

## Depolarization of rotating spins by random walks on lattices

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The depolarization of rotating spins that perform a random walk on a  $d$ -dimensional lattice ( $d=1,2,3$ ) with randomly distributed rotation frequencies is studied by numerical simulations and, especially for  $d=1$ , by analytical methods. For a Gaussian frequency distribution an exponential polarization decay is found in all dimensions for large times  $t$ , or large step numbers  $n$ . The dependence of the decay constant  $\lambda$  on the width  $\sigma$  of the frequency distribution is determined by the dimensionality  $d$ . In  $d=1$ , an approximation that takes the distribution of spans of the random walk into account yields a behavior  $\lambda = \text{const}\sigma^\beta$  with an exponent  $\beta = \frac{4}{3}$  which is in good agreement with the simulations. The exponent  $\beta \approx 1.77$  is numerically obtained for smaller values of  $\sigma$  in  $d=2$ . In  $d=3$  the Gaussian description is appropriate, at least for small  $\sigma$ :  $\lambda = a_0\sigma^2$ , where  $a_0$  depends on the structure of the lattice. These results qualitatively agree with the predictions of an effective-medium theory for the decay constant. Other examples of frequency distributions are considered in  $d=1$  to examine the dependence of the polarization decay on the particular choice of the distribution.

### I. INTRODUCTION

In the study of spin rotation or spin resonance it is a question of special interest how diffusion processes of the particles manifest themselves in the experimentally observed signals. One important quantity is the correlation function that describes the loss of phase coherence or polarization decay of rotating spins. This loss may be caused by interactions of the spins with the environment, and between themselves. A stochastic theory for the polarization decay is easily developed when drastic simplifications on the interaction processes influencing the spin rotation are made. For example, the stochastic theory of the decay of spin polarization developed by Anderson<sup>1</sup> and by Kubo and Tomita<sup>2</sup> in the 1950s assumes that single spins experience additional rotation frequencies  $\omega(t)$ , where  $\omega(t)$  represents a Gaussian stochastic process. In this paper we are concerned with the loss of phase coherence of spin rotation when the stochastic process  $\omega(t)$  should be considered as resulting from the random-walk processes of the spins on a lattice. We point out in the following paragraphs that serious problems arise in the usual stochastic theory, especially when the random walk takes place on a low-dimensional lattice.

The simplest stochastic model of polarization decay is obtained by considering the rotation of a single spin in a transverse magnetic field with uniform frequency  $\omega_0$  (henceforth neglected) and the additional frequency  $\omega(t)$ . The rotation is described in the complex plane, and the ensemble average of the polarization  $P(t)$  at time  $t$  is given by

$$P(t) = \left\langle \exp \left[ -i \int_0^t dt' \omega(t') \right] \right\rangle, \quad (1.1)$$

where the spins were initially polarized along the real axis. The polarization decay is directly observable in NMR as

the free-induction decay of spins turned perpendicular to the axis of the magnetic field by a  $90^\circ$  pulse, and in muon-spin rotation ( $\mu$ SR) by monitoring the decay positions of muons rotating in a transverse field.<sup>3</sup> In the standard stochastic theory of depolarization it is assumed that  $\omega(t)$  represents a stationary Gaussian process with variance  $\sigma^2$  at equal times. The process is then uniquely characterized by its second cumulant  $\langle \omega(t)\omega(0) \rangle$ . When the further assumption of a Markovian process is made, this cumulant can only decay exponentially, as follows from Doob's theorem.<sup>4</sup> It is then easy to evaluate (1.1); we give only the limiting cases:

(i) Slow fluctuations with  $\sigma\tau_c \gg 1$ , where  $\tau_c$  is the correlation time of the frequencies. In this case the decay of polarization is dominated by the behavior at times  $t \ll \tau_c$ , i.e.,

$$P(t) \approx \exp(-\sigma^2 t^2 / 2). \quad (1.2)$$

(ii) Rapid fluctuations with  $\sigma\tau_c \ll 1$ . In this case the large-time behavior dominates the polarization decay, i.e.,

$$P(t) \approx \exp(-\sigma^2 \tau_c t). \quad (1.3)$$

The latter case of exponential decay corresponds to motional narrowing of the line shape.<sup>5</sup> Instead of assuming a Gaussian random process  $\omega(t)$ , one can also introduce a "strong-collision model"<sup>6</sup> where the frequency  $\omega(t)$  changes according to a Poisson process with the event rate  $\tau_c^{-1}$ . After each transition a fixed frequency  $\omega'$  is taken independently out of a (Gaussian) probability distribution. This strong-collision model leads to the same asymptotic results (1.2) and (1.3) as the model of Gaussian modulation; also, the intermediate behavior is very similar.<sup>7</sup> Although the strong-collision model seems to be more similar to a random-walk process, it is, in fact, not really dif-

ferent from the Gaussian model.

As said above, we wish to investigate the stochastic process  $\omega(t)$  that is induced by the random-walk processes of the spins, and its influence on the polarization decay. Consider, e.g., a muon that experiences different rotation frequencies  $\omega_r$  at the interstitial sites  $r$  of a lattice, due to the dipolar interactions with the host nuclei. The muon may perform a random walk on the interstitial lattice, and a random rotation frequency  $\omega(t)$  results.

The stochastic process  $\omega(t)$  induced by this random walk is a Markovian, but evidently not a Gaussian process. Nevertheless, several<sup>8-10</sup> authors applied the assumption of a Gaussian process also to the calculation of polarization decay caused by random walks on low-dimensional lattices, led by the simplicity of this process. The behavior of the second cumulant  $\langle \omega(t)\omega(0) \rangle$  was taken from random-walk theory; it decays especially slowly in  $d=1$  ( $\propto t^{-1/2}$ ), due to the frequent returns of the particle to the same sites. A polarization decay

$$P(t) \sim \exp\left[-\frac{4}{3}(2\pi)^{1/2}\sigma^2\tau_c^{1/2}t^{3/2}\right] \quad (1.4)$$

results from this treatment for diffusion on a linear chain. This result was physically appealing as an intermediate behavior between (1.2) and (1.3) as a consequence of the revisitation effects. It is also in good agreement with experiments on the ESR line shape found in one-dimensional spin diffusion.<sup>8,10</sup> However, the question of the validity of this approximation remains. The assumption of a Gaussian process, the Markovian nature of the random-walk process, and the slow decay of  $\langle \omega(t)\omega(0) \rangle$  are inconsistent in view of Doob's theorem.

In a recent letter<sup>11</sup> we examined the polarization decay caused by discrete random walk of a spin-carrying particle on a linear chain with rotation frequencies taken out of a Gaussian distribution. It was shown by analytical arguments that the polarization decays  $\propto \exp(-\lambda n)$  for large step numbers  $n$ . These results were corroborated by simulations; also, an approximate theory for the polarization decay was given. Here the results of Ref. 11 are developed in greater detail, and extended. In particular, the role of different probability distributions  $g(\omega_r)$  of the rotation frequencies  $\omega_r$  attached to the sites is examined. It turns out that for various distributions there is a range of step numbers where exponential decay prevails. The final decay is governed by the precise form of the frequency distributions. However, for small  $\sigma$ , or short residence time  $\tau$  of a particle on a site in continuous-time random walk, the true asymptotic behavior is shifted to unmeasurably small polarizations. The Gaussian frequency distribution has an idealized character, in that it leads to exponential decay also for the longest times.

Another approximation inherent in the stochastic theory is the single-particle description of polarization decay. Such a picture is appropriate for the discussion of depolarization of diffusing muons; it forms an approximation in the case of many interacting spins. A theory of the spin-correlation function for interacting spins in low-dimensional systems was developed by Reiter and Boucher.<sup>12</sup> They developed a self-consistent theory and found exponential decay of the spin-correlation functions. The reason for the exponential decay could be traced back to

the dipolar-interactions of the spins. In contrast, it is assumed in our derivation that the main or only cause of random rotation frequencies are the spin-host interactions, as exemplified by a muon in a metal. In this work the exponential decay is deduced solely from the detailed nature of the stochastic process leading to depolarization.

In the following section the basic elements of the stochastic theory of spin depolarization by random walks are given. In Sec. III we describe the systematic technique of cumulant expansion, appropriate for short and intermediate times. In Sec. IV the asymptotic polarization decay in  $d=1$  is studied, and in Sec. V the numerical results are presented. The influence of different frequency distributions is examined in Sec. VI. In Sec. VII an effective-medium theory is given, which can be extended to higher dimensions. In Sec. VIII we present the simulation results for  $d=2$  and 3 and their analysis. Section IX contains the concluding remarks.

## II. BASIC PROPERTIES OF THE MODEL

The model we use for a description of the depolarization of spins is defined in the following way: We consider an infinite  $d$ -dimensional lattice which has a spin-rotation frequency  $\omega_r$  associated with each lattice site  $r$  which is taken from a probability distribution  $g(\omega)$ .<sup>13</sup> For convenience we suppress vector notation. With the exception of Sec. VI our considerations will be restricted to the case of a Gaussian distribution with mean  $\langle \omega \rangle = 0$  and variance  $\sigma^2$ , i.e.,

$$g(\omega) = \frac{1}{\sqrt{2\pi\sigma}} \exp\left[-\frac{1}{2}(\omega/\sigma)^2\right]. \quad (2.1)$$

At time  $t=0$  a spin particle is put on a randomly chosen site. The particle performs a simple random walk (RW) on the lattice, i.e., transitions to only nearest-neighbor sites occur with equal probabilities. Two different models will be considered: the continuous-time random walk where the transitions occur according to a Poisson process with event (or transition) rate  $\gamma = \tau^{-1}$  ("continuous model"), and the discrete random walk, which is characterized by a fixed time interval  $\tau$  between successive transitions. Since  $\tau$  defines a natural timescale for both processes, times will be measured in units of  $\tau$  and frequencies in units of  $\gamma$ .

In the static case, where each particle stays on its initial site forever, the decay of polarization depends only on the distribution of the attached frequencies. Introducing the characteristic function  $\hat{g}$  of  $g$ ,

$$\hat{g}(x) = \langle e^{i\omega x} \rangle = \int_{-\infty}^{\infty} e^{i\omega x} g(\omega) d\omega, \quad (2.2)$$

the depolarization is simply given by  $P(t) = \hat{g}(t)$ , and (1.2) is regained for the Gauss distribution (2.1).

In the case of the random walk the ensemble average (1.1) over many spin particles actually must be interpreted as a double average, where one extends over different realizations of the random walk and the other over different configurations  $\{\omega_r\}$ . Thus

$$P(t) = \left\langle \left\langle \exp\left[-i \int_0^t dt' \omega(t')\right] \right\rangle_{\text{RW}} \right\rangle_{\{\omega_r\}}. \quad (2.3)$$

The last average can be performed exactly, if we introduce the total times  $t_r(t)$  that the particle has spent on the particular site  $r$ . The accumulated phase of spin rotation,  $\int_0^t \omega(t') dt'$ , can be written as  $\sum_r \omega_r t_r(t)$ , and we obtain

$$P(t) = \left\langle \left\langle \exp \left[ -i \sum_r \omega_r t_r(t) \right] \right\rangle \right\rangle \\ = \left\langle \prod_r \hat{g}[t_r(t)] \right\rangle, \quad (2.4)$$

i.e., for the distribution (2.1):

$$P(t) = \left\langle \exp \left[ -\frac{\sigma^2}{2} \sum_r t_r^2(t) \right] \right\rangle. \quad (2.5)$$

In the case of the discrete random walk we only consider the depolarization for integer times  $t_n = n$  (i.e., the time when the  $n$ th transition is performed) and the above equations can be rewritten as

$$P_n \equiv P(t_n) = \left\langle \left\langle \exp \left[ -i \sum_r \omega_r h_r(n) \right] \right\rangle \right\rangle \quad (2.6)$$

$$= \left\langle \exp \left[ -\frac{\sigma^2}{2} \sum_r h_r^2(n) \right] \right\rangle, \quad (2.7)$$

where  $h_r(n)$  is the total number of visits on the lattice site  $r$ . For simplicity, it will generally be assumed that random walks start at the origin  $r=0$  of the lattice.

Before we consider the difficulties which are connected with the further evaluation of (2.5) or (2.7), we first discuss the more general aspects of the model.

Obviously, the characteristic property of the stochastic process  $\omega(t)$  induced by the random walk of the particle is that repeated rotations with the same frequency can occur. In comparison with the strong-collision model where a new frequency is taken on with each transition, the number of different frequencies is reduced compared to the number of transitions. These revisiting effects can be considered as a "loss of mobility." One expects, qualitatively, a reduced motional narrowing, or in other words, a somewhat faster polarization decay with time. This effect should be especially strong for random walks on low-dimensional lattices, since revisiting effects become more important with decreasing dimensionality (note that the strong-collision model is "exact in  $d = \infty$ ").

