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Disorder and Dissipation in Quantum Diffusion [and Discussion]

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Disorder and dissipation in quantum diffusion

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During the past 20 years most attention was given to quantum diffusion (QD) in perfect crystals but for the tunnelling motion in extremely narrow bands the question is how often we are dealing with ideal systems in real experiments. It turns out that in many cases QD is strongly affected by crystal disorder even in pure samples, so that the resulting behaviour of the tunnelling object has little in common with the diffusion in a perfect lattice. This point is confirmed by analysing the particle motion in long-ranged potentials.

The other novel feature which was discovered recently in the μSR study of QD is connected with the role of barrier fluctuations at high temperature. Usually this mechanism of the particle interaction with the environment is simply omitted, and the data are fitted by the standard activation type dependence. However, there is a theoretical argument and strong experimental evidence that barrier fluctuations should be included into the consistent analysis of the QD data, especially for the case of a light particle diffusion in a heavy matrix.

The last part of the paper deals with a singular particle state arising from its strong coupling with a dissipative environment. I discuss the problem of overdamped motion and divergent effective mass in dissipative models with the application of the theory to the case of ion dynamics (μ^+–muon can be an example of a positive ion) in normal and superfluid ^{3}He.

1. Introduction

After the notion of quantum diffusion (QD) was introduced some 25 years ago (Andreev & Lifshitz 1969; Guyer *et al.* 1971) an enormous number of different experimental systems was studied and found to demonstrate quantum tunnelling motion at low temperature (see, for example, the book edited by Kagan & Leggett (1992)). Now it is simply impossible to mention all the cases or review the theory in a short talk. Instead, I would rather concentrate on some specific ideas which became important in understanding recent experiments on muon (muonium) QD in insulators and superconductors, along with the new developments in the theory of particle motion in a dissipative environment.

2. Crystal disorder

We used to think about atoms, vacancies, etc., as point defects with only few unit cells around being disturbed. This conclusion usually comes out by compar-

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ing the energy involved in the crystal distortion with the characteristic energies of the solid, Debye temperature Θ_D, or even the Fermi energy ϵ_F, which are quite large. However, from the point of view of the particle dynamics the above mentioned energy scales are rather irrelevant. Consider the particle motion in a narrow band in the presence of impurity potential. The tunnelling band can be extremely narrow while the amplitude of the potential, $U(a)$, may be as large as 10^3 K in a metal or 10–100 K in an insulator (here a is the lattice constant). If the particle approaches the defect from infinity, then at some distance $R \gg a$ where the potential is larger than the kinetic energy (and another resonance is not awaiting right in the next unit cell), it should stop and turn around. In other words, the radius R defines the elastic cross section of the defect potential. It may be difficult to believe, but the fact is that in some cases (like μ^+-muon in copper) more than 10 million unit cells around each defect are inside the radius R, which may be estimated as $R/a \sim [U(a)/\Delta]^{1/3}$ (here Δ is the tunnelling band width).

The well known system of the dilute solution of ^{3}He atoms in the ^{4}He crystal illustrates the point (Richards *et al.* 1972; Allen *et al.* 1982; Grigoriev *et al.* 1973). Here the role of impurity is played by another ^{3}He particle, and at low temperature the diffusion rate is controlled by pair collisions between the hard spheres of radius R. One can easily obtain the cross section from the classical expression for the mean free path $l = 1/n\sigma$ with the result $\sigma \sim 200a^2$ depending on pressure. An obvious question next to ask is: if impurities are so large then at some small concentration their hard-sphere radii must overlap and block their mutual motion completely because no free space is left (Kagan & Maksomov 1983). And this is exactly what happens when ^{3}He concentration is increased to the level of few percent (Mikheev *et al.* 1983). One immediately realizes the connection between this localization and the metal–insulator transition in the Habbard model. In our case, however, the exclusion volume is large and transition takes place at low particle concentration.

