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LOW-TEMPERATURE CLUSTERING OF o-H_2 IMPURITIES IN p-H_2 CRYSTALS

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The presented explanation of anomalies in temperature dependence of pairing time for o-H_2 impurities in p-H_2 quantum crystals is based on an accurate analysis of differences in diffusion trajectories for different mechanisms of quantum diffusion. The transition between different regimes is accompanied by changes in characteristic lengths of pairing trajectories. Later stages of clustering may be influenced by coherent diffusion of pairs and triads.

1. INTRODUCTION

Several years ago puzzling experimental results (1) were reported on quantum diffusion of o-H_2 impurities in solid p-H_2. NMR data revealed a sharp drop in a pairing time for o-H_2 molecules below 0.3K, Fig. 1. Since all reasonable mechanisms of quantum diffusion predict either an increasing pairing time or, at least, a saturation at very low temperatures, the data (1) suggest an existence of some new mechanism of quantum diffusion. However, the existence of a new mechanism at such low temperatures does not seem very plausible.

Nevertheless, the data (1) can be interpreted on the basis of some geometric considerations in the frames of known mechanisms of quantum diffusion: the transition between diffusion regimes is accompanied by changes in patterns of diffusion trajectories with different characteristic lengths. Sometimes this leads to a decreasing pairing time even if the individual hopping rates are temperature independent or increase with the lowering temperature.

2. QUANTUM DIFFUSION AND PAIRING OF o-H_2 MOLECULES

The quantum diffusion, as well as a classical one, is, by definition, a random walk. However, in the process of pairing, two o-H_2 molecules spend most of the time close to each other when the dominant shifts in their energy levels (energy mismatches) on the different lattice sites are due to their electric quadrupole-quadrupole interaction with each other. Since the hopping rates for quantum diffusion strongly depend on energy mismatches (see,e.g.,(2)-(5)), and the mismatches change in a regular manner when the particles approach each other, the quantum diffusion may lose its random character at small separations. And the diffusion (pairing) trajectories do not resemble random walk anymore.

Different hopping mechanisms result in different diffusion trajectories, and the pairing trajectories change with a transition between diffusion mechanisms. There are three basic mechanisms of quantum diffusion, and not all of them favor random walks. The effective hopping rates are determined by the relation between the bare tunneling frequency J_0 and energy mismatches δE between the adjacent sites, and by the means to overcome these mismatches.

In the simplest case of negligible mismatches $J_0 >> \delta E$, one observes a really random diffusion with the diffusion coefficient $D \sim J_0 a l$ (a is the lattice constant, l is the mean free path, and h=1). Other mechanisms (with $\delta E >> J_0$) have effective rates quadratic in J_0.

One of the most important mechanisms of quantum diffusion is associated with two-phonon processes when the mismatches are overcome because of phonons taking care of an energy balance. Then the effective hopping rate is (4),(5) $J = J_0^2 T_0/(T_0^2 + \delta E^2)$, where $T_0 = \theta(T/\xi\theta)^\beta$, θ is the Debye temperature, index β is 7 or 9, and ξ is unknown, $0.1 < \xi < 1$. If $T_0 >> \delta E$, then the effective rate J does not depend on δE, and the diffusion trajectory is random.

As soon as $\delta E << T_0$, J strongly depends on mismatches, and the most probable direction of motion is perpendicular to the energy gradient. If this mechanism dominates, the effective pairing trajectories are very long (much longer than in the case of random walk), especially because the interaction energy - and mismatches - very rapidly increase with decrease in impurity separation. It is possible to show that the effective pairing time for this mechanism is $N^{3\alpha}\delta E_{max}^2/J_0^2 T_0$, where the index α is not universal and lies between 1 and 3, δE_{max} is the maximal value of mismatch (e.g., the energy of EQQ interaction at the smallest pair radius), and the effective path, N, is larger than an initial particles separation, N_0, (in h.c.p. lattice $N > 2N_0$).

At larger mismatches there is a possibility of one-phonon processes when the energy balance is ensured by emission of phonons. Then the motion is irreversible, with the hopping rate $J_0^2(\delta E/\zeta\theta)^3/\theta$, $0.1 < \zeta < 1$. The trajectory is directed almost along the energy gradient, is very short, and has practically no randomness.

