Classification and regularization of spin–spin interactions in magnetically dilute solids

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The spin dynamics in systems with a small concentration of magnetic impurities of the same kind, randomly distributed in a diamagnetic matrix, is considered. All the spins are divided into two groups—those with a discrete spectrum and those with a continuous spectrum. It is assumed that the spins with a discrete spectrum are arranged in clusters, where by an (energy) cluster is meant a group of spins that interact with each other more strongly than with any spin outside the cluster. The spectrum of the spins that do not belong to clusters (after the maximum possible number k_{max} of spins in a cluster has been fixed) is assumed to be continuous. Other ways of defining clusters are also discussed. The contributions of clusters to local fields and the role of clusters in the formation of a magnetic resonance are discussed. In the limit of a continuous medium the usual dipolar specific heat and the second moment of the resonance line are infinite. It is shown that after two- and three-spin clusters have been separated out these quantities become finite (are regularized) and are determined by interactions over intermediate distances. On the basis of numerical modeling the specific heat of the system after such clusters have been separated out is determined, as are the fraction of spins belonging to these clusters and the contribution of these clusters to the resonance line. The process of "hole burning" in the wing of a dipolar-broadened line is considered. The inapplicability of the concept of spin packets to this process is demonstrated, and an explanation of the corresponding experimental results is given.

1. INTRODUCTION

A theoretical analysis of spin-dynamics processes in magnetically dilute solids with a low concentration of spins has been made principally in the context of three groups of topics, viz., the shape of the magnetic-resonance line in a weak alternating field that does not violate the two-temperature equilibrium in the system, the transport of a spin excitation between paramagnetic centers, and the response of the system to an alternating field of amplitude large enough for the system to be strongly nonequilibrium. The first problem has been discussed repeatedly since the classic paper of Anderson,[1] and can be regarded as practically solved (see, e.g., Refs. 2 and 3 and the literature cited therein). The second problem has also been discussed repeatedly (see Refs. 4 and 5), and in it there is now considerable clarity. The third problem has been studied least of all; it rests substantially on the solution of the other two, but is an entirely independent problem.

The question of the kinetics of the establishment of equilibrium in a magnetically dilute spin system with a low concentration of spins is considerably more complicated that in the case of a spatially regular system, and, in certain fundamental aspects, is almost unstudied. We are speaking both of processes of restoration of equilibrium that has been violated by some short-time perturbation and of the establishment of a steady state of the system under conditions of a strong periodic external action. The distinctive features of the system under consideration are linked primarily with the spatial irregularity in the arrangment of the spins in the crystal lattice of the solid and with the resulting nonequivalence of their local environment. The experimental data of Refs. 6 and 7 and the theoretical estimates of Refs. 8 and 9 show that clusters of a small number of closely spaced spins adjust to the basic system extremely slowly, as a consequence of which many states drop out of the observable dynamics, so that a number of questions relating to the magnetic resonance and spin kinetics in such systems require reinterpretation.

In the two-temperature theory of magnetic resonance[10] it is assumed that equilibrium within the Zeeman and dipole reservoirs is established much faster than the mixing of these reservoirs under the influence of the alternating field. For spatially regular systems the consequences of this hypothesis have been confirmed by numerous experiments.[11,12] A number of conclusions of this theory have also been confirmed in the case of strongly dilute systems,[13] but, as later investigations have shown,[6–9] many have not. This is due to the difficulty of migration of spin excitations within the line profile in the presence of spectral nonequilibrium produced by an external perturbation. Possible causes of such nonequilibrium are, e.g., saturation of a wing of the line by steady high-frequency pumping or the pulse saturation used in spin-echo methods.

The theory of stationary saturation has been previously[3,14] extended by us to the case of magnetically dilute systems in the framework of the classical hypothesis of quasiequilibrium in a sufficiently wide temperature range. However, neither the establishment of quasiequilibrium nor a theory of saturation that does not use the above hypothesis in situations in which there are no prior

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grounds for this have been studied (with the exception of a few particular cases,[8,15] which do not provide a complete picture of this complicated phenomenon).

In the present paper we develop the approach proposed in Ref. 16. It starts from division of the spins into several groups and separation of processes that have different time scales and spectra of different natures. It was found that if from the entire system we separate out two- and three-particle clusters as carriers of the discrete spectrum (the method of separation is described below), the remaining spins (the totality of which we call the mass) can be assigned a continuous spectrum. Here, such very important quantities in the theory as, e.g., the specific heat and the second moment of the absorption line of the spins of the mass admit passage to the continuum limit, when in these quantities it is possible to replace lattice sums by integrals over space and the concentration $f \ll 1$ of sites occupied by spins and the lattice constant a appear in observable quantities only through the volume density $c=f/\Omega$ ($\Omega \sim a^3$ is the volume per lattice site). As a result, these quantities are determined by the spin–spin interactions over intermediate distances. We recall that before the clusters are separated out the second moment and specific heat at high temperatures are determined by the interactions of nearest-neighbor spins in the crystal, and in the continuum limit are infinite. This circumstance—the regularization of the spin–spin interactions in the mass—makes it possible to hope that for the mass and for the system as a whole the fastest process will be the establishment of quasiequilibrium in the Zeeman and dipole reservoirs of the mass, for which the two-temperature description is thereby preserved, while the kinetics of the clusters should be constructed with allowance for the discreteness of their spectra. We note that here the spins of the mass are still subject to inhomogeneous broadening from the interaction with the spins of the clusters, and the two-temperature description for them will be valid only over times greater than the time of establishment of equilibrium in this inhomogeneously broadened spectrum.

The magnetic-resonance line in a strongly dilute system with a random uncorrelated distribution of spins has, as is well known,[1,2] extended Lorentzian wings. In accordance with the ideas being developed, these wings are the envelope of the spectra of the individual clusters, while the central part of the line is associated primarily with the mass. The opinion that the line in a strongly dilute system has the nature of the envelope of the spectra of all possible pairs of spins can be traced from the early papers;[17] in particular, in Ref. 6 the situation was likened to inhomogeneous broadening in the case of the description of a spatially regular system in the framework of the spin-packets model. As is well known (see, e.g., Ref. 12, Chapter 6), "classical" inhomogeneous broadening is due to the spread of the g-factors of the various spins that arises from microscopic inhomogeneities of the crystal and (or) electron–nuclear interactions. In contrast to this, the broadening due to dipole–dipole interaction is usually called homogeneous. In the description of inhomogeneous broadening the spin-packets model introduced in Ref. 18 is the most frequently used (see also Ref. 19, Chapter 4, and Ref. 20, Chapter 1). To each packet are assigned spins having one and the same resonance frequency. Energy transfer between packets is possible both as a result of overlap of packets that are very close in frequency (spectral diffusion) and as a result of fluctuational jumps of spins from packet to packet; these processes give rise to an equilibrium between the packets.

Developing here the approach projected in Refs. 6–9 and 16, we investigate the mechanism of spin–spin inhomogeneous broadening in strongly dilute systems, neglecting at this stage its "classical" sources. As the sources of the inhomogeneous broadening of the spectra of the mass we consider clusters distributed randomly in the mass. We prove analytically the regularization of the spin–spin interactions in the mass; combining analytical methods with numerical modeling, we determine the fractions of spins in clusters and in the mass, the magnitudes of the local fields created by the spins at each other, and the relative contributions of the two-particle clusters, three-particle clusters, and the mass to the total absorption line in different parts of the spectrum. The effectiveness of energy transfer over large frequency differences by multispin processes of interaction of clusters with each other and with the mass is estimated, and the experimental results of Ref. 7 on the saturation of the resonance in the case of the inhomogeneous-broadening mechanism under discussion are interpreted.

Everywhere below we confine ourselves to the high-temperature approximation, but we propose to consider the more general case separately on the basis of the technique developed in Refs. 3 and 14.

2. DISTINGUISHING THE CLUSTERS IN A STRONGLY DILUTE SYSTEM

By a spatial k-spin cluster (k_S-cluster) we shall mean a group of k spins such that every spin in the cluster has its $k-1$ nearest neighbors also in the cluster (see Fig. 1). In the language of occupation numbers $n_{\mathbf{q}}$ ($n_{\mathbf{q}}=1$ or 0, depending on whether or not the site q is occupied by a spin; $\langle n_{\mathbf{q}} \rangle_c = f$), we can introduce the object

$$\tilde{z}^{(k)}_{\mathbf{x}_1 \ldots \mathbf{x}_k} = n_{\mathbf{x}_1} n_{\mathbf{x}_2} \ldots n_{\mathbf{x}_k} \prod_{\mathbf{r}}{}' (1-n_{\mathbf{r}}), \tag{1}$$

which we call the occupation number of the k_S-cluster located at the distinct sites $\mathbf{x}_1, \mathbf{x}_2, \ldots, \mathbf{x}_k$. The prime on the product symbol signifies that $\mathbf{r}$ may not coincide with $\mathbf{x}_1, \ldots, \mathbf{x}_k$ and also that the sites $\mathbf{r}$ are in the forbidden volume $V_{\mathbf{x}_1 \ldots \mathbf{x}_k}$ formed by the union of the spheres $S_1, \ldots, S_k$, where the center of S_i lies at $\mathbf{x}_i$ and has radius equal to the largest of the distances from $\mathbf{x}_i$ to the other spins in the cluster. The name "occupation number" is justified by the fact that $\tilde{z}^{(k)}_{\mathbf{x}_1 \ldots \mathbf{x}_k} = 1$ if the sites $\mathbf{x}_1, \ldots, \mathbf{x}_k$ are occupied by spins forming a k_S-cluster, and $\tilde{z}^{(k)}_{\mathbf{x}_1 \ldots \mathbf{x}_k} = 0$ otherwise. The configuration average is

$$\langle \tilde{z}^{(k)}_{\mathbf{x}_1 \ldots \mathbf{x}_k} \rangle_c = f^k \prod_{\mathbf{r}}{}' (1-f) \approx f^k \exp(-f N_{\mathbf{x}_1 \ldots \mathbf{x}_k}), \tag{2}$$

