ISSN 1063-7761, Journal of Experimental and Theoretical Physics, 2017, Vol. 124, No. 1, pp. 151–158. © Pleiades Publishing, Inc., 2017.
Original Russian Text © V.E. Zobov, M.M. Kucherov, 2017, published in Zhurnal Eksperimental'noi i Teoreticheskoi Fiziki, 2017, Vol. 151, No. 1, pp. 174–182.

ORDER, DISORDER, AND PHASE TRANSITION IN CONDENSED SYSTEM

Concentration Dependence of the Wings of a Dipole-Broadened Magnetic Resonance Line in Magnetically Diluted Lattices

V. E. Zobov[a]* and M. M. Kucherov[b]

[a] *Kirenskii Institute of Physics, Siberian Branch, Russian Academy of Sciences, Krasnoyarsk, 660036 Russia*

[b] *Institute of Space and Information Technologies, Siberian Federal University, Krasnoyarsk, 660074 Russia*

**e-mail: rsa@iph.krasn.ru*

Received June 20, 2016

Abstract—The singularities of the time autocorrelation functions (ACFs) of magnetically diluted spin systems with dipole–dipole interaction (DDI), which determine the high-frequency asymptotics of autocorrelation functions and the wings of a magnetic resonance line, are studied. Using the self-consistent fluctuating local field approximation, nonlinear equations are derived for autocorrelation functions averaged over the independent random arrangement of spins (magnetic atoms) in a diamagnetic lattice with different spin concentrations. The equations take into account the specificity of the dipole–dipole interaction. First, due to its axial symmetry in a strong static magnetic field, the autocorrelation functions of longitudinal and transverse spin components are described by different equations. Second, the long-range type of the dipole–dipole interaction is taken into account by separating contributions into the local field from distant and near spins. The recurrent equations are obtained for the expansion coefficients of autocorrelation functions in power series in time. From them, the numerical value of the coordinate of the nearest singularity of the autocorrelation function is found on the imaginary time axis, which is equal to the radius of convergence of these expansions. It is shown that in the strong dilution case, the logarithmic concentration dependence of the coordinate of the singularity is observed, which is caused by the presence of a cluster of near spins whose fraction is small but contribution to the modulation frequency is large. As an example a silicon crystal with different ^{29}Si concentrations in magnetic fields directed along three crystallographic axes is considered.

DOI: 10.1134/S106377611615005X

1. INTRODUCTION

The shape of a magnetic resonance line is an example of a complex dynamical problem of a many-body system without an explicit small parameter. The Fourier transform of the line shape – the free induction decay (FID), is expressed in terms of the time correlation function of a total spin component perpendicular to an external static strong magnetic field [1]. In magnetically diluted systems, additional difficulties appear related to the necessity of taking into account the random inhomogeneous distribution of spins in a lattice and averaging correlation functions over this distribution [1–5]. Nevertheless, such systems are often used for studying the problems of nonequilibrium statistical physics such as the ergodicity, thermalization, spin transport, and many-body localization, which have attracted recent interest [5–8].

As an example of magnetically diluted systems, the systems of electron spins are usually considered because of the high EPR sensitivity [9]. The objects of studies in NMR are often the systems of rare isotopes ^{13}C and ^{29}Si. As an interesting example, we present paper [10] in which the NMR spectra of ^{29}Si in silicon crystals doped with this isotope at different concentrations were measured. Although nuclear spins produce a weaker signal, the spin dynamics is determined only by the dipole–dipole interaction (DDI). In electron spin systems, where along with the DDI the exchange interaction and inhomogeneous broadening play, as a rule, a noticeable role, the problem is more complicated.

For strongly diluted spin systems, Anderson [1, 2] proposed the line shape theory based on the summation of the spectra of spin pairs. His theory clearly explained the transformation of a broad Gaussian line of a regular spin system to a narrow Lorentzian line. This approach was widely used in applications [5, 11, 12]. However, a homogeneous spin system in the Anderson approximation is replaced by the inhomogeneous system of spin pairs. The authors of papers [3, 5, 13] took into account the flip-flop interaction between spins of different pairs which causes the modulation of the local field on spins in pairs, on the one hand, and the establishment of the equilibrium in the

system, on the other hand. In [13, 14], the memory function method was applied in which the memory function was physically substantiated with parameters determined from the first terms of the concentration FID expansion. The FID and NMR spectra were calculated for different spin concentrations and good agreement was obtained between the central part of the spectra and experiments [10, 15].