These expectations are reinforced by the results of the theory which combines the Gaussian assumption for  $\omega(t)$  with frequency correlations appropriate for the random-walk process. For a stationary Gaussian process (1.1) yields

$$P(t) = \exp \left[ -\int_0^t \langle \omega(t') \omega(0) \rangle (t-t') dt' \right]. \quad (2.8)$$

In the case of the continuous random walk,

$$\langle \omega(t) \omega(0) \rangle = \sigma^2 P(0, t) \\ \sim \text{const} \times \sigma^2 t^{-d/2} \quad (t \gg 1), \quad (2.9)$$

where  $P(0, t)$  is the probability that a particle is at the origin  $r=0$  at time  $t$ . Thus the following asymptotic behavior of  $P(t)$  in  $d=1$ —see (1.4)—and  $d=2$  results:

$$P(t) \sim \exp(-\text{const} \times \sigma^2 t^{3/2}) \quad \text{for } d=1, \quad (2.10)$$

$$P(t) \sim \exp[-\text{const} \times \sigma^2 t \ln(t)] \quad \text{for } d=2. \quad (2.11)$$

The Gaussian assumption (2.8) always leads to a simple-exponential decay when a correlation time

$$\bar{\tau} \equiv \langle \omega(0)^2 \rangle^{-1} \int_0^\infty \langle \omega(t) \omega(0) \rangle dt < \infty$$

exists; in this case  $P(t) \sim \exp(-\sigma^2 \bar{\tau} t)$ .<sup>14</sup> This condition is fulfilled for transient random walks; the quantity  $\bar{\tau}$  is given by  $\int_0^\infty P(0, t) dt \equiv a_0$  for the simple random walk in  $d \geq 3$ , e.g.,  $a_0 = 1.516 \dots$  for the simple-cubic lattice.<sup>15</sup> Thus the Gaussian assumption yields

$$P(t) \sim \exp(-\sigma^2 a_0 t) \quad \text{for } d \geq 3. \quad (2.12)$$

In the Introduction we pointed out the principal inconsistency of this treatment. Nevertheless, these results may be used to give an exact bound for the actual polarization decay.

As den Hollander pointed out,<sup>16</sup> Jensen's inequality for convex functions can be applied to the depolarization problem. Note that  $P(t)$  is given in (2.5) as the average of a convex function of the stochastic variable  $\sum_r t_r^2(t)$ ; hence,

$$P(t) \geq \exp \left[ -\frac{\sigma^2}{2} \left\langle \sum_r t_r^2(t) \right\rangle \right]. \quad (2.13)$$

i.e., a rigorous lower bound for the polarization decay is obtained. On the other hand, the expression on the right-hand side of (2.13) is nothing else than another form of (2.8), i.e., the depolarization one obtains by introducing the Gaussian approximation for  $\omega(t)$ . Thus, (2.10)–(2.12) yield lower bounds for the asymptotic decay of  $P(t)$ . The consequences for transient walks are obvious; since an upper bound (for all  $d$ ) is given by the depolarization of the strong-collision model, the actual polarization must thus decay simple-exponentially. The question about the decay in  $d=1$  and 2 can obviously not be answered by these considerations.

According to (2.7) the problem of random-walk averaging is reduced to the problem of determining probabilities  $p_n(\{h_r\})$  for the sets  $\{h_r\}$  of the numbers of visits  $h_r$  on the sites  $r$  by an  $n$ -step random walk. We remark that the knowledge of these probabilities would also be useful for the calculation of  $P(t)$  in the continuous case. Let us briefly comment on the results of random-walk theory concerning the determination of the  $p_n(\{h_r\})$ . For arbitrary dimension  $d$ , Rubin and Weiss have given the generating function of the probability  $p_n(\{h_{r_1}, h_{r_2}, \dots, h_{r_m}\})$  for finite sets of lattice sites.<sup>17</sup> This result is useful in the case where just a few sites  $r_1, r_2, \dots, r_m$  are of interest, but not appropriate for the depolarization problem. Only the simple topology in  $d=1$  allows us to count explicitly all possible random walks with fixed numbers of visits on the different sites. (See van Beijeren and Spohn.<sup>18,19</sup> Their results are still rather complicated, and we were not able to perform the summation over all possible configurations  $\{h_r\}$  in the expressions for the depolarization.)

### III. CUMULANT EXPANSION FOR THE DEPOLARIZATION IN $d=1$

In  $d=1$  the Gaussian assumption for the stochastic frequency modulation  $\omega(t)$  leads to an asymptotic depo-

larization decay proportional to  $\exp(-\text{const} \times t^{3/2})$ , which is in obvious contradiction to the observed simple-exponential decay we briefly mentioned in the Introduction. Nevertheless, it is an interesting question whether or to which extent the Gaussian treatment—or, more generally, a systematic cumulant expansion—can be used as an approximate description. For simplicity, we shall restrict this discussion to the case of the continuous model. Consider the exact frequency-averaged expression (2.5) for the depolarization, which is replaced by (2.13) in the Gaussian approximation. If one considers the probability distribution for the stochastic variable  $\Sigma \equiv \Sigma t_r^2$  generated by the random walk, (2.13) corresponds to its approximation by a simple  $\delta$  distribution, namely  $\delta(\Sigma - \langle \Sigma \rangle)$ . The cumulant expansion of (2.5) provides the possibility to take systematically higher moments of the actual distribution into account; this expansion is given by

$$P(t) = \lim_{m \rightarrow \infty} P_{(m)}(t),$$

$$P_{(m)}(t) \equiv \exp \left[ \sum_{k=1}^m \left[ -\frac{\sigma^2}{2} \right]^k \frac{1}{k!} \langle \Sigma^k(t) \rangle_c \right], \quad (3.1)$$

where each quantity  $\langle \Sigma^k(t) \rangle_c$ , the cumulant of  $k$ th order, is determined by the first  $k$  moments

$$M_1(t) \equiv \langle \Sigma(t) \rangle, \dots, M_k(t) \equiv \langle \Sigma^k(t) \rangle.$$

The Gaussian approximation represents the cumulant expansion in first order.

To investigate the cumulants in more detail, we start from the simple expression for the time  $t_r(t) = \int_0^t n_r(t') dt'$ , where the stochastic variable  $n_r(t')$  is defined to be one if the particle is at site  $r$  at time  $t'$ , and to be zero otherwise. The initial condition of start at the origin  $r=0$  corresponds to  $n_r(0) = \delta_{r,0}$ . The set of all quantities  $n_r(t)$  fully describes the actual random walk. With

$$\Sigma(t) = \sum_r \int_0^t dt_1 \int_0^{t_1} dt_2 n_r(t_1) n_r(t_2),$$

we can write the  $k$ th moment in the form

$$M_k(t) = \sum_{r_1, r_2, \dots, r_k} \int_0^t dt_1 \int_0^{t_1} dt_2 \dots \int_0^{t_{2k-1}} dt_{2k} \langle n_{r_1}(t_1) n_{r_1}(t_2) \times \dots \times n_{r_k}(t_{2k-1}) n_{r_k}(t_{2k}) \rangle. \quad (3.2)$$

It is useful to divide the integral up into  $2k!$  time-ordered integrals, where  $t_1 \geq t_2 \geq \dots \geq t_{2k}$ . If we formally introduce  $2k$  site variables  $r_1, r_2, \dots, r_{2k}$ , which obey the conditions  $r_{k+i} = r_i$  ( $i = 1, 2, \dots, k$ ), and further define  $S(2k)$  to be the set of all permutations  $\Pi$  on the set of indices  $\{1, 2, \dots, 2k\}$ , (3.2) can be written as

$$M_k(t) = \sum_{\Pi \in S(2k)} \sum_{r_1, r_2, \dots, r_k} \prod_{i=1}^k \delta_{r_i, r_{i+k}} \int_0^t dt_1 \int_0^{t_1} dt_2 \dots \int_0^{t_{2k-1}} dt_{2k} \langle n_{r_{\Pi(1)}}(t_1) n_{r_{\Pi(2)}}(t_2) \times \dots \times n_{r_{\Pi(2k)}}(t_{2k}) \rangle. \quad (3.3)$$

For simplicity let us define  $\hat{r}_j \equiv r_{\Pi(j)}$ . The average  $\langle n_{\hat{r}_1}(t_1) n_{\hat{r}_2}(t_2) \dots n_{\hat{r}_{2k}}(t_{2k}) \rangle$  in (3.3) is the joint probability  $p(\hat{r}_1, t_1; \hat{r}_2, t_2; \dots; \hat{r}_{2k}, t_{2k} | r=0, t=0)$  that a particle, starting at time  $t=0$  at the origin  $r=0$ , is at times  $t_j$  at the sites  $\hat{r}_j$  ( $j=1, 2, \dots, 2k$ ). This probability can be written as a product of conditional probabilities  $P(\hat{r}_i, t_i | \hat{r}_{i+1}, t_{i+1}; r=0, t=0)$  that a particle is at site  $\hat{r}_i$  at time  $t_i$ , when it has been at  $\hat{r}_{i+1}$  at  $t_{i+1}$ . Since the random-walk process is a Markovian process, this conditional probability is just the ordinary random-walk probability  $P(\hat{r}_i, t_i | \hat{r}_{i+1}, t_{i+1})$  that a particle, starting at time  $t_{i+1}$  at site  $\hat{r}_{i+1}$ , is at  $\hat{r}_i$  at time  $t_i$ . This can be written as

$$P(\hat{r}_i - \hat{r}_{i+1}, t_i - t_{i+1} | r=0, t=0) \equiv P(\hat{r}_i - \hat{r}_{i+1}, t_i - t_{i+1}).$$

Thus we obtain

$$\langle n_{\hat{r}_1}(t_1) n_{\hat{r}_2}(t_2) \dots n_{\hat{r}_{2k}}(t_{2k}) \rangle = P(\hat{r}_1 - \hat{r}_2, t_1 - t_2) P(\hat{r}_2 - \hat{r}_3, t_2 - t_3) \times \dots \times P(\hat{r}_{2k}, t_{2k}). \quad (3.4)$$

Inserting this expression in (3.3), we obtain a convolution-type integral. Hence the Laplace transform  $\tilde{M}_k(u)$  of  $M_k(t)$  is given by

$$\tilde{M}_k(u) = \frac{1}{u} \sum_{\Pi} \sum_{r_1, r_2, \dots, r_k} \prod_{i=1}^k \delta_{r_i, r_{i+k}} \tilde{P}(\hat{r}_1 - \hat{r}_2, u) \tilde{P}(\hat{r}_2 - \hat{r}_3, u) \times \dots \times \tilde{P}(\hat{r}_{2k}, u), \quad (3.5)$$

where  $\tilde{P}(r, u)$  is the Laplace transform of  $P(r, t)$ .

To derive explicitly  $\tilde{M}_k(u)$ , one has to determine the contributions of the different permutations in (3.5). We refrain from describing the simplifications which can be obtained by using symmetry and factorization arguments.<sup>20</sup>

We point out the moments of successively higher order can actually be computed although with rapidly increasing calculational efforts. Here we restrict the discussion to the first two moments. We note the normalization

$\sum_r P(r,t)=1$ , corresponding to  $\sum_r \tilde{P}(r,u)=u^{-1}$  in the Laplace domain. The Laplace transform of the one-dimensional probability  $P(r,t)$  is given by<sup>21</sup>

$$\tilde{P}(r,u) = \frac{[u+1-\sqrt{u(u+2)}]^{|r|}}{\sqrt{u(u+2)}} \equiv \frac{[A(u)]^{|r|}}{C(u)}. \quad (3.6)$$

The calculation of the first moment is trivial, and (3.5) yields

$$\tilde{M}_1(u) = \frac{2}{u^2} \tilde{P}(0,u) = \frac{2}{u^2 \sqrt{u(u+2)}}, \quad (3.7)$$

which corresponds to the following expression in the time domain:

$$M_1(t) = \langle \Sigma(t) \rangle = \int_0^t t' \exp(-t') [I_0(t') + I_1(t')] dt',$$

where  $I_0(t), I_1(t)$  are modified Bessel functions.<sup>22</sup> For  $t \gg 1$  one finds the asymptotic behavior given in (1.4) or (2.10). We also consider the second moment  $\tilde{M}_2(u)$ . In the evaluation of (3.5) for  $k=2$ , three different contributions (with equal weights  $2^{k!}=8$ ) must be calculated; the result is

$$\tilde{M}_2(u) = \frac{8}{u^2} \left[ \frac{1}{u} \tilde{P}^2(0,u) + \tilde{P}(0,u) \sum_r \tilde{P}^2(r,u) + \sum_r \tilde{P}^3(r,u) \right], \quad (3.8)$$

especially for  $d=1$ ,

$$\tilde{M}_2(u) = \frac{8}{u^2 C(u)} \left[ \frac{1}{u} + \frac{1}{C(u)} \frac{1+A^2(u)}{1-A^2(u)} + \frac{1}{C(u)} \frac{1+A^3(u)}{1-A^3(u)} \right]. \quad (3.9)$$

