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Non-Markovian spin dynamics driven by quantum coherence

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Abstract. The F–μ–F state provides a unique signature in muon spin rotation data and is observed in many fluorides. It gives rise to a characteristic oscillatory muon polarization that results from entanglement between the spins of the muon and nearby fluorine nuclei. If the muon hops from site to site, then the relaxation of this oscillatory signal can increase. The usual method to treat muon hopping in fluorides assumes the strong-collision approximation, which is then used to dynamicize the standard F–μ–F relaxation function. This approach presupposes Markovian dynamics and so neglects the quantum entanglement between a muon and a neighbouring spin to which it may still be strongly coupled, even after a hopping event. Entanglement that persists between a hopping event becomes even more important to include in the case when a muon subsequently hops back to its initial position. This article demonstrates the effect of including these coherent effects which results in dynamics that depart from those that are calculated using the strong-collision approximation.

1 Introduction

A muon implanted in a fluoride, even one which is not magnetically ordered, will show a coherent oscillatory muon polarization that results from the formation of a F–μ–F state [1] and which is produced by the quantum entanglement between the spin of the muon ($S = \frac{1}{2}$)and those of nearby fluorine (^{19}F, $I = \frac{1}{2}$) nuclei [2, 3]. The detailed shape of the oscillations is highly dependent on the local arrangement of fluorine ions (see e.g. [4, 5]), as demonstrated in Fig. 1, which shows the form of the oscillations that would result from the muon being implanted with different hypothetical local geometries. Measuring the shape of such oscillations in experiment therefore provides a fingerprint that can identify the muon site and provide a test of the accuracy of muon site calculations [6, 7]. The muon response is dominated by the arrangement of the very closest fluorine ions, but coupling with more distant fluorine nuclei can affect the long-time relaxation of the oscillations, as has been demonstrated particularly clearly in the case of CaF_2 and NaF, for which the time dependence can be very accurately modelled by including the effect of the more distant fluorine nuclei [3]. It is instructive to consider the simple linear F–μ–F state, whose polarization is shown in Fig. 2. If we could also measure the nearest-neighbour fluorine nuclear spin polarization, we would see that this also develops an oscillatory signal, as also shown in Fig. 2, as would the next-nearest-neighbour fluorines which also acquire a polarization, and hence exhibit an oscillatory signal, albeit one with much lower amplitude. This can be interpreted in terms of quantum information [8]: the muon is implanted in a pure state and acts like a qubit; subsequent interaction with nearby fluorine nuclei (initially unpolarized and hence in a mixed state) lead to the quantum information spreading out through the network of nearby nuclear spins, resulting in decoherence [3].

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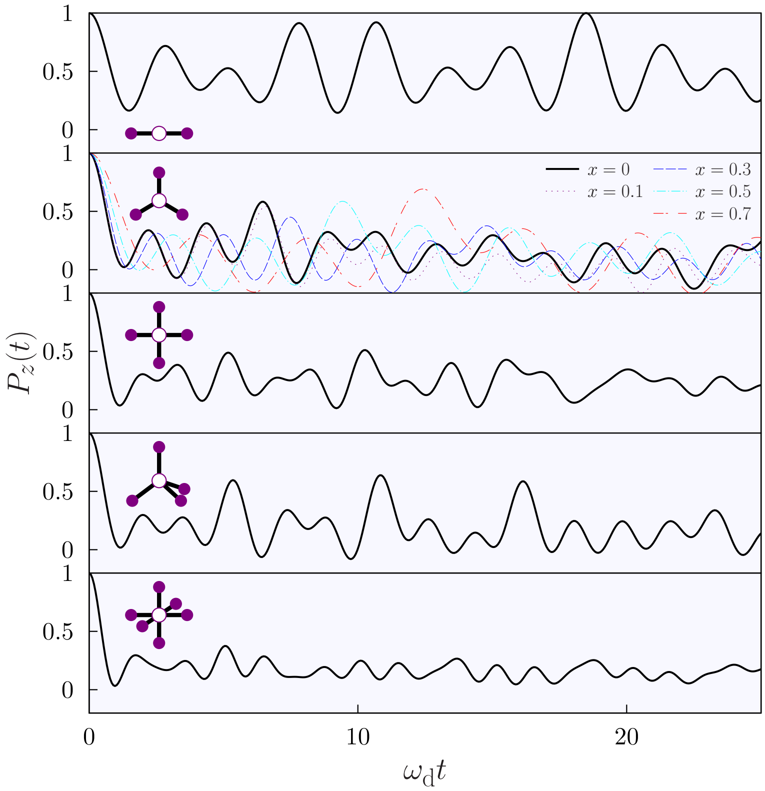