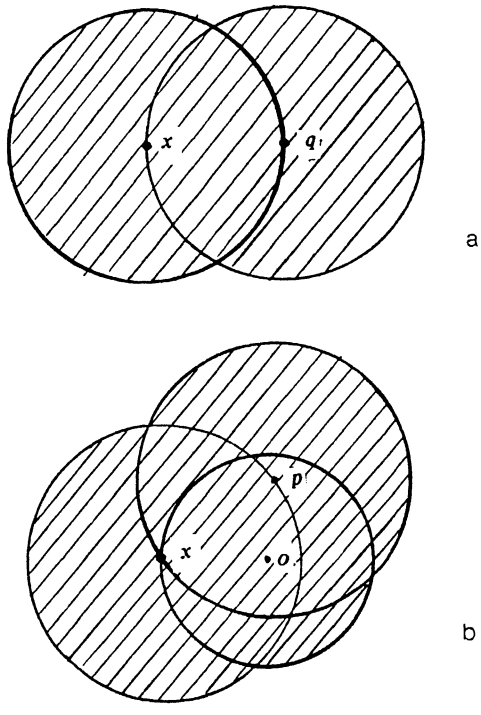

FIG. 1. a) Forbidden volume of a 2_S-cluster; b) forbidden volume of a 3_S-cluster (the figure shows sections cut by the plane in which the spins forming the cluster lie); • denotes the position of a spin.

where $N_{\mathbf{x}_1\ldots\mathbf{x}_k} = V_{\mathbf{x}_1\ldots\mathbf{x}_k}/\Omega$ is the number of sites in the volume $V_{\mathbf{x}_1\ldots\mathbf{x}_k}$. Here and below, it is assumed that the continuum limit is taken. In deriving (2) we have taken into account that $\langle n_{\mathbf{x}_1}\rangle_c = f \ll 1$, $\langle n_{\mathbf{x}_1} n_{\mathbf{x}_2}\rangle_c = f^2$ for $\mathbf{x}_1 \neq \mathbf{x}_2$, etc. Details of working with the occupation numbers $n_{\mathbf{q}}$ can be found in Refs. 2 and 14.

In physical investigations it is more natural to base the classification of clusters not on distances but on the energies of interaction of the spins. By an energy k-spin cluster (k_E-cluster) we shall mean a group of k spins such that the energy of interaction of any spin not belonging to this group with each spin in the group is smaller than the interaction of this spin in the group with every other spin in the group. The nontrivial dependence of the dipole–dipole interaction of a pair of spins in a strong magnetic field H_0, which has the form

$$(\mathscr{H}'_d)_{jk} = A_{jk}\left(S_j^z S_k^z - \frac{1}{4}S_j^+ S_k^- - \frac{1}{4}S_j^- S_k^+\right), \qquad (3)$$

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

θ_{jk} is the angle between $\mathbf{r}_{jk}$ and $\mathbf{H}_0$, leads to a rather complicated shape of forbidden volume (see Fig. 2) and, correspondingly, to significant complications in the use of k_E-clusters. In actual calculations, however, we have convinced ourselves repeatedly that the simpler identification of k_S-clusters often leads to practically the same results as the identification of k_E-clusters (the most important of the exceptions known to us is shown in Fig. 3 below). Therefore, in some of the analytical calculations below we shall use division into k_S-clusters.

The above-described classification of spins in a disordered system admits a number of natural generalizations. It is apparent that the most important of these may be based on comparison of intracluster and extracluster interactions multiplied by scale factors, which could be refined by detailed analysis of any particular spin-kinetics process.

In applications it is convenient to have a division in which each spin belongs to no more than one cluster. This is obtained if we fix the rank $k_{\max}$ of the largest cluster, and then exclude from the k-clusters with $k<k_{\max}$ all those clusters that also belong to clusters of higher rank q ($k<q\leqslant k_{\max}$). It is natural to designate the clusters thus obtained as orthogonal, while the clusters considered above will henceforth be called primary clusters. The occupation number of an orthogonal k-cluster with $k<k_{\max}$ is

$$z_{\mathbf{x}_1\ldots\mathbf{x}_k} = \tilde{z}^{(k)}_{\mathbf{x}_1\ldots\mathbf{x}_k} \prod_{m=1}^{k_{\max}-k}\left(1-\frac{1}{m!}\sum_{\mathbf{r}_1\ldots\mathbf{r}_m}\tilde{z}^{(k+m)}_{\mathbf{x}_1\ldots\mathbf{x}_k\mathbf{r}_1\ldots\mathbf{r}_m}\right), \qquad (4)$$

and, obviously, $z_{\mathbf{x}_1\ldots\mathbf{x}_k} = \tilde{z}^{(k)}_{\mathbf{x}_1\ldots\mathbf{x}_k}$ for $k=k_{\max}$. To prove Eq. (4) we note that if the spins at sites $\mathbf{x}_1,\ldots,\mathbf{x}_k$ do not form a k-cluster, then $z_{\mathbf{x}_1\ldots\mathbf{x}_k} = 0$ as it should. If the spins at $\mathbf{x}_1,\ldots,\mathbf{x}_k$ do form a k-cluster, then the sum

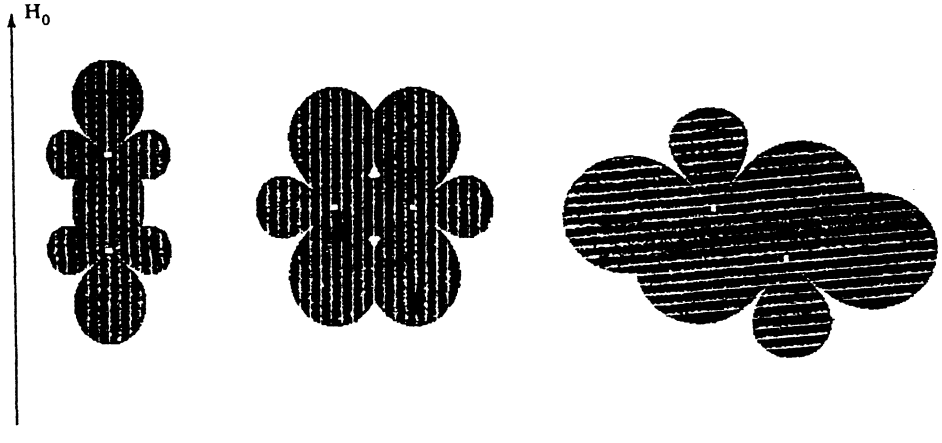

FIG. 2. Forbidden volume of a 2_E-cluster for different orientations with respect to $\mathbf{H}_0$ (the figure shows sections cut by the plane in which the spins forming the cluster lie).

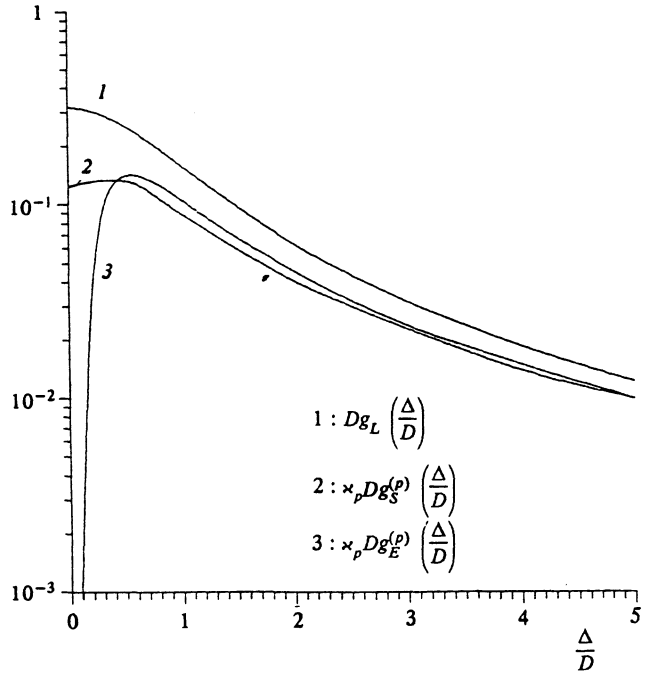

FIG. 3. Form of the functions $g_E^{(p)}(\Delta)$, $g_S^{(p)}(\Delta)$, and $g_L(\Delta)$ in the central part of the line (see the text).

$$\sigma_m=\frac{1}{m!}\sum_{\mathbf{r}_1\ldots\mathbf{r}_m}\tilde{z}^{(k+m)}_{\mathbf{x}_1\ldots\mathbf{x}_k\mathbf{r}_1\ldots\mathbf{r}_m}$$

can contain a contribution from only one cluster of rank $k+m$, which, if it exists, incorporates the k-cluster $\mathbf{x}_1,\ldots,\mathbf{x}_k$, while the factor $1/m!$ cancels the contribution of the identical permutations of the coordinates $\mathbf{r}_1,\ldots,\mathbf{r}_m$. Therefore, the entire sum σ_m takes the value 0 or 1, and the factor $1-\sigma_m$ in (4) is equal to zero if the k-cluster $\mathbf{x}_1,\ldots,\mathbf{x}_k$ is contained in any of the $(k+m)$-clusters and is equal to unity otherwise. Correspondingly, according to (4), $z_{\mathbf{x}_1\ldots\mathbf{x}_k}=0$ if the k-cluster $\mathbf{x}_1,\ldots,\mathbf{x}_k$ is incorporated in any of the q-clusters ($k<q\leqslant k_{\max}$). Otherwise, $z_{\mathbf{x}_1\ldots\mathbf{x}_k}=\tilde{z}^{(k)}_{\mathbf{x}_1\ldots\mathbf{x}_k}$, as it should.

