

Random walks of a particle on lattices with traps

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The diffusion of a particle in a lattice is considered in the presence of absorbing traps. It is shown that for a small concentration of traps the problem can be reduced to that of an electron in a random field. The probability $\bar{W}(t)$ that the particle will not be absorbed by a trap is determined for long times in the one-, two-, and three-dimensional cases. It is shown that allowance for fluctuations of the trap density results in a much slower decrease of $\bar{W}(t)$ with time than in the usual gas approximation. The results are extended to the case of arbitrary values of the coefficient of attachment to a trap.

1. Random walks of a particle on lattices that include traps have been studied in^[1-5]. This problem has several applications, such as to the diffusion of vacancies of interstitial atoms in crystals containing impurity centers. In a recent paper^[5] Ryazanov determined the probability, in the so-called gas approximation, that a particle will "survive" for a long time. This approximation, which is equivalent to the first term of the expansion in powers of the concentration as employed for the problem of state density in disordered systems,^[6] does not take into account the fluctuations of trap density. It is clear, however, that particle survival for long periods of time will occur only in sufficiently large trap-free regions.

The present paper is concerned mainly with the effect of fluctuations in the trap distribution on the asymptotic survival probability $\bar{W}(t)$ after a long time t . It is shown here that for a small trap concentration $c \ll 1$ our problem is reduced to that of the spectrum and wave functions of a particle in a field of randomly situated impurities. For the considered small values of c an exact solution is obtained in the one-dimensional case. In the two-dimensional and three-dimensional cases an exponentially correct asymptotic form of $\bar{W}(t)$ is determined by means of a method similar to that which Lifshitz employed^[6] to find the density of states near the edge of a band. The considered fluctuation mechanism is shown to govern the case of large t , and the limits of applicability of the gas approximation are obtained.

2. Let us consider the random walks of particles on a lattice containing randomly located traps at some sites. The density of the particles performing the random walks will be assumed to be much smaller than the trap concentration, so that we may neglect collisions between the particles or saturation of the traps. For simplicity, we shall discuss the random walk of a single particle. Let $\tilde{W}_t(\mathbf{r})$ represent the probability that after t steps the particle is located at a site \mathbf{r} ; the unit in which t is measured is the time required for a jump between two neighboring sites. The equation of motion for $\tilde{W}_t(\mathbf{r})$ is

$$\tilde{W}_{t+1}(\mathbf{r}) = \sum_{\mathbf{r}'} p(\mathbf{r}-\mathbf{r}') (1-\delta_{\mathbf{r}'}) \tilde{W}_t(\mathbf{r}') + \delta_{\mathbf{r}} \tilde{W}_t(\mathbf{r}), \quad (1)$$

Here $p(\mathbf{r}-\mathbf{r}')$ is the probability of a transition from a site \mathbf{r}' to a neighboring site \mathbf{r} in a lattice without traps; the summation of $p(\mathbf{r}-\mathbf{r}')$ with respect to \mathbf{r}' equals unity. When a trap is located at a site \mathbf{r} the value of $\delta_{\mathbf{r}}$ is unity, whereas it vanishes for "pure" sites. The factor $(1-\delta_{\mathbf{r}'})$ in (1) forbids the departure

of the particle from a trap; the last term in the equation corresponds to a trapped particle. It is also seen from (1) that the summation of $\tilde{W}_t(\mathbf{r})$ over all sites is independent of t ; this corresponds to the conservation of the normalization.

We shall hereafter be interested only in $\tilde{W}_t(\mathbf{r})$ for pure sites, i.e., the probability that the particle does not fall into a trap. It is therefore convenient to introduce the expression $W_t(\mathbf{r}) = (1-\delta_{\mathbf{r}})\tilde{W}_t(\mathbf{r})$, which coincides with $\tilde{W}_t(\mathbf{r})$ at pure sites and vanishes at sites that are occupied by traps. The equation of motion for $W_t(\mathbf{r})$ is obtained when (1) is multiplied by $(1-\delta_{\mathbf{r}})$ and can be written as

$$W_{t+1}(\mathbf{r}) = D \sum_{\rho} \eta(\mathbf{r}, \mathbf{r}+\rho) W_t(\mathbf{r}+\rho). \quad (2)$$

Here D is the probability of a jump from a neighboring site to a given site in a pure lattice; the summation in (2) goes over nearest neighbors, the number of which is $1/D$. The factor $\eta(\mathbf{r}, \mathbf{r}+\rho)$ is unity when \mathbf{r} and $\mathbf{r}+\rho$ are pure sites, and vanishes when either \mathbf{r} or $\mathbf{r}+\rho$ is a trap site.