From the expansion of  $\tilde{M}_2(u)$  for small  $u$  one finds the asymptotic behavior of  $M_2(t)$  for  $t \rightarrow \infty$ :  $M_2(t) \sim 11/9t^3$ . This is a special case of the general result  $M_k(t) \sim \text{const} \times t^{3k/2}$  ( $t \rightarrow \infty$ ), which can be proven rigorously. Omitting numerical factors, we can represent the result of the second-cumulant approximation as

$$P_{(2)}(t) \sim \exp(\text{const} \times t^3) \text{ as } t \rightarrow \infty. \quad (3.10)$$

Since the constant in the exponent is positive, this asymptotic expression obviously is irrelevant for the description of the asymptotic depolarization decay; nevertheless, the second-cumulant approximation makes an important contribution at intermediate times. The explicit calculation of  $P_{(2)}(t)$  for arbitrary times  $t$  can be performed by numerical inversion of (3.9). Figure 1 shows the behavior of  $P_{(1)}(t)$  and  $P_{(2)}(t)$  for a typical value of  $\sigma$ . Comparison with the simulation shows that the actual decay is relatively well approximated up to the order of  $10^{-1}$  by the first- and up to the order of  $10^{-2}$  by the second-cumulant approximation. This observation gives an explanation for the fact that experimental results were found in relatively good agreement with the theory based on the Gaussian approximation.<sup>8</sup> Already the rough approximation given by (1.4) yields times for decay to  $1/e$  which typically deviate only about 30% from the simulated values. This allows

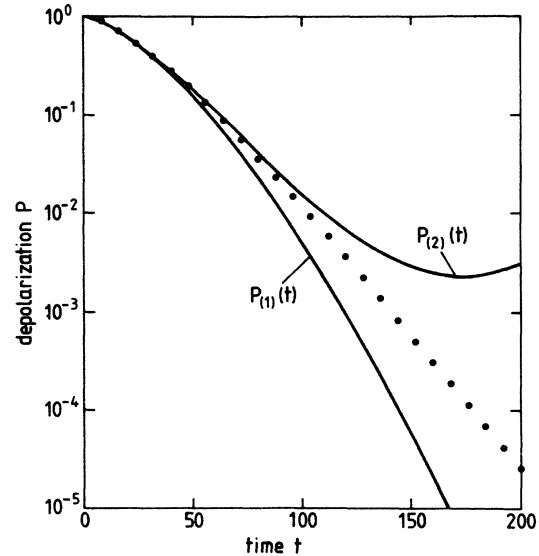


FIG. 1. Depolarization of the continuous random walk in  $d=1$  for  $\sigma=0.1$ . Comparison of the first- and second-cumulant approximation,  $P_{(1)}(t)$  and  $P_{(2)}(t)$ , respectively, with the result of the numerical simulation (points).

the conclusion that the linewidth derived from the Gaussian treatment should describe quite well the correct behavior (see the experimental results in Ref. 8). We note that a similar conclusion was drawn by Reiter and Boucher for the model with interacting spins.<sup>12</sup>

The cumulant expansion obviously is an expansion for short times. The time region, in which the decay of polarization is quite well approximated, increases with decreasing parameter  $\sigma$ , which can be considered as a measure of disorder. The larger the disorder, i.e.,  $\sigma$ , the earlier finer details of the random-walk process, which actually probes the disorder, become important. Finer details enter via higher correlations, i.e., averages of the form  $\langle n_{r_1}(t_1) n_{r_1}(t_2) \cdots n_{r_k}(t_{2k}) \rangle$ , in this description, i.e., via higher moments or cumulants. At least in  $d=1$ , correlations due to visits on a fixed number  $k$  of lattice sites cannot sufficiently describe the situation in the asymptotic regime, where arbitrarily many sites and returns to all of them are involved. The advantage of the cumulant method is that, principally, the initial decay of polarization can be approximated as far and as well as one wishes by successive expansion. We note that for values  $\sigma \gg 1$  the Gaussian-type decay, given by (1.2), yields a good description for the behavior of the depolarization, since the details of the frequency modulation  $\omega(t)$  are effectively irrelevant.

#### IV. ASYMPTOTIC POLARIZATION DECAY IN $d=1$

While a description of the initial decay of the depolarization may profit from the fact that in the initial time region visits of only a restricted number of lattice sites are important, a theory appropriate for the asymptotic behavior must also take into consideration that principally an increasing number of lattice sites will be visited with increasing time. Let us consider the number  $s$  of distinct

sites visited by a discrete random walk with  $n$  steps (in  $d=1$  the "span" of the walk); for fixed  $n$  this quantity can take on values between  $s=2$  and  $s=n$ . The two extreme cases illustrate the totally different behavior of "contracted" (small  $s$ ) and "extended" (large  $s$ ) walks: Their individual contributions are given by a Gaussian-type decay  $\exp(-\sigma^2 n^2/4)$  for  $s=2$  (if  $n$  even), and the simple exponential  $\exp(-\sigma^2 n/2)$  for  $s=n$ . The latter corresponds to the depolarization in a discrete version of the strong-collision model, since with each new step a new site or frequency is visited. For arbitrary  $s$  the contributions of the different walks cannot easily be determined, but an approximative expression is obtained from the assumption<sup>11</sup> that for fixed  $n$  each of the  $s$  sites is equally often visited, namely  $n/s$  times. Equation (2.7) then reduces to

$$P_n \approx \langle C_n(s) \rangle, \quad C_n(s) \equiv \exp \left[ -\frac{\sigma^2 n^2}{2s} \right] \quad (4.1)$$

where now the average extends over all possible values of  $s$ . The further, crude approximation that each  $n$ -step walk visits exactly  $\langle s \rangle$  sites, where  $\langle s \rangle \sim (8n/\pi)^{1/2}$  for  $n \rightarrow \infty$  in  $d=1$ ,<sup>22</sup> leads to the result

$$P_n \sim \exp \left[ -\frac{\sigma^2}{2} \left[ \frac{\pi}{8} \right]^{1/2} n^{3/2} \right]. \quad (4.2)$$

This result is analogous to the decay given by (2.10). Similarly, for  $d=2$ , with  $\langle s \rangle \sim \text{const} \times n \ln(n)$ ,<sup>23</sup> a decay analogous to (2.11) is obtained; again for  $d \geq 3$ , where  $\langle s \rangle \sim \text{const} \times n$ ,<sup>23</sup> a simple exponential decay results, analogous to (2.12). A better evaluation of the approximate expression (4.1) must take the actual distribution  $W_n(s)$  of the spans of the random walk into account. In  $d=1$ ,  $W_n(s)$  is exactly known.<sup>24</sup> Since we are interested in the asymptotic depolarization decay, we directly use an asymptotic expression for the span distribution. Weiss and Rubin have given two expressions in the form of infinite series, which differ in their convergence on the left (small  $s$ ) or right wing (large  $s$ ) of the distribution.<sup>24</sup> Since extended walks provide the largest individual contributions  $C_n$ , we use the form which converges rapidly in the region beyond the maximum:

$$W_n(s) = \frac{8}{\sqrt{2\pi n}} \sum_{j=1}^{\infty} (-1)^{j+1} j^2 \exp(-\frac{1}{2} j^2 s^2/n). \quad (4.3)$$

In the average in (4.1) and for large  $n$  it suffices to take the first term in (4.3) only, since the other terms all lead to exponentially small contributions. The summation of the product  $f_n(s) \equiv W_n(s)C_n(s)$  can be performed by saddle-point integration. The maximum or the saddle-point of  $f_n$ —see Fig. 2—is located at  $s_m \simeq \sigma^{2/3} 2^{-1/3} n$ , with a relative width around the maximum of the order of  $n^{-1/2}$ . Since  $f_n(s)$  is not defined for arguments  $s > n$ , the derivation is restricted to values of  $\sigma$  where  $s_m$  is smaller than  $n$ , i.e., roughly for values of  $\sigma$  not exceeding unity. The result is

$$P_n \sim \frac{8}{\sqrt{3}} \exp(-3 \times 2^{-5/3} \sigma^{4/3} n). \quad (4.4)$$

Thus a simple exponential decay of the depolarization is

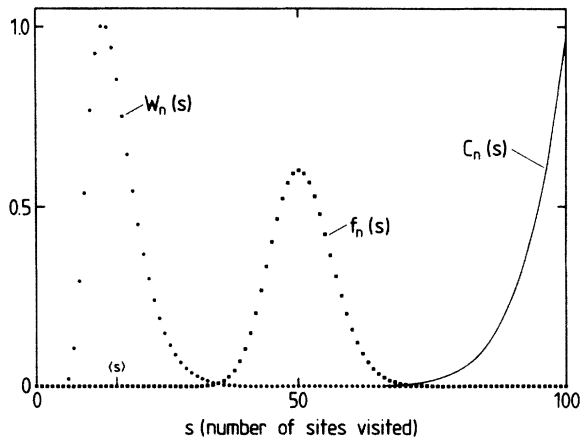


FIG. 2. Distribution  $W_n(s)$  of the number  $s$  of distinct sites visited in a one-dimensional  $n$ -step random walk,  $C_n(s) = \exp(-\sigma^2 n^2/2s)$ , and  $f_n(s) = W_n(s)C_n(s)$  for  $n=100$  and  $\sigma=0.5$ . (The maxima of  $W_n$  and  $C_n$  are normalized to 1, and the maximum of  $f_n$  to 0.6.)

obtained. We note that the approximation (4.1) yields an upper bound of the polarization decay, since for each individual walk the inequality  $\sum_r h_r^2 \geq n^2/s$  holds.

The comparison with the results of numerical simulation (see Sec. V) shows that (4.4) is indeed a good description of the actual polarization decay in  $d=1$ . Nevertheless, the role of (4.4) as a simple exponential upper bound for  $P_n$  is not satisfactory since it cannot exclude the possibility of a polarization decay with a power of  $n$  higher than  $n^1$  in the exponent. It is useful to construct also an appropriate lower bound, from which relevant conclusions can be drawn. Consider now the class of all random walks which end at a fixed site  $r$ . Its minimal individual contribution to  $P_n$  is given by a walk with maximum value of  $\sum_r h_r^2$ . Such a walk is characterized by two properties ("maximum degree of contraction"); see Fig. 3: The number  $s$  of distinct sites visited is as small as possible, i.e.,  $s=r$ , and a minimum number of these  $r$  sites, which corresponds to two neighboring sites, is visited as frequently as possible. If for simplicity only the case of even  $n=2m$  is considered, one easily obtains for each possible site  $r=2k$  ( $|k|=0,1,2,\dots,m$ )

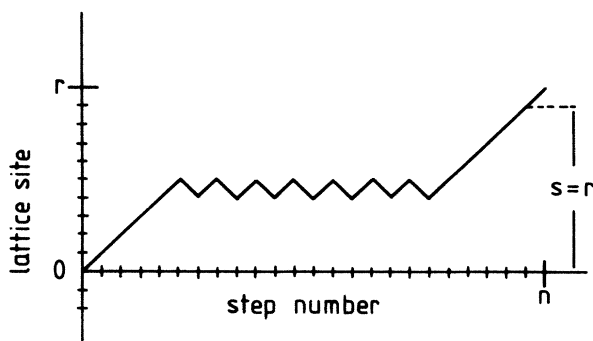


FIG. 3.  $n$ -step walk which ends at site  $r$  with "maximum degree of contraction" (see text).

$$\sum_{r'} h_r^2 \leq L(k) \equiv 2|k| + 1 - 2(m - |k| + 1)^2,$$

where the equality holds for all  $k \neq 0$ . Obviously,

$$P_n > \tilde{P}_n \equiv \sum_{k=-m}^m Q_{2m}(2k) \exp\left[-\frac{\sigma^2}{2} L(k)\right], \quad (4.5)$$

where  $Q_{2m}(2k)$  denotes the well-known probability that a walk with  $n = 2m$  steps ends at site  $r = 2k$ . For large  $n$  the distinction between even and odd  $n$  becomes irrelevant, and starting from (4.5)  $\tilde{P}_n$  can be evaluated using the asymptotic probability density  $Q_{2m}(2k) \sim (m\pi)^{-1/2} \exp(-k^2/m)$ . The result of this straightforward evaluation is given by (for values of  $\sigma$  not exceeding unity)

$$\tilde{P}_n \sim \exp\left[-\frac{1}{2}(\sigma^2 + 1)n\right], \quad (4.6)$$

i.e., again by a decay with the power  $n^1$  in the exponent. In summary, we have shown the asymptotically  $P_n$  cannot decay faster than given by (4.6); hence, a simple exponential decay of  $P_n$  follows.