Besides the objects introduced above, it is convenient to use occupation numbers specifying the occupation of individual sites or groups of sites by the mass or by clusters. For example, a natural classification of contributions to any single-particle operator $U_1=\Sigma_{\mathbf{x}}n_{\mathbf{x}}U^{(1)}_{\mathbf{x}}$ or two-particle operator $U_2=\Sigma_{\mathbf{xq}}n_{\mathbf{x}}n_{\mathbf{q}}U^{(2)}_{\mathbf{xq}}$ is obtained on the basis of the relations

$$n_{\mathbf{x}}=n^c_{\mathbf{x}}+n^m_{\mathbf{x}},\quad n^c_{\mathbf{x}}=\sum_{k=2}^{k_{\max}}n^{c(k)}_{\mathbf{x}},\quad n^{c(k+1)}_{\mathbf{x}}=\frac{1}{k!}\sum_{\mathbf{r}_1\ldots\mathbf{r}_k}z_{\mathbf{x}\mathbf{r}_1\ldots\mathbf{r}_k},\tag{5}$$

where $n^c_{\mathbf{x}}$ and $n^m_{\mathbf{x}}$ are the occupation numbers of site x by clusters and the mass, respectively, and $n^{c(k)}_{\mathbf{x}}$ is the occupation number of site x by orthogonal k-clusters. The occupation numbers by clusters and the mass for groups of sites are also useful; a relevant example is contained in Eq. (32).

At first sight, the division into orthogonal clusters depends in an essential way on $k_{\max}$. For example, for $k_{\max}=N$, where N is the total number of spins in the sample, there exists only one orthogonal cluster. It has rank N and incorporates all other clusters. However, it will be seen below that in the theory the important k are $k\leqslant k_0\sim 3$. If $k\ll k_{\max}\ll N$, then $z_{\mathbf{x}_1\ldots\mathbf{x}_k}$ is almost independent of $k_{\max}$, since the probability of formation of clusters of rank $q\sim k_{\max}$ decreases [as $\exp(-N^{(q)})$, where $N^{(q)}$ is the number of sites in the corresponding forbidden volume].

In an entirely analogous way we can construct the occupation numbers of neighbors of a specified site. The occupation number of site $\mathbf{x}$ by the spin closest to site $\mathbf{y}$ is

$$n^{(1)}_{\mathbf{xy}}=n_{\mathbf{x}}\prod'_{|\mathbf{r}-\mathbf{y}|<|\mathbf{x}-\mathbf{y}|}(1-n_{\mathbf{r}}).\tag{6}$$

Next, by induction, the occupation number of site $\mathbf{x}$ by the spin that is the kth neighbor of site $\mathbf{y}$ is

$$n^{(k)}_{\mathbf{xy}}=n_{\mathbf{x}}\sum_{0\neq|\mathbf{z}-\mathbf{y}|<|\mathbf{x}-\mathbf{y}|}n^{(k-1)}_{\mathbf{zy}}\prod_{|\mathbf{x}-\mathbf{y}|>|\mathbf{r}-\mathbf{y}|>|\mathbf{z}-\mathbf{y}|}(1-n_{\mathbf{r}}).\tag{7}$$

These formulas presuppose subsequent passage to the continuum limit, when we can neglect configurations in which several neighbors turn out to be at an equal distance from site $\mathbf{y}$. The averages

$$\langle n^{(k)}_{\mathbf{xy}}\rangle_c=\frac{f^k}{(k-1)!}(S_{\mathbf{xy}})^{k-1}\exp(-fS_{\mathbf{xy}}),$$

$$S_{\mathbf{xy}}=\frac{4\pi}{3}\frac{|\mathbf{x}-\mathbf{y}|^3}{\Omega},$$

coincide, as they should, with a Poisson distribution. Equation (6) was proposed earlier by V. E. Shestopal during the preparation of Ref. 21.

We note that the construction described in this section makes it possible to obtain many results directly in quadratures, in contrast to methods dating back to Ref. 22, based on the formulation of integral equations (see, e.g., Ref. 23).

The fraction of spins belonging to k_S-clusters for $k_{\max}=2$ can be calculated analytically if we neglect the details of the lattice structure. We shall consider a crystal containing $\tilde{N}$ sites, in which the N spins are distributed randomly ($N/\tilde{N}=f\ll 1$). The occupation number of site $\mathbf{x}$ by a spin from a 2_S-cluster is equal to $\Sigma_{\mathbf{q}}\tilde{z}^{(2)}_{\mathbf{xq}}$. Accordingly, in the 2_S-clusters there are $N_p=\Sigma_{\mathbf{xq}}\tilde{z}^{(2)}_{\mathbf{xq}}$ spins, and the fraction that they constitute of the total number of spins is equal to

$$\varkappa_p=\frac{N_p}{N}=\sum_{\mathbf{x}}\langle\tilde{z}^{(2)}_{\mathbf{x}0}\rangle_c$$

$$=f\sum_{\mathbf{x}}(1-f)^{N_{\mathbf{x}0}}\approx c\int d^3x\exp(-cV_{\mathbf{x}0}),\tag{8}$$

where $V_{\mathbf{x}0}$ is the forbidden volume. Since, in the given case, $V_{\mathbf{x}0}=9\pi x^3/4$, after calculation of the integral we obtain $\varkappa_p=16/27\approx 0.59$. Thus, for $k_{\max}=2$ just 11/27 of the total number of spins remain in the mass.

We shall make an approximate estimate of the fraction of spins that belong to k_E-clusters for $k_{max}=2$. In this case too Eq. (8) is valid, if by V_{x0} we understand the forbidden volume corresponding to the interaction (3). It is obvious that $V_{x0} \sim x^3$. Performing a numerical investigation of the angular dependence of the forbidden volume, we find that, if we represent it in the form $V_{x0} = \alpha(\theta)x^3/|1 - 3\cos^2\theta|$, the function $\alpha(\theta)$ varies slowly in the range 5.8–4.7, and for estimates of the integrals it is natural to replace it by the average value $\alpha_e = \int_0^{\pi/2}\alpha(\theta)\sin\theta d\theta = 5.43$. Now the integral (8) is easily calculated, and gives $\varkappa_p = 16\pi/9\sqrt{3}\alpha_e$. The numerical value $\varkappa_p \approx 0.58$ thus determined corresponds to the value $\alpha_e = 5.55$.

Obviously, the fraction of spins in the mass decreases with increase of k_{max}, but analytical calculations with $k_{max} > 2$ encounter great difficulties, even for k_S-clusters. In view of this, numerical modeling has been undertaken. This has been performed both on a lattice and in the continuum approximation. In the former case, N spins (of concentration $f = N/M^3$) are distributed randomly over a cubic lattice in a cube containing $M \times M \times M$ sites, the necessary clusters are distinguished, and the total number of spins appearing them is found. Although the fractions of spins belonging to clusters of different kinds are configurationally self-averaging quantities, the relatively small number N that is accessible in practice for modeling does not give configurationally averaged values, and so the distributions of the spins have been repeated many times until stable averages have been obtained. The same procedure has also been used for the numerical modeling, described below, of other quantities. The statistical processing has been performed using the Student distribution; the parameter uncertainties given below correspond to a confidence level of 0.95 (see, e.g., Ref. 24). To take account of the special conditions of the spins situated near the boundary of the cube, we selected a smaller cube concentric with the given cube and conducted the search for clusters only in the inner cube, using the spins outside the inner cube to check whether the group under inspection was a cluster. The same procedure was used for the modeling in the continuum limit, but the spins were distributed randomly in the same volume.

For $k_{max}=2$ the fraction of spins in k_S-clusters, within the error bars of the modeling that was used as a control (since the exact result is known), amounted (for $f = 1\times10^{-3}$) to 0.59 ± 0.01 (the calculations were performed for $N = 227, 531, 1000$). The result practically coincided with the exact result $16/27 \approx 0.593$. The modeling performed in the continuum limit gave the same result. We note that the calculated value of $\varkappa_p$ does not depend on the choice of boundary conditions within the limits of the indicated accuracy of the calculations.

An analogous calculation of the fraction of spins in k_E-clusters for $k_{max}=2$ for pairs interacting in accordance with the law (3) gave, in the continuum limit, $\varkappa_p = 0.58\pm0.01$, which agrees well with the analytical estimate given above. In the case of modeling on a lattice $\varkappa_p$ oscillates in the range (0.57 ± 0.01)–(0.59 ± 0.01), depending on the orientation of the axes relative to H_0.

The fractions of spins in orthogonal 2_E- and 3_E-clusters for $k_{max}=3$, obtained by numerical modeling, amounted to $\varkappa_p = 0.51\pm0.01$ and $\varkappa_t = 0.11\pm0.01$ (both the results in the continuum limit and the results on a lattice with different orientations of the field lie within the indicated error bars). In all cases, the fraction of spins in 2-clusters for $k_{max}=2$ is very close to the total fraction of spins in 2- and 3-clusters for $k_{max}=3$. This weak dependence of the total number of spins in clusters on k_{max} illustrates the above-noted stability of the properties of clusters of rank $k \leqslant k_0 < k_{max}$ to increase of k_{max}.

We note also the obvious relation

$$\varkappa_k^{(k+1)} + \frac{k}{k+1}\varkappa_{k+1}^{(k+1)} = \varkappa_k^{(k)},$$

where $\varkappa_q^{(k)}$ is the fraction of spins in q-clusters for $k_{max}=k$. For example, in the results given above, $\varkappa_2 + 2\varkappa_3/3 = 0.58\pm0.01$ for $k_{max}=3$ is equal to $\varkappa_2 = 0.58\pm0.01$ for $k_{max}=2$. This result is useful, in particular, for monitoring the results of numerical modeling.