Figure 1: Muon response for several local geometries of F ions. From the top, these are the linear geometry (the F–μ–F bond), the triangular geometry (also shown is the effect of moving the muon a distance x out of the plane, where x is measured in units of the μ–F bond length), the square geometry, the tetrahedral geometry, and the octahedral geometry. Here $\omega_{\rm d} = \mu_0 \hbar \gamma_\mu \gamma_{\rm F}/(4\pi r_{\mu {\rm F}}^3)$, and all the symbols take their usual meanings.

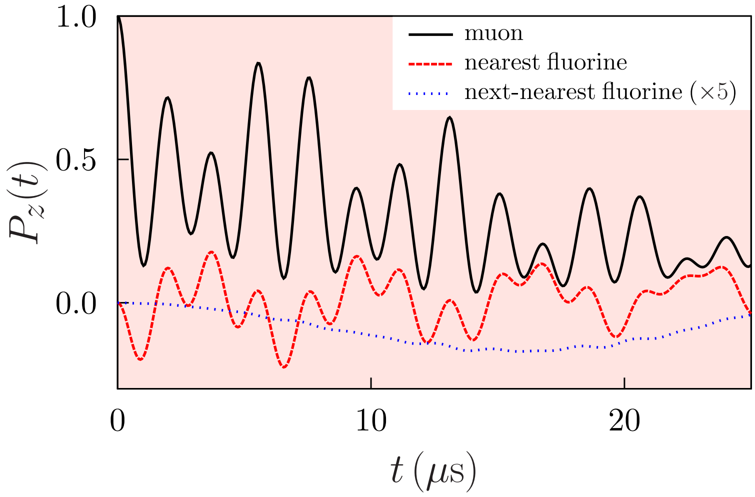

Figure 2: The calculated spin-polarization in a fluoride (such as CaF_2) following muon implantation. The muon is initially spin polarized, whereas the nearest-neighbour fluorines and next-nearest neighbour fluorines are unpolarized. As time increases, the fluorine ions begin to acquire spin polarization and this oscillates back and forth between the fluorine nuclei and the muon.

A separate question is the effect of muon hopping, which can occur at high temperature when thermally activated movement of the muon from one stable site to another can occur. This can result in increased relaxation of the oscillations in the muon polarization (see e.g. [9]. The effect of hopping is conventionally modelled using the strong collision approximation [10], which assumes that each hopping event is a Markov process in which the muon finds itself in an entirely new environment, uncorrelated with that of its previous location. This model assumes that muon relaxation follows two different time dependences. First, under normal situations between hopping events, the muon follows a static relaxation function $g_z(t)$, which in the present case would be a F–μ–F relaxation function, as given in Fig. 1. Second, at each hopping event (which is assumed to occur at a rate $\nu = \tau^{-1}$) the muon polarization is frozen, and then begins to relax again from this point. Any quantum information transferred to nearby nuclear spins is supposed to be instantly and irretrievably lost at the hopping event, never to return. So, if the hopping event occurs at time $t = t_1$, where the muon polarization has dropped to $g_z(t_1)$, the subsequent $(t > t_1)$ polarization function is assumed to be $g_z(t_1)g_z(t - t_1)$; the clock restarts at t_1, and the polarization can never again rise above $g_z(t_1)$. The dynamic relaxation function $P_z(t)$ is then given by

$$P_z(t) = \mathrm{e}^{-\nu t}\left(g_z(t) + \nu\int_0^t \mathrm{d}t_1\, g_z(t_1)g_z(t-t_1) + \nu^2\int_0^t \mathrm{d}t_2\int_0^{t_2}\mathrm{d}t_1\, g_z(t_1)g_z(t_2-t_1)g_z(t-t_2) + \cdots\right), \tag{1}$$

which is a sum of terms representing the effect of no hops up to time t, a single hop (at time t_1) up to time t (the single integral), a double hop (at times t_1 and t_2) up to time t, etc. There are methods for computing these integrals [10, 11] and the strong collision model has been used to describe the effect of hopping of muons in NaF [12].

