due solely to a truncated dipolar coupling between the spins, i.e., we have no effects from lattice defects and motions, quadrupole coupling, and paramagnetic impurities.

The choice of $I=\infty$, which was made for practical reasons, allows a test of the theories in the limiting case of infinite spin. This is of course not possible by any real experiment. Moreover, the tail of the FID curve is tracked for a longer time in our molecular-dynamics experiments than in most real experiments. This is a considerable advantage, since it must be expected that the inherent approximations in models of the types considered here will be reflected most strongly in the long-time predictions from the models. The present investigation demonstrates that this holds for the theory of Gade and Lowe and for Abragam's trial function. The discrepancy between our FID shapes and those calculated from the expressions of Gade and Lowe and Abragam becomes most significant beyond the first minimum of the curves.

Apart from constituting tests for theoretical FID curves, molecular-dynamics experiments may supplement real FID experiments in a more direct way. FID shapes derived from GLII[5] indicate that the FID is not very dependent upon I, for $I\geq 1$. To the extent this is true; our FID shapes represent the properties of a system with a finite spin $I\geq 1$, where the dipolar interaction is limited to the 26 nearest neighbors. Accordingly, molecular-dynamics experiments may provide insight into the properties of real spin systems if the dipolar interaction between an appropriate number of neighboring spins is taken into account.

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Determination of the Contact Charge Density of 4*s* Electrons in Fe Metal from an Internal-Conversion Experiment

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A clearly resolved spectrum of the outer-shell conversion was obtained for the 14.4-keV nuclear transition in ^{57}Fe with sources consisting of ^{57}Co diffused into Fe metal. From the ratio of the 4*s*-conversion-electron intensity to that of the 3*s*, the contact charge density of 4*s* electrons of Fe metal was uniquely determined to be $\rho_{4s}(0) = 5.53 \pm 0.46\ a_0^{-3}$, which is compared with various theoretical values. Although these are generally smaller than the experimental value, the calculation of Wakoh and Yamashita gives the closest value of $5.18 a_0^{-3}$. The 3*p*-to-3*s* conversion-intensity ratio was also obtained, as 0.0828 ± 0.0038.

The recent study by Pleiter and Kolk[1] on the internal conversion spectrum of the 14.4-keV transition in ^{57}Fe diffused in Fe metal shows the possibility that electron configurations in the outer shells of transition metals may be determined by internal conversion. The energy spectrum measured by them, however, exhibits too much broadening, so that it is rather difficult to analyze the data in order to determine the contribution to the contact charge density at the Fe nuclear site from the *s*-like electrons of each shell. The purpose of the present paper is to show that careful sample preparation can lead to a successful measurement of the electron energy spectrum with such a high resolution that one can obtain clear profiles of the contributions from 4*s*, 3*s*, and even 3*p* electrons. With such a technique, there is no reason why further information on other transition metals and their dilute alloys cannot be investigated. As pointed out by Watson *et al.*,[2] the measurement of internal-conversion electrons probes the contact charge density associated with the Bloch orbitals of band electrons below the Fermi energy, and it should serve as a crucial test for the existing band calculations of the metal, since the conversion experiment can select energetically the valence electrons; and, specifically, for nuclear magnetic dipole (*M*1) transitions effectively only the *s*-like electrons are ejected.[3] The latter selection rule gives to (*M*1) internal conversion a unique superiority in obtaining the contact charge density of *s* electrons in each shell.

Although internal-conversion electrons from the outermost shells of the atom, or from the valence band of the crystal, have been measured for some nuclear transitions and the so-called chemical effects have been detected,[4] the data of Pleiter and Kolk[1] for the 4*s*-electron conversion of the 14.4-keV *M*1 transition in ^{57}Fe diffused into Fe metal are the first to have been subject to comparison[2] with the band calculation of the metal. According to Watson *et al.*,[2] however, the value of the contact charge density of the valence band of Fe metal obtained by Pleiter and Kolk is too large to be explained by the existing band theory of Fe metal. Recently a similar experiment[5] gives for ^{57}Fe in Co metal a smaller value, but direct comparison with theory is impossible, since there is no cal-