## V. NUMERICAL RESULTS IN $d = 1$

Before we turn to the verification of the results of Sec. IV by numerical simulations, we give a brief description of how these simulations were performed:

In a first method, corresponding to the simulation of (2.6)—or the analogous formula in the continuous case—a linear chain of typically  $5 \times 10^5$  sites with random frequency configuration  $\{\omega_r\}$  is generated, on which many random walks (typically  $5 \times 10^4$ ) with random starting positions are performed. The accumulated spin-rotation phase  $\Phi$  of each walker is observed. After repeating this scheme some  $10^1$  times for different frequency configurations, the depolarization is computed as the mean value  $\langle \cos \Phi \rangle$ , averaged over all generated random walks. This method is time consuming and not very accurate, since when  $P_n$  is of the order of  $10^{-2}$ – $10^{-3}$ , where typically the asymptotic regime is not yet reached, the statistical scatter of the data becomes too large.

The second method uses Eq. (2.7), which is now estimated by the simulation. Since the average over the rotation frequencies is exact, only an average over different random walks has to be performed. With this method the decay of polarization could be followed up to  $10^{-8}$  (and in some instances far beyond) for a wide range of parameters  $\sigma$  between 2 and  $10^{-3}$ . The number of generated walks has been of the order of  $10^4$ – $10^6$ . We note that for values  $\sigma \geq 2$  the decay of polarization becomes so fast that no relevant information is obtained by computing the discrete depolarization  $P_n$ . Although this decay can, in principle, be followed in the continuous case, this range of the parameter  $\sigma$  is not of special interest, since the polarization becomes extremely small long before asymptotic properties of the random walk play their specific role.

Simulations of the continuous model, which have also been performed by both methods, have some specific disadvantages compared to the discrete case, mainly the necessity of the additional simulation of the waiting-time distribution. Hence, these simulations were not per-

formed to such an extent as in the discrete case, which provides the simpler possibility of investigating a wide range of parameters. The relationship of the two models will be discussed below.

The simulations show that for all values of  $\sigma$  the asymptotic decay indeed approaches a simple exponential; in Fig. 4 the results for the discrete as well as the continuous depolarization are given for two typical values of  $\sigma$ ; for comparison, some data points of the first method are included.

Since (4.4) only describes the asymptotically leading behavior of the depolarization, based on the approximation (4.1), the appropriate quantity for comparison with the numerical simulations is the decay constant

$$\lambda = 3 \times 2^{-5/3} \sigma^{4/3} \quad (5.1)$$

of the exponential (4.4). Figure 5 shows the dependence of the decay constant on the width of the frequency distribution. The slope of the straight line through the data points is  $1.30 \pm 0.02$ , which is close to the value  $\frac{4}{3}$  in (5.1). The observed decay constants are larger than (5.1)—in agreement with the fact that (4.4) is a lower bound for  $P_n$ . The absolute deviation can be described by a numerical factor which is approximately given by  $\approx 1.3$ .

The question arises as to which extent the result (4.4) can be used to describe the polarization decay in the continuous model. We now compare the depolarization of both models. It is tempting to assume that  $P_n$ , for large step numbers  $n$ , can be translated into  $P(t)$  by identifying  $n$  with  $t/\tau$ . However, the results of the simulations—see Figs. 4 and 5—show that this identification cannot be made; the results of the continuous model and of the discrete model are different, in that  $P(t)$  has a larger decay constant. Such a difference already appears in the strong-collision model, where a different frequency is taken at each step. We have mentioned (cf. Sec. IV) that

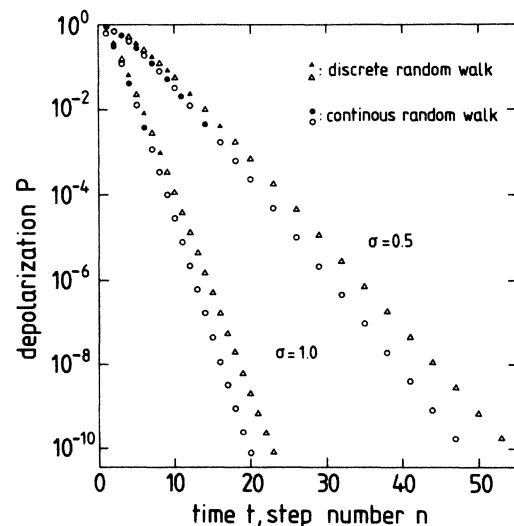


FIG. 4. Depolarization of discrete and continuous random walk ( $d = 1$ ) for two typical values of  $\sigma$ . Solid symbols, results of simulations performed by the first method described in text; open symbols, second method.

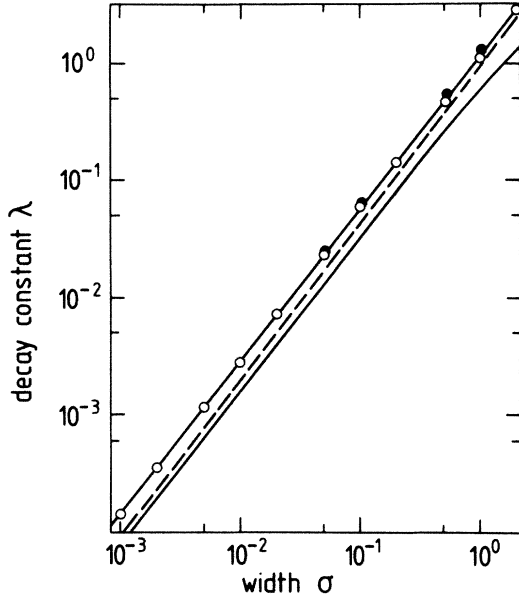


FIG. 5. Dependence of the decay constant  $\lambda$  in  $P_n \sim \exp(-\lambda n)$  on the width  $\sigma$ . Results of the simulations represented by open (discrete model) and solid circles (continuous model). Dashed line, Eq. (5.1); lower curve, result of effective-medium theory (see Sec. VII).

$P_n = \exp[-(\sigma\tau)^2 n/2]$ , while the continuous model yields  $P(t) \approx \exp(-\sigma\tau t)$ . This difference can be understood by the following arguments.

For the Poisson jump process in the continuous model, the probability that  $n$  steps are performed in the time interval  $[0, t]$  is

$$\psi_t(n) \equiv \frac{1}{n!} (t/\tau)^n \exp(-t/\tau);$$

it has a sharp maximum at  $n = t/\tau$  for  $t \gg \tau$  with a relative width  $\Delta n/n = (t/\tau)^{-1/2}$ . Thus in the asymptotic limit only those walks which perform  $n = t/\tau$  jumps contribute. For the depolarization the accumulated phase of spin rotation is important. This quantity depends sensitively on the actual waiting times between successive transitions, and thus the contribution of all walks with  $n$  jumps in  $[0, t]$ , which will be denoted  $P(t|n)$ , cannot simply be replaced by  $P_n$  with  $n = t/\tau$ . We decompose the time  $t_j$  of rotation with the  $j$ th frequency,  $t_j = \tau + \Delta_j$ , where  $\tau$  is the mean value and  $\Delta_j$  the deviation ( $j = 1, 2, \dots, n$ ). With

$$P(t|n) = \left\langle \exp \left[ -\frac{\sigma^2}{2} \sum_j t_j^2 \right] \right\rangle_{\{t_j\}}$$

and

$$\sum_j \Delta_j = 0,$$

we obtain

$$P(t) \approx P(t|n) = \left\langle \exp \left[ -\frac{\sigma^2}{2} \sum_j \Delta_j^2 \right] \right\rangle_{\{\Delta_j\}} P_n < P_n. \quad (5.2)$$

If we assume that for large  $n$  correlations between the different  $\Delta_j$  become more and more irrelevant, the average in (5.2) can be replaced by a product of  $n$  identical averages  $\langle \exp[-(\sigma^2/2)\Delta^2] \rangle$ . This average can be performed exactly, leading to a rather complicated expression; for  $\sigma\tau \ll 1$  the result is

$$\exp \left[ -\frac{\sigma^2}{2} \langle \Delta^2 \rangle \right] = \exp \left[ -\frac{\sigma^2}{2} \tau^2 \right].$$

Thus we obtain, from (5.2), replacing  $n$  by  $t/\tau$ ,

$$P(t) \approx \exp(-\sigma^2 \tau t) \quad (\sigma\tau \ll 1), \quad (5.3)$$

which is the result of the strong-collision model.

The argumentation cannot directly be applied to the random-walk problem where the actual numbers of visits on each site play a crucial role, but if we again assume that all visited sites are equally often visited, we obtain, by analogous derivations,

$$P(t) \approx P_n \exp[-(\sigma\tau)^2 n/2] \quad (\sigma\tau \ll 1). \quad (5.4)$$

The resulting decay constant is  $\lambda + \sigma^2/2$  ( $\tau \equiv 1$ ), where  $\lambda$  is given by (5.1), i.e., proportional to

$$\sigma^{4/3} (1 + \text{const} \times \sigma^{2/3}). \quad (5.5)$$

This makes it plausible that the decay constants of the continuous model approach the values of the discrete model from above when  $\sigma$  decreases. Thus for sufficiently small  $\sigma$  we also expect for the continuous model the dependence

$$\lambda = \text{const} \times \sigma^\beta \quad (5.6)$$

with  $\beta \approx \frac{4}{3}$ . This exponent  $\beta$  should be contrasted to the value  $\beta = 2$  of the standard Gaussian treatment or the strong-collision model. If we describe the polarization decay on the real time scale, the decay constant corresponding to (5.6) is given by  $\lambda = \text{const} \times \sigma(\sigma\tau)^{\beta-1}$ , i.e., here,  $\tau$  enters with the exponent  $\approx \frac{1}{3}$ . The principal experimental relevance is the following: If one knows the temperature dependence of  $\tau$ , e.g., from independent measurements of the diffusion constant, the possibility arises of measuring the dependence of the decay constant on  $\sigma$ , i.e., to estimate the value of  $\beta$ . The dimensionless combination  $(\sigma\tau)^{1/3}$  can also be identified in the result of the Gaussian treatment (cf. Ref. 9) and also appears in the theory of Reiter and Boucher.<sup>12</sup>

## VI. DIFFERENT FREQUENCY DISTRIBUTIONS

Up to now we have assumed that the local rotation frequencies  $\omega_r$  are taken from the Gaussian distribution (2.1). It was pointed out by Van Vleck<sup>25</sup> that this approximation gives a good description of the actual distribution of local magnetic fields in a crystal. Since we are discussing rather detailed behavior of the polarization (e.g., decay down to  $10^{-9}$ ), it is a relevant question whether this behavior depends on the explicit assumption of a Gaussian distribution. We hence consider other types of distributions in this section.

Let us first briefly consider the example of the Cauchy



or Lorentz distribution  $g(\omega) = (\delta/\pi)(\delta^2 + \omega^2)^{-1}$  ( $\delta > 0$ ). It is known<sup>26</sup> that in this case the decay of polarization is totally independent of how many different frequencies are taken on—and how they are taken on; the decay is always given by  $P(t) = \exp(-\delta t)$ . (In the context of spin-resonance phenomena this effect has been called “total absence of motional narrowing.”) Hence, the question of how the random-walk process influences the polarization decay is completely irrelevant for the Cauchy distribution.

In the following we shall consider three examples of distributions, which all have mean  $\langle \omega \rangle = 0$  and variance  $\sigma^2$ , but differ according to their (finite) higher moments  $\langle \omega^k \rangle$ . We first discuss the symmetric exponential distribution

$$g(\omega) = \frac{1}{\sqrt{2}\sigma} \exp(-\sqrt{2}|\omega|/\sigma). \quad (6.1)$$

We will investigate the polarization decay due to this distribution in more detail.<sup>27</sup> The two other examples are the rectangular (constant) and the dichotomic distribution, and these will be discussed later.

#### A. Symmetric exponential distribution

The polarization decay of an ensemble of spins with the distribution (6.1) in the static case is easily derived by calculating the characteristic function  $\hat{g}(x)$ , cf. (2.2). We have

$$\hat{g}(t) = (1 + \frac{1}{2}\sigma^2 t^2)^{-1}. \quad (6.2)$$

The asymptotic decay is now algebraic, i.e., much slower than in the Gaussian case. The initial decay of  $P(t)$  for the exponential distribution is given by  $1 - \sigma^2 t^2/2$ , valid for  $t \ll \sigma^{-1}$ . The same initial decay results from the Gaussian frequency distribution of width  $\sigma$ .

