Yu. M. Kagan. *Quantum diffusion in nonideal crystals.* 1. In an ideal crystal, at $T = 0$, any defect can in principle be delocalized. This concept was systematically developed by Andreev and Lifshitz,[1] who introduced the notion of quasiparticle band motion of defects in a quantum crystal.

The extremely weak tunneling coupling between equivalent positions in the crystal leads to the fact that already at a very low temperature T and low density of defects x, the diffusing particle interacts strongly with dynamic and statistical fluctuations, resulting in a distinct tendency toward localization of particles.

2. For any elementary tunneling act (transition into the interstitial lattice, exchange transition between neighboring atoms, involving also an entire group of atoms of the matrix, exchange of atoms between neighboring molecules, etc.), the usual two-well model turns out to be adequate. As is well known, tunneling in this model leads to a splitting Δ_0 of initially degenerate levels and the band arising in the regular crystal has a width $\Delta \approx z\Delta_0$. For the class of phenomena examined, the condition $\Delta \ll \omega_D$ is always satisfied (ω_D is the characteristic energy of phonons) and, as a rule, $\Delta < T$. It is precisely the presence of a small energy parameter that permits constructing a theory of quantum diffusion in the presence of a strong interaction with fluctuations of the medium[2–5]

3. For $T \neq 0$, due to oscillations of atoms of the matrix, the energy levels in neighboring wells fluctuate relative to one another. If the amplitude of these fluctuations is small compared to Δ, then band motion of particles with weak scattering by phonons occurs. However, even for very low T, the relative displacement of the levels begins to exceed Δ and dynamic breakdown of the band occurs.[2,3] The particle motion completely loses its band character, although true localization does not occur: at the moment that the fluctuating levels coincide, a particle can coherently (without excitation of phonons accompanying tunneling) go over into the neighboring well. It is interesting that the effective mean-free path l turns out to be small, in this case, compared to the interatomic distance a. The general solution of the problem, found in Refs. 2 and 3, for an arbitrary ratio of l and a leads to a diffusion coefficient of the form

$$D = \frac{Za^2}{3}\frac{\Delta_0^2}{\Omega(T)}. \qquad (1)$$

Phonon fluctuations here are characterized by the quantity $\Omega(T)$: the damping rate of correlations of the positions of particles in neighboring wells. At low T, two-phonon processes are the determining ones and

$$\Omega(T) \approx 10^6 \omega_D \left(\frac{T}{\omega_D}\right)^9. \qquad (2)$$

For $\Omega \ll \Delta$, when the motion has a band character ($l \gg a$), expression (1) coincides with the expression found in Ref. 1.

4. The small width of the band causes the coherent quantum diffusion to be anomalously sensitive to breakdown of the ideality of the crystal. Indeed, an individual point defect gives rise to a static displacement of the levels, exceeding Δ in a large region of radius $r_0 \gg a$, determined by the relation $\varepsilon_{im}(r_0) = \Delta$. Slowly decreasing perturbations present the greatest danger. In dielectric crystals, this is the field of deformations around a defect and in a metal, a weakly decreasing perturbation of the electron density (Friedel oscillations) are added to it. In both cases, in order of magnitude, we have

$$\varepsilon(r) \approx U_0 \left(\frac{a}{r}\right)^3. \qquad (3)$$

Already with a low density of defects x, spheres of radius r_0 begin to overlap and, beginning with some value x_c, as $T \to 0$, localization characteristic of the classical theory of percolation appears. From (3), it is easy to conclude that

$$x_c \approx \frac{\Delta}{U_0}\nu \ll 1, \qquad (4)$$

where ν is a numerical coefficient, which contains the characteristic percolation factor.

For $x \ll x_c$ and $T = 0$, band diffusion accompanied by scattering by regions of "excluded volume" with radius r_0, occurs. In this case, $D_0 \sim \Delta_0^{5/3}/x$.

Near the critical concentration we have

$$D(x) \approx D_0 Q(x), \quad Q(x) \approx \left(\frac{x_c - x}{x_c}\right)^t. \qquad (5)$$

Keeping in mind the fact that the local mean free path length $l \sim r_0 \gg a$, it can be shown that the main drop in D near x_c will be determined by the critical percolation index $t \sim 1.8$. The transition into the quantum localization regime with a corresponding change in the critical index occurs only in a very narrow region near x_c, whose scale is determined by the small parameter a/r_0.