In regular (magnetically concentrated) lattices, for example, formed by ^{19}F nuclei in CaF_2 [1], the shape of the NMR spectrum is not described by a Gaussian but is close to the convolution of a Gaussian with a rectangle. In this case, the FID is described by the expression [1]

$$F(t) = \exp\left(-\frac{p_1^2 t^2}{2}\right)\frac{\sin(p_2 t)}{p_2 t},$$

in which parameters can be expressed in terms of the second and fourth moments of the spectrum as

$$M_2 = p_1^2 + p_2^2/3, \quad M_4 = 3p_1^4 + 2p_1^2 p_2^2 + p_2^4/5.$$

The FID oscillations are caused by the contribution from cross correlation functions of spins at two different lattice sites, whereas autocorrelation functions (ACFs) of a spin in one site decay without oscillations. The simplified calculation [13] of cross correlation functions by changing the coefficient at the memory function does not provide an accurate description of FID oscillations. These oscillations are better described by integral equations [16] based on the predominant consideration of the interaction between spin components parallel to a strong magnetic field (the z axis). The FID theory was further developed in papers [17, 18] in which kinetic equations were derived for the densities of dipoles with the certain projection of the local field on the z axis. These and other papers on the line shape are analyzed in our paper [19].

The authors of these papers [13–18] calculated the FID and the central part of the NMR spectrum. To calculate the wings of spectra in regular lattices, a method of accounting for the modulation of local fields was developed, which leads to self-consistent equations for autocorrelation functions [20–24]. The wings of the spectrum have the exponential frequency dependence with the exponent determined through the coordinate of the nearest singularity of autocorrelation functions on the imaginary time axis. In turn, this coordinate can be calculated through the radius of convergence of a power series in time for autocorrelation functions. After the inclusion of corrections taking into account the properties of real lattices, good agreement was obtained with experiments both for homonuclear [19, 25] and heteronuclear [26–28] systems with the DDI. The wings of spectra of correlation functions play an important role in the determination of the rate of slow equilibrium-establishment processes in spin systems. Their role increases in inhomogeneous systems. We estimated [29] the concentration dependence of the coordinates mentioned above for a magnetically diluted system with the isotropic spin–spin interaction. However, to make comparison with experiments, it is also important to solve a more complicated problem about the change in the coordinate of the singularity of autocorrelation functions in DDI systems during their magnetic dilution, which was not considered so far.

In this paper, we obtained equations for autocorrelation functions in the self-consistent fluctuating field approximation taking into account the axial DDI symmetry and calculated the change in the coordinate of the ACF singularity on the imaginary time axis during magnetic dilution. The equations were derived by performing averaging over the independent occupation of lattice sites taking into account the separation of contributions from nearest neighbors and other spins in the initial regular lattice. This separation is necessary due to different manifestations of contributions from near spins in calculations of different spectral regions. At the line center, their contribution is averaged to zero with increasing the real time due to fast oscillations, whereas for the high-frequency asymptotics of ACFs, their contribution monotonically increases with increasing the imaginary time.

2. EQUATIONS FOR TIME AUTOCORRELATION FUNCTIONS

Consider the initial regular lattice by replacing magnetic atoms by diamagnetic ones at randomly chosen lattice sites. We assume that the occupation of different sites is independent and the probability of a magnetic atom retaining at its site is equal to the mean concentration $c = N_M/N$, where N_M is the number of magnetic atoms or spins and N is the total number of lattice sites. We will use the occupation number representation n_j for the site j: $n_j = 1$ if the site j is occupied by a spin, and $n_j = 0$ if the site is occupied by a nonmagnetic atom [3–5, 13]. After averaging, we obtain $\langle n_j \rangle = c$.

The main reason for broadening of the NMR absorption spectrum in nonmetal diamagnetic solids is known to be the secular part of the internuclear DDI [1], which completely determines the dynamics of the nuclear spin system:

$$H = \sum_{i \neq j} n_i n_j b_{ij}[I_i^z I_j^z - \xi(I_i^x I_j^x + I_i^y I_j^y)]. \quad (1)$$

Here, $b_{ij} = \gamma^2\hbar\,(1 - 3\cos^2\theta_{ij}/2r_{ij}^3$ is the DDI constant, $\mathbf{r}_{ij}$ is a vector connecting spins i and j, θ_{ij} is the angle formed by the vector $\mathbf{r}_{ij}$ with a static external magnetic field, I_i^α is the α component ($\alpha = x, y, z$) of the vector spin operator in the site i, $I = 1/2$, ξ is a parameter introduced for the convenience of a theoretical analysis: $\xi = 1/2$ in the DDI case under study and $\xi = -1$ in

systems with the isotropic spin–spin interaction [29]. Hereafter, energy is expressed in frequency units.

