ELECTRONIC PROPERTIES
OF SOLIDS

The Coordinate of a Singular Point of the Time Correlation Functions for a Heteronuclear Spin System of a Crystal

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Abstract—The singularities of the time autocorrelation functions (ACFs) for a heteronuclear spin system of a crystal are investigated. Exact expressions are obtained for ten moments of the spectra of ACFs in the approximation of a self-consistent fluctuating field (SCFF) with arbitrary axial symmetry. These expressions are applied to determine the coordinate of the lowest singular point of these functions on the imaginary-time axis for a spin system with a dipole–dipole interaction (DDI). The leading corrections to this coordinate due to the correlation of local fields in real crystals are calculated. These corrections are determined by lattice sums with triangles of four bonds and pairs of four bonds. Numerical values of the coordinate are obtained for a LiF crystal in a magnetic field directed along three crystallographic axes. An increase in the coordinate of the singular point, which follows from the theory and leads to a faster falloff of the wings of the ACF spectra, qualitatively agrees with experiment. © 2005 Pleiades Publishing, Inc.

1. INTRODUCTION

Nuclear magnetic systems with controllable states whose variations can be observed by an NMR method [1] are of great interest for nonequilibrium statistical physics. Heteronuclear systems (i.e., systems that consist of nuclei with different Larmor frequencies in a strong magnetic field) open up new possibilities for studying relaxation processes. This fact was demonstrated in [2] with an example of cross relaxation between subsystems. It is also important that heteronuclear systems are encountered more frequently than homonuclear systems and that there are many methods developed for studying them; the informativeness of these methods depends on the development level of the theory. Finally, systems consisting of nuclei with different resonance frequencies have recently attracted the attention of researchers in relation to the problem of addressing spins in quantum computation [3].

The modern dynamic theory of dense spin systems in solids is based on the concept of a time-fluctuating random local magnetic field [4–9] whose properties are close to those of a Gaussian random field. In heteronuclear systems, this field has several (according to the number of different types of nuclei) components. This fact complicates the construction of a theory. The use of the self-consistency conditions [5] for the time correlation functions of the field and the spins has made it possible to construct a theory [10, 11] that qualitatively

explains many experimental data. This theory implies that the spin correlation functions have singularities on the imaginary-time axis that are responsible for the exponential wings of the spectra of these functions, which are observed by magnetic-resonance methods. An important consequence of this result is that the wings of the spectra of different types of nuclei are universal because the coordinates of the singular points of the time correlation functions, which are coupled due to the interaction, must coincide. A comparison with experiment has shown that the correlation of local fields weakens their fluctuation; in particular, it increases the coordinate of the singular point. In [11], such a correlation was taken into account phenomenologically.

In the present paper, we develop a microscopic approach to the calculation of correlation phenomena in the theory of a self-consistent fluctuating field (SCFF). The validity of this approach has recently been demonstrated by a simpler example of a homonuclear system [12]. First of all, we obtain general expressions for moments up to the tenth order inclusive after generalizing a diagrammatic series for the memory function [6] to the heteronuclear case. Then, based on these moments, we calculate the coordinate of the lowest singular point of the correlation functions on the imaginary-time axis. Finally, we determine a correction to the moments due to the correlation of local fields and apply them to calculate the corresponding shift in the coordinate of the singular point.

2. EQUATIONS
FOR AUTOCORRELATION FUNCTIONS

Equations for the autocorrelation functions (ACFs) of a spin precessing in an anisotropic Gaussian random field were derived in [6]. If we take into account that, in the case of a heteronuclear spin system, such equations should be written out for spins of each type, we arrive at the system of integral equations

$$\frac{d}{dt_1}\Gamma_{\alpha q}(t) = -\int_0^t G_{\alpha q}(t-t_1)\Gamma_{\alpha q}(t_1)dt, \qquad (1)$$

where the subscript α denotes the spin projections x, y, and z and q enumerates the subsystems. The memory functions $G_{\alpha q}(t)$ are represented as series in irreducible dressed skeleton diagrams with increasing number of vertices:

$$G_{\alpha q}(t) = \sum_{m=1}^{\infty} G_{\alpha q}^{2m}(t). \qquad (2)$$

For the case of a field with arbitrary anisotropy in a homonuclear system, all diagrams with 2, 4, 6, and 8 vertices are presented in [6]. In the heteronuclear case, the form of a diagram remains the same; however, in the explicit expressions for these diagrams, one should associate with the zz lines (dashed lines) a sum over contributions to the longitudinal field rather than a single term:

$$g_{zq}(t) = \sum_p \Delta_{qp}^2 \Gamma_{zp}(t). \qquad (3)$$

The xx and yy lines correspond to a single contribution as before, but this contribution is different for different types of nuclei:

$$g_{xq}(t) = \Delta_{xq}^2\Gamma_{xq}(t), \quad g_{yq}(t) = \Delta_{yq}^2\Gamma_{yq}(t).$$

Here, we express the correlation functions of a Gaussian random field in terms of the time-dependent spin ACFs $\Gamma_{xq}(t)$, $\Gamma_{yq}(t)$, and $\Gamma_{zq}(t)$ in a self-consistent way. At high temperatures, the ACF of the α component of the spin located at site i of the lattice is given by

$$\Gamma_{\alpha q}(t) = \frac{Sp\{\exp(i\mathcal{H}t)I_{i\alpha}^{(q)}\exp(-i\mathcal{H}t)I_{i\alpha}^{(q)}\}}{Sp\{(I_{i\alpha}^{(q)})^2\}}, \qquad (4)$$

where $\mathcal{H}$ is the Hamiltonian of the secular part of the dipole–dipole interaction (DDI) [1, 2] in a strong magnetic field. The mean squares of different contributions to the longitudinal field are

$$\Delta_{qp}^2 = I^{(p)}(I^{(p)}+1)\frac{4}{3}\sum_k b_{ik}^{(qp)^2},$$
$$b_{ij}^{(qp)} = \frac{\gamma_q\gamma_p\hbar}{2r_{ij}^3}(1-3\cos^2\theta_{ij}), \qquad (5)$$

where θ_{ij} is the angle between the internuclear vector $\mathbf{r}_{ij}$ and the direction of the static magnetic field. In the axially symmetric case, we have

$$\Delta_{xq}^2 = \Delta_{yq}^2 = \Delta_{qq}^2/4. \qquad (6)$$

Each term of series (2) is expressed in terms of a multiple time integral of the products of ACFs [6].