5. For $T \neq 0$, the interaction with phonons leads to delocalization of particles. If the relative splitting of the levels at neighboring nodes satisfies the inequality $\Delta < \varepsilon_{\mathbf{r},\mathbf{r}+\mathbf{g}} < T$, then we have the following expression for the probability of a coherent hop from the node $\mathbf{r}$ to the node $\mathbf{r} + \mathbf{g}$[4,5]

$$W_{\mathbf{r},\mathbf{r}+\mathbf{g}} = \frac{2\Delta_0^2 \Omega(T)}{(\varepsilon_{\mathbf{r},\mathbf{r}+\mathbf{g}})^2 + \Omega^2(T)}. \qquad (6)$$

In the region of strong localization $x > x_c'$,[4,5] where x_c' is found from the condition that spheres with radius r_0' overlap, determined by the relation

$$a\left|\left(\frac{\partial \varepsilon}{\partial r}\right)_{r=r_0'}\right| = \Delta,$$

the diffusion problem actually reduces to the problem of the conductivity of a three-dimensional network with random distribution of conductivities of separate links, determined by (6). The value of D depends on the form of the defects and on their distribution. However, the temperature dependence in the most interesting cases can be established directly. Indeed, for low T, when $\Omega(T) \ll \varepsilon_{\mathbf{r},\mathbf{r}+\mathbf{g}}$, it follows directly from (6) that

$$D = \frac{Za^2}{3}\frac{\Delta_0^2}{(\delta\varepsilon)^2}\Omega(T), \quad \delta\varepsilon = \alpha U_0 x^{4/3}, \qquad (7)$$

and only the numerical coefficient α depends on averaging. From here, substituting (2), we have

$$D \sim T^9. \qquad (7')$$

As a result, instead of the dependence $D \sim T^{-9}$, following from (1) for $x \ll x_c$, the inverse temperature dependence appears.

At higher temperatures, when $\Omega(T) \gg \varepsilon_{\mathbf{r},\mathbf{r}+\mathbf{g}}$, a universal dependence on T coinciding with (1) is again satisfied. For some intermediate value of T_m, the diffusion coefficient passes through a sharp maximum and only the behavior in the narrow interval near T_m actually depends on the averaging. The general form of $D(T)$ is shown in Fig. 1. The ascending branch on the right corresponds to activation processes (classical above barrier or noncoherent subbarrier diffusion, vacancy diffusion). The dependence $D(T)$ for $x \ll x_c$ (curve 2) is shown for comparison.

For sufficiently low T or large splitting $\delta\varepsilon$, single-phonon processes, for which $\Omega(T) \sim T(\delta\varepsilon)^4$, become important. As a result, on the initial section of curve 1, $D \sim T$.

6. There exists an entire class of phenomena, for which particles diffusing in a crystal approach each other to within interatomic distances. However, such close approach, due to the interaction of the particles, is unavoidably accompanied by passage through a region in which levels in neighboring wells are strongly displaced. For this reason, subbarrier diffusion through such a region will be the limiting effect for the kinetics of such phenomena at low T. It is interesting that at large distances, the displacements of the levels are small and diffusion slows down as convergence occurs according to (7), but at close distances, a transition occurs to the single phonon regime and diffusion begins to accelerate. Thus a "bottleneck" appears, which is what determines the kinetics. The corresponding theory was developed in Ref. 6. Experimentally, quantum subbarrier diffusion under such conditions was observed in investigations of the recombination of hydrogen atoms in a molecular hydrogen matrix at low T.[7,8] It was found that instead of the exponential dependence on T, the dependence $D \sim T$ occurs in agreement with the theory.

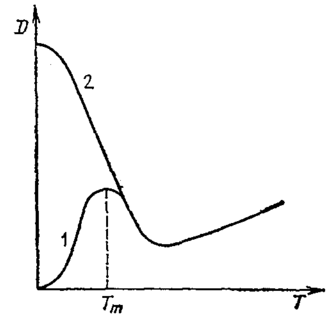
7. In studying the diffusion of μ^+ mesons in Bi,[9] an anomalous temperature dependence was observed, which gave rise to the generally used term "strange diffusion." As analysis showed, the picture found is a direct reflection of the anomalous behavior of $D(T)$ described by curve 1 in Fig. 1 (see Ref. 4).

8. Quantum diffusion of interacting atomic particles in an ideal crystal is of special interest. As in the case of external defects, in the case of neutral particles, the interaction has the form (3) and leads to an excluded volume with a large radius r_0. In such a system, at some critical value of the particle density $x_p^c \ll 1$, localization must occur.[4] *This new type of localization is entirely due to the discreteness of the space* (lattice of the matrix) and is absent, for example, in a liquid. The discreteness leads to a number of fundamental properties:

a) tunneling occurs only over a finite distance a; the number of locations z in the nearest coordination sphere is finite;

b) presence of strongly pronounced/transfer processes for $T \sim \Delta$;

c) suppression of collective motions. A cluster of N particles has a transition amplitude $\sim \Delta(\Delta/\delta\varepsilon)^{N-1}$. As a result, an ensemble of particles decays into a subsystem of stationary clusters and mobile separate particles. When the volume occupied by the stationary clusters exceeds some critical value, localization will occur for the remaining mobile particles and the picture will be analogous to the one described above for the case of external defects. In particular, an analogous temperature dependence and the transition with increasing x_p from curve 2 to a curve of the form 1 in Fig. 1 will remain. An approximate expression for D, valid in the entire x_p, T plane and encompassing both band motion and localization and phonon-assisted delocalization of particles was obtained in Ref. 5.



The phenomenon of localization examined above was first observed experimentally in investigations of diffusion of He^3 atoms in a He^4 matrix.[10] It was found that delocalization, due to the interaction with phonons, leads to the temperature dependence (7) and (7').

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