We define the time-dependent ACFs of a spin located at the lattice site i at high temperature by the expression

$$\Gamma_{\alpha i}(t) = \frac{\mathrm{Tr}[e^{iHt} I_i^{\alpha} e^{-iHt} I_i^{\alpha}]}{\mathrm{Tr}[(I_i^{\alpha})^2]}. \quad (2)$$

The ACFs can be expanded in a power series in time

$$\Gamma_{\alpha i}(t) = \sum_{n=0}^{\infty} \frac{(-1)^n}{(2n)!} M_{2n\alpha} t^{2n}. \quad (3)$$

It is known [1] that the coefficient $M_{2n\alpha}$ is the $2n$-order moment of the spectral density of the corresponding ACF. In particular,

$$M_{2z} = 2\xi^2 c S_1, \quad M_{2x} = (1+\xi^2) c S_1, \quad S_1 = \sum_j b_{ij}^2. \quad (4)$$

Exact expressions are obtained only for a few first-order moments. Because of this, ACFs are calculated by various approximate methods. In the axially symmetric Hamiltonian (1), the zz interaction (the interaction of spin components parallel to a magnetic field) plays the main role. By neglecting the second term in Eq. (1) (the interaction of transverse spin components or flip–flop interaction), assuming $\xi = 0$, we find

$$\Gamma_{xi}(t) = \prod_j \cos(n_j b_{ij} t). \quad (5)$$

After averaging over the independent site occupation [3–5, 13], we obtain

$$\Gamma_x(t) = \prod_j [(1-c) + c\cos(b_{ij} t)]. \quad (6)$$

In regular lattices ($c = 1$) with a great number of neighbors, we obtain from (5) a Gaussian

$$\Gamma_x(t) \approx \exp(-S_1 t^2/2). \quad (7)$$

For a strong dilution ($c \ll 1$), we can obtain from (6) [3, 13] the exponential decay

$$\Gamma_x(t) \approx \exp(-Bt), \quad (8)$$

to which the Lorentzian line shape corresponds. Here, $B = (2/3)D_A$, $D_A = 2\pi^2\gamma^2\hbar c/3\sqrt{3}\Omega$ is the Anderson width of a NMR line [1, 2], and Ω is a volume per crystal lattice site.

Anderson used the isolated spin pair approximation. For the correlation function of the total moment of the pair $I_1^x + I_2^x$, we find

$$\Gamma_x(t) = \cos[(1+\xi) b_{12} t], \quad (9)$$

whereas the ACF of one of the spins of the pair has the form

$$\Gamma_{x1}(t) = \cos(b_{12} t)\cos(\xi b_{12} t). \quad (10)$$

The difference of these two functions is caused by the cross correlation function of operators I_1^x and I_2^x:

$$\Gamma_{x12}(t) = -\sin(b_{12} t)\sin(\xi b_{12} t). \quad (11)$$

The Anderson linewidth D_A is obtained for $\xi = 1/2$.

The Anderson theory [1, 2] well describes the narrowed center of the spectrum. However, the wing of the Lorentzian line in relation (8) should be cut off because function (6) has finite moments. The cutoff frequency for one pair of spins is $(1 + \xi)b$, where b is the constant of the DDI spins at the minimal distance in the lattice. The study of regular systems showed that the wing extends well above the resonance frequency of the interaction pair of neighboring spins in the lattice. The high frequency at the wing is obtained due to summation of spin precession frequencies in local fields. The summation occurs through the modulation of these fields due to flip–flop processes. This mechanism is also preserved after magnetic dilution. We will describe a change in the orientation of spins producing a local filed by the ACF $\Gamma_{zj}(t)$ (2) and, by introducing the flip–flop interaction, we will replace the cosine in product (5) by the function $F_{ij}^{(1)}(t)$ specified by the equation

$$\frac{d}{dt} F_{ij}^{(1)}(t) = -b_{ij}^2 \int_0^t \Gamma_{zj/i}(t') F_{ij}^{(1)}(t-t')dt', \quad (12)$$

where $\Gamma_{zj/i}(t)$ is the ACF (2) z component of the spin j in which the interaction with the spin i, on which the field is considered, in excluded.

We neglect in (12) the fact that the interaction between transverse spin components will increase the second ACF moment (4). This effect can be taken into account by renormalizing the DDI constants [19], i.e., by replacing b_{ij}^2 with $(1 + \xi^2)b_{ij}^2$ in Eq. (12). At the same time, the ACF shape in an isolated pair will change so that the product of cosines (10) will appear instead of cosine. The reason is in the transition of polarization on the adjacent spin and its return. This process plays an important role in an isolated pair, but in the presence of a large number of neighbors, the polarization return can be neglected. On the other hand, the interaction between transverse spin components will lead to correlation between contributions from different spins in product (5). For lattices with a large coordination number, such a correlation effect is insignificant and can be taken into account by renormalizing constants b_{ij}, as shown in [19]. This is explained by the fact that in the case of many neighbors, the decay time of ACFs is determined by their combined action. The correlation from interaction of two spins has no time to reveal itself at such times. This reason is also preserved in the calculation of the coordinate of the nearest singularity on the imaginary time axis because its value is

Table

Orientation	Z	$b^2/M_2(1)$	$\delta^2/M_2(1)$
[111]	1	0.653	0.365
[110]	4	0.204	0.187
[100]	4	0.067	0.732

also a collective effect. We will renormalize parameters and take into account corrections later on during comparison with experiments. In this paper, we consider only the simplest equations because focus our attention on the qualitative aspect of the problem.