It can be shown that an arbitrary frequency distribution  $g(\omega)$  with mean  $\langle \omega \rangle = 0$  and variance  $\sigma^2$  leads to an initial polarization decay  $P(t) \approx 1 - \sigma^2 t^2/2$  for  $t \ll \sigma^{-1}$ . Under these assumptions the cumulant expansion of the characteristic function  $\hat{g}(x)$  can be written as

$$\begin{aligned} \ln \hat{g}(x) &= \sum_{m=1}^{\infty} \frac{(-1)^m}{(2m)!} K_{2m}(\sigma x)^{2m} \\ &= -\frac{\sigma^2}{2} x^2 + \frac{1}{4!} K_4(\sigma x)^4 - + \dots, \end{aligned} \quad (6.3)$$

where  $K_{2m} \equiv \langle \omega^{2m} \rangle_c / \sigma^{2m}$ . It can be used for an approximation of  $\hat{g}(x)$  for  $\sigma x \ll 1$ , and  $P(t) \equiv \hat{g}(t)$  according to (2.2). As (6.2) exemplifies, the asymptotic polarization decay depends on the precise form of the frequency distribution in the static case. The question is how this fact influences the polarization decay in the case of random walk on the spin particles. For simplicity, we shall restrict our considerations to discrete random walk, where  $P_n = \langle \prod_r \hat{g}(h_r) \rangle$ ; see (2.7).

We will discuss the properties of  $P_n$  in the frame of the approximation we introduced in Sec. IV, i.e.,  $h_r$  is replaced by  $n/s$  for all  $s$  different visited sites. We thus start with (4.1),

$$P_n \approx \sum_{s=2}^n W_n(s) C_n(s), \quad (4.1')$$

where  $W_n$  is the span distribution, and  $C_n(s)$  the contribution of a walk with span  $s$ , in this approximation. For an arbitrary distribution,

$$C_n(s) = [\hat{g}(n/s)]^s = \exp[-s \ln \hat{g}(n/s)]. \quad (6.4)$$

For the Gauss distribution,  $C_n(s)$  is given by  $\exp[-\sigma^2 n^2/(2s)]$ , a monotonically increasing function of  $s$ . This property is reflected in the tendency that increasing “mobility” leads to a slower polarization decay. For arbitrary  $g$  we can use (6.3) to obtain an approximation for  $\sigma n/s \ll 1$ ,

$$C_n(s) \approx \exp \left[ -\frac{1}{2} \sigma^2 \frac{n^2}{s} + \frac{1}{4!} K_4 \sigma^4 \frac{n^4}{s^3} + s O((\sigma n/s)^6) \right]. \quad (6.5)$$

Only for  $\sigma \ll 1$  is there a certain range of  $s$  values ( $\sigma n \ll s < n$ ) where  $C_n(s)$  behaves similar to the Gaussian case. For  $\sigma \gg 1$ , where  $s \ll \sigma n$  for all  $s$ , the behavior of  $C_n$  is determined by  $\hat{g}(x)$  for  $x \gg 1$ . Let us explicitly consider the case of the exponential distribution (6.1); with (6.2),

$$C_n(s) = \exp \left[ -s \ln \left[ 1 + \frac{1}{2} \sigma^2 \frac{n^2}{s^2} \right] \right]. \quad (6.6)$$

We first note that  $C_n$  has a (single) minimum at  $s_0 = k\sigma n$ ; the constant  $k$  can be calculated numerically and is given by  $k \approx 0.357$ .  $C_n(s)$  increases for  $s < s_0$  with decreasing  $s$  and for  $s > s_0$  with increasing  $s$ . The question is which part of  $C_n(s)$  is important for the behavior of  $P(t)$ ; this requires a detailed discussion of separate cases.

#### 1. $\sigma \gg 1$

Here (or more precisely for all  $\sigma \geq k^{-1}$ , where  $s_0 \geq n$ ),  $C_n$  is monotonically decreasing over the entire range of possible  $s$  values. Contrary to the Gaussian case, contracted walks now yield the most important individual contributions. Since  $C_n$  is also convex on  $\{2, 3, \dots, n\}$ , a lower bound for  $P_n$  can be obtained by applying Jensen's inequality:  $P_n = \langle C_n(s) \rangle \geq C_n(\langle s \rangle)$ . With  $\langle s \rangle \sim (8n/\pi)^{1/2}$  ( $n \gg 1$ ) in  $d = 1$ , this inequality reads

$$\begin{aligned} P_n &\geq \exp \left[ - \left[ \frac{8n}{\pi} \right]^{1/2} \ln \left[ 1 + \frac{\pi \sigma^2}{16} n \right] \right] \\ &\approx \exp[-\text{const} \times n^{1/2} \ln(\sigma^2 n)], \quad n \gg 1. \end{aligned} \quad (6.7)$$

We thus find the interesting result that the symmetric exponential distribution leads to an asymptotic polarization decay which is slower than simple-exponential. To investigate the actual decay of  $P_n$  in our approximation for  $n \gg 1$ , we take advantage of the fact that in leading order this decay depends on the position  $s_m$  of the maximum of the product  $f_n = C_n W_n$ . Since, here, contracted walks are most important, we use a form for  $W_n(s)$  which is especially suited for a description of the span distribution for  $s \leq \langle s \rangle$ . It suffices to take the first term of the sum in Eq. (23) of Ref. 24,

$$W_n(s) \sim \frac{8n}{s^3} \left[ \frac{\pi^2 n}{s^2} - 1 \right] \exp \left[ -\frac{\pi^2 n}{2s^2} \right]. \quad (6.8)$$

To determine the maximum of  $f_n$ , we formally write  $f_n(s) = \exp[-\varphi(s)]$ ;  $\varphi'(s_m) = 0$  yields  $s_m$ . Only considering contributions which are relevant for  $n \rightarrow \infty$ , we obtain

$$\varphi(s) \approx \frac{\pi^2 n}{2s^2} + s \ln \left[ \frac{\sigma^2 n^2}{2s^2} \right], \quad (6.9)$$

$$\frac{\pi^2}{s_m^3} n \approx \ln \left[ \frac{\sigma^2 n^2}{2s_m^2} \right].$$

Saddle-point integration yields  $P_n \sim f_n(s_m) = \exp[-\varphi(s_m)]$  to leading order in  $n$ . Introducing  $x \equiv n/s_m^2$ ,  $\varphi(s_m)$  can be written as  $(3\pi^2/2)x$ , and  $x$  is determined by

$$\pi^2 x^{3/2} = n^{1/2} \ln \left( \frac{1}{2} \sigma^2 n x \right). \quad (6.10)$$

With the ansatz  $x = \pi^{-4/3} n^{1/3} h(y)$ , where  $y \equiv \sigma^{3/2} n$ , (6.10) reduces to

$$h^{3/2}(y) = \ln \left[ \frac{1}{2} y^{4/3} h(y) \right], \quad (6.11)$$

which yields the leading behavior of  $h$ :

$$h(y) \sim \left( \frac{4}{3} \right)^{2/3} \ln^{2/3}(y). \quad (6.12)$$

We thus obtain, for  $n \rightarrow \infty$ ,

$$P_n \sim \exp \left[ - (6\pi^2)^{1/3} n^{1/3} \ln^{2/3}(\sigma^{3/2} n) \right], \quad (6.13)$$

i.e., an extreme slow decay for large step numbers  $n$ , compared to the asymptotic decay  $\exp(-\lambda n)$ . However, the polarization  $P_n$  is already extremely small for step numbers  $n$  where the approximation leading to (6.13) is justified. Hence, we hardly expect that this asymptotic behavior can be verified by numerical simulation. Nevertheless, the qualitative tendency should be visible.

## 2. $\sigma \ll 1$

For  $\sigma \ll 1$  the situation is not as simple as for  $\sigma \gg 1$ , since now the minimum  $s_0$  of  $C_n$  is somewhere in the range of possible span values  $s = 2, 3, \dots, n$ , so that, in principle, both wings of  $C_n$  can become important. We can distinguish two different cases, which depend on the position of  $s_0$  relative to the maximum of  $W_n$ , which is at about  $\langle s \rangle \sim (8n/\pi)^{1/2}$ . Let  $n^*$  be the step number where both coincide, i.e., where  $s_0 \approx \langle s \rangle$ :

$$n^* = \frac{8}{\pi k^2} \sigma^{-2} \approx 20 \sigma^{-2}. \quad (6.14)$$

(i) For  $n \ll n^*$  (and, consequently,  $s_0 \ll \langle s \rangle$ ),  $f_n$  has a pronounced maximum beyond the maximum of  $W_n$ . This situation is shown in Fig. 6(a) for the parameters  $\sigma = 0.2$ ,  $n = 200$  ( $n^* = 500$ ). To determine the value of  $f_n$  at the maximum, i.e.,  $f_n(s_m)$ , we use the approximate form

$$W_n(s) \sim \frac{8}{\sqrt{2\pi n}} \exp(-s^2/n),$$

in analogy to our analysis in Sec. IV. If, in addition, the  $s$

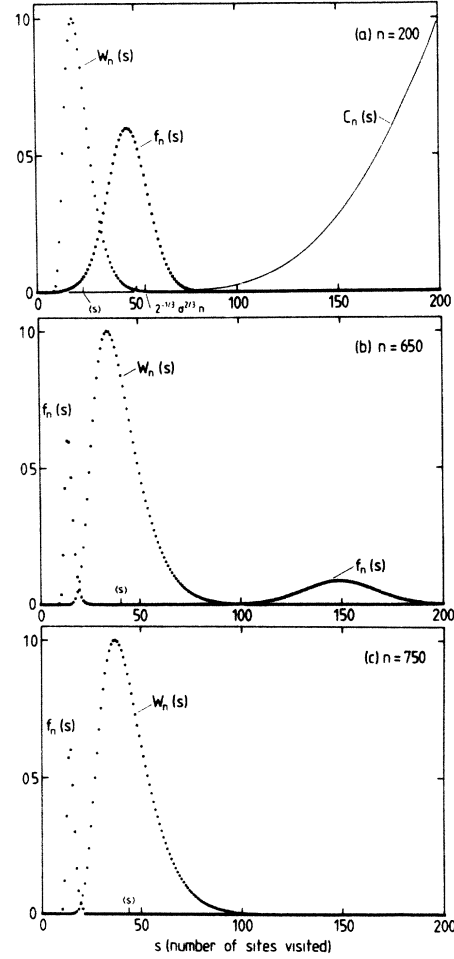


FIG. 6. Distribution  $W_n(s)$  of the number  $s$  of different sites visited in an  $n$ -step random walk ( $d=1$ ),  $C_n(s)$  [given by Eq. (6.6)], which is only shown in (a), and  $f_n(s) = W_n(s)C_n(s)$ , for the exponential frequency distribution with width  $\sigma=0.2$ . In (a)–(c) different step numbers are considered. See Fig. 2 for normalization.

values of interest ( $s$  larger than  $\langle s \rangle$ ) obey the condition  $\sigma n/s \ll 1$ , the calculation of  $f_n(s_m)$  can be simplified by using the expansion (6.5). (Note that this last assumption is at least valid, if already  $\sigma n/\langle s \rangle \ll 1$ .) With  $C_n(s)$ , to lowest order, approximated by  $\exp(-\sigma^2 n^2/2s)$ , we obviously obtain the same results as obtained in Sec. IV for the Gaussian frequency distribution, cf. (4.4),

$$s_m \approx 2^{-1/3} \sigma^{2/3} n, \quad (6.15)$$

$$P_n \sim \exp(-\lambda n), \quad \lambda = 3 \times 2^{-5/3} \sigma^{4/3}.$$

The influence of the next term in the expansion (6.5) for  $C_n(s)$  can be treated approximately, i.e., we can calculate the corrections to (6.15) to lowest order in  $\sigma$ . The result for the position of the maximum is

$$s'_m \approx s_m \left[ 1 - \frac{1}{4!} K_4 2^{5/3} \sigma^{2/3} \right]. \quad (6.16)$$

Since for our distribution (6.1),  $K_4 = \langle \omega^4 \rangle_c / \sigma^4 = 3 > 0$ , at

least (6.16) yields a qualitatively correct picture; e.g., see Fig. 6(a). We obtain the decay constant

$$\lambda' = \lambda \left[ 1 - \frac{K_4}{9} 2^{-1/3} \sigma^{2/3} \right], \quad (6.17)$$

i.e., in the frame of our approximation, the decay constant, which describes the exponential polarization decay in a certain range of step numbers  $n \ll n^*$ , is expected to be somewhat smaller than the decay constant in the case of the Gaussian distribution.

(ii) Let us now consider the case of larger  $n$ . When  $n \geq n^*$ , a second maximum of  $f_n$ , at the left-hand side of the maximum of  $W_n$ , becomes more and more pronounced, while the first maximum slowly vanishes, see Fig. 6(b). This signals that now the influence of contracted walks begins to dominate the decay of  $P_n$ . For  $n \gg n^*$ , i.e., when the minimum of  $C_n$  is far beyond the maximum of  $W_n$ , only these contracted walks contribute, see Fig. 6(c), and we have the situation we already discussed for  $\sigma \gg 1$ . Thus, also for small values of  $\sigma$  we expect an asymptotic polarization decay proportional to  $\exp[-\text{const} \times n^{1/3} \ln^{2/3}(\sigma^{3/2} n)]$ , see (6.13).