The exact formal solution of (2) is obtained conventionally by means of its expansion in terms of the eigenfunctions $\varphi_{\nu}(\mathbf{r})$ corresponding to the eigenvalues λ_{ν} :

$$D \sum_{\mathbf{r}'} \eta(\mathbf{r}, \mathbf{r}') \varphi_{\nu}(\mathbf{r}') = \lambda_{\nu} \varphi_{\nu}(\mathbf{r}). \quad (3)$$

Since the kernel of (3) is real and symmetric, the eigenvalues λ_{ν} are real. It is also clear from physical considerations that the functions $\varphi_{\nu}(\mathbf{r})$ comprise a complete system for the set of pure lattice sites:

$$\sum_{\mathbf{r}} \varphi_{\nu}^*(\mathbf{r}) \varphi_{\mu}(\mathbf{r}) = \delta_{\nu\mu}, \quad \sum_{\mathbf{r}} \varphi_{\nu}^*(\mathbf{r}) \varphi_{\nu}(\mathbf{r}') = \delta_{\mathbf{r}\mathbf{r}'}. \quad (4)$$

Here \mathbf{r} represents pure sites; from (3) we have $\varphi_{\nu}(\mathbf{r}) = 0$ for trap sites. Multiplying (3) by $\varphi_{\nu}^*(\mathbf{r})$ and summing over \mathbf{r} and ν , we obtain the sum rule for λ_{ν} :

$$\sum_{\nu} \lambda_{\nu} = 0. \quad (5)$$

The general solution of (2) is expressed as follows in terms of λ_{ν} , $\varphi_{\nu}(\mathbf{r})$, and the initial distribution $W_0(\mathbf{r})$:

$$W_t(\mathbf{r}) = \sum_{\nu} A_{\nu} \varphi_{\nu}(\mathbf{r}) \lambda_{\nu}^t, \quad (6a)$$

$$A_{\nu} = \sum_{\mathbf{r}} \varphi_{\nu}^*(\mathbf{r}) W_0(\mathbf{r}). \quad (6b)$$

At $t=0$, Eq. (6a) becomes an identity as a consequence of (4).

Let us assume that at $t = 0$ the particle is located with equal probability at any one of the lattice sites. Here we have $\bar{W}_0(\mathbf{r}) = 1/N$, where N is the total number of sites. For $\bar{W}(t)$, which is the total probability that the particle is not captured by a trap, we thus have

$$\bar{W}(t) = \sum_{\mathbf{r}} W_t(\mathbf{r}) = \frac{1}{N} \sum_{\nu} \lambda_{\nu}^t \left| \sum_{\mathbf{r}} \varphi_{\nu}(\mathbf{r}) \right|^2. \quad (7)$$

The summation with respect to \mathbf{r} in this equation goes over only pure sites, so that $\bar{W}(0) = 1 - c$, where c is the trap concentration. It is clear from the bounds of $W_t(\mathbf{r})$ and $\bar{W}(t)$ that the absolute value of λ_{ν} cannot exceed unity: $-1 \leq \lambda_{\nu} \leq 1$.

Equations (6) and (7) provide us with an exact formal solution of our problem. Unlike the thermodynamic properties of systems containing impurities, which we have studied previously^[7] and which were determined from only the energy spectrum of the appropriate equations, the present random-walk problem requires knowledge of both the eigenvalues λ_{ν} and the eigenfunctions $\varphi_{\nu}(\mathbf{r})$. Although our problem is therefore, generally speaking, more complex, certain limiting cases can be investigated.