The DDI constant in (1) decreases with distance by a power law, and therefore each spin interacts, formally speaking, with all others. However, the interaction with Z nearest neighbors is much stronger than that with the far environment. We will consider the contribution from a great number of far neighbors in the form of contribution from the Gaussian local field

$$D_i(t) = \exp\left[-\sum_{i\notin Z} n_j b_{ij}^2 \int_0^t\int_0^{t'} \Gamma_{zj}(t'')dt'dt''\right]. \quad (13)$$

The action of the nearest neighbors is taken into account in the approximation used in [19] assuming that contributions are independent of each of Z nearest neighbors:

$$\Gamma_{xj}(t) = \prod_f^Z [(1-n_f) + n_f F_{jf}^{(1)}(t)]D_j(t), \quad (14)$$

where $F_{jf}^{(1)}(t)$ is contribution (12) to the change in the correlation function of a spin in the site j from the direction in the lattice leading to the spin f.

For $\Gamma_{zj}(t)$ and $\Gamma_{zj/i}(t)$, we will take equations derived in [19] and transform them, taking magnetic dilution into account, to the form

$$\begin{aligned} \frac{d}{dt}\Gamma_{zj}(t) &= -\frac{1}{2}\sum_f b_{jf}^2 n_f \\ &\times\int_0^t \Gamma_{xf/j}(t')\Gamma_{xj/f}(t')\Gamma_{zj}(t-t')dt', \\ \frac{d}{dt}\Gamma_{zj/i}(t) &= -\frac{1}{2}\sum_{f(\neq i)} b_{jf}^2 n_f \\ &\times\int_0^t \Gamma_{xf/ji}(t')\Gamma_{xj/fi}(t')\Gamma_{zj/i}(t-t')dt'. \end{aligned} \quad (15)$$

The system of equations (12)–(15) develops concepts of papers [3, 13] considering the local field modulation in magnetically diluted systems. The self-consistent consideration of ACFs that we introduced allows us to analyze the nonlinear properties of the system on the imaginary time axis in addition to the analysis of the ACF relaxation on the real time axis performed in [13]. On passing to the averaging of Eqs. (12)–(15) over the random spin arrangement, we note that the separation boundary between near and far spins depends on the problem being solved. The center of the spectrum is calculated by studying the correlation function decay on the real time axis. Here, the scale is the width D_A of the spectrum linear in concentration and equal by the order of magnitude to the interaction of spins on the mean distance [1–5, 10, 13]. Contributions to the correlation function from terms containing pairs at smaller distances ("near" pairs) will decay due to fast oscillations in time. Therefore, the number of such sites in the initial regular lattice (the number of "near" spins) will increase during magnetic dilution and will exceed many times the coordination number of this lattice. On the contrary, we calculated spectral wings by studying the growth of the correlation function on the imaginary time axis. In this case, the contribution from near pairs monotonically increases and therefore the nearest neighbors of the initial lattice should be taken as near spins.

Based on the above discussion, consider a simple model spin system in which each spin has a small number Z of nearest neighbors with equal interaction constants b and a great number of weakly interacting far neighbors with the total contribution δ^2 to the lattice sum S_1 (4): $\delta^2 = S_1 - Zb^2$. We will use below the notation

$$M_2(c) = cZb^2 + c\delta^2, \quad M_2(1) = Zb^2 + \delta^2 = S_1$$

for the concentration-dependent second moment of the ACF. Model parameters used for calculating the ACF for the ^{29}Si system in a silicon crystal [10, 15] for magnetic fields directed along three crystallographic axes [111], [110], and [100] are presented in the table. Calculations were performed using lattice sums from [31]. By comparing contributions to the second moment from the DDI spins at different distances, we set $Z = 1$ for the [111] orientation and $Z = 4$ for the [110] and [100] orientations.

By averaging Eqs. (12)–(15) over the random spin arrangement in the independent site occupation approximation for the model system considered above, we obtain equations for averaged functions

$$\begin{aligned} \Gamma_x(t) &= \langle\Gamma_{xj}(t)\rangle = [Y(t)]^Z D(t), \\ \Gamma_z(t) &= \langle\Gamma_{zj}(t)\rangle, \quad \Gamma_z'(t) = \langle\Gamma_{zj/i}(i)\rangle, \end{aligned} \quad (16)$$

where

$$D(t) = \exp\left[-c\delta^2\int_0^t\int_0^{t'}\Gamma_z(t'')dt'dt''\right], \quad (17)$$

and the function

$$Y(t) = [1 + c(\langle F_{ij}^{(1)}(t)\rangle - 1)] \quad (18)$$

satisfies the equation

$$\frac{d}{dt}Y(t) = -b^2\int_0^t \Gamma_z'(t_1)[Y(t-t_1)-(1-c)]dt_1. \quad (19)$$