3. CONTRIBUTIONS OF THE CLUSTERS AND MASS TO THE LOCAL FIELDS

To estimate the influence of clusters on the mass we must calculate the root-mean-square local fields produced by the spins of the clusters at spins of the mass and by the spins of the mass at each other.

After distributing the spins randomly over the lattice, as described above, and identifying the orthogonal 3_E- and 2_E-clusters ($k_{max}=3$), we calculated the second moments (squares of the local fields) produced by spins of group "a" at spins of group "b," where a,b take one of the three values m,p,t (mass, pair, triplet):

$$M_2^{a\to b} = \frac{9}{16}\frac{1}{N_b}\sum_{i_b}\sum_{j_a}{}' \frac{(\gamma^2\hbar)^2}{r_{i_b j_a}^6}(1-3\cos^2\theta_{i_b j_a})^2. \quad (9)$$

The prime on the sum over j_a signifies that for $b=p$ and $a=p$ or $b=t$ and $a=t$ we exclude from the sum over j_a the contributions from the cluster to which i_b belongs; N_b is the number of spins in group "b."

Three variants of the calculations of the partial moments were performed (see Table I). Comparison of variants 1 and 3 shows extremely small differences in the classification of the clusters by energies and by distances. Comparison of variants 1 and 2 makes it possible to estimate the concentration dependence of the partial moments, which differs sharply from the usual proportionality of the total M_2 to the concentration of spins. For a discussion of this question, see Sec. 5 below.

In all cases the local fields produced at the mass by pairs is appreciably greater than the fields produced by the spins of the mass at each other, i.e., there are indeed grounds for speaking of substantial inhomogeneous broadening of the line of the mass by pairs. The role of the triplets in this process is appreciably smaller, and the inhomogeneous broadening due to the triplets is smaller than the homogeneous broadening. In exactly the same way, the

TABLE I. Contributions of clusters and the mass to the local fields. The lattice is cubic, and the field $\mathbf{H}_0 \| [001]$; $M_2^{a\to b}=f^2(\gamma^2\hbar/a^3)^2\mu_2^{a\to b}$.

Classification of clusters	by energies	by energies	by distances
Concentration	$1\cdot10^{-3}$	$5\cdot10^{-4}$	$1\cdot10^{-3}$
$\mu_2^{m\to m}$	5,5 ± 1,1	5,6 ± 1,2	6,8 ± 1,2
$\mu_2^{p\to m}$	16,0 ± 3,0	15,2 ± 2,8	14,8 ± 3,0
$\mu_2^{t\to m}$	0,95 ± 0,18	1,24 ± 0,24	1,2 ± 0,2
$\mu_2^{m\to p}$	12,0 ± 2,3	12,0 ± 2,4	10,1 ± 2,1
$\mu_2^{p\to p}$	4,6 ± 0,9	4,8 ± 0,8	5,7 ± 1,0
$\mu_2^{t\to p}$	0,75 ± 0,15	0,80 ± 0,16	1,0 ± 0,2
$\mu_2^{m\to t}$	3,0 ± 0,6	4,0 ± 0,8	2,9 ± 0,6
$\mu_2^{p\to t}$	3,2 ± 0,6	3,4 ± 0,7	3,5 ± 0,6
$\mu_2^{t\to t}$	0,25 ± 0,05	0,27 ± 0,06	0,32 ± 0,06

triplets give rise to substantially smaller local fields at the pairs than do the pairs at each other or than does the mass at the pairs. This gives grounds to expect that the role of the triplets in the spin dynamics is small, and in the first approximation the triplets can be disregarded. However, identification of the triplets as well as the pairs, i.e., the adoption of $k_{\max}=3$, is of fundamental importance when one takes into account the correlations in the spatial arrangement of the spins of the mass, since only in this case, as noted above, can the mass be considered in the continuum limit. In the next section this will be demonstrated in more detail.

The extent to which our ideas on the structure of the energy levels of the spin system change after the clusters have been separated out can be seen from the following relations. The root-mean-square local field, from the magnitude of which one can estimate the broadening of the levels of individual spins in the ordinary "quasi-one-spin" picture of the spectrum of a many-spin system,[11-13] and which is formally defined as $H_L = \gamma\omega_L = \gamma(\mathrm{Tr}\mathscr{H}_d'^2/\mathrm{Tr}S_z^2)^{1/2}$, is related to M_2 by $H_L=\gamma(M_2/3)^{1/2}$ (Ref. 11). When the 3_E- and 2_E-clusters are separated out, with $f=1\times10^{-3}$ and $k_{\max}=3$, the root-mean-square local fields at the mass, at the spins of pairs, and at the spins of triplets are related to H_L as follows: $H_L^{(m)}/H_L\approx0.05$, $H_L^{(p)}/H_L\approx0.05$, and $H_L^{(t)}/H_L\approx0.03$, where $H_L^{(m)}=H_L^{m\to m}+H_L^{p\to m}+H_L^{t\to m}$, etc. This emphasizes once again that, for a random distribution of spins with $f\ll1$, it is practically impossible to judge the structure of the energy spectrum from the magnitude of H_L.

4. CONTRIBUTION OF THE CLUSTERS TO THE MAGNETIC-RESONANCE LINE

The expression for the shape of the magnetic-resonance line in a system of spins $S=1/2$ coupled by dipole–dipole interaction can be represented in the form[3]

$$g(\Delta)=\frac{1}{2\pi N}\int_{-\infty}^{\infty}dt\langle[S_+(t),S_-]_+\rangle e^{i\Delta t}, \qquad (10)$$

where $S_+(t) = \exp(i\mathscr{H}_d't)S_+\exp(-i\mathscr{H}_d't)$, and $\Delta=\omega_0-\omega$ is the frequency shift relative to the center ω_0 of the line ($\langle(...)\rangle=\mathrm{Tr}(...)/\mathrm{Tr}1$).

First we shall perform an analysis of the contribution of pairs (2-clusters) to the absorption for $k_{\max}=2$, neglecting the interaction between the clusters and between the clusters and the mass. Each 2-cluster gives contributions to the absorption at frequencies

$$\pm\frac{3}{4}\frac{\gamma^2\hbar|1-3\cos^2\theta_{jk}|}{\Gamma_{jk}^3}.$$

We shall find the envelope of these contributions (the absorption line of the 2-clusters) by averaging these frequencies with allowance for their statistical weights; this procedure should lead to the exact result, at least in the Anderson model, which forms the basis of the statistical theory of the line shape in a strongly dilute system. It is well known that the line shape in this model, in which one confines oneself to the anisotropic part of the spin–spin interactions (i.e., replaces the Hamiltonian in (3) by $\frac{3}{2}A_{jk}S_j^zS_k^z$), is very close to the actual line shape that arises from the interactions (3).

Proceeding in exactly the same way as in Sec. 2 to determine of the number of spins appearing in pairs, we obtain the number $dn_p(\Delta)$ of pairs that make a contribution in the range of frequencies from Δ to $\Delta+d\Delta$ ($\Delta>0$; the line is symmetric about $\Delta=0$). We represent the forbidden volume in the form $V_{x0}=\varphi(\theta)x^3$, where, for the spatial classification of clusters, $\varphi(\theta)=9\pi/4$, and, for the energy classification, $\varphi(\theta)=\alpha(\theta)/|1-3\cos^2\theta|$. Then

$$dn_p(\Delta)=\frac{1}{16}NC\frac{\gamma^2\hbar}{\Delta^2}\int_0^{\pi}d\theta\sin\theta|1-3\cos^2\theta| \times\exp\left\{-\frac{3}{4}\varphi(\theta)\left|1-3\cos^2\theta\right|\frac{c\gamma^2\hbar}{\Delta}\right\}. \qquad (11)$$

With the spatial classification, after normalization, we obtain

TABLE II. Integral absorption intensities in different parts of the spectrum on account of 2_S- and 2_E-clusters ($k_{max}=2$) and for the corresponding Lorentzian line.

	Part of the spectrum						
	$0-0{,}3D$	$0{,}3D-D$	$D-3D$	$3D-10D$	$10D-30D$	$30D-100D$	$>100D$
2_S cluster	0,039	0,083	0,091	0,055	0,019	0,0072	0,0028
2_E cluster	0,0075	0,089	0,104	0,059	0,020	0,0073	0,0032
g_L	0,093	0,157	0,148	0,071	0,021	0,0074	0,0032

$$g_S^{(p)}(\Delta)=\frac{\mu}{2}\frac{D}{\Delta^2}\int_0^1 dx\,|1-3x^2|\exp\left\{-\mu\left|1-3x^2\right|\frac{D}{\Delta}\right\}, \tag{12}$$

where $D=(2\pi^2/3\sqrt{3})c\gamma^2\hbar$ is the half-width of the Lorentzian line

$$g_L(\Delta)=\frac{1}{\pi}\frac{D}{D^2+\Delta^2}, \tag{13}$$

predicted for a strongly dilute system of dipole-interacting spins in the continuum approximation in the Anderson model, and $\mu=(3/2)^5(1/\pi\sqrt{3})$. As was shown earlier,[2,3] this model reproduces the line shape in a system with real interactions, except, possibly, in the region of small frequencies $|\Delta|<D$. For modeling on a lattice, there are no grounds to expect a line shape of the form (13) even in the region of very large frequences $\Delta\approx\Delta_{max}$, where the continuum approximation is no longer applicable; in order of magnitude, Δ_{max} is equal to $\gamma^2\hbar/a^3$, where a is the lattice constant.

The high-frequency asymptotic form of the function (12) for $\Delta\gg D$ is

$$g_S^{(p)}(\Delta)\approx\frac{27}{16}\frac{1}{\pi}\frac{D}{\Delta^2}. \tag{14}$$

Recognizing that the fraction $\varkappa_p$ of spins that are in pairs is in this case equal to 16/27, we find that the quantity $\varkappa_p=g_S^{(p)}(\Delta)$ at high frequencies completely exhausts the line. We note that for $\Delta\approx0$ the quantity $g_S^{(p)}(\Delta)$ is appreciably different from zero ($Dg_S^{(p)}(0)=(16\pi/243)\approx0.21$).