Even more striking results were observed resently (Kiefl *et al.* 1990; Karlsson *et al.* 1993) in the study of the μ^+-muon QD in superconducting Al. Well below $T_c \approx 1.2$ K when all inelastic processes are completely suppressed (at least during the time of the μSR measurement) muons should form two independent ensembles of fast and static (those stopped inside the radius R around the nearest defect) particles, and the fraction of fast moving muons should disappear at very low defect concentration due to enormous elastic cross section. Experimentally the two-component form of the polarization function $P(t) = P_f + P_{st}(t)$ was clearly observed at temperatures $T < 0.2$ K (Karlsson *et al.* 1993) (the fast component P_f does not decay because of the motional narrowing effect). In the pure sample with only *ca.* 1 ppm Li content the polarization function ceases to depend on time at all, thus proving coherent delocalization of all muons. This result should be contrasted to the case of 18 ppm Li sample where $P(t)$ is a clear superposition of the classical Kubo–Toyabe relaxation and constant term with almost equal amplitudes. Finally in the 75 ppm Li-doped aluminium all muons undergo static relaxation – the behaviour which is consistent with the estimated critical concentration of overlapping defects $n_{im} \approx 40$ ppm (Kagan & Prokof'ev 1991).

The preliminary analysis of the T_1 relaxation data in KCl with 0.27%-Na and 2.7%-Na doping (Kiefl *et al.*, this volume) is also consistent with the above picture. μSR spectra have a distinctive two-component behaviour at low temperatures. It is strongly believed that in the pure KCl the plateau at low temperature is due to the coherent band motion with the coherent delocalization time *ca.* 2×10^{-11} s. In the 0.27%-Na sample the first component which has an amplitude around 0.3 has exactly the same correlation time as that found in a pure sample, unambiguously proving that this contribution comes from coherently delocalized muonium. At the same time the other fraction with 0.7 amplitude is moving extremely slow ($\tau \sim 10^{-6}$ s at $T = 4$ K). At a higher level of doping the coherent fraction disappears.

Of course, the formation of two independent ensembles of particles holds only at very low temperature when inelastic interaction with the thermal excitations is weak enough. At higher T some of the particles will be able to gain a sufficient amount of energy from the crystal to overcome the defect potential and to move even if the tunnelling states are out of resonance. Now, particles can exchange places and move inside and outside the radius R during the experiment, thus making the two-ensemble separation at least a much less precise procedure. On the other hand, the energy shift which the particle has to overcome between the initial and final states is much larger close to the defect centre, and the diffusion rate strongly depends on this energy shift. Even when particles at distances of order R from impurities start moving, those deep inside the radius R are still static. Increasing the temperature we change continuously the ratio between the moving and static components. To be more precise, at higher T there is a spatial distribution of the muon hop rates in the impure crystal. The basic features of this distribution can be easily obtained from the approximate expression for the hop rate (see Kagan & Leggett 1992 for more details)

$$\tau^{-1}(\xi, T) \approx \frac{8\overline{\Delta}_0^2 \mathit{\Gamma}(\xi, T)}{\mathit{\Gamma}^2(\xi, T) + \xi^2} \qquad (\xi < T), \tag{2.1}$$

where $\overline{\Delta}_0$ is the renormalized tunnelling amplitude (temperature dependent in the general case, e.g. in a metal), $\mathit{\Gamma}$ is the level broadening due to the particle coupling with the environment, and $\xi = E(\boldsymbol{r}) - E(\boldsymbol{r} + \boldsymbol{a})$ is the energy difference between the initial and final states due to the crystal structure and disorder. Note, that in the metallic case one usually has $\mathit{\Gamma}(T) \sim T$, so that the diffusion rate as derived from (2.1) is homogeneous (for $\xi > T$ the particle may be considered as being trapped already). In insulators and superconductors there is a wide parameter space where (2.1) is valid because in this case $\mathit{\Gamma} \ll T$.

The inhomogeneity of QD has a direct consequence for the muon depolarization process. Firstly, both T_2 and T_1 relaxation rates are now functions of the muon position in the crystal. The most naive way of dealing with this problem is simply to sum polarization functions corresponding to different hop rates

$$P(t) = \langle P(t, \tau(\boldsymbol{r}))\rangle, \tag{2.2}$$

where $\langle \ldots \rangle$ stands for the volume average $\int_{\mathrm{vol}} \mathrm{d}V$. This approach will certainly work if the particle moves only slightly (few atomic distances) during the characteristic relaxation time or the time of experiment. Clearly (2.2) gives a multicomponent polarization function. On the other hand, if the hop rate is fast enough or the relaxation time is very long, so that the particle changes most of the energy configurations during the characteristic time, then another averaging procedure might be appropriate,