However, it is worth questioning whether the Markov approximation is valid in this case. In a fluoride, a muon often sits between two fluorine ions [6], and so in a hopping event it will move to sit at another site between two fluorine ions. One of these two fluorine ions will be the same as at the original site, and so the muon will still be strongly coupled to it, and their mutual entanglement will naturally persist. Some of the polarization transferred to that fluorine has the potential, at least, to be transferred back to the muon, even though some polarization might also be transferred to more distant nuclei. Is it therefore really correct to completely ignore this fact and pretend that the muon's new environment is entirely uncorrelated with its old one? Clearly not. This paper examines what effect including these persisting entanglements could have.

2 Coherent model

The coupling of the muon with its local environment of n fluorine nuclear spins can be considered fully quantum mechanically in a Hilbert space of dimensionality $D = 2^{n+1}$. The density matrix of the system at $t = 0$ is written as

$$\rho_0 = \frac{1}{D}(\mathbb{1}_\mu + \sigma_z^\mu) \otimes \left(\bigotimes_{i=1}^{n} \mathbb{1}_i\right). \tag{2}$$

This means that the muon is initially in a pure state (polarized along z) and the n ^{19}F nuclear spins are in mixed states (with $\mathbb{1}_i$ being a 2×2 unit matrix). Markovian hopping demands that the *fluorine coherences are zeroed at every hopping event*, and the density matrix is reset to $P_z^\mu(t)\rho_0$, clearly a gross simplification.

The new model preserves all the coherences. All that happens at a hopping event is that the muon moves. This means that the vectors joining the muon and neighbouring spins need to be updated appropriately, and the full dipolar Hamiltonian $\mathcal{H}$ (and hence the time evolution operator $\mathrm{e}^{-\mathrm{i}\mathcal{H}t}$) is similarly updated. The time-evolved muon polarization after implantation in the absence of hopping is $P_z^\mu(t) = \frac{1}{D}\mathrm{Tr}\left[\mathrm{e}^{-\mathrm{i}\mathcal{H}t}\sigma_z^\mu \mathrm{e}^{\mathrm{i}\mathcal{H}t}\sigma_z^\mu\right]$ [3], but with hopping one simply evolves the density matrix according to the continually-updated Hamiltonian. Full angular averages of the local geometry are performed in all the calculations presented in order to demonstrate the effect of polycrystalline averaging. The simulations will assume $n = 4$, so that the muon is located on one side of a square of fluorine ions, appropriate for CaF_2; the Hilbert space dimensionality is $D = 2^{n+1} = 64$, so the calculations are straightforward to perform. The bond-length parameters used are for CaF_2, including the muon-induced distortion that pulls the nearest-neighbour fluorine ions closer to the muon [1, 6, 3].

3 Results and discussion

To illustrate the difference this effect makes, Fig. 3 shows the calculation for no hopping, Markov hopping and coherent hopping, under the unrealistic assumption that hopping events occur every 5 μs. Unsur-

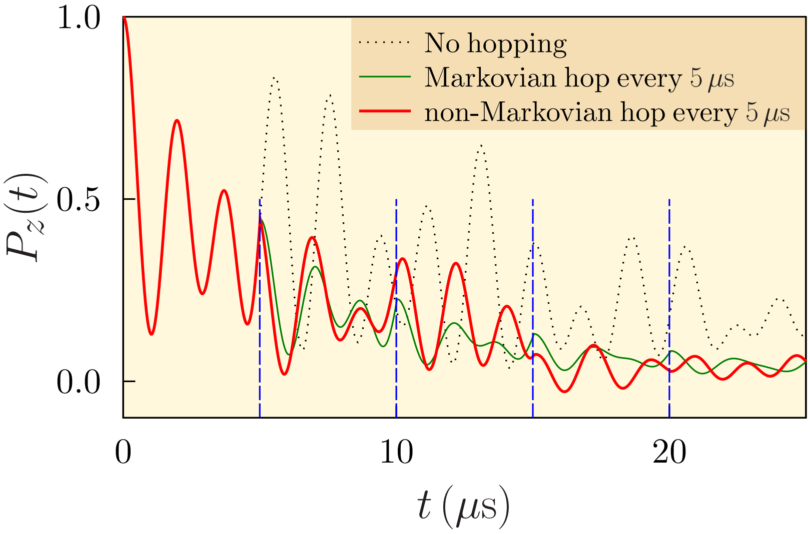

Figure 3: The effect of Markovian and coherent hopping. To illustrate the effect, the hopping is artificially forced to occur every 5 μs