With the energy classification of the clusters the calculation is simplified considerably if we take $\alpha(\theta)=\alpha=$const (see Sec. 2). We obtain

$$g_E^{(p)}(\Delta)=\frac{\nu}{2}\frac{D}{\Delta^2}\exp\left(-\nu\frac{D}{\Delta}\right),\quad \nu=\frac{9\sqrt{3}}{8\pi^2}\alpha. \tag{15}$$

At high frequences the asymptotic forms of $g_E^{(p)}(\Delta)$ and $g_S^{(p)}(\Delta)$ coincide, but at the center of the line ($\Delta\lesssim2D$) they have noticeably different behavior (see Fig. 3). The physical meaning of the difference in the asymptotic forms at low frequencies is the following. The principal contribution to the low-frequency asymptotic form in the case of spatial clusters is given by spins located at an arbitrary distance but close to $\theta=\arccos1/\sqrt{3}$. But the probability of formation of an energy cluster by these spins is exponentially small, since the forbidden volume for them is inversely proportional to the frequency and is large. In all cases, however, the following conclusion is valid: Asymptotically on the wings of the line, the absorption arises entirely from 2-clusters (for $k_{max}=2$).

To illustrate the extent to which clusters and the mass are important in the absorption in different regions of the spectrum, in Table II we give the results of calculations [using Eqs. (12) and (15)] of the integral intensities

$$I_i^{(p)}=\varkappa_p\int_{\Delta_i}^{\Delta_{i+1}}g^{(p)}(\Delta)d\Delta, \tag{16}$$

where Δ_i are the boundaries of the frequency intervals. For these boundaries we have taken 0, $0.3D$, D, $3D$, $10D$, $30D$, $100D$. In accordance with (16) we have $\Sigma_i I_i^{(p)}=\frac{1}{2}\varkappa_p$. It can be seen that above $\Delta\approx10D$ the absorption arises almost entirely from 2-clusters.

For $k_{max}>2$, analytical estimates of the contributions of clusters of different ranks to the absorption are difficult. However, estimates of these contributions are possible by numerical modeling. Writing the average in (10) in the representation in which the Hamiltonian of the dipole–dipole interactions is diagonal, and using an integral form of the δ-function, we bring (10) to the form

$$g(\Delta)=\frac{2}{N}\frac{1}{\mathrm{Tr}\,1}\sum_{m,n}\langle m|S_+|n\rangle\langle n|S_-|m\rangle \times\delta(E_m-E_n-\omega_0+\Delta), \tag{17}$$

where E_m are the energy levels of the system of interacting spins in a strong field H_0 ($\omega_0=\gamma H_0$). In our approach, in which the system is divided up into orthogonal clusters, we represent (17) in the form

$$g(\Delta)=\sum_{k=1}^{k_{max}}\varkappa_k g^{(k)}(\Delta), \tag{18}$$

where $g^{(k)}(\Delta)$ is the normalized "line-shape function" of the clusters of rank k ($k=1$ corresponds to the mass):

$$g^{(k)}(\Delta)=\frac{1}{2^{k-1}N_k}\sum_{\mu(k)}\sum_{n_\mu m_\mu}\langle m_\mu|S_+|n_\mu\rangle \times\langle n_\mu|S_-|m_\mu\rangle f_\mu^{(k)}(E_m^\mu-E_n^\mu-\omega_0+\Delta). \tag{19}$$

The summation over μ runs over all clusters of rank k, and N_k is the number of spins appearing in them. The normalized functions $f_\mu^{(k)}$ describe the broadening of the levels of the clusters on account of their interactions with each other and with the mass (without allowance for these interactions they would be transformed into δ-functions); E_m^μ and $|n_\mu\rangle$ are the energies and states of the clusters.

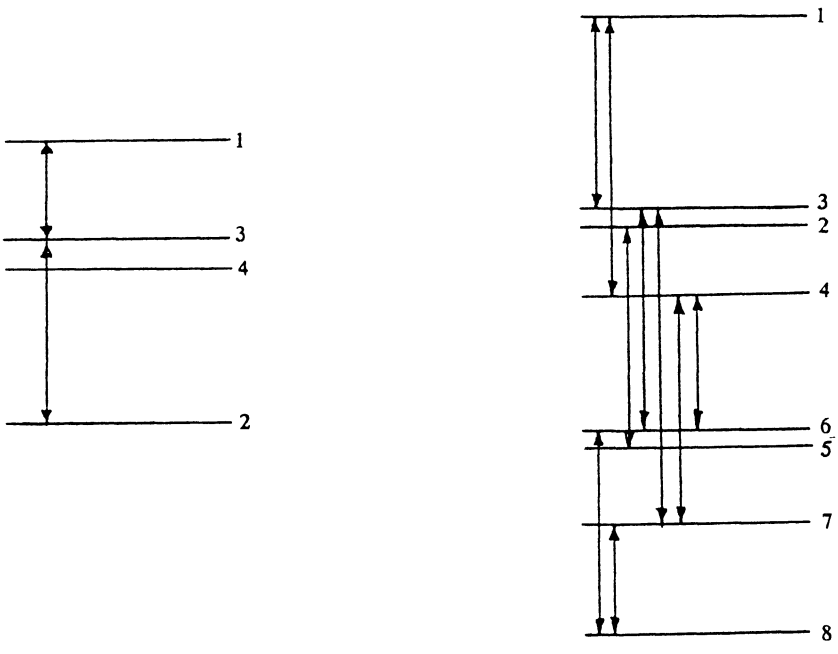

FIG. 4. Transitions induced by an alternating field in the spectra of a pair and a triplet (see the text).

We note that the division used in Eq. (18) is exact for the Anderson model, in which there is no transfer of polarization from spin to spin, and also that the spins in different clusters precess mainly with different velocities, making the transmission of magnetization from one group to another difficult. Therefore, cross terms of the type $g^{(t,m)}$, $g^{(m,p)}$, etc. in $g(\Delta)$ should be comparatively small.

Numerical modeling turns out to be possible because the spectrum and wave functions of relatively small clusters can be determined in practice; for clusters of ranks 2 and 3 this can be done analytically, while for large k ($k \lesssim 10$) numerical diagonalization of matrices of order 2^k (since $S=1/2$) is technically fully realizable.

The spectrum of a pair of spins that are coupled by the interaction (3), and the corresponding wave functions, can be obtained easily:

$$E_1=\omega_0+A_{jk}/4, \quad E_2=-\omega_0+A_{jk}/4, \quad E_3=-A_{jk}/2,$$
$$E_4=0, \quad |1\rangle=|++\rangle, \quad |2\rangle=|--\rangle, \tag{20}$$
$$|3,4\rangle=\frac{1}{\sqrt{2}}(|+-\rangle \pm |-+\rangle).$$

Here, $|++\rangle=|+\rangle_j|+\rangle_k$, and $|+\rangle_j$ and $|-\rangle_j$ are the normalized eigenfunctions of the operator S_j^z.

Substantially more cumbersome is the calculation of the states and spectrum of a three-particle cluster ($S=1/2$) for an arbitrary relative arrangement of the spins. Its Hamiltonian is

$$\mathscr{H}=\omega_0(S_j^z+S_k^z+S_l^z)+(\mathscr{H}'_d)_{jk}+(\mathscr{H}'_d)_{jl}+(\mathscr{H}'_d)_{kl}. \tag{21}$$

The spectrum has the form

$$E_1=3\tau+2a, \quad E_2=\tau, \quad E_3=\tau-a+b, \quad E_4=\tau-a-b,$$
$$E_5=-\tau, \quad E_6=-\tau-a+b, \quad E_7=-\tau-a-b, \tag{22}$$
$$E_8=-3\tau+2a,$$

where

$$\tau=\frac{\omega_0}{2}, \quad a=\frac{1}{2}(x+y+z),$$
$$b=\sqrt{\frac{9}{4}(x^2+y^2+z^2)-\frac{3}{2}(xy+xz+yz)}, \tag{23}$$
$$x=\frac{A_{jk}}{4}, \quad y=\frac{A_{jl}}{4}, \quad z=\frac{A_{kl}}{4}.$$

The states $|1\rangle$ and $|8\rangle$ have the form $|1\rangle=|+++\rangle$ and $|8\rangle=|---\rangle$. The states in the group $|2\rangle$, $|3\rangle$, $|4\rangle$ are linear combinations of the states $|++-\rangle$, $|+-+\rangle$, and $|-++\rangle$, while the states in the group $|5\rangle$, $|6\rangle$, $|7\rangle$ are linear combinations of the states $|--+\rangle$, $|-+-\rangle$, and $|+--\rangle$.

In Fig. 4 the spectra of a pair and of a triplet are depicted schematically; the arrows indicate transitions for which the matrix elements of the operators $S_\pm$ are nonzero, i.e., those which are induced (in leading order in the interaction) by an alternating field perpendicular to H_0. If we represent

$$n\rangle=\alpha_n|++-\rangle+\beta_n|+-+\rangle+\gamma_n|-++\rangle \quad n=2,3,4,$$
$$n\rangle=\mu_n|--+\rangle+\nu_n|-+-\rangle+\zeta_n|+--\rangle \quad n=5,6,7, \tag{24}$$

then

$$\alpha_n=\Delta_1^{(n)}/D_n, \quad \beta_n=\Delta_2^{(n)}/D_n, \quad \gamma_n=\Delta_3^{(n)}/D_n,$$
$$D_n^2=\Delta_1^{(n)2}+\Delta_2^{(n)2}+\Delta_3^{(n)2}, \tag{25}$$

where $\Delta_1^{(n)}$, $\Delta_2^{(n)}$, and $\Delta_3^{(n)}$ can be represented, in particular, in the form

$$\Delta_1^{(2)}=y-z, \quad \Delta_2^{(2)}=z-x, \quad \Delta_3^{(2)}=x-y,$$

TABLE III. Integral absorption intensities in different parts of the spectrum in energy clusters, obtained by numerical modeling: 1) $k_{max}=2$; 2) $k_{max}=3$ in the Anderson model; 3) $k_{max}=3$ with the real interaction.