$$P(t) = P(t, \langle \tau(\boldsymbol{r}) \rangle), \quad \text{or} \quad P(t) = P(t, \langle \tau^{-1}(\boldsymbol{r}) \rangle), \tag{2.3}$$

depending on whether the relaxation rate is defined by τ or τ^{-1}. Even at this stage of discussion we see the problem in the simple-minded (2.2) and (2.3): what if in some regions the relaxation time is defined by τ and in another regions by τ^{-1}; or if the relaxation time itself is short in some parts of the crystal and very long in another. Obviously, both (2.2) and (2.3) fail to account for such a situation, because the actual relaxation of $P(t)$ may be due to the muon/muonium diffusion flow between different parts of the sample. The proper description of the problem requires the solution of the kinetic equation for the particle motion and depolarization in the inhomogeneous medium. The particular case of the muon T_2-relaxation under these conditions was considered and solved in (Kagan & Prokof'ev 1991). The other possibility is to use a computer simulations of the relaxation process. Whatever the complexity of the problem might be, the general trend is the transformation of the two-component $P(t)$ at low temperature into some multicomponent function at intermediate T and eventually a simple exponential relaxation and high temperature.

There is no doubt that at intermediate T the polarization function both in the superconducting Al and in doped KCl has a multicomponent relaxation. Although the qualitative explanation of the peculiar temperature dependence of the muon relaxation rate in Al below T_c was given in (Kagan & Prokof'ev 1991) the real question is to what extent the theory of QD can account for the details of $P(t)$. The results of the computer simulations (Karlsson *et al.* 1993) show that the agreement between the theory and the experiment is quite remarkable taking into account that only few constants were used to describe the multidimensional set of curves $P(t, T, n_{\text{im}})$ in superconducting Al. The advantage of the T_1-measurement in KCl is that one can easily follow the temperature variation of the particle hop rates many orders in magnitude, and the system parameters are extremely well characterized (comparing the uncertainty of our knowledge about QD in most cases). In this case one may hope to describe (or to fail – both outcomes have comparable value!) the experiment in the doped KCl without adjustable parameters at all.

3. Barrier fluctuations

At temperatures close to Θ_D the diffusion rate increases exponentially with T. It may be due to various reasons, for example due to vacancy diffusion, classical overbarrier motion or small polaron quantum hopping. Except may be the case of quantum crystals of helium and hydrogen the first mechanism can be ruled out by extremely high activation energy for the vacancy creation; the other argument may be that there is no reason for the heavy vacancy to move faster than a light muon. The crossover to the classical overbarrier motion takes place at a temperature of order $T_* \approx \omega_0/2\pi$, where ω_0 is the zero point vibration frequency in the potential well obtained by inverting the potential barrier between the two tunnelling states. Given a very large ratio between the muon mass and masses of atoms forming the lattice one expects that there should exist a wide temperature interval even above Θ_D in which small polaron hopping still dominates over the classical transitions.

There are two competing effects in the tunnelling problem when one includes the particle–phonon interaction. The first one is connected with the lattice distortion around the particle sitting in a potential well. In the tunnelling process not only the particle has to tunnel to the neighbouring well, but so has the lattice distortion. As a result the tunnelling amplitude is suppressed exponentially. At high temperature most of the polarization cloud may be shook off during the transition which leads to the activation-type dependence of the hop rate (Flynn & Stoneham 1970),

$$\tau^{-1} \sim \Delta_0^2 (ET)^{-1/2} \mathrm{e}^{-E/T}. \tag{3.1}$$

In this expression the pre-exponential term depends on the tunnelling amplitude, and, therefore, is much less than the classical attempt frequency.

The second effect is absolutely the opposite to the polaronic narrowing. It arises from the particle interaction with the lattice in the underbarrier region. Due to vibrations of the host atoms the barrier height and shape constantly fluctuate around their equilibrium values, and so does the tunnelling action. Obviously, the particle prefers to tunnel when the potential barrier is the lowest, so that the actual tunnelling rate corresponds to the optimal fluctuation (Kagan & Klinger 1976). This interaction tends to increase the tunnelling amplitude exponentially, and due to the fact that thermal vibrations of the lattice increase linearly with T the resulting effect is much more pronounced at $T > \Theta$. With barrier fluctuations included the hop rate in (3.1) acquires a form (Kagan & Klinger 1976)

$$\tau^{-1} \sim T^{-1/2} \mathrm{e}^{-E/T + T/E_1}. \tag{3.2}$$

Provided this expression is valid in a wide temperature range, the last term in the exponent will dominate in the exponential dependence at high T.