3. Let us first consider the exactly solvable problem of a one-dimensional walk. In the limiting case of low trap concentration $c \ll 1$ and long time $t \gg 1$ we can go from a discrete to a continuous description. Along a segment of length l between two neighboring traps ($l \gg 1$ for small c) the particle obeys the equation (2) for the motion of a free particle. For the large values of t and l that we are considering, this equation becomes the ordinary diffusion equation

$$\partial W(t, x) / \partial t = D \partial^2 W(t, x) / \partial x^2. \quad (8)$$

Here we have the dimensionless diffusion coefficient $D = 1/2$, the lattice constant is taken to equal unity, and $W = 0$ at the limits of the segment.

Solving (8) for the region $x_1 \leq x \leq x_{i+1}$ (x_1 and x_{i+1} are the coordinates of neighboring traps) with the boundary conditions

$$W(t, x_i) = W(t, x_{i+1}) = 0, \quad W(0, x) = (1-c)/L \approx 1/L,$$

where L is the total length of the chain, we obtain

$$W_i(t, x) = \frac{4}{L} \sum_{n=0}^{\infty} \exp\left\{-\frac{1}{2} k_n^2 t\right\} \frac{\sin k_n(x-x_i)}{k_n l_i}. \quad (9)$$

Here $k_n = (2n+1)\pi/l_i$, $l_i = |x_1 - x_{i+1}|$. The desired quantity $\bar{W}(t)$, which equals the integral of $W(t, x)$ over all values of x , is represented by the sum of a large number N_i of random quantities $\bar{W}_i(t)$:

$$\bar{W}_i(t) = \int_{x_i}^{x_{i+1}} W_i(t, x) dx. \quad (10)$$

($N_i = cN$ is the total number of traps). Therefore, in accordance with the law of large numbers,

$$\bar{W}(t) = \sum_i \bar{W}_i(t) = N_i \langle \bar{W}_i(t) \rangle, \quad (11)$$

where $\langle \dots \rangle$ denotes averaging over the lengths of segments (the distances l between impurities). For a random distribution of traps the distribution function $f(l)$ has the Poisson form: $f(l) = c \exp(-cl)$, where $c = N_i/N = N_i/L$, and from (9)–(11) after some transformations we obtain

$$\bar{W}(t) = \frac{4}{\pi^2} \int_0^{\infty} \exp\left\{-\frac{a}{\xi^2}\right\} \frac{\xi d\xi}{\text{sh } \xi}, \quad a = \frac{c^2 \pi^2 t}{2}. \quad (12)$$

At relatively short times $t \ll c^{-2}$ (but, as previously, $t \gg 1$) the decrease of $\bar{W}(t)$ is represented by a power function:

$$\bar{W}(t) = 1 - 2 \left(\frac{2}{\pi} c^2 t\right)^{1/2} + \dots, \quad (13)$$

and for $t \gg c^{-2}$ by an exponential function:

$$\bar{W}(t) = 8 \left(\frac{2c^2 t}{3\pi}\right)^{1/2} \exp\left[-\frac{3\pi^{1/2}}{2} (c^2 t)^{1/2}\right]. \quad (14)$$

The aforementioned gas approximation corresponds to the substitution $l_i \rightarrow \bar{l} = c^{-1}$ in (9). For $t \ll c^{-2}$ this yields (13), and for $t \gg c^{-2}$ it yields

$$\bar{W}_g(t) \sim \exp(-\pi^2 c^2 t/2). \quad (15)$$

A comparison of (15) with (14) shows that for $t \gg c^{-2}$ the fluctuation mechanism is dominant, due to the absence of the region where (15) is applicable.

4. We shall now consider random walks in the two-dimensional and three-dimensional cases, assuming, as previously, $c \ll 1$ and $t \gg 1$. It is here convenient to go from the discrete equation (3) to a continuous equation. Under the considered condition $t \gg 1$ the main contribution to (6) and (7) comes from values of λ_{ν} that are close to unity, so that $1 - \lambda_{\nu} \equiv \epsilon_{\nu} \ll 1$. The procedure employed in^[7] can be followed for the transition to a continuous equation. In regions without traps we can express (3) in terms of gradients, thus obtaining the Schrödinger equation for a free particle, with $\epsilon = 1 - \lambda$ in the role of the energy. The presence of traps is represented by introducing the potentials of the impurities, so that (3) becomes

$$\Delta \varphi(\mathbf{r}) + k^2 \varphi(\mathbf{r}) = \sum_i V(\mathbf{r}-\mathbf{r}_i) \varphi(\mathbf{r}), \quad k = (2\epsilon)^{1/2}. \quad (16)$$

Here the index ν has been omitted, the lattice constant is unity as previously, and the summation in the right-hand side goes over the coordinates \mathbf{r}_i of the traps.