№	x_t	x_p	Part of the spectrum						
			$0-0.3D$	$0.3D-D$	$D-3D$	$3D-10D$	$10D-30D$	$30D-100D$	$>100D$
1	—	0,58 ± 0,01	$I^{(p)}=0,0080\pm 0,0005$	0,093 ± 0,002	0,105 ± 0,002	0,056 ± 0,002	0,018 ± 0,001	0,0066 ± 0,0004	0,0027 ± 0,0003
2	0,11 ± 0,01	0,50 ± 0,01	$I^{(t)}=0,0088\pm 0,0004$	0,0133 ± 0,0005	0,0155 ± 0,0005	0,0107 ± 0,0005	0,0042 ± 0,0003	0,0018 ± 0,0002	0,0008 ± 0,0002
			$I^{(p)}=0,0068\pm 0,0004$	0,0816 ± 0,0013	0,0933 ± 0,0015	0,0488 ± 0,0011	0,0150 ± 0,0006	0,0050 ± 0,0004	0,0018 ± 0,0002
			$I^{(t+p)}=0,0156\pm 0,0006$	0,0949 ± 0,0014	0,1088 ± 0,0016	0,0595 ± 0,0012	0,0192 ± 0,0007	0,0068 ± 0,0005	0,0026 ± 0,0003
3	0,11 ± 0,01	0,50 ± 0,01	$I^{(t)}=0,0042\pm 0,0002$	0,0055 ± 0,0003	0,0146 ± 0,0006	0,0166 ± 0,0007	0,0076 ± 0,0005	0,0035 ± 0,0004	0,0018 ± 0,0003
			$I^{(p)}=0,0069\pm 0,0004$	0,0821 ± 0,0013	0,0914 ± 0,0015	0,0498 ± 0,0013	0,0145 ± 0,0006	0,0050 ± 0,0004	0,0019 ± 0,0002
			$I^{(t+p)}=0,0111\pm 0,0005$	0,0876 ± 0,0014	0,1060 ± 0,0016	0,0664 ± 0,0015	0,0221 ± 0,0008	0,0085 ± 0,0006	0,0037 ± 0,0004

$$\Delta_1^{(3,4)}=\frac{x-y-z}{2}\pm\frac{b}{3},\quad \Delta_2^{(3,4)}=\frac{y-x-z}{2}\pm\frac{b}{3},\quad \Delta_3^{(3,4)}=\frac{z-x-y}{2}\pm\frac{b}{3}. \tag{26}$$

The quantities μ_n, ν_n, and ζ_n are given by formulas analogous to (25).

Using Eqs. (20)–(26) it is easy to obtain for pairs and triplets the matrix elements $\langle m|S_\pm|n\rangle$ that appear in Eqs. (19) for $g^{(p)}(\Delta)$ and $g^{(t)}(\Delta)$.

In the Anderson model, which was discussed above, formulas analogous to (20)–(26) can be obtained considerably more simply, since the Hamiltonian is already diagonal in the starting representation. To save space we do not write them out explicitly. We note that the frequencies and probabilities of transitions for pairs in this model coincide with those for the real interaction, and this lies at the basis of the model; for triplets we no longer have this coincidence.

Numerical modeling was performed for $k_{max}=3$ in the manner described above; the frequencies of the transitions indicated in Fig. 4, and the matrix elements of the operators in Eq. (19), were found for each of the clusters. The region of frequencies from 0 to Δ_{max} (the line is symmetric about the center) is divided into intervals in which the contributions to $g(\Delta)$ from the pairs and triplets separately are summed as in Eqs. (19). The sizes of these intervals are certainly greater than the characteristic widths of the functions $f_\mu^{(p)}$ and $f_\nu^{(t)}$ appearing in Eq. (19), and, therefore, the concrete form of these functions is of little importance for the histogram illustrating the frequency distribution of the quantities $I^{(p)}$, $I^{(t)}$, and $I^{(t+p)}=I^{(t)}+I^{(p)}$.

As a control, the modeling was first performed for the case $k_{max}=2$ in the continuum limit, with $f=1\times10^{-3}$, with the aim of comparing with the results given in Table II [obtained on the basis of Eq. (11)], studying the influence of the number of spins in the sample on the results of the modeling, and comparing different ways of choosing the boundary conditions. The modeling was carried out for $N=531$ (a cube of $81\times81\times81$ sites) and $N=1000$ (a cube of $100\times100\times100$ sites). In Table III (variant 1) we give the results obtained with the "energy" classification of pairs and boundary conditions of the "cube with a wall" type, in which, in the "large" cube, one selects a concentric cube embedded in it and investigates whether the spins of the inner cube belong to clusters, the spins of the wall being used only to check the clustering. For $k_{max}=2$ the wall thickness has only a very weak effect on the results, and, below, is equal to the average spacing between spins for the given concentration. The contribution of 2_E clusters to the absorption for the same frequency ranges as in Table II is given in Table III; the results are in fully satisfactory agreement.

In Table III we also give the results of the modeling for $k_{max}=3$. These were all obtained for $N=1000$ and a "wall" thickness equal to twice the average interspin spacing, by averaging over 1500 realizations. Variant 2 is the Anderson model in the continuum limit, and variant 3 is the real interaction in the continuum limit.

Comparison of the results with the analogous results corresponding to a Lorentzian (Table II) and with each other permits us to draw the following conclusions. Far out on the wings the functions $g^{(t)}(\Delta)$ and $g^{(p)}(\Delta)$ are quasi-Lorentzian ($\sim\Delta^{-2}$). Their total contribution to the absorption $I^{(t+p)}$ is also quasi-Lorentzian, and, for $\Delta>30D$, practically exhausts it. This can be seen especially clearly from variant 3, since only in this case (the Anderson model) do we have exact information on the line shape (it is Lorentzian over the whole frequency range). As noted previously,[2,3] the actual shape of the line can differ from Lorentzian in the region of small frequencies $|\Delta|\lesssim D$, as a result of flip-flop processes. The actual absorption at the center on account of 3_E-clusters is somewhat smaller than in the Anderson model. For a concentration $f\lesssim10^{-3}$ allowance for the concrete structure of the lattice (the corresponding modeling has also been carried out) leaves the results obtained in the continuum limit almost unchanged (we recall, however, that we are speaking only of the integral contributions to the absorption in very wide frequency intervals).

The main result of the numerical modeling for $k_{max}=3$ is the following: The partial contributions of triplets and

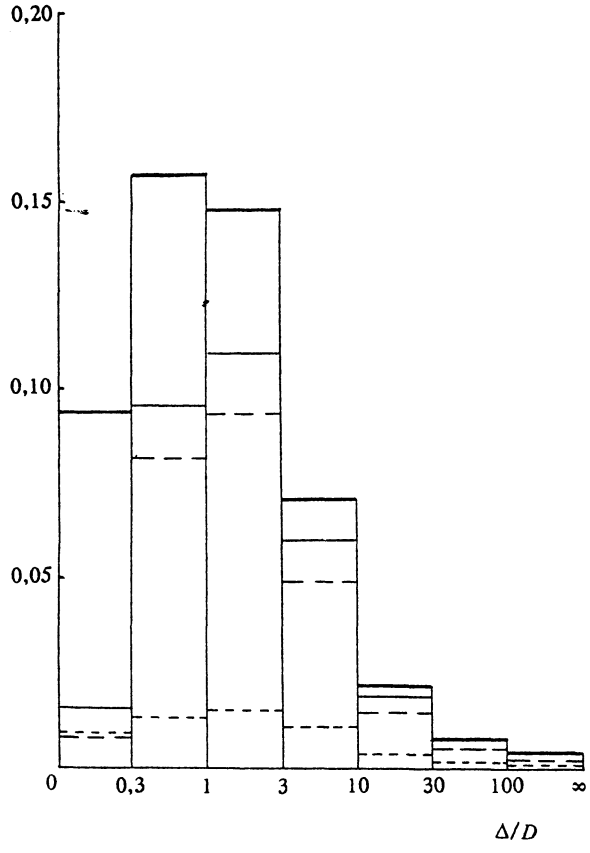

FIG. 5. Partial contributions to the absorption from (a) 3_E-clusters (the short-dashed line), (b) 2_E-clusters (the long-dashed line), and (c) 3_E- and 2_E-clusters jointly (the thin solid line); (d) the Lorentzian function (the thick solid line).

pairs to the absorption are comparable over the whole range of frequencies, fall off in the wings as $\sim\Delta^{-2}$, and, together, practically exhaust the absorption.

In Fig. 5 the partial contributions to the absorption are represented graphically in the form of a histogram corresponding to variant 2. The contribution of the 3_E-clusters is depicted by a short-dashed line, that of the 2_E-clusters by a long-dashed line, and the total contribution of the pairs and triplets by a thin solid line; a thick solid line denotes the result of integration of the Lorentzian function. The results can be interpreted in an obvious way: With increase of Δ the contribution of clusters to the total absorption increases. Whereas for $|\Delta|<0.3D$ it is of order 1/6 (the remainder is the contribution of the mass), for $0.3D<\Delta<2/3$ it is of order 2/3, for $D<\Delta<3D$ it is of order 3/4, for $3D<\Delta<10D$ it is of order 6/7, and for $\Delta>30D$ it practically exhausts the absorption. Of course, the contribution of the mass to the absorption is manifested to a small extent at any high frequency, but an estimate of this contribution lies beyond the possibilities of our modeling. Also beyond these possibilities is the estimate of the asymptotic form of the frequency dependence of the absorption for $\Delta\gtrsim\Delta_{\max}$; for a strongly dilute system this question needs a separate, preferably not numerical, investigation.