We point out that the step number  $n^*$  is a rough estimate of the actual step number, where [in the approximation (4.1')] the crossover to the non-simple-exponential decay appears. Nevertheless, we can make a rough guess of the magnitude of  $P_n$  and its dependence on  $\sigma$  at this crossover point. From (6.15) combined with (6.14) we deduce the estimate

$$P_{n^*} \approx \exp(-3 \times 2^{1/3} \pi^{-1} k^{-2} \sigma^{-2/3}). \quad (6.18)$$

Thus the crossover can be shifted to unmeasurably small values of  $P_n$  by reducing  $\sigma$ .

### B. Other distributions

We now briefly consider the two other examples of frequency distributions. The rectangular distribution is defined by

$$g(\omega) = \begin{cases} \frac{1}{2}(\sqrt{3}\sigma)^{-1}, & -\sqrt{3}\sigma \leq \omega \leq \sqrt{3}\sigma \\ 0, & \text{otherwise.} \end{cases} \quad (6.19)$$

Its characteristic function is given by

$$\hat{g}(x) = \frac{\sin(\sqrt{3}\sigma x)}{\sqrt{3}\sigma x}. \quad (6.20)$$

The dichotomic distribution describes the case where the local rotation frequency can only take the values  $\omega = \pm\sigma$ :

$$g(\omega) = \frac{1}{2}[\delta(\omega + \sigma) + \delta(\omega - \sigma)], \quad (6.21)$$

$$\hat{g}(x) = \cos(\sigma x). \quad (6.22)$$

It is a characteristic property of both distributions that they lead to oscillations of  $P(t) = \hat{g}(t)$ , the depolarization in the static case. We note that the cumulant expansion (6.3) of  $\hat{g}$  is well defined for arguments  $x$  with  $\hat{g}(x) > 0$ ; the range of convergence is given by  $|x| < x_c$ , with  $x_c = \pi(\sqrt{3}\sigma)^{-1}$  (rectangular) or  $x_c = \pi(2\sigma)^{-1}$  (dichotomic). Comparison with (6.2) further shows that the decay

of the amplitudes of  $\hat{g}(t)$  for  $t \rightarrow \infty$  is even slower than the decay in the case of the exponential distribution (6.1); for the dichotomic distribution (6.21) there is no decay of the amplitude with time at all.

It is important for the discussion of the polarization decay in the case of random walks that the individual contribution of a single walk with span  $s$ ,  $C_n(s) = [\hat{g}(n/s)]^s$ , may also take on negative and positive values. We distinguish two regimes: for  $s > s_c \equiv n x_c^{-1}$ , i.e., where the behavior of  $\hat{g}(x)$  for  $x < x_c$  determines the behavior of  $C_n$ , the function  $C_n$  is positive and monotonically increasing from  $C_n(s_c) = 0$  to  $C_n(n) = [\hat{g}(1)]^n$ . For  $s < s_c$  the behavior of  $C_n$  is mainly dominated by "oscillations," which—roughly speaking—are becoming more and more rapid when  $s$  decreases. Thus, again, two different situations, now distinguished by the relative position of  $s_c$  and  $\langle s \rangle$ , are relevant for the behavior of  $P_n$ . In analogy to the definition of the step number  $n^*$  in the preceding subsection, we introduce a step number  $n_c$  where  $s_c \approx \langle s \rangle$ ;  $n_c$  is given by  $(8\pi/3)\sigma^{-2}$  (rectangular) or  $2\pi\sigma^{-2}$  (dichotomic). The situation where  $s_c \gg \langle s \rangle$ , i.e., where  $n \gg n_c$ , is relevant for the asymptotic decay of  $P_n$  for arbitrary  $\sigma$  (and especially for the polarization decay for  $\sigma \gg 1$ ). Since here the oscillations of  $C_n$  play an essential role, simple saddle-point arguments cannot be applied.  $P_n$  also takes on negative as well as positive values. At least we may conclude that the decay of the amplitudes will be slower than the asymptotic decay obtained for the exponential distribution.

For  $\sigma \ll 1$  and  $n \ll n_c$  ( $s_c \ll \langle s \rangle$ ) the situation is similar to that of  $\sigma \ll 1$  and  $n \ll n^*$  discussed above; actually, the same arguments can be applied. If, in addition,  $\sigma n / \langle s \rangle \ll 1$ , the expansion (6.5) for  $C_n$  can again be used. We directly refer to the result (6.17), in which—at least to lowest order in  $\sigma$ —differences between the different kinds of distributions appear in the decay constant of the resulting exponential decay  $\exp(-\lambda'n)$ :  $\lambda' = \lambda(1 - \text{const} \times K_4 \sigma^{2/3})$ . For the rectangular distribution (6.19),  $K_4 = -\frac{6}{5}$ ; for the dichotomic distribution (6.21),  $K_4 = -2$ . Thus, contrary to the case of the exponential distribution (6.1), we now expect decay constants which are somewhat larger than the decay constant  $\lambda$  resulting from the Gaussian frequency distribution.

### C. Comparison with simulations

Our preceding considerations are all based on the approximation (4.1'). In the case of the Gaussian distribution this approximation leads to a qualitatively (but not quantitatively) correct description of the actual polarization decay. The question remains of whether the same is true for the three other distributions. We have simulated the depolarization caused by discrete random walks, for the three distributions with different values of  $\sigma$ . It is not our aim to make an equally extensive and detailed comparison as in Sec. V for the Gaussian distribution; we are mainly interested in a qualitative verification of our predictions.

In Fig. 7(a) the depolarization  $P_n$  for the exponential distribution and  $\sigma = 2.0$  is shown in comparison to  $P_n$  for the Gaussian one. It is clearly seen that, already for small step numbers  $n$ , the decay is slower than simple-

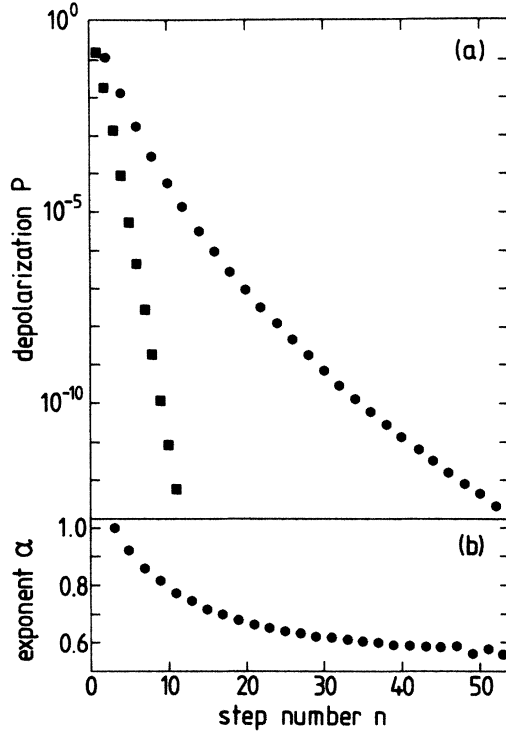


FIG. 7. (a) Depolarization of spin rotation for random walks on a linear chain with rotation frequencies taken from the exponential distribution (6.1), circles (simulation), compared to the results of the simulation for the Gaussian frequency distribution (2.1), squares; the width  $\sigma$  was 2.0. (b) Apparent exponent  $\alpha$  of decay law  $\ln P_n \propto n^\alpha$ .

exponential; also see Fig. 7(b), which shows the dependence of the apparent exponent  $\alpha$ ,  $P_n \propto \exp(-\text{const} \times n^\alpha)$ , on  $n$ . The expected asymptotic decay, given by (6.13), corresponds to an asymptotic value  $\alpha = \frac{1}{3}$ . Figure 7(b) exemplarily shows the difficulty to see—and thus to verify—this behavior by simulations. In Fig. 8 the analogous results are plotted for  $\sigma = 0.5$ . For this value of  $\sigma$  there is only a small range of step numbers where the polarization decay can be approximated by a simple exponential (at about  $n = 45$ ,  $\alpha(n) \approx 1.0$ ; note that  $n^* = 80$ ). The result for  $\sigma = 0.05$  (see Fig. 9) shows a nearly exponential decay over many decades; as expected, the nonexponential decay is shifted to very small values of  $P_n$ . The decay constant  $\lambda$  in the regime of exponential decay has been determined for several values of  $\sigma$ . Figure 10 shows that these values are smaller than the decay constants obtained for the Gaussian distribution, in qualitative agreement with (6.17) with positive  $K_4$ ; the difference decreases with decreasing  $\sigma$ .

For the other two distributions, (6.19) and (6.21), the results of the simulations show qualitatively the behavior we expect from our previous considerations. For  $n$  larger than a crossover step number, which increases with decreasing  $\sigma$ , superimposed oscillations emerge in the polarization decay, such that finally  $P_n$  takes on negative as well as positive values; the amplitudes decay rather slowly

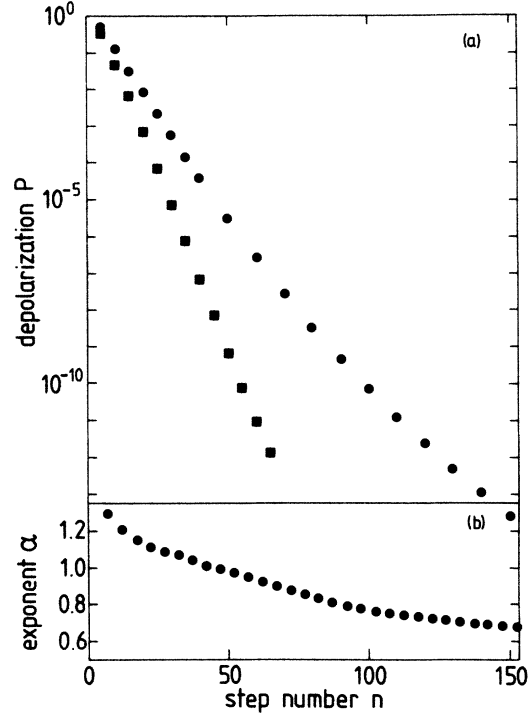


FIG. 8. See Fig. 7; here the width  $\sigma$  is 0.5.

compared to the polarization decay of the first example. For small  $\sigma$  the initial (“undisturbed”) decay can indeed be approximated by a simple exponential; see the data in Fig. 9. Again, in agreement with (6.17), this decay is faster in the case of the dichotomic distribution ( $K_4 = -2$ ) than in the case of the rectangular distribution ( $K_4 = -\frac{6}{5}$ ), and both are faster than the decay of the Gaussian distribution.

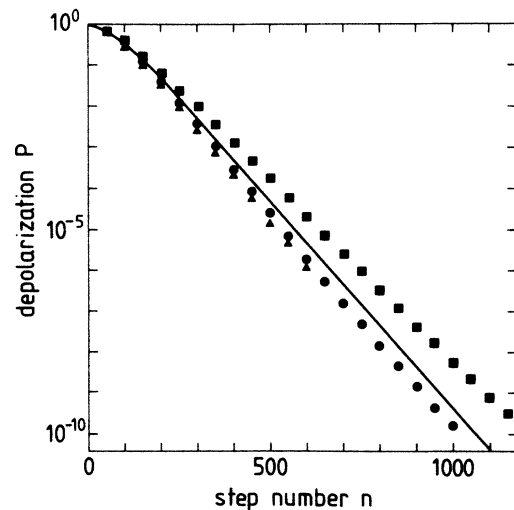


FIG. 9. Depolarization for random walks on a linear chain with exponential frequency distribution (6.1), squares, rectangular distribution (6.19), circles, and dichotomic distribution (6.21), triangles; the data are results of simulations. The results for the Gaussian distribution (2.1) are represented as a continuous curve. The width  $\sigma$  was 0.05.

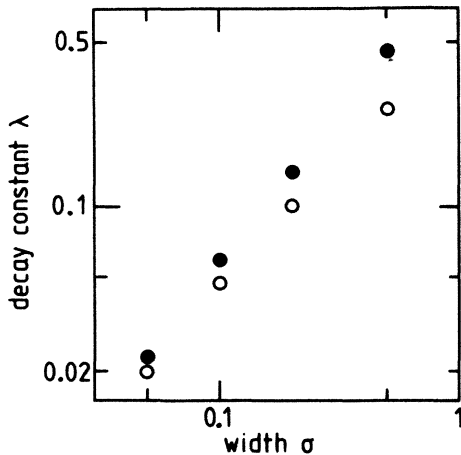


FIG. 10. Decay constant  $\lambda$  in the regime of simple-exponential polarization decay,  $P_n \approx \exp(-\lambda n)$ , for random walks on a linear chain with frequency distribution (6.1); the results of simulations for several values of the width  $\sigma$  are represented as open circles. The solid circles denote the Gaussian distribution.

The results of this section demonstrate that it is indeed a special property of the Gaussian frequency distribution that it leads to an *asymptotic* simple-exponential polarization decay. While for the other distributions a similar decay results over a certain range of step numbers (when  $\sigma \ll 1$ ), the asymptotic decay always depends on the actual form of the distribution, normally slower than exponential.