The two other functions satisfy equations

$$\frac{d}{dt}\Gamma_z(t) = -\frac{1}{2}cZb^2\int_0^t Y^{2Z-2}(t_1)D^2(t_1)\Gamma_z(t-t_1)dt_1$$
$$-\frac{1}{2}c\delta^2\int_0^t Y^{2Z}(t_1)D^2(t_1)\Gamma_z(t-t_1)dt_1,$$
$$\frac{d}{dt}\Gamma_z'(t) = -\frac{1}{2}c(Z-1)b^2\int_0^t Y^{2Z-3}(t_1)D^2(t_1)\Gamma_z'(t-t_1)dt_1 \quad (20)$$
$$-\frac{1}{2}c\delta^2\int_0^t Y^{2Z-1}(t_1)D^2(t_1)\Gamma_z'(t-t_1)dt_1.$$

Our previous studies [22–28] of nonlinear equations for ACFs in regular lattices ($c = 1$) showed the presence of singularities on the imaginary time axis. In the vicinity of singularities nearest to the origin the ACF has the form

$$\Gamma_\alpha(t) \approx \frac{A_\alpha}{(it \pm \tau_0)^2}. \quad (21)$$

In particular, for the system of equations (17) and (20) for $b = 0$, the singularity coordinate was found to be $\tau_0^2 = 6.81/M_2(1)$. If $M_2(1)$ is replaced formally by $M_2(c) = c\delta^2$, this solution can describe a change in the singularity coordinate upon weak dilution.

In the general case for $b \neq 0$ and magnetic dilution, the form of functions (16)–(20) changes. However, they still will have a singularity at the common point on the imaginary axis, because all these functions are coupled by the system of equations. We will express the singularity coordinate τ_0 in terms of the radius of convergence of power series in time (3) for these functions. To do this, we substitute these series into Eqs. (16)–(20) to obtain the recurrent equations for coefficients

$$D_{n+1} = c\delta^2\sum_{k=0}^{n}\binom{2n+1}{2k}D_{n-k}G_k,$$
$$Y_{n+1} = b^2\sum_{k=0}^{n}K_{n-k}Y_k - b^2(1-c)K_n,$$
$$G_{n+1} = cb^2Z\frac{1}{2}\sum_{k=0}^{n}P_{n-k}^{(2Z-2)}G_k$$
$$+c\delta^2\frac{1}{2}\sum_{k=0}^{n}P_{n-k}^{(2Z)}G_k, \quad (22)$$

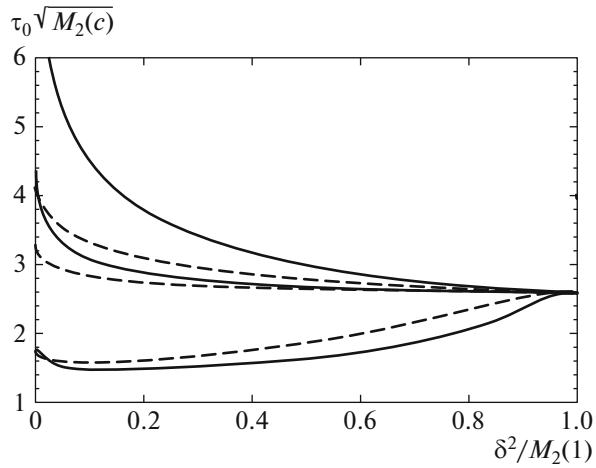

Fig. 1. Dependences of the coordinate of the ACF singularity (in $M_2^{-1/2}(c)$ units) on $\delta^2/M_2(1)$ for $Z = 1$ (solid curves) and $Z = 4$ (dashed curves) at three concentrations (from top to bottom, $c = 1, 0.1, 0.01$).

$$K_{n+1} = cb^2(Z-1)\frac{1}{2}\sum_{k=0}^{n}P_{n-k}^{(2Z-3)}K_k$$
$$+c\delta^2\frac{1}{2}\sum_{k=0}^{n}P_{n-k}^{(2Z-1)}K_k,$$

where Y_n and D_n are the $2n$-order coefficients in series (3) of similar functions, G_n is the corresponding coefficient for $\Gamma_z(t)$, K_n – for $\Gamma_z'(t)$, and $P_n^{(m)}$ is the coefficient for the product $Y^m(t)D^2(t)$.

3. CALCULATION AND DISCUSSION

By solving numerically Eqs. (22), we will calculate the coordinate of the nearest singularity using the d'Alembert formula as the limit of the ratio of adjacent terms in series (3) for $\Gamma_z(t)$. The calculation results are presented in Figs. 1 and 2.