5. REGULARIZATION OF THE SPIN–SPIN INTERACTIONS IN THE MASS

We shall consider such important characteristics of the spin system as the dipolar specific heat C_{dd} and the second moment M_2 of the resonance line. The quantity C_{dd}, defined, as is customary in the theory of magnetic resonance, by differentiation of the dipole energy with respect to the inverse temperature, is, in the high-temperature approximation, simply related to M_2 (see, e.g., Ref. 3):

$$C_{dd}=\frac{1}{12}M_2,\quad M_2=\frac{9}{16}f\sum_{\mathbf{x}}A_{\mathbf{x}0}^2. \tag{27}$$

The main contribution to C_{dd} and M_2 is given by clusters of closely spaced spins, since $A_{\mathbf{x}0}^2\sim|\mathbf{x}|^{-6}$; in this case, $C_{dd}\sim D^2/f$. We shall prove that, if we eliminate from C_{dd} the intracluster contributions of the 3- and 2-clusters for $k_{\max}=3$, the remaining expression will be finite in the continuum limit and proportional to the square of the interaction energy of the spins over intermediate distances.[16]

We rewrite M_2 in the form

$$M_2=\frac{9}{16N}\sum_{\mathbf{xq}}n_{\mathbf{x}}n_{\mathbf{q}}A_{\mathbf{xq}}^2,\quad N=\sum_{\mathbf{x}}n_{\mathbf{x}}. \tag{28}$$

We shall consider first the quantity obtained from M_2 by elimination of the contribution of pairs for $k_{\max}=2$ (i.e., pairs are the only form of clusters separated out):

$$\tilde{M}_2=\frac{9}{16N}\left(\sum_{\mathbf{xq}}n_{\mathbf{x}}n_{\mathbf{q}}A_{\mathbf{xq}}^2-\sum_{\mathbf{xq}}\tilde{z}_{\mathbf{xq}}^{(2)}A_{\mathbf{xq}}^2\right), \tag{29}$$

where $\tilde{z}_{\mathbf{xq}}^{(2)}$ is the occupation number of a rank-2 cluster (for $k_{\max}=2$ we have $\tilde{z}_{\mathbf{xq}}^{(2)}=z_{\mathbf{xq}}$). As was shown in Sec. 2, the configuration average is

$$\langle\tilde{z}_{\mathbf{xq}}^{(2)}\rangle_c=f^2\exp/-cV_{\mathbf{xq}})\quad(f\ll1), \tag{30}$$

where $V_{\mathbf{xq}}$ is the forbidden volume depicted in Figs. 1a and 2. Thus,

$$\tilde{M}_2=\frac{9}{16}f\sum_{\mathbf{x}}(1-\exp(-cV_{0\mathbf{x}}))A_{0\mathbf{x}}^2. \tag{31}$$

For $\mathbf{x}\to0$ we have $1-\exp(-cV_{0\mathbf{x}})\to cV_{0\mathbf{x}}\sim cx^3$, and there is a substantial weakening of the short-distance singularity in comparison with the initial M_2 (the power dependence $M_2\sim ca^{-3}$ is replaced by a logarithmic singularity. We note that $\tilde{M}_2$ contains a contribution not only from the interactions in the mass but also from the interactions of the spins of the mass with pairs.

We shall show that extraction of the contribution of the triplets (as well as the contribution of the pairs) eliminates completely the dependence of the second moment and dipolar specific heat on the shortest distance between the spins. This circumstance is nontrivial, since even after the elimination of the pairs and triplets spins at the shortest distance in the lattice continue to be present in the mass, though with smaller statistical weight. We set $k_{\max}=3$ and introduce the quantity $\tilde{z}_{\mathbf{xq}}^{(3)}=\Sigma_{\mathbf{p}}\tilde{z}_{\mathbf{xpq}}^{(3)}$ (the occupation number of a rank-3 cluster; see Sec. 2). The quantity $\tilde{z}_{\mathbf{xq}}^{(3)}$ is indeed an occupation number, i.e., can take only the value 0 or 1, since in the sum defining it not more than one term

can be nonzero. We shall consider the contribution to M_2 from the interactions of spins not belonging to the same cluster (pair or triplet):

$$\tilde{\tilde{M}}_2=\frac{9}{16N}\sum_{\mathbf{xq}} n_{\mathbf{x}}n_{\mathbf{q}}(1-\tilde{z}^{(2)}_{\mathbf{xq}})(1-\tilde{z}^{(3)}_{\mathbf{xq}})A^2_{\mathbf{xq}}$$

$$=\frac{9}{16N}\sum_{\mathbf{xq}} n_{\mathbf{x}}n_{\mathbf{q}}(1-\tilde{z}^{(2)}_{\mathbf{xq}}-\tilde{z}^{(3)}_{\mathbf{xq}}+\tilde{z}^{(2)}_{\mathbf{xq}}\tilde{z}^{(3)}_{\mathbf{xq}})A^2_{\mathbf{xq}}. \quad (32)$$

After configurational averaging we have

$$\langle\tilde{\tilde{M}}_2\rangle_c=\frac{9}{16}f\sum_{\mathbf{x}}\Bigg[1-\exp(-cV_{\mathbf{x}0})-f\sum_{\mathbf{q}}\exp(-cV_{\mathbf{x}0\mathbf{q}})$$

$$+f\sum_{\mathbf{q}\in V_{\mathbf{x}0}}\exp(-cV_{\mathbf{x}0\mathbf{q}})\Bigg]A^2_{\mathbf{x}0}$$

$$=\frac{9}{16}f\sum_{\mathbf{x}}\Bigg[1-\exp(-cV_{\mathbf{x}0})$$

$$-f\sum_{\mathbf{q}\in V_{\mathbf{x}0}}\exp(-cV_{\mathbf{x}0\mathbf{q}})\Bigg]A^2_{\mathbf{x}0}. \quad (33)$$

Here, $V_{\mathbf{x}0\mathbf{q}}$ is the forbidden volume of a three-spin cluster. In the case of a 3_S-cluster it is depicted in Fig. 1b. For $\mathbf{x}\to 0$ we have

$$1-\exp(-cV_{\mathbf{x}0})-f\sum_{\mathbf{q}\in V_{\mathbf{x}0}}\exp(-cV_{\mathbf{x}0\mathbf{q}})$$

$$\to 1-\left(1-cV_{\mathbf{x}0}+\frac{1}{2}c^2V^2_{\mathbf{x}0}\right)-c\left(V_{\mathbf{x}0}-f\sum_{\mathbf{q}\in V_{\mathbf{x}0}}V_{\mathbf{x}0\mathbf{q}}\right)$$

$$\sim c^2V^2_{\mathbf{x}0}\sim c^2x^6. \quad (34)$$

Thus, under the summation sign of (33) there are indeed no short-distance singularities, and $\tilde{\tilde{M}}_2\sim D^2$. Again we note that $\tilde{\tilde{M}}_2$ contains a contribution not only from the interactions of the spins of the mass with each other but also from their interactions with the clusters. It is clear that finiteness of the quantities M_2 and C_{dd} in the continuum limit is found also after elimination of the contributions of clusters for $k_{\max}=3$.

The results of numerical modeling of the partial second moments $M_2^{a\to b}$ were given above in Sec. 3 and confirm their quadratic dependence on the concentration of spins. This does not contradict the fact that the total second moment is proportional to the concentration, since the overwhelming part of it (see the discussion in Sec. 3) is due to the intracluster contributions, which were eliminated from all the $M_2^{a\to b}$.

6. SATURATION ON THE WINGS OF A DIPOLE-BROADENED LINE

A direct experimental investigation of the "hole burning" (saturation of the resonance) in a dipole-broadened line was carried out in Ref. 7. The principal result was interpreted in the framework of the hypothesis of spin packets, and is as follows. If the spin system is characterized by an inhomogeneously broadened line $G(\omega-\omega_0)$ consisting of homogeneous spin packets with form factors $g(\omega-\omega_0)$, the area of the hole that is formed upon saturation at the frequency $\omega_0+\Delta$ is equal to[7]

$$m=\int_{-\infty}^{\infty}d\omega G(\Delta-\omega)[1-\exp(-\pi\omega_1^2 g(\omega)t_p], \quad (35)$$

where $\omega_1=\gamma H_1$ (H_1 is the amplitude of the alternating field) and t_p is the pulse duration. The best agreement in the experimental dependences of m on the power of the ultra-high-frequency pulse has been obtained with the assumption that the wings of the packet have an exponential shape:

$$g(\omega)=(2\delta_{l0})^{-1}\exp(-|\omega|/\delta_{l0}) \quad (36)$$

(rather than a Lorentzian or Gaussian shape).

The spin packet is a global concept, characterizing the entire spin system as a whole, and its shape does not depend on the intensity of the field H_1. But the absorption of the external field occurs locally, and has an essential dependence on the concrete configuration of the spins that participate in each elementary act. Unlike the shape of a spin packet, the line-shape function describing the homogeneous broadening of these spins is not a self-averaging quantity, and only directly observable quantities are subject to configurational averaging. In accordance with this, we shall examine the results of Ref. 7 starting from the ideas developed above.