Table 1. Exact values of the ACF moments in a LiF crystal in the SCFF approximation when the magnetic field is applied along three crystallographic axes ($X_{2n}^{(q)} = M_{2nX}^{(q)}\Delta_{FF}^{-2n}$ and $Z_{2n}^{(q)} = M_{2nZ}^{(q)}\Delta_{FF}^{-2n}$)

| | $H_0 \parallel [111]$ | | $H_0 \parallel [110]$ | | $H_0 \parallel [100]$ | |
	F	Li	F	Li	F	Li
X_2	1.5565	0.243175	3.2059	0.573075	10.9709	2.126295
X_4	7.2621246	0.2062389	30.122726	1.1550313	357.05369	14.396791
X_6	60.240334	0.4462115	476.06509	5.7148923	19329.186	193.62608
X_8	785.97935	2.6795506	10916.055	73.047633	1469814.4	5748.5288
X_{10}	15296.507	36.221363	342734.27	2018.0911	144788238	429421.26
Z_2	0.5	0.07275	0.5	0.07275	0.5	0.07275
Z_4	1.9315	0.0433208	3.5809	0.0913213	11.3459	0.3173148
Z_6	17.917389	0.0669163	67.435923	0.3356018	739.17481	4.1540520
Z_8	277.83143	0.1998922	2104.5260	2.4024558	80039.257	96.558982
Z_{10}	6249.6440	1.1415668	92520.012	31.813793	12125388	3612.1025

ACFs (4) can be expanded in power series,

$$\Gamma_{\alpha q}(t) = \sum_{n=0}^{\infty} \frac{(-1)^n M_{2n\alpha}^{(q)} t^{2n}}{(2n)!}. \tag{7}$$

It is well known [1] that $M_{2n\alpha}^{(q)}$ is a moment on the order of $2n$ of the spectral density of the corresponding ACF. From similar equations for the homonuclear case [6], after appropriate modifications according to (3), we obtained recurrence equations for the moments for the heteronuclear case. Henceforth, we will assume for definiteness that there are only two types of spins in the system. In the Appendix, we present expressions for the moments of the tenth order inclusive for a general axially symmetric case. As an example, we calculated the moments for a LiF crystal (see Table 1); the contributions Δ_{qp}^2 (5) for this crystal were taken from Table 2.

Equations (1) were derived for an interaction with arbitrary magnetic anisotropy. The application of an axially symmetric Hamiltonian of the DDI essentially improves the convergence of the series for the memory function [10–15]. Therefore, it is expedient to transform Eqs. (1) in order to maximally take into consideration the longitudinal component of the local field and minimally take into account the transverse component. In this approximation, we obtain the following system of nonlinear integral equations for the ACFs of a LiF crystal:

$$\Gamma_{zL}(t) = 1 - \frac{1}{2}\Delta_{LL}^2$$

$$\times \int_0^t \int_0^{t'} dt' dt'' \Gamma_{xL}^2(t'-t'') \Gamma_{zL}(t''),$$

$$\Gamma_{xL}(t) = \Gamma_{AL}(t) - \frac{k_L}{4}\Delta_{LL}^2 \int_0^t dt' \Gamma_{AL}(t-t')$$

$$\times \int_0^{t'} dt'' \Gamma_{xL}(t'-t'') \Gamma_{zL}(t'-t'') \Gamma_{xL}(t''),$$

$$\Gamma_{zF}(t) = 1 - \frac{1}{2}\Delta_{FF}^2$$

$$\times \int_0^t \int_0^{t'} dt' dt'' \Gamma_{xF}^2(t'-t'') \Gamma_{zF}(t''),$$

$$\Gamma_{xF}(t) = \Gamma_{AF}(t) - \frac{k_F}{4}\Delta_{FF}^2 \int_0^t dt' \Gamma_{AF}(t-t')$$

$$\times \int_0^{t'} dt'' \Gamma_{xF}(t'-t'') \Gamma_{zF}(t'-t'') \Gamma_{xF}(t''),$$

(8)

Table 2. Mean squares of homo- and heteronuclear contributions to the longitudinal local fields, renormalization parameters, and the coordinates of the singular point of the ACF in LiF for a magnetic field directed along three crystallographic axes

	$H_0 \parallel$ [111]	$H_0 \parallel$ [110]	$H_0 \parallel$ [100]
Δ_{FF}^2, (rad/s)^2	$2838 \cdot 10^6$	$2581 \cdot 10^6$	$1809 \cdot 10^6$
$\Delta_{FL}^2/\Delta_{FF}^2$	0.3065	1.9559	9.7209
$\Delta_{LF}^2/\Delta_{FF}^2$	0.0613	0.3912	1.94442
$\Delta_{LL}^2/\Delta_{FF}^2$	0.1455	0.1455	0.1455
$\tau_0(\lambda_q=1)\Delta_{FF}$	2.372	1.843	1.127
$\tau_0(\lambda_q^2=5/4)\Delta_{FF}$	2.263	1.783	1.113
λ_F	1.101	1.0911	1.0855
λ_L	1.099	1.0896	1.0850
$\tau_0(\lambda_q)\Delta_{FF}$	2.33	1.82	1.12
$\tau_0(\lambda_q)\sqrt{M_{2F}}$	3.72	3.74	3.87
$\delta\tau_c/\tau_0$	0.158	0.215	0.161
$\tau_0(\lambda_q)+\delta\tau_c$, µs	51	44	31
$\tau_c(\lambda_q)\sqrt{M_{2F}}$	4.31	4.53	4.50

where

$$\Gamma_{AL}(t) = \exp\left\{-\lambda_L^2\Delta_{LL}^2 \int_0^t \int_0^{t'} dt' dt'' \Gamma_{zL}(t'')\right.$$

$$\left. -\Delta_{LF}^2 \int_0^t \int_0^{t'} dt' dt'' \Gamma_{zF}(t'')\right\},$$

$$\Gamma_{AF}(t) = \exp\left\{-\Delta_{FL}^2 \int_0^t \int_0^{t'} dt' dt'' \Gamma_{zL}(t'')\right.$$

$$\left. -\lambda_F^2\Delta_{FF}^2 \int_0^t \int_0^{t'} dt' dt'' \Gamma_{zF}(t'')\right\}$$