## VII. EFFECTIVE-MEDIUM THEORY FOR THE DECAY CONSTANT

Returning to the model with Gaussian frequency distribution, we now present an effective-medium theory for the decay constant  $\lambda$  of the exponential polarization decay,

$$P(t) \sim \exp(-\lambda t) \text{ as } t \rightarrow \infty, \quad (7.1)$$

in the continuous model. It is a special advantage of this theory that results can be obtained for higher dimensions quite as simply as for the one-dimensional case. We note that, since already in  $d=1$  the asymptotic decay is simple-exponential, the same will be true in all higher dimensions (for  $d \geq 3$  this property could already be deduced from Jensen's inequality; see Sec. II).

Here we want to restrict ourselves to the basic ideas of the effective-medium theory; more details can be found in the Appendix. The theory is introduced in the following way: The asymptotic polarization decay (7.1) can formally be described as resulting from a random-walk process on a homogeneous lattice—the “effective medium”—where on each site the local amplitudes experience an exponential damping of strength  $\lambda$ . To obtain an approximation for  $\lambda$  we replace the actual, completely disordered lattice with configuration  $\{\omega_r\}$  by a less disordered lattice, which is constructed in the way that only the site  $r=0$  retains its random local rotation frequency  $\omega_0$  while all oth-

er sites are considered to belong to the effective medium. The corresponding depolarization amplitudes can be calculated exactly and the condition that the decay of the total depolarization—after averaging over all possible values of  $\omega_0$ —is again given by (7.1) leads to a self-consistency equation which determinates  $\lambda$  [Eq. (A12) of the Appendix]. Since the properties of the random-walk process only enter via the probability of return to the origin, the decay constant can be calculated in principle for each type of lattice for which this probability (or its Laplace transform) is known.

Let us first consider the case  $d=1$ . The result for  $\lambda$ —also see Fig. 11—has been included in Fig. 5 and must be compared with the results of the simulations of the continuous model. For the parameters considered the decay constant in this approximation is about a factor 1.8–2.1 too small. This tendency is plausible, if one considers the basic properties of our approximation. Compared to the actual random-walk depolarization problem, where correlations due to returns to all lattice sites are responsible for the fast polarization decay, we have only included the effect of correlations due to returns to the origin. In the log-log plot the slope of  $\lambda(\sigma)$  yields the exponent  $\beta$ , appearing in  $\lambda(\sigma) \sim \sigma^\beta$ , which should be compared to  $\beta = \frac{4}{3}$ , e.g., for  $\sigma=0.1$  one obtains  $\beta=1.29$ , and for  $\sigma=0.01$ ,  $\beta=1.32$ . Actually, for  $\sigma \ll 1$  the exponent  $\beta$  finally seems to approach the value  $\frac{4}{3}$ .

The results for  $d=2$  (square lattice) and  $d=3$  (simple-cubic lattice) are shown in Fig. 11; for comparison we also included the result for  $d=1$ . First, as one expects, the decay constant decreases with increasing dimension. If in

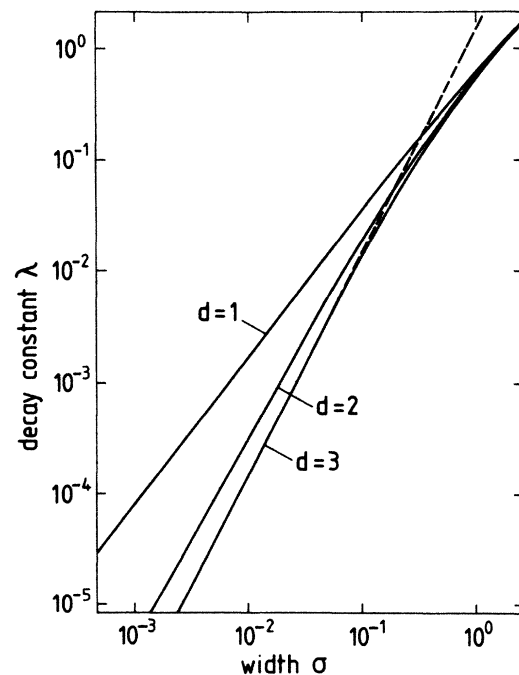


FIG. 11. Dependence of the decay constant  $\lambda$  in  $P(t) \sim \exp(-\lambda t)$  on the width  $\sigma$ . The solid curves represent the results of the effective-medium theory, i.e., the solutions of Eq. (A12), for  $d=1,2,3$ . The dashed line represents the result of Gaussian theory for  $d=3$ ; see Eq. (2.12).

analogy to the discussion for  $d=1$  we consider the slopes of the curves in Fig. 11, we obtain an estimate for the actual dependence of the decay constant on  $\sigma$ , i.e., for the exponents  $\beta$ . In  $d=2$  the theory yields values of  $\beta$  of about 1.66 for  $\sigma=0.1$  and 1.82 for  $\sigma=0.01$ , while in  $d=3$  the corresponding values are 1.86 and 1.99. As a more detailed consideration for  $d=3$  shows, the effective-medium result  $\lambda(\sigma)$  tends to  $\lambda=a_0\sigma^2$  for  $\sigma \ll 1$ , with  $a_0=\tilde{P}(0, u=0)=1.516\dots$ , i.e., to the decay constant one obtains in the Gaussian approximation; see Eq. (2.12). While the ladder yields an upper bound for the actual decay constant, we expect—see our arguments above—that the effective-medium theory yields a lower bound. We thus conjecture that in the limit  $\sigma \ll 1$  the actual value should also be given by  $\lambda=a_0\sigma^2$ . In the following section we shall present the results of numerical simulations for  $d=2$  and 3; it will be an interesting question whether the qualitative features of this theory correspond to the actual polarization decay.

### VIII. RESULTS FOR $d \geq 2$

In this section the numerical results of the spin depolarization due to a random walk on a square and on a simple-cubic lattice will be presented and compared with theory. For simplicity, the numerical simulations have been restricted to the case of the discrete random walk. They have been performed in analogy to the second method described in Sec. V for the one-dimensional random walk, i.e., we estimated

$$P_n = \left\langle \exp \left[ -\frac{\sigma^2}{2} \sum_r h_r^2 \right] \right\rangle$$

by averaging over a large number of  $n$ -step walks. Values of  $\sigma$  between 2.0 and 0.01 have been considered. Figure 12 shows the decay of polarization for a typical value of  $\sigma$

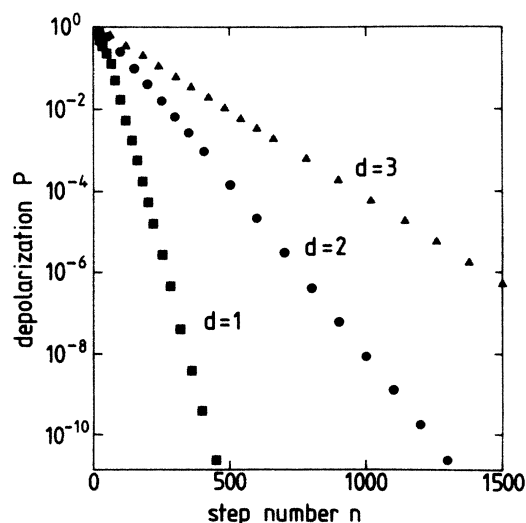


FIG. 12. Depolarization  $P$  as a function of the step number  $n$  for random walks on a linear chain ( $d=1$ ), on a square ( $d=2$ ), and on a simple-cubic ( $d=3$ ) lattice. The width  $\sigma$  was 0.1.

in one, two, and three dimensions; it shows the slower decay for higher dimension.

For the one-dimensional case the analytical treatment based on the approximation (4.1) yielded a qualitatively good description of the dependence of the decay constant on  $\sigma$ . The analogous treatment seems not to be possible for higher dimensions since much less is known about the distribution  $W_n(s)$  of the number of distinct sites visited. In Fig. 13 the numerically obtained decay constants  $\lambda$  are plotted versus  $\sigma$ ; for comparison, the data points for  $d=1$  have been included. We first consider the case  $d=2$ : if we assume a dependence  $\lambda \propto \sigma^\beta$ , the data show that the value of  $\beta$  increases slightly with decreasing  $\sigma$ ; for the smaller values,  $\beta \approx 1.77$ . Remember that the effective-medium theory yielded quite similar values for the exponent  $\beta$ .

For  $d=3$  it is useful to compare the data with bounds which can be given for the decay constant. The first one is the trivial lower bound  $\lambda = \sigma^2/2$ , given by the depolarization of the model where no revisiting effects are present. An upper bound can be obtained from Jensen's inequality; in analogy to (2.13),

$$P_n \geq \exp \left[ -\frac{\sigma^2}{2} \left\langle \sum_r h_r^2 \right\rangle \right]. \quad (8.1)$$

As den Hollander<sup>16</sup> has explicitly shown, for transient walks  $\langle h_r^2 \rangle \sim (2a_0 - 1)n$  ( $n \gg 1$ ). Thus the expression on the right-hand side of (8.1)—the Gaussian approximation for  $P_n$ —decays asymptotically with the decay constant  $\lambda = \sigma^2(a_0 - \frac{1}{2})$ , i.e.,  $\lambda \approx 1.016\sigma^2$  for the sc lattice in  $d=3$ . Note that the difference to the analogous decay constant in the continuous model,  $\lambda = \sigma^2 a_0$  [Eq. (2.12)], is given by

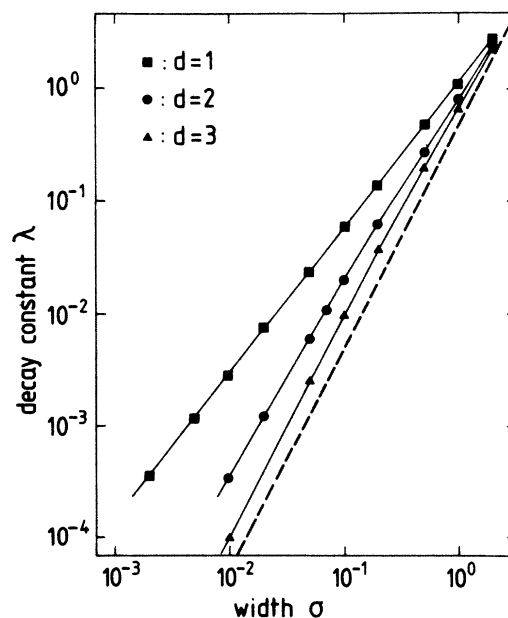


FIG. 13. Dependence of the decay constant in  $P_n \sim \exp(-\lambda n)$  on the width  $\sigma$ ; results of simulations for random walks on a linear chain ( $d=1$ ), on a square ( $d=2$ ), and on a simple cubic ( $d=3$ ) lattice. The dashed line denotes the lower bound  $\lambda = \sigma^2/2$ .

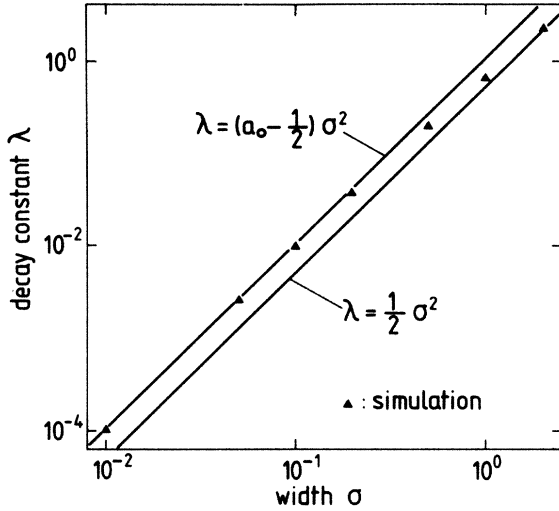


FIG. 14. Dependence of the decay constant  $\lambda$  on the width  $\sigma$  for random walks on a simple-cubic lattice ( $d=3$ ): The lines represent upper and lower bounds for the decay constant, see (8.2); the upper bound is the result of the Gaussian theory.

$\sigma^2/2$ —in agreement with our considerations at the end of Sec. V; see (5.4).

The decay constant, hence, must lie between these two bounds, i.e.,

$$\sigma^2/2 \leq \lambda \leq \sigma^2(a_0 - \frac{1}{2}). \quad (8.2)$$

Figure 14 shows the validity of this relation for the numerical result. Moreover, we see that for  $\sigma \ll 1$  the values of  $\lambda$  approach the upper bound  $\sigma^2(a_0 - \frac{1}{2})$ . From this behavior we conclude that for the continuous random walk the decay constant approaches the value  $\sigma^2 a_0$  for  $\sigma \ll 1$ , just as expected from the result of the effective-medium theory.