prisingly, all three traces agree before the first hopping event occurs at $t = 5$ μs. In both the Markovian and coherent models, the effect of hopping is to add relaxation, and so most of the structure seen in the "No hopping" case is reduced. (Note that even the "No hopping" simulation has some relaxation, due to the effect of the two next-nearest neighbour spins that are included in the calculation.) However, the key takeaway is that Markovian hopping relaxes much faster than the coherent case. At each hopping event, the Markov model restarts the characteristic F–μ–F relaxation function, but at a reduced level (their form slightly changes due to an angular averaging effect). The non-Markovian coherent model retains more long-time structure because the fluorine coherences are not artificially zeroed.

The main results of this paper are shown in Fig. 4 and for these simulations, the hopping times follow an exponential distribution with mean hopping time τ (which is the inverse of the fluctuation rate $\nu = \tau^{-1}$). The first simulations in Fig. 4(a) show the Markovian model which assumes the strong-collision approximation and zeros the fluorine coherences at every hopping event. When $\tau = \infty$ we obtain the static relaxation function for the system (which will be identical in all models), but for finite τ the oscillations become progressively damped as τ decreases, until the relaxation becomes close to exponential. Thereafter, further decreases in τ lead to the relaxation rate falling, as we enter the well-known fast-fluctuation limit [10]. This observed behaviour is entirely consistent with earlier work on muon hopping in NaF [12].

The new model allows the muon to hop and keeps all the coherences intact. In Fig. 4(b), the results of a 2-state model are shown in which the muon hops from one site to another and back again. When τ is large it looks quite similar to the Markovian case, but for small τ we no longer reach the fast-fluctuation limit, but new oscillatory structure develops. This is essentially because the muon is becoming strongly entangled with all spins in the system, particularly with the fluorine on the top-left corner (to which it is always a nearest neighbour) but also with two others (top-right and bottom-left, to which it is a nearest-neighbour half of the time). If we allow the muon to hop all the way around the square, each hop either clockwise or anticlockwise with a 50% probability, we obtain the 4-state model shown in Fig. 4(c), which shows something very similar. Now at short τ the muon becomes entangled equally with all four nuclei, so the resulting polarization also contains oscillatory structure, but at a lower frequency than in Fig. 4(b), since now the muon is a nearest-neighbour to each of the four spins only one half of the time on average.

These results clearly demonstrate the effect that retaining entanglement can have and results in a less drastic relaxation of the coherent oscillations than is found in the fully Markovian model. Nevertheless, this oscillatory behaviour is not seen in data on real inorganic fluorides and is an artefact of the finite size of the system being considered in this case. The problem is of course that the quantum information has no possibility of escape and has to reside in some distribution over the five spins (muon plus four fluorines)