(9)

is the ACF of a spin rotating in the local field that has only a longitudinal component, similar to the function used in Anderson's model [4]. Unlike the previous work [11], we introduce a renormalization parameter for the longitudinal local field into Eq. (9). The value of
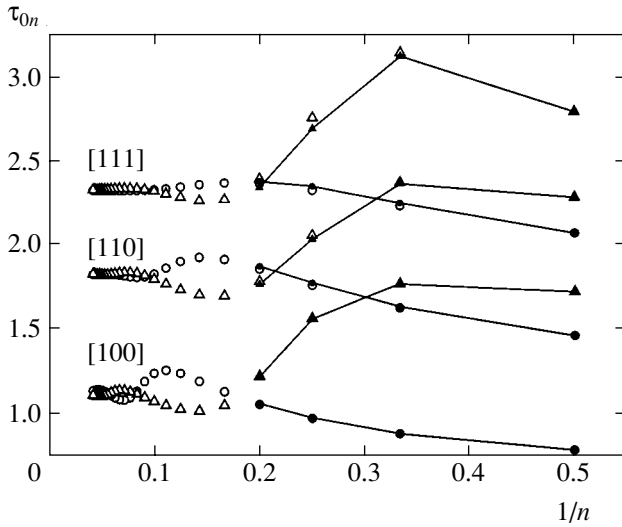
Ratios of moments (12) for the ACF of the x components of the spins of fluorine (circles) and lithium (triangles) nuclei in a LiF crystal when the magnetic field is applied along three crystallographic axes indicated in the figure. The results obtained by the moments of solutions to Eqs. (8) and (9) are indicated by open symbols, and the results obtained by the moments from Table 1 are indicated by closed symbols connected by straight lines.

this parameter is determined from the moments. The correctness of the second moments is guaranteed by the strict relation between the parameters k_q and λ_q that enter Eqs. (8) and (9):

$$k_q = 5 - 4\lambda_q^2.$$

Let us determine the fourth moment of the solution to Eqs. (8) and (9):

$$\frac{M_{4x}^{(q)}}{\Delta_{qq}^4} = 2\lambda_q^2 - \frac{5}{4}\lambda_q + \frac{15}{4}$$

$$+ \frac{\Delta_{qp}^2}{\Delta_{qq}^2}\left(3\lambda_q + \frac{15}{4} + \frac{\Delta_{pp}^2}{2\Delta_{qq}^2}\right) + 3\left(\frac{\Delta_{qp}^2}{\Delta_{qq}^2}\right)^2.$$

From the equality of this moment to its exact value (A1), we determine the parameters

$$\lambda_q = \frac{15}{6} - \frac{3D_q}{4} + \frac{1}{4}\left(9D_q^2 + \frac{47}{2}D_q + \frac{161}{16}\right)^{1/2}, \quad (10)$$

where $D_F = \Delta_{FL}^2/\Delta_{FF}^2$ and $D_L = \Delta_{LF}^2/\Delta_{LL}^2$. The values of the parameters calculated by formula (10) are presented in Table 2.

The solutions of Eqs. (8) and (9) have singular points on the imaginary-time axis. The principal parts

of the ACFs in the neighborhoods of these points are given by

$$\Gamma_{zq}(t) \approx \frac{C_{zq}}{(\tau_0 + it)^{\zeta_q}}, \quad \Gamma_{xq}(t) \approx \frac{C_{xq}}{(\tau_0 + it)^{\chi_q}},$$

$$\Gamma_{Aq}(t) \approx \frac{C_{Aq}}{(\tau_0 + it)^{\delta_q}} \quad (q = \mathrm{F}, \mathrm{Li}). \tag{11}$$

The exponents were determined in [10] by the Painelvé method for three orientations of a LiF crystal. For the F nuclei, all exponents equal 2, whereas, for the Li nuclei, only the exponents for the orientation [100] are equal to 2; for two other orientations, the exponents are as follows: $\delta = \chi = 0.123$ for [111] and $\delta = \chi = 0.784$ for [110] (we do not need the exponent ζ).

For a known value of the exponent of the lowest singular point, its coordinate, equal to the convergence radius with respect to moments (7), can be evaluated as the limit of the sequence of relations

$$\tau_{0n}^2 = \frac{M_{2(n-1)\alpha}^{(q)}\Gamma(2n + \chi)}{M_{2n\alpha}^{(q)}\Gamma(2n - 2 + \chi)}, \tag{12}$$

as $n \longrightarrow \infty$, where $\Gamma(x)$ is the gamma function. These sequences are shown in the figure. The calculations are performed by the exact values of ten moments from Table 1 and by 50 moments of the solutions to approximate Eqs. (8) and (9) for the values of the parameters λ_q given in Table 2. In the homonuclear case, the convergence of the sequence of relations is better [6]; this allowed us to determine τ_0 by the first ten moments to a sufficiently high degree of accuracy. In the heteronuclear case, the convergence deteriorates because the interaction between lithium nuclei is weak ($\Delta_{LL}^2 = 0.15\Delta_{FF}^2$). Therefore, it takes some time for the system of Li nuclei to adjust to the system of F nuclei. The figure shows that the first terms in the sequence of the ratios of moments of the solution to system (8), (9) are close to the ratios of exact moments. The approximate equations have allowed us to follow up how the ratios of moments pass to the limit (see Table 2). This approach represents the development of the simple estimate of [6]. Applying it to the homonuclear case for $\lambda = 1.105$, we arrive at the value $\tau_0\Delta_Z = 2.48$, which was determined earlier by ten moments. Note that an estimate for τ_0 was obtained in [10] by using Eqs. (8) and (9) for $\lambda = 1$ (which is also shown in Table 2). The variation of λ_q from 1 to 1.1 leads to variation of the coordinate τ_0 by less than 2%, which is indicative of the accuracy of its determination.