The experimental confirmation of (3.2) is rather difficult. Usually, the data concerning the high temperature diffusion are available only in a narrow temperature range making the results of the fitting ambiguous, or not very convincing. As a rule an additional information concerning the value of the polaron energy, E, or barrier fluctuation energy, E_1, is not available independently. This situation changed drastically since the discovery of the coherent mounium motion in ionic and semiconducting crystals KCl (Kiefl *et al.* 1989), NaCl (Kadono *et al.* 1990*a*), GaAs (Kadono *et al.* 1990*b*) and CuCl (Schneider *et al.* 1992). The point is that knowing the pre-exponential factor and activation energy in (3.1) one can obtain not only the bare value of the tunnelling amplitude but the scale of the polaronic narrowing as well. Now, the result of such an analysis should be compared with the observed coherent band width at low T. In GaAs all the data are in good agreement with the standard small polaron theory. On the other hand, starting from (3.1) and applying the same procedure to the case of muonium diffusion in KCl it was found in (Kagan & Prokof'ev 1990) that the data can not be described consistently in the polaron picture alone. The consistency was achieved by assuming a dominant role of the underbarrier interaction in the formation of the coherent band. The contrast between the two limiting cases is very sharp leaving little doubt that the second mechanism is operating more effectively in KCl, and $\overline{\Delta}_0 > \Delta_0$.

No matter how strong was the argument in favor of the barrier fluctuations effect on tunnelling in KCl, it was only a theoretical one because the high temperature data on their own were not sufficient to reveal the linear term in the exponent of (3.2). The idea of studding a material with the heaviest lattice atoms in order to expand the temperature range were (3.2) is supposed to work was realized in solid xenon (Storchak *et al.* 1993). For the first time the data on quantum hopping became available on a scale (τ was measured to vary between 2×10^{-7} s and 4×10^{-11} s) allowing a direct test of the theory. The observed dependence $\tau(T)$ was an obvious superposition of two terms in the exponent (3.2). Both the activation-type exponent and the linear dependence alone fail to describe the curve $\tau(T)$ above 80 K and below 60 K correspondingly. The discrepancy was almost an order in magnitude. One should admit that an extremely good fitting obtained for the sum of two terms in the exponent is now a strong experimental argument demonstrating the effect of barrier fluctuations on tunnelling at $T > \Theta_D$. I believe that the phenomenon is quite universal and any treatment of the high temperature results in QD has to deal with the full expression (3.2).

4. Dissipative environments

Conventionally the term QD is connected with the tunnelling motion of particles. Still, there are cases where quantum interference and quantum corrections essentially modify transport properties of the system. The well known example is the weak localization problem, where localization occurs due to the interference of multiple scattering events from crystal disorder (Lee & Ramakrishnan 1985). This is a remarkable phenomenon, but it is not that unexpected that the particle gets localized in a static external potential, even if it is a random one. In the most general way one can argue that any object should delocalize in the homogeneous liquid at $T = 0$ simply because there are no excitations in the ground state to scatter it. Note that one assumes *a priori* a finite effective mass for the particle. What is more surprising is that there is a possibility to localize the particle in a homogeneous liquid (no static potential) provided it is coupled strongly enough to the liquid excitations. It turns out that if the liquid is a so called Ohmic dissipative environment (Caldeira & Leggett 1981) the particle dynamics is overdamped down to zero temperature (with the effective mass being ill defined but diverging), and the diffusion rate is zero in the ground state. Even worse, if the liquid is a 'sub-Ohmic' environment then at $T = 0$ the particle placed at the origin at the initial moment of time stays at the origin forever!

In the standard picture of the particle transport through the medium one assumes random scattering events with a free particle propagation in between. What will happen if scattering events are so frequent that they overlap with each other, and formally the particle is scattered many times on the scale of its wavelength (or the scattering rate is larger then the particle thermal energy)? The answer is that the effective mass will start to diverge, thus 'absorbing' most of the scattering into the virtual renormalization and keeping the rest of the interaction in balance with the uncertainty principle. The following simple consideration illustrates the point.