Figure 1 shows the dependences of the singularity coordinate in the imaginary time axis for the solution of the system of equations (16)–(20) $\tau_0\sqrt{M_2(c)} = \tau_0\sqrt{c(Zb^2+\delta^2)}$ on $\delta^2/M_2(1)$ for different concentrations. For $\delta^2/M_2(1) \to 1$, the curves converge to the limit $\tau_0 = \sqrt{6.81/M_2(c)}$ (21). In this limit analyzed above, the concentration dependence is determined by the corresponding dependence of the second moment and has the form $\tau_0 \propto 1/\sqrt{c}$. In this case, the dependence on Z vanishes because $b \to 0$. The dependence $\tau_0 \propto 1/\sqrt{c}$ is observed in Fig. 2 for the [100] orientation at sufficiently high concentrations.

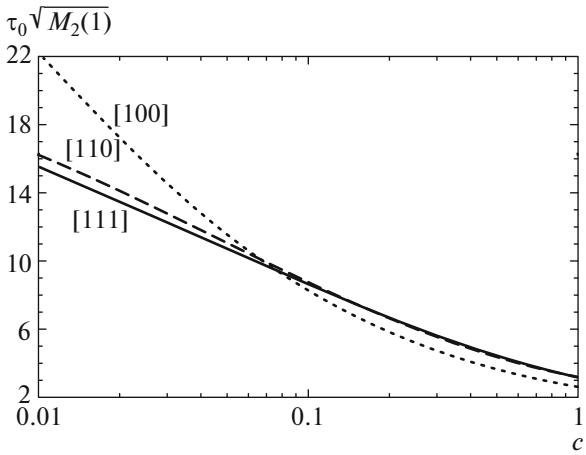

Fig. 2. Dependences of the coordinate of the ACF singularity (in $M_2^{-1/2}(1)$ units) on the ^{29}Si concentration in a silicon crystal with parameters presented in table for a magnetic field directed along the [111], [110], and [100] crystallographic axes.

For $\delta^2/M_2(1) \to 0$, the limit of the value $\tau_0\sqrt{M_2(c)} = \tau_0 b\sqrt{cZ}$ depends on the number of neighbors. In this limit for $Z = 1$, we obtain a spin system consisting of isolated pairs (10). The corresponding oscillating functions have no singularities on the imaginary time axis and therefore $\tau_0 \to \infty$ for $\delta^2/M_2(1) \to 0$. For $Z = 4$ in this limit, τ_0 tends to a finite value because nonlinear self-consistent equations are preserved for the ACF. Similar equations obtained in the Bethe lattice approximation were earlier studied for isotropic spin–spin interaction in our paper [29].

Figure 1 shows that the concentration dependence of τ_0 is not reduced to the concentration dependence of the second moment if we deviate from the limiting value $\delta^2/M_2(1) = 1$. Let us find the form of this dependence for the relation $Zb^2 \gg \delta^2$ at which near spin pairs play an important role. Their contribution $\langle F_{jf}^{(1)}(t)\rangle \sim \cos(b\tau) \sim \cos(bt)$ rapidly oscillating on the imaginary time axis $t = i\tau$ transforms to the rapidly growing contribution $\langle F_{jf}^{(1)}(t)\rangle \propto \cosh(b\tau) \propto e^{b\tau}/2$. However, one isolated pair will not produce a singularity. A singularity of the solution of nonlinear equations (17)–(20) appears when signals from many pairs are combined. As long as $\Gamma_z(t) \sim 1$ and $\Gamma_z'(t) \sim 1$, Eqs. (17) and (19) remain linear. The nonlinear character of the equations and the rapid growth of functions is manifested for $\Gamma_z(t) > 1$. According to Eq. (20), this growth begins for

$$\frac{1}{2}cZb^2\int_0^\tau\int_0^{\tau_1} Y^{2Z-2}(\tau_2)D^2(\tau_2)d\tau_1 d\tau_2 + \frac{1}{2}c\delta^2\int_0^\tau\int_0^{\tau_1} Y^{2Z}(\tau_2)D^2(\tau_2)d\tau_1 d\tau_2 \geq 1. \quad (23)$$

To make the estimate, we write the integrand in (23) in the form $(c/2)^m\exp(2mb\tau + c\delta^2\tau^2)$, where $m = Z - 1$ in the first integral and $m = Z$ in the second integral. By calculating approximately the integrals, we obtain the equation

$$W(Z)\exp[a(Z)\tau_0 + c\delta^2\tau_0^2] = 1, \quad (24)$$

where

$$a(Z) = \begin{cases} 2b, & Z = 1, \\ 2(Z-1)b, & Z > 1, \end{cases}$$
$$W(Z) = \begin{cases} \dfrac{\delta^2}{4b^2}\left(\dfrac{c}{2}\right)^3, & Z = 1, \\ \dfrac{Z}{4(Z-1)^2}\left(\dfrac{c}{2}\right)^{2Z-1}, & Z > 1. \end{cases} \quad (25)$$

By solving Eq. (24), we find the required estimate of the singularity coordinate on the imaginary time axis:

$$\tau_0 = \frac{-2\ln W(Z)}{a(Z) + [a^2(Z) - 4c\delta^2\ln W(Z)]^{1/2}}. \quad (26)$$