3. CALCULATION OF A CORRECTION TO THE COORDINATE OF A SINGULAR POINT OF THE CORRELATION FUNCTIONS

The SCFF approximation corresponds to the limit $d \longrightarrow \infty$. As is shown in [12] by an example of homonuclear systems, for $d = 3$, the coordinate of the singular point of the ACF on the imaginary-time axis increases due to the correlation of local fields,

$$\tau_c = \tau_0 + \delta\tau_c, \tag{13}$$

and the main contribution to $\delta\tau_c$ is given by the corrections to the moments that can be represented by trees of double bonds with a built-in pair of fourfold interaction or triangle of four bonds. In the heteronuclear case, the degree of correlation is characterized by the following ratios of lattice sums:

$$S_2/(S_1)^2, \quad S_2'/(S_1)^2, \quad S_3/(S_1)^2, \quad S_3'/(S_1 S_1'), \tag{14}$$

where

$$S_1 = \sum_j b_{ij}^2, \quad S_2 = \sum_j b_{ij}^4, \quad S_3 = \sum_{k,j} b_{kj}^2 b_{ik} b_{ji} \tag{15}$$

with summation over the sites occupied by nuclei of one type with a nucleus at site i. In the primed sums, the summation is performed over the sites occupied by nuclei of another type. The numerical values of ratios (14) for three orientations of a LiF crystal are presented in [11]. The small value of these ratios corresponds to the real smallness parameter. For convenience, we introduce a formal parameter ε; in finite formulas, we set this parameter equal to zero. Contributions with lattice sums (14) are already contained in the fourth moment (A.3). The corresponding decrease in M_{4x} can be ascribed to the SCFF if one reduces λ_q. For example, for the orientation [110], we obtain $\lambda_F = 0.94$ and $\lambda_L = 0.75$. Such a variation leads to an increase in the coordinate τ_0 by about 2%. As is shown in [12, 16] in the homonuclear case, the incorporation of such fragments of (14) into large trees of bonds corresponding to higher order moments produces a more significant effect. Such contributions can be determined from Eqs. (1) for the ACFs.

Assuming that the correction $\varepsilon\delta\tau_c$ is small, we estimate it by taking a simplified version of equations in which the zz interactions are predominant. Take

$$\Gamma_{\alpha q}(t) = \Gamma_{\alpha q0}(t) - \varepsilon\Gamma_{\alpha q1}(t)$$

and substitute it into an integral equation with appropriate correction terms for the memory function

$$G_{\alpha q}(t) = G_{\alpha q0}(t) - \varepsilon G_{\alpha q1}(t).$$

In view of the form of the equations, it is more convenient to pass to an equation for the squared transverse ACF:

$$\Gamma_{xq}^2(t) = Y_q(t) = Y_{q0}(t) - \varepsilon Y_{q1}(t). \tag{16}$$

For the first correction $Y_{F1}(t)$, we find the equation

$$\frac{d}{dt}Y_{F1}(t) = 2\langle FF\rangle Y_{F1}(t)\int_0^t \Gamma_{zF0}(t_1)dt_1$$

$$+ 2\langle FL\rangle Y_{F1}(t)\int_0^t \Gamma_{zF0}(t_1)dt_1$$

$$+ \frac{4}{5}\langle FF\rangle Y_{F0}(t)\int_0^t\int_0^{t_1}\int_0^{t_2} Y_{F1}(t_3)dt_1dt_2dt_3$$

$$+ \frac{4}{5}\langle FL\rangle Y_{F0}(t)\int_0^t\int_0^{t_1}\int_0^{t_2} Y_{L1}(t_3)dt_1dt_2dt_3$$

$$+ 2\langle FL\rangle^2 S_2' Y_{F0}(t)\int_0^t \Gamma_{zL0}(t_1)dt_1\int_0^t\int_0^{t_1}\Gamma_{zL0}(t_2)dt_1dt_2$$

$$+ 2S_2\langle FF\rangle^2 Y_{F0}(t)\int_0^t \Gamma_{zF0}(t_1)dt_1\int_0^t\int_0^{t_1}\Gamma_{zF0}(t_2)dt_1dt_2$$

$$+ \frac{4}{5}\langle FF\rangle^2(S_2 + S_3)Y_{F0}(t)\int_0^t\int_0^{t_1}\int_0^{t_2} Y_{F0}(t_3)dt_1dt_2dt_3$$

$$+ \frac{4}{5}\langle LF\rangle\langle LL\rangle S_3' Y_{F0}(t)\int_0^t\int_0^{t_1}\int_0^{t_2} Y_{L0}(t_3)dt_1dt_2dt_3 \tag{17}$$

$$+ \langle FF\rangle^3\frac{8}{5}\left(\frac{3}{2}S_2 + S_3\right)Y_{F0}(t)$$

$$\times \int_0^t\int_0^{t_1}\int_0^{t_2} Y_{F0}(t_3)dt_1dt_2dt_3\int_0^{t_3}\int_0^{t_4}\Gamma_{zF0}(t_5)dt_4dt_5$$

$$+ \langle LL\rangle^2\langle FL\rangle\frac{8}{5}(S_2 + S_3)Y_{F0}(t)$$

$$\times \int_0^t\int_0^{t_1}\int_0^{t_2} Y_{L0}(t_3)dt_1dt_2dt_3\int_0^{t_3}\int_0^{t_4}\Gamma_{zL0}(t_5)dt_4dt_5$$

$$+ \langle LL\rangle\langle FL\rangle\langle LF\rangle\frac{4}{5}(S_2' + 2S_3')Y_{F0}(t)$$