To determine the form of $V(\mathbf{r})$ we must, as in^[7], obtain a solution for the scattering of a plane wave in a discrete lattice by a single trap located, say, at the site $\mathbf{r} = 0$. We isolate the term in (3) that corresponds to a pure lattice and we transfer the inhomogeneity to the right side. The resulting equation is solved by means of a Fourier transformation, yielding

$$\varphi(\mathbf{r}) = e^{i\mathbf{k}\mathbf{r}} - G(\mathbf{r})/G(0). \quad (17)$$

Here G is the Green's function for scattering in a discrete lattice:

$$G(\mathbf{r}) = \sum_{\mathbf{r}'} \frac{e^{i\mathbf{p}\mathbf{r}'}}{\lambda_{\mathbf{p}} - \lambda_{\mathbf{k}} + i\delta}, \quad \delta \rightarrow +0, \quad \lambda_{\mathbf{k}} = \sum_{\mathbf{r}} p(\mathbf{r}) e^{i\mathbf{k}\mathbf{r}}, \quad (18)$$

where $p(\mathbf{r}) = D\eta(\mathbf{r}, 0)$ as in (1) and (2). Values of $\lambda_{\mathbf{k}}$ for different lattices are given in^[1,2], for example. As we noted in^[7], the form of the potential $V(\mathbf{r})$ in (16) is not unique because agreement of the scattering amplitudes in the discrete problem (17) and the continuous problem (16) is required only within the region of interest $k \ll 1$, $r \gg 1$. We therefore select the potential $V(\mathbf{r})$ in the very simple form of an impenetrable barrier with radius r_0 . Then, comparing the solution of the Schrödinger equation with (17), we find that for agreement of the results when $k \ll 1$, $r \gg 1$ the potential radius r_0 in the two-dimensional and three-dimensional cases must be

$$r_0^{(2)} = \frac{e^{-c}}{2\sqrt{2}} \approx 0.20, \quad r_0^{(3)} = \frac{3}{2\pi I_1} \approx 0.31 \quad (19)$$

for a square and a simple cubic lattice, respectively.

Here C is Euler's constant and $I_1 = 1.516$ is Watson's integral.^[1]

The random walk problem has thus been reduced to determining the energy spectrum and eigenfunctions of a particle in a random field of impurities.^[1] Specifically, $\bar{W}(t)$ of (11) can be expressed using the Green's function of the particle [from the time-dependent equation corresponding to (16)]:

$$\bar{W}(t) = \frac{1}{V} \int d\mathbf{r} \int d\mathbf{r}' G(\mathbf{r}, \mathbf{r}'; -it). \quad (20)$$

The features of the energy spectrum of a particle in a random field have been considered by several writers, in^[6] for example. However, to our knowledge the properties of the eigenfunctions in such systems have not been treated. It is therefore difficult to investigate (6), (7), and (20) directly.

However, an exponentially correct asymptotic form of $\bar{W}(t)$ can be obtained. For this purpose we follow the method that Lifshitz^[6] employed to determine the density of states near the edge of a band. For large values of t a particle will survive only inside sufficiently large regions that are free of traps. Within such regions the particle obeys the ordinary diffusion equation that follows from (2) for $t \gg 1$, $r \gg 1$. The attenuation of $W(t, r)$ at a boundary occurs within a distance smaller than or of the same order as the average distance between the impurities (see^[8], for example); this distance is much smaller than the dimension l of the region. Therefore it is sufficiently accurate for our purposes to assume $W(t, r) = 0$ at the boundary. Then for the probability that the particle will not be captured by traps in a region of volume V (or of area S in the two-dimensional case), when t is large we obtain from the diffusion equation the result

$$W(t) \sim \exp(-Dk_0^2 t). \quad (21)$$

Here k_0 , the smallest possible wave number for the given region, is of the order $V^{-1/3}(S^{-1/2})$. The desired quantity $\bar{W}(t)$ is the sum of expressions having the form of (21) for all the fluctuation regions, i.e., it can be represented, as in (11), by the sum of a large number of random quantities. Therefore $\bar{W}(t)$ is obtained from (21) by averaging $W(t)$ over the regions with the aid of a distribution function $f(V)$.