A detailed theory of magnetic-resonance saturation that takes into account the hierarchy of evolution times of clusters that exists in a magnetically dilute system with $f\ll 1$ should incorporate kinetic equations describing the absorption of energy of the high-frequency field by the mass and by clusters and the process of cross relaxation between them with allowance for the correlations in the spatial distribution of clusters with different transition frequencies and broadenings of the spectral lines; in the later stage of the evolution it should also take spin-lattice relaxation into account. Leaving this complicated problem aside, we shall perform simple estimates of the rates of the elementary processes. We shall assume that the line width determined by the interactions of the spins of the system with the spins of the mass is of order $D_m<D$. Suppose that pumping is carried out on a wing of the line, i.e., the detuning Δ satisfies the condition $D\ll\Delta\ll\Delta_{\max}$. Energy is absorbed principally by clusters having a splitting $3A_\mu=\Delta$ (A_μ is the interaction constant in the μth pair). The rate of the corresponding induced transition is $W^{(1)}\sim\omega_1^2/D_m$. The rate of the four-spin process that restores the equilibrium of a cluster with the mass is $W\sim(D_m^2/A_\mu)^2/D_m$ (Ref. 8); of the same order is the rate of the resonance four-spin process of energy transfer from a pair on a wing to a pair in the region of frequencies $|\Delta|\sim D$. The rates of nonresonance processes are exponentially small in $|A_\mu/D_m|$ (estimates of the rates of many-spin processes are based on the application of averaging methods[11,25–27]). Therefore, the transfer of dipole energy (and, partially, of magnetization) occurs diffusively by small ($\lesssim D_m$) steps through the spectrum. It follows from all this that for

$\omega_1^2 \gg D_m^4/\Delta^2$ and with pumping at frequencies $\Delta \gg D$ two-temperature equilibrium in the large cannot be established, and this agrees with the results of Ref. 7.

The fields from the spins of the mass that produce the homogeneous broadening of the spectra of clusters are naturally regarded as a normal random process in the spirit of Anderson–Weiss theory. In fact, we shall consider the case when the free-induction signal of a spin located at the point $\mathbf{x}$ can be approximated by the expression

$$g_{\mathbf{x}}(t)=\int_{-\infty}^{\infty} d\omega g_{\mathbf{x}}(\omega)\cos\omega t$$
$$\approx\left\langle \exp\left(i\int_0^t d\tau\omega_{l\mathbf{x}}(\tau)\right)\right\rangle$$
$$=\exp\left[-M_2(\mathbf{x})\int_0^t d\tau(t-\tau)K(\tau)\right], \qquad (37)$$

where the second moment $M_2(\mathbf{x})=\frac{9}{16}\Sigma_{\mathbf{q}}n_{\mathbf{q}}A_{\mathbf{qx}}^2$, and the function $K(\tau)$ does not depend on the configuration of the spins. Then,

$$\langle g_{\mathbf{x}}(t)\rangle_c=\exp\left\{-1.13D\left[\int_0^t d\tau(t-\tau)K(\tau)\right]^{1/2}\right\}. \qquad (38)$$

This expression is derived in the same way as, e.g., Eqs. (22) in Ref. 14. If the local fields do not depend on time, it is obvious that we obtain a Lorentzian line with a width equal to 0.8 of the exact value. But if the local fields fluctuate in time, so that $K(\tau)$ falls off over an interval $\tau_c=\int_0^\infty d\tau K(\tau)$, we obtain for $t\gg\tau_c$ the asymptotic form

$$\ln\langle g_x(t\gg\tau_c)\rangle_c=-1.13D\sqrt{t\tau_c} \qquad (39)$$

in agreement with the earlier prediction of Ref. 2. Thus, the proposed approximation leads to surprisingly good results even when the actual distribution of the local fields is still not very close to a normal distribution. After the 3_E- and 2_E-clusters have been separated out, the distribution of the local fields should become closer to a normal distribution, and this justifies the proposed approximation. An important point is that Eq. (37) leads to a practically exponential wing of $g_x(\omega)$ for any reasonable form of the correlator $K(t)$; an example of the corresponding analysis is contained in Ref. 28.

We now consider hole burning at a distance $\Delta \gg D$ from the center of a dipole-broadened line in the experiment of Ref. 7. The wing of the dipole-broadened line

$$g_s(\omega\gg D)=\frac{1}{\pi}\frac{D}{\omega^2}\left(1+O\left(\frac{D}{|\omega|}\right)\right)$$

is due entirely to the first term (of order f^1) of the concentration expansion,[2,3] which can be expressed in terms of the solution of the problem of the evolution of two isolated spins. After division of the system into clusters and the mass, a contribution of order f^1 will arise only from the primary 2_E-clusters with interaction $|A_{ij}|\gg D$, since all other contributions contain additional powers of f from the forbidden volume or from the rank ($k>2$) of the cluster. Correspondingly, the leading term in the above-indicated asymptotic form of g_S ($\omega\gg D$) is completely determined by the contribution of the 2_E-clusters. Another proof of this fact, based on Eq. (11), is outlined in Sec. 4.

As was explained above, the change of the populations of the states of the cluster under the influence of interactions with the mass and other clusters occurs slowly by virtue of the large magnitude of the inhomogeneous broadening induced by intracluster interaction. Thus, the hole burning is the saturation of 2-cluster transitions in resonance with the alternating external field. Here, Eq. (35) remains valid if it is understood locally, i.e., if it applies to one cluster, and we recognize that the functions $G(\Delta)$ and $g(\omega)$ are determined by the concrete arrangement of both the spins in the absorbing cluster and the nearest spins of the mass. If we approximate the fields induced by the spins of the mass and by a normal random process with second moment M_2 and $K(t)=\exp(-\frac{1}{2}\delta_l^2t^2)$, where the fluctuation rate $\delta_l=(\langle\dot{\omega}_l^2\rangle/M_2)^{1/2}$, then, in the case of strong saturation, when the rate of hole burning substantially exceeds the rate of hole covering, the observed area of the hole is

$$m(\Delta)=\left\langle\int_{-\infty}^{\infty}d\omega G(\Delta-\omega)(1-\exp(-\pi\omega_1^2g(\omega)t_p))\right\rangle_c$$
$$=\left\langle\frac{2\delta_lLG(\Delta)}{\sqrt{2\ln(1+\delta_l^2L/M)}}\left(1+O\left(\frac{1}{\ln L}\right)\right)\right\rangle_c, \qquad (40)$$

$$L=\ln(\sqrt{\pi/2M_2}\omega_1^2t_p).$$

In the absence of division of the spins into the mass and clusters, we would find that, in the continuum limit, $\langle\delta_l\rangle_c=\infty$ and Eq. (40) would be wrong. When the division is taken into account, $\langle\delta_l\rangle_c$ is finite, and the distribution of δ_l is substantially narrower than in the absence of division. In fact, $\langle\delta_l\rangle_c$ could diverge only because in our construction the fourth moment $\langle M_4\rangle_c$ diverges, in contrast to $\langle M_2\rangle_c$. However, such a divergence could stem only from the action of spins of a resonance cluster on the surroundings—only this gives dangerous terms of the type A_{ij}^4 in $\langle\dot{\omega}_l^2\rangle$. But this perturbation is smaller than Δ by virtue of the very construction of the cluster, and, consequently, there is no divergence. To summarize, if we take into account the slowness of the variation of the logarithm, all the averages in (40) can be decoupled and, as a result,

$$m(\Delta)\approx\frac{2\langle\delta_l\rangle_c\langle G(\Delta)\rangle_c\ln\bar{W}t_p}{\sqrt{2\ln\ln\bar{W}t_p}},\quad \bar{W}=\sqrt{\frac{\pi}{2}\left\langle\frac{1}{M_2}\right\rangle_c}\omega_1^2t_p. \qquad (41)$$

This formula is in good qualitative agreement with the results of Ref. 7. A numerical analysis is necessary to elucidate the quantitative agreement, since the pre-asymptotic terms in (40) are important in the entire region $\bar{W}t_p\lesssim 10^5$ that is practically accessible in experiment.

We draw attention especially to the fact that when Eqs. (35) and (36) are used we also find that

$$m(\Delta)\sim\ln\bar{W}t_p, \qquad (42)$$

but, in contrast to (36), $\langle g(\omega)\rangle\sim\omega^{-4}$ and the correct logarithmic dependence of $m(\Delta)$ on $\bar{W}t_p$ is obtained only

upon averaging not of $g(\omega)$ but of the directly observable quantity $m(\Delta)$. It is evident that the deviations from Eq. (42) that were observed in Ref. 7 at large values of $\bar{W}t_p$ can be due to direct pumping on a wing of the line of the spins of the mass. The rough estimate $\langle g_m(\Delta)\rangle_c \sim (1/D_m)(D_m/\Delta)^4$, corresponding to $k_{\max}=3$, does not contradict this hypothesis. A more detailed investigation of this question can give information on $k_{\max}$, independently of the above-mentioned quasi-thermodynamic condition $k_{\max}=3$ formulated in Sec. 5.

An important point is that the result (41) is extremely stable to the choice of correlator $K(t)$, and answers close to (41) and (42) are obtained with the use of the more realistic approximations for $g(\omega)$ that were indicated in Ref. 4.

7. CONCLUSION

The approach developed above, based on the physically clear idea of the isolation of clusters in a strongly dilute spin system, has made it possible to achieve regularization of the spin–spin interactions in the remaining part (the mass) and to justify, qualitatively and quantitatively, the idea originally proposed in Refs. 6 and 7 concerning a nonstandard mechanism of inhomogeneous broadening of the magnetic-resonance line in such a system. We have combined, simplified, and improved the primary cluster-analysis elements that were indicated independently, and in different directions, in Refs. 8 and 9. The classification of orthogonal clusters and the technique of working with their occupation numbers, introduced in Ref. 16 and developed in the present paper, make it possible to obtain many results analytically as well as numerically, and to make further progress in the study of spin kinetics. An important point is that the structure of the crystal lattice is eliminated from the proposed theory in the limit of small concentrations, i.e., the theory has a physically well-defined continuum limit, as it should for the group of phenomena under consideration.

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