$$\times \int_0^t\int_0^{t_1}\int_0^{t_2} Y_{L0}(t_3)dt_1dt_2dt_3\int_0^{t_3}\int_0^{t_4}\Gamma_{zF0}(t_5)dt_4dt_5$$

$$+ \langle FF\rangle^2\langle FL\rangle\frac{8}{5}S_3' Y_{F0}(t)$$

$$\times \int\limits_{0}^{t}\int\limits_{0}^{t_1}\int\limits_{0}^{t_2} Y_{F0}(t_3)dt_1 dt_2 dt_3 \int\limits_{0}^{t_3}\int\limits_{0}^{t_4} \Gamma_{zL0}(t_5)dt_4 dt_5 - R_F(t),$$

in which we introduced dimensionless imaginary time $t' = -it(5\Delta_{FF}^2/4)^{1/2}$; omitted the prime; denoted

$$\langle FF \rangle = 1, \quad \langle FL \rangle = \frac{4\Delta_{FL}^2}{5\Delta_{FF}^2},$$

$$\langle LL \rangle = \frac{\Delta_{LL}^2}{\Delta_{FF}^2}, \quad \langle LF \rangle = \frac{4\Delta_{LF}^2}{5\Delta_{FF}^2},$$

and denoted ratios (14) by S_2, S_2', S_3, S_3'. Equation (17) differs from that considered in the homonuclear case [12] by the contributions of nuclei of different types and by the change in the form of the correction $R_q(t)$ (because lithium nuclei have spin 3/2) due to the permissible fourfold interaction of nearest neighbors:

$$R_F(t) = 2S_2\langle FF\rangle^2 Y_{F0}(t)\int\limits_{0}^{t}\Gamma_{zF0}(t-t_1)dt_1$$

$$\times \int\limits_{0}^{t_1}\int\limits_{0}^{t_2}\Gamma_{zF0}(t_3)dt_2 dt_3$$

$$+ 2S_2'\langle FL\rangle^2 Y_{F0}(t)\int\limits_{0}^{t}\Gamma_{zL0}(t-t_1)dt_1$$

$$\times \int\limits_{0}^{t_1}\int\limits_{0}^{t_2}\left[1 + \frac{16}{15}\Gamma_{zL0}(t_1-t_2)\right]\Gamma_{zL0}(t_3)dt_2 dt_3,$$

$$R_L(t) = 2S_2'\langle LF\rangle^2 Y_{L0}(t)\int\limits_{0}^{t}\Gamma_{zF0}(t-t_1)dt_1$$

$$\times \int\limits_{0}^{t_1}\int\limits_{0}^{t_2}\Gamma_{zF0}(t_3)dt_2 dt_3$$

$$+ 2S_2\langle LL\rangle^2 Y_{L0}(t)\int\limits_{0}^{t}\Gamma_{zL0}(t-t_1)dt_1$$

$$\times \int\limits_{0}^{t_1}\int\limits_{0}^{t_2}\left[1 + \frac{16}{15}\Gamma_{zL0}(t_1-t_2)\right]\Gamma_{zL0}(t_3)dt_2 dt_3.$$

A change in the remaining part of Eq. (17) when passing to $Y_{L1}(t)$ reduces to a change in the subscripts $F \longrightarrow L$ and $L \longrightarrow F$. For the functions $Y_{q0}(t)$ and $\Gamma_{zq0}(t)$ of zeroth-order approximation, we use a simple set of four

equations that is obtained from (8) for $\lambda_q^2 = 5/4$. Moreover, we neglect the time variation ($\Gamma_{zq}(t'') \sim \Gamma_{zq}(0) = 1$) compared with ($\Gamma_{xq}(t' - t''))^2$ in the integrand; as is shown in [12], this leads to a slight decrease in the moments but significantly simplifies the calculations. For comparison, the coordinate of the singular point $\tau_0(\lambda_q^2 = 5/4)\Delta_{FF}$ of the solution to such a system is shown in Table 2. We had to simplify the equation in order to increase the numerical positions up to 50. In turn, such a large mantissa is required to sum up the moments that strongly differ in magnitude.

Using the equations, we calculate the moments of functions (16) up to $n = 70$. The coordinate of the singular point is determined from the ratio of moments:

$$\tau_c^2 = \tau_0^2 \lim_{n\to\infty} \frac{1 - \varepsilon Y_{F2(n-1)}^{(1)}/Y_{F2(n-1)}^{(0)} + \cdots}{1 - \varepsilon Y_{F2n}^{(1)}/Y_{F2n}^{(0)} + \cdots}.$$

Then,

$$2\frac{\delta\tau_c}{\tau_0} = \lim_{n\to\infty}\left(\frac{Y_{F2n}^{(1)}}{Y_{F2n}^{(0)}} - \frac{Y_{F2(n-1)}^{(1)}}{Y_{F2(n-1)}^{(0)}}\right). \tag{18}$$

Extrapolating the ratios obtained, we determine the values presented in Table 2.

Let us compare the results for hetero- and homonuclear systems. In the homonuclear case, we have $\tau_0\Delta_Z = 2.48$ for any orientation, whereas, in the heteronuclear case, Table 2 shows that $\tau_0\Delta_{FF}$ decreases by a factor of 2 under rotation from [111] to [100]. Using the units of the total moment of the NMR spectrum of fluorine nuclei,

$$M_{2F} = \Delta_{FF}^2(1 + X_2^{(F)}),$$

we obtain $\tau_0\sqrt{M_{2F}} = 3.72$ for the [111] orientation. This result coincides with that in the homonuclear case, where the coordinate increases to 3.87 under the rotation to the [100] orientation. Such an increase is associated with an increase in the contribution of the heteronuclear zz interaction to M_{2F}. In the homonuclear case, a similar increase in the coordinate of the singular point of the ACF with the ratio Δ_Z^2/Δ_X^2 was observed in [17].