We shall assume that the traps are scattered at random, so that $f(V)$ is a Poisson distribution. Since for a given V (or S) this distribution does not depend on the shape of the region, we must, as a general rule, first sum (21) over all possible configurations of the boundaries of a fixed volume (or area). However, to derive an exponentially correct form of $\bar{W}(t)$ it is sufficient to confine ourselves to the shape of the region yielding the smallest value $(k_0)_{\min}$; from symmetry this region is a sphere (or circle).^[6] For a sphere of radius R we have

$$k_0 = \pi/R = \pi(4\pi/3)^{1/3} V^{-1/3},$$

and for a circle we have $k_0 = \mu_0/R = \mu_0 \sqrt{\pi} S^{-1/2}$, where $\mu_0 \approx 2.405$ is the first zero of the Bessel function $J_0(\mu)$. Averaging (21) on the basis of the Poisson distribution $f(V) = c \exp(-cV)$, in the three-dimensional case we obtain

$$\bar{W}(t) \sim \exp[-\alpha_3 (c^{1/3} t)^{3/2}], \quad \alpha_3 = 5 \left(\frac{\pi}{3} \right)^{1/2} 2^{-1/2} \approx 4.7, \quad t \gg c^{-1/3}. \quad (22)$$

Analogously in the two-dimensional case, when $f(S) = ce^{-cS}$ we obtain

$$\bar{W}(t) \sim \exp[-\alpha_2 (ct)^{1/2}], \quad \alpha_2 = \sqrt{\pi} \mu_0 \approx 4.3, \quad t \gg 1/c. \quad (23)$$

To determine the pre-exponential factors in (22) and (23) it would be necessary to perform the aforementioned averaging of W with respect to boundary configurations close to a sphere (or circle).

Let us now compare these results with the gas approximation, in which trap density fluctuations are neglected. Thus the true potential in (16) is here replaced by an average potential; the consequent electron energy shift is $\sim c$ in the three-dimensional case. For large t in (7) it is sufficient to confine ourselves to the minimum value $\lambda_{\min} = 1 - \epsilon_{\min}$, where $\epsilon_{\min} \sim c$. In conjunction with the equality $\lambda^t = (1 - \epsilon)^t \approx e^{-\epsilon t}$ this procedure gives us

$$\bar{W}_g(t) \sim \exp(-ct). \quad (24)$$

Numerical factors of the order of unity have been omitted in the arguments of the exponentials in (24) and (25). A comparison of (24) with (22) shows that the "fluctuation" mechanism of survival is dominant for $t \gg c^{-3/2}$, so that (24) can be applied for $c^{-1} \ll t \ll c^{-3/2}$.

For two-dimensional walks, as was shown by Ryazanov, the average energy shift in the gas approximation is determined from the relation $\epsilon \ln(1/\epsilon) \sim c$, whence, with logarithmic correctness, we have $\epsilon \sim c [\ln(1/c)]^{-1}$. Correspondingly, from (7) we obtain^[5]

$$\bar{W}_g(t) \sim \exp\left[-\frac{c}{\ln(1/c)} t\right]. \quad (25)$$

According to (23) and (25), the fluctuation mechanism becomes dominant when $t \gg c^{-1} \ln^2(1/c)$. Thus the exponential law (25) is actually applicable to only a small region:

$$\frac{1}{c} \ln \frac{1}{c} \ll t \ll \frac{1}{c} \ln^2 \frac{1}{c}. \quad (26)$$