As a result, both the hopping rate and the length of the pairing trajectory depend on mismatches. On the final stages

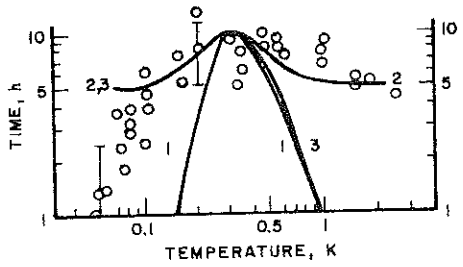
of pairing, the situation is clear: the potential relief and the mismatches δE depend only on EQQ interaction of the pairing particles. Then the hopping rate and the trajectory are determined only by the current pair radius, r: $\delta E = \delta E(r)$, $J = J(r)$.

At larger separations, the interactions with all other impurities are equally important, and the values of δE are random filling some energy interval E_0 with the constant density of states. Then it is possible to show that both the unassisted tunneling through the "windows" of the bare width J_0 and two-phonon processes averaged over the whole interval E_0 give the same hopping rate $J \sim J_0^2/E_0$, and lead to a random diffusion while the one-phonon processes are negligible.

The problem is to understand the temperature dependence of the pairing times at $0.05K < T < .2K$ and o-H_2 concentration $0.1\% < x < 1\%$ when the average distance between impurities is about 4 ÷ 5 lattice constants. At the distances less than 2 ÷ 3 the mismatches are due to the interaction within the given pair, and at larger separations the mismatches may be considered as random. [Except for high concentrations, x > 1%, none of the above regimes can alone account for the whole pairing process: the pairing corresponds to a combination of different regimes on different stages of pairing. At higher concentrations when the whole pairing process takes place with the mismatching caused by all other, randomly distributed, impurities]. Definitely, most of the pairing time is spent in overcoming the large mismatches at small distances. The only thing which can happen at $T < 0.3K$ is the transition from the two-phonon regime with $J \sim J_0^2 T_0/\delta E^2$ to the one-phonon mechanism, $J \sim J_0^2(\delta E/\zeta\theta)^3/\theta$. If one does not consider the pairing trajectory carefully enough, this transition results only in a disappearance of the temperature dependence of the pairing time. The account of the diffusion trajectories makes the results different.

All hopping rates discussed above contain one or more unknown constants. The calculation of the pairing time by means of simple numerical simulations of the diffusion problem with all these constants as fitting parameters provides an easy fit to the experimental data because of the large total number of parameters. The situation is even worse: the h.c.p. structure of the lattice dictates the doubling of the number of parameters.Therefore one may prefer to reduce a number of unknown parameters by simplifying the diffusion problem. Two such approaches are described below.

The first one involves only the transition from the two-phonon to the one-phonon regime with the drastic shortening of pairing trajectories. The set of unknown parameters is reduced to only two: one describes the product of J with the length, N, of the trajectory for the two-phonon regime, and the second - the ratio of coefficients ξ and ζ (curve 1 in Fig. 1). The first of these parameters is not very important being responsible for the overall scale. The second one was used to fit the position of maximum. The form of the curve



was independent of parameters. This mean-field, or continual, limit is not supposed to provide a very good agreement with the experiment: it does not use neither the exact lattice structure important at small pairs' radii (low-temperature wing), nor the randomness of mismatches responsible for the high-temperature wing of the curve.

The second approach is more accurate describing the diffusion in hexagonal lattice at small pairs' radii in combination with an approximate description of behavior at larger radii. The fitting parameters J and ξ/ζ gave the location of the maximum. An additional parameter described the ratio of jumps made at small and large separations (curve 2 in Fig. 1 corresponds to the case when nearly all time is spent at small separations, curve 3 - to the opposite case); the low-temperature side of the curve is independent of this parameter. One may improve the agreement with experiment, but the price would be the introduction of additional parameters: on the low-temperature side one should use the exact description of jumps including the second sublattice, and to increase the range of exact description (some admixture of the low-temperature wing of curve 1); on the high-temperature side, the improvement demands a better description of transition from random towards regular mismatches.

The lack of concentration dependence also gets a simple explanation.

3. SUMMARY

The large drop in the pairing time for ortho-impurities is due not only to changes in diffusion mechanisms, but also to a large difference in lengths and forms of diffusion trajectories associated with these mechanisms. The most important diffusion problem corresponds to the two-phonon mechanism of quantum diffusion. The fit to the experimental data may be achieved by an accurate analysis of transitions between the regimes with random and determinate mismatches.

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