This gives the logarithmic concentration dependence of the singularity coordinate of the ACF in the form

$$\tau_0 \sim \frac{v}{b}\ln\frac{1}{c} + R. \quad (27)$$

In semilogarithmic coordinates in Fig. 2, this dependence corresponds to the segments of straight lines which we observe in the strong dilution region $c \ll 1$. Coefficients v and R, determined according to (26), give correct qualitative dependences of τ_0 on parameters b, δ^2, and Z observed in Figs. 1 and 2. First, τ_0 decreases with increasing δ^2 and Z and, second, the dependence of τ_0 on δ^2 weakens with increasing Z. Unfortunately, our rough estimate does not provide a quantitative agreement with the results of numerical solution of complex nonlinear equations.

Thus, upon magnetic dilution, the shape of the central part of the ACF spectrum is transformed from a Gaussian with exponential wings to a Lorentzian with exponential wings. It is known that the width of the spectrum linearly depends on the concentration [1–5, 10, 13] and is determined by spins located at distances close to the average distance. Such spins—"mass" spins—represent the majority and they play an important role in the establishment of equilibrium between the parts of the system. We obtained above that the exponent of the wing ($\omega > 0$) has the logarithmic concentration dependence:

$$g(\omega) \sim \exp(-\tau_0\omega) \sim \exp\left(-\frac{v\omega}{b}\ln\frac{1}{c}\right) = c^{v\omega/b}. \quad (28)$$

Physically, this means that the high frequency appears in the system due to the combination of frequencies of many close spin pairs (the number of pairs is $k \sim \omega/b$) caused by the local field modulation. The probability of formation of one pair is c and that of k pairs is c^k. Although this probability is small, the contribution to the modulation frequency is large and therefore the far wing of the spectrum is determined by the rare clusters of near spins. Note for comparison that, as the modulation frequency is successively increased in the regular lattice, $k \sim \omega/\omega_{loc}$ of mean local frequencies $\omega_{loc} \sim \sqrt{M_2}$ are added.

Based on the theory presented above, we will analyze the transformation of the NMR spectra of a silicon crystal observed in experiments during magnetic dilution [10, 15]. In the magnetically concentrated case, the shape of the spectrum strongly depends on the crystal orientation in a magnetic field. Note that because the silicon lattice is isomorphic to the diamond lattice, the NMR spectra of ^{13}C in diamond enriched with this isotope have the same dependence [32, 33]. The NMR spectrum of ^{29}Si in the [111] orientation has the form of a Pake doublet with the 1250-Hz splitting. In the [100] and [110] orientations, spectra with widths 800 and 2000 Hz are observed, respectively, which are close in shape to the well-known NMR spectra of ^{19}F in fluorite [1] in the [111] and [110] orientations, respectively [34]. Our earlier analysis [25] of experimental spectra in fluorite showed that exponential asymptotics (28) begins approximately with frequencies $2.2\sqrt{M_2}$ measured from the line center.

The NMR line narrows down upon magnetic dilution. In samples with the natural ^{29}Si content (c = 0.047), the linewidth in all orientations decreases down to 100 Hz, of which a significant part (70 Hz) in the opinion of the authors of papers [10, 15] is caused by the magnetic field inhomogeneity. According to the Anderson theory [2], the shape of this line should be Lorentzian. The wing of the Lorentzian ends at frequencies equal to the interaction of two spins at the minimal distance for the lattice. These frequencies are 550 Hz for the [111] orientation and 225 Hz for the [110] orientation. At these frequencies, weak peaks are observed in spectra [10, 15] with ratios of their area to that of the central line equal to 0.06 for the [111] orientation and 0.22 for the [110] orientation. The corresponding theoretical values 0.047 and 0.047 × 4 are equal to the probabilities of formation of close spin pairs for the random ^{29}Si distribution in the lattice [15]. Frequencies in the spectrum exceeding these limiting values will be formed due to the interaction of many spins and can be described by the theory presented above. Unfortunately, the ^{29}Si signal at low concentrations in this spectral region is weak and is masked by noise in experiments [10, 15]. To make quantitative calculations of the reliably observed part of the spectrum, it is necessary to add equations for cross correlation functions to the system of equations for the ACF, to pay great attention to the value of coefficients in equations and to perform numerical calculations similar to these in [13, 14, 19, 27].