5. Let us now consider a somewhat more complex trap model that allows for the possible presence of a potential barrier against the capture of a particle by an impurity. We shall describe this effect by introducing a "sticking coefficient" that differs from unity, $1 - \eta$. A particle that has reached an impurity site can in the next instant jump from the trap (be reflected) with the probability η . But if the particle is captured it can no longer leave the trap, so that the probability of escape from the trap is zero during the following instants. Instead of (1), the equation for the time-dependent probability is now

$$\begin{aligned} \mathcal{W}_{i+1}(\mathbf{r}) = \sum_{\mathbf{r}'} p(\mathbf{r}-\mathbf{r}') \left[(1-\delta_{\mathbf{r},\mathbf{r}'}) \mathcal{W}_i(\mathbf{r}') + \eta \delta_{\mathbf{r},\mathbf{r}'} \sum_{\mathbf{r}''} p(\mathbf{r}'-\mathbf{r}'') \mathcal{W}_{i-1}(\mathbf{r}'') \right] \\ + \delta_{\mathbf{r},\mathbf{r}'} \left[\mathcal{W}_i(\mathbf{r}) - \eta \sum_{\mathbf{r}'} p(\mathbf{r}-\mathbf{r}') \mathcal{W}_{i-1}(\mathbf{r}') \right]. \end{aligned} \quad (27)$$

The notation is here the same as in (1). Since we were considering the application to the case of low concentrations, for simplicity we neglected configurations with two or more neighboring traps. The above-considered case of fully effectual sticking corresponds to $\eta = 0$, and the absence of capture (in a pure lattice) corresponds to $\eta = 1$, when the solution (27), as is easily verified, represents free-particle walks, which are solutions of (1) without terms containing $\delta_{\mathbf{r}}$. It is also easily verified that (27), like (1), satisfies the condition that normalization is conserved.

As in the foregoing, it is convenient to introduce the quantity $W_t(\mathbf{r}) = (1 - \delta_{\mathbf{r}}) \bar{W}_t(\mathbf{r})$, for which we write the equation

$$W_{i+1}(\mathbf{r}) = \sum_{\mathbf{r}'} p_1(\mathbf{r}, \mathbf{r}') W_i(\mathbf{r}') + \eta \sum_{\mathbf{r}'} p_2(\mathbf{r}, \mathbf{r}') W_{i-1}(\mathbf{r}'),$$

$$p_1(\mathbf{r}, \mathbf{r}') = (1 - \delta_r) p(\mathbf{r} - \mathbf{r}') (1 - \delta_{r'}), \quad (28)$$

$$p_2(\mathbf{r}, \mathbf{r}') = (1 - \delta_r) \sum_{\mathbf{r}''} p(\mathbf{r} - \mathbf{r}'') \delta_{r''} p(\mathbf{r}'' - \mathbf{r}') (1 - \delta_{r'}).$$

Instead of (3) we shall now have

$$\lambda_v \dot{\varphi}_v(\mathbf{r}) = \lambda_v \sum_{\mathbf{r}'} p_1(\mathbf{r}, \mathbf{r}') \varphi_v(\mathbf{r}') + \eta \sum_{\mathbf{r}'} p_2(\mathbf{r}, \mathbf{r}') \varphi_v(\mathbf{r}'). \quad (29)$$

The solution of (28) subject to the additional initial condition $W_{-1}(\mathbf{r}) = 0$ can be written in the form of (6a), where, as previously, (6b) can be used for A_v in the given case of small c .

In the case of small c and sufficiently large t we go, as previously, from (29) to a continuous equation. Then we revert to (16), where the potential barrier (for a simple cubic lattice) now has the radius

$$r_0^{(3)}(\eta) = \frac{3}{2\pi} \frac{g}{1 + gI_1}, \quad g = \frac{1 - \eta}{\eta}, \quad (30)$$

where I_1 is the same quantity as in (19).

The subsequent discussion remains the same as above and leads again to (22), but with a region of applicability that now depends on the value of g . For $g \gtrsim 1$ the gas approximation yields (24), as previously, so that the foregoing calculations are retained. However, for small $g \ll 1$ [$g \ln(cg)^{-1} \ll 1$ in the two-dimensional case] the gas approximation applies to a wider region. The gas approximation here leads to (24) with the substitution $c \rightarrow cg$, while the fluctuation contribution retains the form of (22), as is proved by reasoning similar to that of Zaslavskii and Pokrovskii^[8] in their

analysis of state density. Therefore the fluctuation mechanism becomes the main mechanism only for $t \gg c^{-3/2} g^{-5/2}$ in the three-dimensional case, and for $t \gg (cg^2)^{-1}$ in the two-dimensional case.

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¹⁾The mathematical equivalence of these problems was noted by Ryazanov. [5]

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