Let us pass to the correction $\delta\tau_c$. We calculated $\delta\tau_c/\tau_0$ for a homonuclear face-centered cubic lattice by the formula obtained in [12] for three orientations and obtained the following results: 0.17 for [111], 0.37 for [110], and 0.22 for [100]. A comparison of these results with the values presented in Table 2 shows that, in both cases, the correction attains its maximal value for the [110] orientation and is primarily associated with the large value of the parameter $S_2/(S_1)^2 = 0.225$. The addi-

tion of a heteronuclear interaction reduces the correction and smoothes out its dependence on orientation.

Thus, we have obtained, theoretically, the required increase in the coordinate of the singular point of an ACF due to the correlation of local fields, which was revealed in [11] by analyzing experimental spectra from the viewpoint of the SCFF theory. A quantitative comparison of the theory and experiment requires that one should determine a variation in the preexponential factor due to the correlation in the motion of spins. An appreciable effect of this factor was also shown in [11], which testifies to the fact that the observed detuning from the center of the spectrum is less than the mathematical asymptotics. Finally, publications do not contain all experimental conditions that are necessary for a successful comparison of the results.

In conclusion, note that we have performed calculations for a LiF crystal when a magnetic field is directed along three crystallographic axes. The theory allows us to obtain results for other heteronuclear systems and orientations. To this end, one should substitute the lattice sums and the contributions to the squared local fields into the formulas and perform the calculations described in this paper.

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APPENDIX

Here, we present expressions for the moments up to the tenth order for a system consisting of two types of spins for an axially symmetric case $\Delta_{xq}^2 = \Delta_{yq}^2$ with an arbitrary relation between Δ_{xq}^2 and Δ_{qq}^2 (below, for definiteness, we will use subscripts "F" and "L" in place of q and p). The moments are calculated by the recurrence relations obtained from the equations of the homonuclear case [6] after the above-mentioned modifications. The results are exact in the SCFF approximation, which corresponds to infinite-dimensional lattices:

$$M_{2X}^{(F)} = \Delta_{FF}^2 + \Delta_{FL}^2 + \Delta_{XF}^2,$$

$$M_{4X}^{(F)} = 3\Delta_{FL}^4 + 3\Delta_{FF}^4 + (4\Delta_{XF}^2 + 6\Delta_{FF}^2 + 2\Delta_{XL}^2)\Delta_{FL}^2 + 5\Delta_{XF}^4 + 6\Delta_{FF}^2\Delta_{XF}^2,$$

$$M_{6X}^{(F)} = 15\Delta_{FL}^6 + 15\Delta_{FF}^6 + (21\Delta_{XF}^2 + 45\Delta_{FF}^2 + 30\Delta_{XL}^2)\Delta_{FL}^4 + 51\Delta_{XF}^6 + 73\Delta_{FF}^2\Delta_{XF}^4 + 55\Delta_{FF}^4\Delta_{XF}^2$$
$$+ (45\Delta_{XF}^4 + 45\Delta_{FF}^4 + 10\Delta_{XL}^4 + 76\Delta_{FF}^2\Delta_{XF}^2 + 30\Delta_{FF}^2\Delta_{XL}^2 + 4\Delta_{LF}^2\Delta_{XL}^2 + 4\Delta_{LL}^2\Delta_{XL}^2 + 18\Delta_{XF}^2\Delta_{XL}^2)\Delta_{FL}^2,$$

$$M_{8X}^{(F)} = 105\Delta_{FL}^8 + (144\Delta_{XF}^2 + 420\Delta_{FF}^2 + 420\Delta_{XL}^2)\Delta_{FL}^6$$
$$+ (112\Delta_{XL}^2\Delta_{LL}^2 + 448\Delta_{XF}^4 + 112\Delta_{XL}^2\Delta_{LF}^2 + 630\Delta_{FF}^4$$
$$+ 840\Delta_{FF}^2\Delta_{XL}^2 + 360\Delta_{XF}^2\Delta_{XL}^2 + 420\Delta_{XL}^4$$
$$+ 988\Delta_{FF}^2\Delta_{XF}^2)\Delta_{FL}^4 + \{1678\Delta_{FF}^2\Delta_{XF}^4 + 92\Delta_{XL}^6$$
$$+ 1544\Delta_{FF}^4\Delta_{XF}^2 + 420\Delta_{XL}^6 + 24\Delta_{XL}^2\Delta_{LL}^4$$
$$+ (72\Delta_{XF}^2\Delta_{XL}^2 + 78\Delta_{XL}^4 + 48\Delta_{XL}^2\Delta_{LF}^2 + 112\Delta_{FF}^2\Delta_{XL}^2)\Delta_{LL}^2$$
$$+ 112\Delta_{FF}^2\Delta_{XL}^2\Delta_{LF}^2 + 24\Delta_{XL}^2\Delta_{LF}^4 + 180\Delta_{XF}^2\Delta_{XL}^4$$
$$+ 284\Delta_{XF}^4\Delta_{XL}^2 + 648\Delta_{FF}^2\Delta_{XF}^2\Delta_{XL}^2 + 70\Delta_{XL}^4\Delta_{LF}^2$$
$$+ 80\Delta_{XF}^2\Delta_{XL}^2\Delta_{LF}^2 + 914\Delta_{XF}^6 + 280\Delta_{FF}^2\Delta_{XL}^4$$
$$+ 420\Delta_{FF}^4\Delta_{XL}^2\}\Delta_{FL}^2 + 1470\Delta_{FF}^2\Delta_{XF}^6 + 861\Delta_{XF}^8$$
$$+ 105\Delta_{FF}^8 + 1378\Delta_{FF}^4\Delta_{XF}^4 + 700\Delta_{FF}^6\Delta_{XF}^2,$$