In the normal ^{3}He the mobility μ of the negative ion (electron bubble) was measured to be independent on T down to the superfluid transition temperature (Ahonen *et al.* 1978), or, using Einstein relation the diffusion rate was $D \propto \langle v^2 \rangle \tau \propto \mu T$, where $\langle v^2 \rangle$ is the mean square velocity. Multiplying both sides by M^{eff} and assuming that the thermodynamic relation $M^{\text{eff}} \langle v^2 \rangle \sim T$ holds, we find quite generally

$$M^{\text{eff}} \sim \tau e / \mu \tag{4.1}$$

(where e is the particle charge). Now, the uncertainty principle requires that the scattering rate can not be defined better then the particle thermal energy, so that there is an upper bound $\tau > 1/T$. Under this conditions the temperature independent result for the mobility inevitably leads to the divergent effective mass $M^{\text{eff}} > e/(\mu T)$. Of course, this consideration is nothing but a qualitative arguing, especially due to the fact that for $\tau T \sim 1$ any coherence is completely suppressed in the overdamped particle dynamics.

The other way to look at the problem is to say that polarization waves in the liquid produced by the particle in the past relax so slowly that at any given moment of time the particle is, in its turn, localized in the random potential created by these polarization waves. This potential is not static but is changing constantly in time, and it is only due to the presence of the particle itself. Within the Feynmann path integral approach we can describe this picture quantitatively by writing an influence functional in the form,

$$\frac{e}{\mu} \iint_0^{1/T} \mathrm{d}t\mathrm{d}t' \frac{(\boldsymbol{r}(t) - \boldsymbol{r}(t'))^2}{(t-t')^{\nu}}. \tag{4.2}$$

The power exponent ν tells us how long the environment keeps track of the particle position back in time. If ν is very large then only the nearest points in time are important. At the critical value $\nu = 2$, which corresponds to the 'Ohmic' environment, all points in time are coupled strongly provided $e/\mu > MT$. In most cases the mobility itself is a strong function of temperature and diverges as $T \to 0$. However, for $\mu =$ const. in (4.2) the strong coupling condition is always satisfied at sufficiently low T. The case of sub-Ohmic environment mentioned above corresponds here to $\nu < 2$.

Until now the notion of the sub-Ohmic environment is only a theoretical one (the most probable candidate might be the so called 'gauge liquid' (Halperin *et al.* 1993)). Fortunately, ions in the normal ^{3}He at low temperature are exactly the Ohmic strong coupling case (Prokof'ev 1993, 1994*a*). To reveal the 'latent' divergence of the effective mass unambiguously one should go to the superfluid state of ^{3}He, where the energy gap cuts off the divergence and restores the ballistic motion between rare scattering events. A precise calculation of the effective mass in ^{3}He-B was done in Prokof'ev (1993, 1994*b*); the prediction is that M^{eff} for the electron bubble is as large as about 20 000 ^{3}He-atomic masses at zero pressure – some 100 times heavier than that in ^{4}He!

References

Allen, A. R., Richards, M. G. & Schratter, J. 1982 *J. Low Temp. Phys.* **47**, 289.

Andreev, A. F. & Lifshitz, I. M. 1969 *Zh. eksp. teor. Fiz.* **52**, 2057. (*Sov. Phys. JETP* **29**, 1107.)

Ahonen, A. I., Kokko, J., Paalanen, M. A., Richardson, R. C., Schoepe, W. & Takano, W. Y. 1978 *J. Low Temp. Phys.* **30**, 205.

Caldeira, A. O. & Leggett, A. J. 1981 *Phys. Rev. Lett.* **46**, 211.

Caldeira, A. O. & Leggett, A. J. 1983 *Ann. Phys. (N.Y.)* **149**, 374.

Flynn, C. P. & Stoneham, A. M. 1970 *Phys. Rev.* B **1** 3966.

Grigoriev, V. N., Eselson, B. N., Mikheev, V. A. & Shul'man, Yu. E. 1973 *Zh. eksp. teor. Fiz.* **17**, 25. (*Sov. Phys. JETP Lett.* **17**, 161.)

Guyer, R. A., Richardson, R. C. & Zane, L. I. 1971 *Rev. mod. Phys.* **43**, 532.