The shape of the far wing can be estimated from the rate of slow saturation processes by the resonance field and relaxation similar to these observed in [9]. The presence of the modulation wing for mass spins predicted by the theory presented above can explain the observed exponential shape of spin packets [9]. This theory allows us to treat the saturation mechanism at the wing of the spectrum in a somewhat different way. It was assumed earlier [5, 11] that the resonance field applied at the wing of the spectrum acts on the resonance pair of close spins. After the change of the resonance frequency of the field to the opposite performed in experiments [9], this field acts on the same or similar pair. The rate of this process is limited by the energy exchange between pairs and mass. In the new mechanism, the resonance field applied to the wing of the spectrum acts on the wing of the mass spin line. Its modulation frequency corresponds to some close pair (or pairs) related to the given mass spin. The probability of rotation is small because the number of pairs is small. After the change of the resonance frequency of the field to the opposite, this field acts on another mass spin coupled to the same pair (or close) and turns to the initial state. This process is accompanied by the energy exchange between mass spins and does not require energy exchange between pairs and mass. To compare quantitatively our theory with experiments, it is necessary to take into account the inhomogeneous broadening playing an important role in EPR spectroscopy.

REFERENCES

1. A. Abraham, *The Principles of Nuclear Magnetism* (Clarendon, Oxford, 1961).
2. P. W. Anderson, Phys. Rev. **82**, 342 (1951).
3. F. S. Dzheparov, A. A. Lundin, and T. N. Khazanovich, Sov. Phys. JETP **65**, 314 (1987).
4. E. B. Fel'dman and S. Lacelle, J. Chem. Phys. **104**, 2000 (1996).
5. F. S. Dzheparov, J. Phys.: Conf. Ser. **324**, 012004 (2011).
6. J. Eisert, M. Friesdorf, and C. Gogolin, Nat. Phys. **11**, 124 (2015).
7. R. Nandkishore and D. A. Huse, Ann. Rev. Condens. Matter. Phys. **6**, 15 (2015).
8. D. A. Abanin, W. de Roeck, and F. Huveneers, Phys. Rev. Lett. **115**, 256803 (2015).
9. V. A. Atsarkin, G. A. Vasneva, and V. V. Demidov, Sov. Phys. JETP **64**, 898 (1986).
10. H. Hayashi, K. M. Itoh, and L. S. Vlasenko, Phys. Rev. B **78**, 153201 (2008).

11. F. S. Dzheparov and E. B. Fel'dman, Izv. Akad. Nauk SSSR, Ser. Fiz. **52**, 455 (1988).
12. A. A. Nevzorov and J. H. Freed, J. Chem. Phys. **112**, 1425 (2000).
13. F. S. Dzheparov, D. V. L'vov, and M. A. Veretennikov, JETP Lett. **98**, 484 (2013).
14. F. S. Dzheparov, D. V. L'vov, and M. A. Veretennikov, J. Low Temp. Phys. **41**, 9 (2015).
15. A. S. Verhulst, D. Maryenko, Y. Yamamoto, and K. M. Itoh, Phys. Rev. B **68**, 054105 (2003).
16. G. Sauermann and M. Wiegand, Physica B **103**, 309 (1981).
17. G. E. Karnaukh, A. A. Lundin, B. N. Provotorov, and K. T. Summanen, Sov. Phys. JETP **64**, 1324 (1986).
18. B. N. Provotorov, T. P. Kulagina, andb G. E. Karnaukh, J. Exp. Theor. Phys. **86**, 527 (1998).
19. V. E. Zobov, M. A. Popov, Yu. N. Ivanov, and A. I. Lifshits, J. Exp. Theor. Phys. **88**, 157 (1999).
20. P. Borckmans and D. Walgraef, Physica **35**, 80 (1967), Phys. Rev. **167**, 282 (1968).
21. M. Blume and J. Hubbard, Phys. Rev. B **1**, 3815 (1970).
22. V. E. Zobov, Phys. Lett. A **119**, 315 (1986).
23. V. E. Zobov, Sov. J. Theor. Math. Phys. **77**, 1299 (1988), Sov. J. Theor. Math. Phys. **84**, 751 (1990).
24. A. A. Lundin, A. V. Makarenko, and V. E. Zobov, J. Phys.: Condens. Matter **2**, 10131 (1990).
25. V. E. Zobov and M. A. Popov, J. Exp. Theor. Phys. **97**, 78 (2003).
26. V. E. Zobov and A. A. Lundin, J. Exp. Theor. Phys. **79**, 595 (1994).
27. V. E. Zobov, A. A. Lundin, and O. E. Rodionova, J. Exp. Theor. Phys. **93**, 542 (2001).
28. V. E. Zobov and M. A. Popov, J. Exp. Theor. Phys. **100**, 775 (2005).
29. V. E. Zobov and M. M. Kucherov, JETP Lett. **103**, 687 (2016).
30. A. A. Lundin and B. N. Provotorov, Sov. Phys. JETP **43**, 1149 (1976).
31. H. T. Stokes and D. C. Ailion, Phys. Rev. B **15**, 1271 (1977).
32. K. Lefmann, B. Buras, E. J. Pedersen, et al., Phys. Rev. B **50**, 15623 (1994).
33. K. Schaumburg, E. S. Shabanova, and J. P. F. Sellschop, J. Magn. Res. A **112**, 176 (1995).
34. J. Jensen, Phys. Rev. B **52**, 9611 (1995).

Translated by M. Sapozhnikov