$$M_{10X}^{(F)} = (1245\Delta_{XF}^2 + 4725\Delta_{FF}^2 + 6300\Delta_{XL}^2)\Delta_{FL}^8$$
$$+ (2520\Delta_{XL}^2\Delta_{LL}^2 + 5856\Delta_{XF}^4 + 2520\Delta_{XL}^2\Delta_{LF}^2$$
$$+ 9450\Delta_{FF}^4 + 18900\Delta_{FF}^2\Delta_{XL}^2 + 6060\Delta_{XF}^2\Delta_{XL}^2$$
$$+ 12600\Delta_{XL}^4 + 15120\Delta_{FF}^2\Delta_{XF}^2)\Delta_{FL}^6 + \{36858\Delta_{FF}^2\Delta_{XF}^4$$
$$+ 4830\Delta_{XL}^4\Delta_{LF}^2 + 8340\Delta_{XL}^6 + 37890\Delta_{FF}^4\Delta_{XF}^2$$
$$+ 9450\Delta_{FF}^6 + 1080\Delta_{XL}^2\Delta_{LL}^4 + (2280\Delta_{XF}^2\Delta_{XL}^2$$
$$+ 5190\Delta_{XL}^4 + 2160\Delta_{XL}^2\Delta_{LF}^2 + 5040\Delta_{FF}^2\Delta_{XL}^2)\Delta_{LL}^2$$
$$+ 5040\Delta_{FF}^2\Delta_{XL}^2\Delta_{LF}^2 + 16676\Delta_{XF}^6 + 1080\Delta_{XL}^2\Delta_{LF}^4$$
$$+ 8280\Delta_{XF}^2\Delta_{XL}^4 + 9140\Delta_{XF}^4\Delta_{XL}^2 + 2700\Delta_{FF}^2\Delta_{XF}^2\Delta_{XL}^2$$
$$+ 2656\Delta_{XF}^2\Delta_{XL}^2\Delta_{LF}^2 + 18900\Delta_{FF}^2\Delta_{XL}^4$$
$$+ 18900\Delta_{FF}^4\Delta_{XL}^2\}\Delta_{FL}^4 + \{27643\Delta_{XF}^8 + 1442\Delta_{XL}^8$$
$$+ 57858\Delta_{XF}^6\Delta_{FF}^2 + 64744\Delta_{XF}^4\Delta_{FF}^4 + 35400\Delta_{XF}^2\Delta_{FF}^6$$
$$+ 4725\Delta_{FF}^8 + 1080\Delta_{XL}^2\Delta_{FF}^2\Delta_{LF}^4 + 4140\Delta_{XL}^6\Delta_{FF}^2$$
$$+ 2852\Delta_{XF}^2\Delta_{XL}^6 + 6300\Delta_{XL}^2\Delta_{FF}^6 + 7492\Delta_{XF}^6\Delta_{XL}^2$$
$$+ 2520\Delta_{XL}^2\Delta_{FF}^4\Delta_{LF}^2 + 9940\Delta_{XF}^2\Delta_{XL}^4\Delta_{FF}^2$$
$$+ 3150\Delta_{XL}^4\Delta_{FF}^2\Delta_{LF}^2 + 4352\Delta_{XF}^2\Delta_{XL}^2\Delta_{FF}^2\Delta_{LF}^2 \quad \text{(A.1)}$$

$$+ 20940\Delta_{XF}^2\Delta_{XL}^2\Delta_{FF}^4 + 1532\Delta_{XL}^6\Delta_{LF}^2$$

$$+ 240\Delta_{XL}^2\Delta_{LF}^6 + 240\Delta_{XL}^2\Delta_{LL}^6 + 756\Delta_{XL}^4\Delta_{LF}^4$$

$$+ 6300\Delta_{XL}^4\Delta_{FF}^4 + 4560\Delta_{XF}^4\Delta_{XL}^4$$

$$+ 2418\Delta_{XL}^4\Delta_{XF}^2\Delta_{LF}^2 + 984\Delta_{XF}^2\Delta_{XL}^2\Delta_{LF}^4$$

$$+ 2112\Delta_{XF}^4\Delta_{XL}^2\Delta_{LF}^2 + 18996\Delta_{XF}^4\Delta_{XL}^2\Delta_{FF}^2$$

$$+ (3976\Delta_{XF}^2\Delta_{XL}^2\Delta_{FF}^2 + 720\Delta_{XL}^2\Delta_{LF}^4$$

$$+ 1824\Delta_{XF}^4\Delta_{XL}^2 + 2520\Delta_{XL}^2\Delta_{FF}^4 + 2160\Delta_{XL}^2\Delta_{FF}^2\Delta_{LF}^2$$

$$+ 1768\Delta_{XL}^4\Delta_{LF}^2 + 3510\Delta_{XL}^4\Delta_{FF}^2$$

$$+ 1728\Delta_{XF}^2\Delta_{XL}^2\Delta_{LF}^2 + 1820\Delta_{XL}^6 + 2418\Delta_{XF}^2\Delta_{XL}^4)\Delta_{LL}^2$$

$$+ (1080\Delta_{XL}^2\Delta_{FF}^2 + 1012\Delta_{XL}^4 + 720\Delta_{XL}^2\Delta_{LF}^2$$

$$+ 744\Delta_{XF}^2\Delta_{XL}^2)\Delta_{LL}^4\}\Delta_{FL}^2 + 945\Delta_{FL}^{10} + 43989\Delta_{FF}^2\Delta_{XF}^8$$

$$+ 21847\Delta_{XF}^{10} + 945\Delta_{FF}^{10} + 48498\Delta_{FF}^4\Delta_{XF}^6$$

$$+ 33742\Delta_{FF}^6\Delta_{XF}^4 + 11385\Delta_{FF}^8\Delta_{XF}^2,$$

$$M_{2Z}^{(F)} = 2\Delta_{XF}^2,$$

$$M_{4Z}^{(F)} = 4\Delta_{XF}^2\Delta_{FL}^2 + 10\Delta_{XF}^4 + 4\Delta_{FF}^2\Delta_{XF}^2,$$

$$M_{6Z}^{(F)} = 24\Delta_{XF}^2\Delta_{FL}^4 + 92\Delta_{XF}^6 + 78\Delta_{FF}^2\Delta_{XF}^4$$

$$+ 24\Delta_{FF}^4\Delta_{XF}^2 + (70\Delta_{XF}^4 + 48\Delta_{FF}^2\Delta_{XF}^2 + 8\Delta_{XF}^2\Delta_{XL}^2)\Delta_{FL}^2,$$