Halperin, B. I., Lee, P. A. & Read, N. 1993 *Phys. Rev.* B **47**, 7312.

Kadono, R., *et al.* 1990*a* *Phys. Rev. Lett.* **64**, 665.

Kadono, R., *et al.* 1990*b* *Hyperfine Interact.* **64**, 635.

Kagan, Yu. & Klinger, M. I. 1976 *Zh. eksp. teor. Fiz.* **70**, 255. (*Sov. Phys. JETP* **43**, 132.)

Kagan, Yu. & Maksimov, L. A. 1983 *Zh. eksp. teor. Fiz.* **84**, 792. (*Sov. Phys. JETP* **57**, 459.)

Kagan, Yu. & Prokof'ev, N. V. 1990 *Phys. Lett.* A **150**, 320.

Kagan, Yu. & Prokof'ev, N. V. 1991 *Phys. Lett.* A **159**, 289.

Kagan, Yu. & Leggett, A. (eds) 1992 *Quantum tunneling in condensed matter.* Elsevier.

Karlsson, E., *et al.* 1994 *Hyperfine Interact.* (In the press.)

Kiefl, R. F., *et al.* 1989 *Phys. Rev. Lett.* **62**, 792.

Kiefl, R. F., *et al.* 1990 *Hyperfine Interact.* **64**, 737.

Lee, P. A. & Ramakrishnan, T. V. 1985 *Rev. mod. Phys.* **57**, 287.

Mikheev, V. A., Mikhin, N. P. & Maydanov, V. A. 1983 *Fizika Nizk. Temp.* **9**, 901. (*Sov. J. Low Temp. Phys.* **9**, 4615.)

Prokof'ev, N. V. 1993 *Int. J. mod. Phys.* B **7**, 3327.

Prokof'ev, N. V. 1994*a* *Hyperfine Interact.* **85**, 56.

Prokof'ev, N. V. 1994*b* *Phys. Rev. Lett.* (Submitted.)

Richards, M. J., Pope, J. & Widom, A. 1972 *Phys. Rev. Lett.* **29**, 708.

Schneider, J. W., *et al.* 1992 *Phys. Rev. Lett.* **68**, 3196.

Storchak, V., Brewer, J. H., Morris, G. D., Arseneau, D. J. & Senba, M. 1994 *Hyperfine Interact.* **85**, 34.

Discussion

A. M. STONEHAM (*Harwell Laboratory, Oxfordshire, U.K.*). (i) The $\exp(T/E)$ form of the barrier fluctuation mechanism depends on first how the μ^+–host interaction is represented; the Flynn–Stoneham analysis for FCC hosts gave an activated form. For KCE and NaCl, good calculations could be done.

(ii) For NaCl, KCl there are the self-trapped (electronic) holes. These seem to show activated behaviour (the data are not very good) and it would be nice to understand why they behave differently.

(iii) Dr Prokof'ev mentioned metals at high temperatures. The data may not be easy to interpret because of other mechanisms (e.g. for H in liquid Fe it is reasonably clear that relatively long diffusion jumps are occuring).

N. V. PROKOF'EV. (i) I do agree that the coefficient in the high temperature dependence of the barrier fluctuations effect, which is material and lattice structure dependent quantity, can be calculated in simple ionic crystals with reasonable accuracy allowing its comparison with the experimental data.

(ii) The problem is whether we can discriminate between different possibilities. Usually data are insufficient to test the linear-T term in the exponent because of the narrow temperature range where quantum hopping is observed. I am not aware of any attempt to analyse self-trapped electronic holes in NaCl and KCl in this respect.

(iii) I am not at all sure that the theory can be applied directly to the liquid case (in a sense there is no barrier in a liquid, or alternatively, fluctuations are of the order of the barrier height itself). What I mentioned was the possibility (by no means the only one) to see the effect of barrier fluctuations for the high temperature hydrogen diffusion in metals.

H. R. Schober (*KFA, Julich*). Given the adiabatic decoupling between H and the lattice motion, how are the anharmonic couplings between localized H vibrations and lattice vibrations accounted for?

N. V. Prokof'ev. I assumed in this approach that the difference in mass between the muon/proton and lattice atoms is big enough that we can ignore this coupling. I believe this is certainly a good approximation for the muon in metals.