## IX. CONCLUSION

We have investigated the decay of phase coherence of transverse spin rotation due to the random walk of the spins on lattices with random rotation frequencies. In the main part of this work it was assumed that these frequencies are taken from a Gaussian distribution with mean zero and width  $\sigma$ . The frequency modulation  $\omega(t)$  induced by the random walk is characterized by a slow, i.e., algebraic, decay of correlations  $\langle \omega(t)\omega(0) \rangle$ , due to revisiting effects which become especially important for the low-dimensional lattices. In  $d=1$  and 2 no finite correlation time exists; thus the theory based on the Gaussian assumption for  $\omega(t)$  leads to a nonexponential asymptotic polarization decay, e.g., to a decay with an exponent proportional to  $t^{3/2}$  in  $d=1$ . For comparison, the “standard” (Gaussian) theory, where an exponential decay of frequency correlations is assumed, leads to a simple-exponential decay with decay constant  $\sigma^2 \tau_c$ , where  $\tau_c$  is the correlation time; in the case of fast modulation ( $\sigma \tau_c \ll 1$ ) the same result is obtained from the strong-collision model.

The process  $\omega(t)$  induced by random walks is actually

not a Gaussian process. For  $d=1$  we analyzed the role of this assumption within the discussion of the more general cumulant expansion for the depolarization  $P(t)$ . The approximations based on this systematic expansion are appropriate for the description of the initial decay of  $P(t)$ , but not for the asymptotic regime.

For the discrete random walk we were able to show that the polarization decay is simple exponential already in  $d=1$ —in contrast to the prediction of the Gaussian theory. A theory which takes the distribution of the span  $s$  of the one-dimensional walk into account elucidates the basic physical reason for this result: The decay proportional to the step number  $n$  in the exponent is due to the contributions of walks which are “extended” compared to the average walks with  $s \propto n^{1/2}$ , i.e., which visit of the order of  $n^1$  different sites ( $s \propto n^1$ ). Our derivations indicate that the specific properties of the one-dimensional random walk lead to a characteristic dependence of the decay constant  $\lambda$  on the width  $\sigma$ ,  $\lambda = \text{const} \times \sigma^\beta$ ; the result is valid for values of  $\sigma$  not exceeding unity. The exponent  $\beta \approx 1.3$  which has been obtained from numerical simulations is in good agreement with the theoretical result  $\beta = \frac{4}{3}$ .

Comparison between the depolarization  $P_n$  of the discrete and the depolarization  $P(t)$  of the continuous random walk showed that the two quantities are not identical when the identification  $n = t/\tau$  for larger step numbers  $n$  is made. The faster decay of  $P(t)$  could be understood by taking the actual distribution of waiting times into account. However, for small values of  $\sigma$  the dependence of the decay constant on  $\sigma$  can be described by the same exponent as in the discrete model. This is supported by the result of the effective-medium theory for the one-dimensional continuous random walk.

For  $d=1$  also other examples of frequency distributions (with mean zero and width  $\sigma$ ) were considered in order to answer the question to which extent our previous results depend on the particular choice of a Gaussian distribution. In the discrete model and for  $\sigma \ll 1$  there is a certain range of step numbers  $n$  for each distribution where exponential polarization decay similar to that in the Gaussian case prevails. With decreasing  $\sigma$ , the actual asymptotic decay, which indeed depends on the particular distribution, is shifted to extremely small values of the depolarization. For instance, for the example of the symmetric exponential distribution we deduced an asymptotic decay proportional to  $\exp[-n^{1/3} \ln^{2/3}(n)]$ . It appears to be a specific property of the Gaussian distribution that it leads to a simple-exponential decay for all large step numbers  $n$ .

In which way are the results on  $P_n$  experimentally relevant, e.g., the presence of an extended range with simple-exponential decay for  $\sigma \ll 1$ ? Random walk of particles takes place as a continuous-time process in various physical systems. Hence the mean time  $\tau$  between two transitions should be restored;  $\sigma$  substituted by  $\sigma\tau$  and  $n$  by  $t/\tau$ . [The difference between  $P_n$  and  $P(t)$  discussed above will not be considered further.] The condition  $\sigma \ll 1$  of the discrete model translates into the condition  $\sigma\tau \ll 1$  of the continuous model. This condition may be easily fulfilled by adjusting the residence time  $\tau$  of the particles, for instance, by temperature variation. Further,

small  $\tau$  corresponds to large step numbers  $n$  for fixed time  $t$ . Hence the case  $\sigma \ll 1, n \gg 1$  of the discrete model can be experimentally realized in the corresponding continuous model.

We furthermore investigated the polarization decay for random walks in two and three dimensions. In agreement with the result of the effective-medium theory for the square lattice, the dependence of the decay constant on  $\sigma$ , as it was observed in the simulations in  $d=2$ , could quite well be described by  $\lambda = \text{const} \times \sigma^\beta$  with  $\beta \approx 1.77$  in the regime of smaller values of  $\sigma$ .

In  $d=3$  (simple-cubic lattice) the decay constant of the effective-medium theory for the continuous random walk equals the decay constant of the Gaussian theory, given by  $\lambda = \sigma^2 a_0$  ( $a_0 = 1.516 \dots$ ), in the limit  $\sigma \ll 1$ . Our simulations for the discrete random walk showed that the decay constants approach the value of the corresponding Gaussian theory at smaller values of the parameter  $\sigma$ . We expect that for  $d \geq 3$  the Gaussian theory is the appropriate description in the case of fast frequency modulation.

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#### APPENDIX: EFFECTIVE-MEDIUM THEORY

In this appendix we show how the basic idea of the effective-medium theory already presented in Sec. VII actually leads to the self-consistency equation for the decay constant and how the results for  $d=1-3$  are obtained.

First, we consider spin particles performing a continuous-time random walk on an infinite lattice with a given frequency configuration  $\{\omega_r\}$ , where the initial condition is that at time  $t=0$  all particles start on site  $r=0$ , and we introduce the average spin polarization of these particles, denoted by  $P_{\{\omega_r\}}(t)$ , where we explicitly indicate the fixed frequency configuration. This quantity may formally be written as

$$P_{\{\omega_r\}}(t) = \sum_r A(r, t) . \quad (\text{A1})$$

where the quantities  $A(r, t)$ , which we call "local depolarization amplitudes," contain the contributions of all particles which are at site  $r$  at time  $t$ . Averaging over all frequency configurations yields the depolarization  $P(t)$ , i.e.,

$$P(t) = \langle P_{\{\omega_r\}}(t) \rangle_{\{\omega_r\}} = \left\langle \sum_r A(r, t) \right\rangle_{\{\omega_r\}} . \quad (\text{A2})$$

The amplitudes  $A(r, t)$  can be described by a system of differential equations, in analogy to the master-equation formalism for the elementary random-walk probabilities  $P(r, t)$ . The derivation of these equations is straightforward, and one obtains

$$\dot{A}(r, t) = -i\omega_r A(r, t) + \sum_{r'} p(r-r') A(r', t) - A(r, t) , \quad (\text{A3})$$

where the initial condition is given by  $A(r, t=0) = \delta_{r,0}$ .

The quantity  $p(r-r')$  is the probability for a particle at a site  $r'$  to jump to the site  $r$ , e.g.,  $p(r-r') = \frac{1}{2}(\delta_{r,r'+1} + \delta_{r,r'-1})$  for the one-dimensional simple symmetric random walk.

Let us now consider the effective medium itself, characterized by an exponential damping of the local amplitudes of strength  $\lambda$  on each site. For distinction we denote these amplitudes by  $E(r, t)$ . They are described by

$$\dot{E}(r, t) = -\lambda E(r, t) + \sum_{r'} p(r-r') E(r', t) - E(r, t) , \quad (\text{A4})$$

with the initial condition  $E(r, 0) = \delta_{r,0}$ . For  $\lambda=0$  this equation reduces to the ordinary master equation for the probability  $P(r, t)$ . Thus one easily obtains  $E(r, t) = \exp(-\lambda t) P(r, t)$ . It follows that the depolarization of the effective medium is given by [see (7.1)]

$$P_E(t) \equiv \sum_r E(r, t) = \exp(-\lambda t) . \quad (\text{A5})$$

We now consider the lattice, where—compared to the actual problem—only the site  $r=0$  retains its random rotation frequency  $\omega_0$ , while all other sites are considered to belong to the effective medium. The depolarization amplitudes obey the following equation:

$$\begin{aligned} \dot{A}(r, t) = & -\lambda A(r, t)(1 - \delta_{r,0}) - i\omega_0 A(0, t)\delta_{r,0} \\ & + \sum_{r'} p(r-r') A(r', t) - A(r, t) . \end{aligned} \quad (\text{A6})$$

Now the true depolarization, determined by (A2) and (A3), is approximated by  $\langle \sum_r A(r, t) \rangle_{\omega_0}$ , which still depends on the constant  $\lambda$ . Asymptotically,  $P(t)$  should be given by  $P_E(t)$ , which itself is characterized by  $\lambda$ ; thus, this approximation leads to the following self-consistency equation:

$$\left\langle \sum_r A(r, t) \right\rangle_{\omega_0} \sim P_E(t) = \sum_r E(r, t) \text{ as } t \rightarrow \infty . \quad (\text{A7})$$

This equation yields the decay constant  $\lambda$ .

Let us return to (A6). Laplace transformation,

$$\tilde{A}(r, u) = \int_0^\infty \exp(-ut) A(r, t) dt ,$$

and Fourier transformation,

$$\tilde{A}_q(u) = \sum_r \exp(iqr) \tilde{A}(r, u) ,$$

of the amplitudes  $A(r, t)$  yields, with (A4) and the analogous transformations of the quantities  $E(r, t)$ ,

$$\tilde{A}_q(u) = \tilde{E}_q(u) + (\lambda - i\omega_0) \tilde{A}(0, u) \tilde{E}_q(u) . \quad (\text{A8})$$

Fourier backtransformation gives

$$\tilde{A}(r, u) = \tilde{E}(r, u) + (\lambda - i\omega_0) \tilde{A}(0, u) \tilde{E}(0, u) , \quad (\text{A9})$$

which allows the determination of  $\tilde{A}(0, u)$ :

$$\tilde{A}(0, u) = [\tilde{E}^{-1}(0, u) + i\omega_0 - \lambda]^{-1} . \quad (\text{A10})$$

Since the self-consistency equation (A7) can be formulated in the Laplace domain as

$$\langle \tilde{A}_{q=0}(u) \rangle_{\omega_0} = \tilde{E}_{q=0}(u) \text{ as } u \rightarrow 0 ,$$



it is equivalent to

$$\langle (\lambda - i\omega_0)\tilde{A}(0, u) \rangle_{\omega_0} = 0$$

[see (A8)], and thus equivalent to  $\langle \tilde{A}(r, u) \rangle = \tilde{E}(r, u)$  [see (A9)]. With the choice  $r=0$ , i.e.,

$$\langle \tilde{A}(r=0, u) \rangle = \tilde{E}(0, u) \text{ as } u \rightarrow 0,$$

we obtain, with (A10),

$$\langle [\tilde{E}^{-1}(0, u) + i\omega_0 - \lambda]^{-1} \rangle_{\omega_0} = \tilde{E}(0, u) \text{ as } u \rightarrow 0, \quad (\text{A11})$$

which only involves the quantity  $\tilde{E}(0, u)$ . This quantity is simply given by  $\tilde{P}(0, u + \lambda)$ , the Laplace transform of the probability  $P(0, t)$  with shifted argument, since  $E(0, t) = \exp(-\lambda t)P(0, t)$ . Taking the average in (A11) for  $u=0$ , we obtain the final form of the self-consistency equation:

$$\frac{\pi^{1/2}}{\sqrt{2}\sigma} \exp[B^2(\lambda)] \operatorname{erfc}[B(\lambda)] = \tilde{P}(0, \lambda), \quad (\text{A12})$$

where  $B(\lambda) \equiv [\tilde{P}^{-1}(0, \lambda) - \lambda]/2\sigma^2$ . This equation can be numerically solved when  $\tilde{P}(0, u)$  is known.

In  $d=1$ ,  $\tilde{P}(0, u)$  is given by  $[u(u+2)]^{-1/2}$ ; see Eq. (3.6). In the case of the square lattice ( $d=2$ ), this quantity can also be written in closed form:<sup>28</sup>

$$\tilde{P}(0, u) = (1+u)^{-1} \frac{2}{\pi} K[(1+u)^{-2}], \quad (\text{A13})$$

where  $K$  is the complete elliptic integral of the first kind; for the numerical calculation appropriate approximations and/or expansions<sup>22</sup> can be applied.  $\tilde{P}(0, u)$  cannot be written in closed form for  $d=3$ , but with the application of two different series expansions given for the sc-lattice Green function [Eq. (7) in Ref. 29, Eq. (5.24) in Ref. 30],  $\tilde{P}(0, u)$  can be calculated with sufficient accuracy for all values  $u > 0$  and (A12) can be solved for all interesting values of  $\sigma$ .

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<sup>27</sup>We should like to mention that Th. M. Nieuwenhuizen called our attention to this interesting distribution.

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