$$M_{8Z}^{(F)} = 240\Delta_{XF}^2\Delta_{FL}^6 + (240\Delta_{XF}^2\Delta_{XL}^2 + 756\Delta_{XF}^4$$

$$+ 720\Delta_{XF}^2\Delta_{FF}^2)\Delta_{FL}^4 + (720\Delta_{XF}^2\Delta_{FF}^4 + 1768\Delta_{XF}^4\Delta_{FF}^2$$

$$+ 16\Delta_{XF}^2\Delta_{LF}^2\Delta_{XL}^2 + 16\Delta_{XF}^2\Delta_{LL}^2\Delta_{XL}^2 + 1532\Delta_{XF}^6$$

$$+ 248\Delta_{XF}^4\Delta_{XL}^2 + 240\Delta_{XF}^2\Delta_{FF}^2\Delta_{XL}^2 + 40\Delta_{XF}^2\Delta_{XL}^4)\Delta_{FL}^2$$

$$+ 1820\Delta_{XF}^6\Delta_{FF}^2 + 1012\Delta_{XF}^4\Delta_{FF}^4$$

$$+ 240\Delta_{XF}^2\Delta_{FF}^6 + 1442\Delta_{XF}^8,$$

$$M_{10Z}^{(F)} = 3360\Delta_{XF}^2\Delta_{FL}^8 + (6720\Delta_{XF}^2\Delta_{XL}^2 + 10956\Delta_{XF}^4$$

$$+ 13440\Delta_{XF}^2\Delta_{FF}^2)\Delta_{FL}^6 + (20160\Delta_{XF}^2\Delta_{FF}^4 \qquad \text{(A.2)}$$

$$+ 40580\Delta_{XF}^4\Delta_{FF}^2 + 896\Delta_{XF}^2\Delta_{LF}^2\Delta_{XL}^2$$

$$+ 896\Delta_{XF}^2\Delta_{LL}^2\Delta_{XL}^2 + 28752\Delta_{XF}^6 + 11516\Delta_{XF}^4\Delta_{XL}^2$$

$$+ 13440\Delta_{XF}^2\Delta_{FF}^2\Delta_{XL}^2 + 3360\Delta_{XF}^2\Delta_{XL}^4)\Delta_{FL}^4$$

$$+ \{13440\Delta_{XF}^2\Delta_{FF}^6 + 368\Delta_{XF}^2\Delta_{XL}^6$$

$$+ 896\Delta_{XF}^2\Delta_{XL}^2\Delta_{FF}^2\Delta_{LF}^2 + 48292\Delta_{XF}^4\Delta_{FF}^4$$

$$+ 46702\Delta_{XF}^8 + 840\Delta_{XF}^4\Delta_{XL}^2\Delta_{LF}^2$$

$$+ 72348\Delta_{XF}^6\Delta_{FF}^2 + 13788\Delta_{XF}^4\Delta_{XL}^2\Delta_{FF}^2$$

$$+ 6720\Delta_{XF}^2\Delta_{XL}^2\Delta_{FF}^4 + (192\Delta_{XF}^2\Delta_{XL}^2\Delta_{LF}^2 + 808\Delta_{XF}^4\Delta_{XL}^2$$

$$+ 312\Delta_{XF}^2\Delta_{XL}^4 + 896\Delta_{XF}^2\Delta_{XL}^2\Delta_{FF}^2)\Delta_{LL}^2$$

$$+ 2240\Delta_{XF}^2\Delta_{XL}^4\Delta_{FF}^2 + 9048\Delta_{XF}^6\Delta_{XL}^2$$

$$+ 96\Delta_{XF}^2\Delta_{XL}^2\Delta_{LL}^4 + 96\Delta_{XF}^2\Delta_{XL}^2\Delta_{LF}^4 + 280\Delta_{XF}^2\Delta_{XL}^4\Delta_{LF}^2$$

$$+ 2020\Delta_{XF}^4\Delta_{XL}^4\}\Delta_{FL}^2 + 18668\Delta_{XF}^4\Delta_{FF}^6 + 58138\Delta_{XF}^8\Delta_{FF}^2$$

$$+ 3360\Delta_{XF}^2\Delta_{FF}^8 + 44748\Delta_{XF}^6\Delta_{FF}^4 + 35492\Delta_{XF}^{10}.$$

Expressions for the moments of the nuclei of the second type are obtained by changing the subscripts $F \longrightarrow L$ and $L \longrightarrow F$.

For comparison, we present an exact expression for the fourth moment of the ACF $\Gamma_{xF}(t)$ (4) for a system consisting of nuclei of two types with a DDI for a real LiF crystal, which was derived from the results for a similar moment of the NMR spectrum [18], and an expression for the fourth moment of the ACF of a homonuclear system [19]:

$$\frac{M_{4X}^{(F)}}{\Delta_{FF}^4} = \frac{77}{16} - \frac{9S_2}{4S_1^2} + \frac{S_3}{2S_1^2}$$

$$+ \frac{\Delta_{FL}^2}{\Delta_{FF}^2}\left(7 + \frac{\Delta_{LL}^2}{2\Delta_{FF}^2} + \frac{S_3'}{2S_1S_1'}\left(1 - \frac{\Delta_{LL}^2}{\Delta_{FF}^2}\right)\right) \qquad \text{(A.3)}$$

$$+ \left(\frac{\Delta_{FL}^2}{\Delta_{FF}^2}\right)^2\left(3 - \frac{34S_2'}{(5S_1')^2}\right).$$

A similar moment for the Li nuclei is obtained by the change of the subscripts $F \longrightarrow L$ and $L \longrightarrow F$ and by the simultaneous change of the numerical coefficients: $9S_2/4S_1^2$ to $1.27S_2/S_1^2$ and $34S_2'/(5S_1')^2$ to $2S_2'/(S_1')^2$, which is associated with the difference in the spin quantum numbers of the F and Li nuclei.

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