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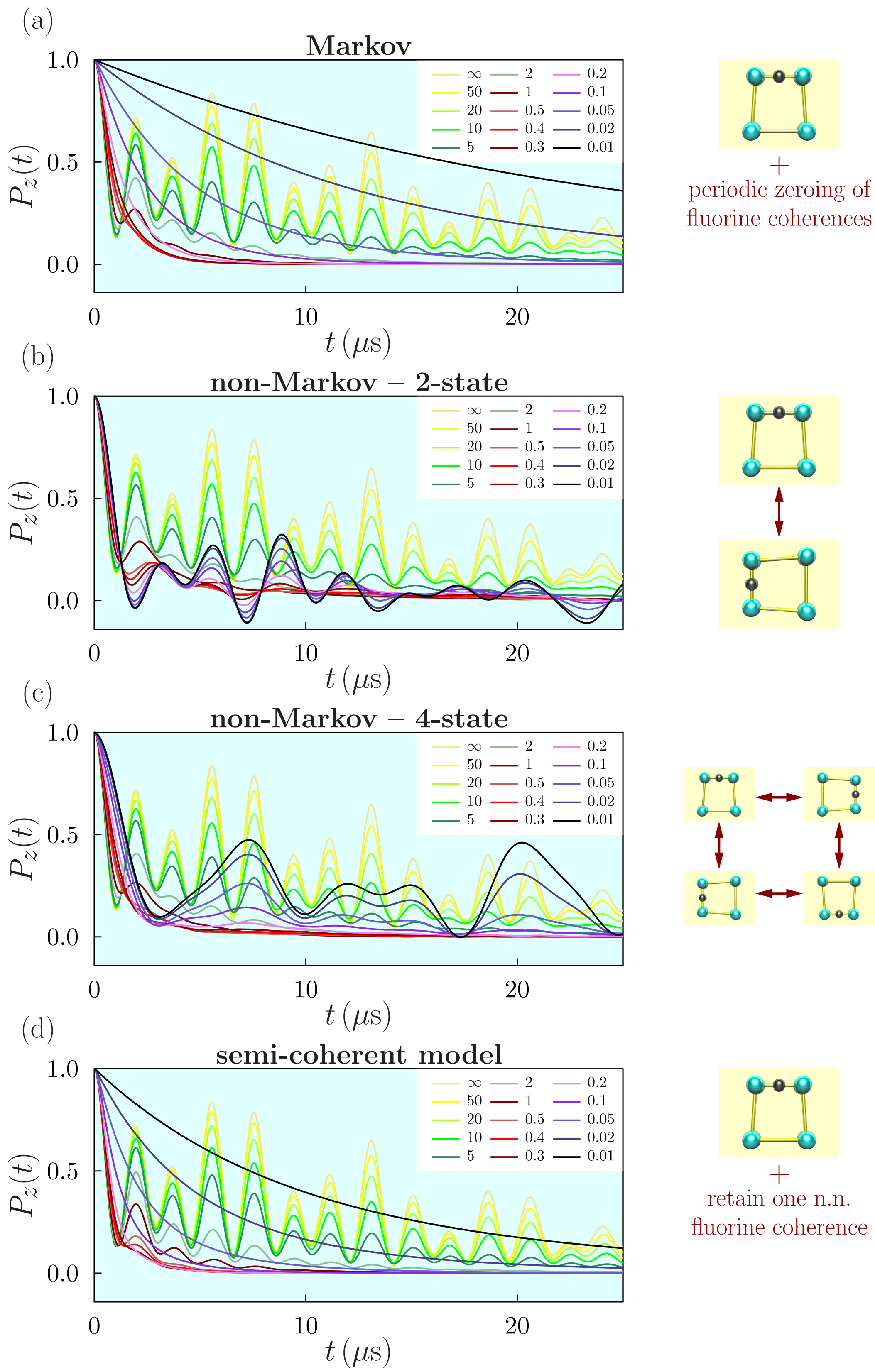

Figure 4: Various hopping models plotted for different values of the hopping time $\tau = \nu^{-1}$ measured in μs. (a) The Markov model assumes that the fluorine coherences are zeroed at every hopping event. (b) Coherence is now retained but the muon position hops back and forth between two adjacent sites. (c) The same as (b) but the muon hops between four sites. (d) For the semi-coherent model, only a single fluorine coherence is zeroed.

in this model; in a real three-dimensional lattice, it would have the option to leak away. To obtain a more realistic model, it is necessary to set the problem on an infinite lattice, but then it is not possible to include the coherence with every nearby spin as the Hilbert space becomes too big. Nevertheless, in what can be called the semi-coherent model, the infinite lattice is included (dropping the restriction to study coherences on a finite part of the lattice) and this leads to the polarization shown in Fig. 4(d). For every hopping event in this model we choose one nearest-neighbour fluorine at random and keep its coherence with the muon; all other coherences are zeroed. This therefore accounts for the fact that in a hopping event the muon will retain one nearest-neighbour fluorine and will maintain coherence with it (for those familiar with celidhs, one can think of this as a "Scottish-country-dancing effect", in which each dancer has two nearest-neighbour partners, but swaps out one of them at the end of a verse). Zeroing of the coherences on a fluorine is performed by tracing out the density matrix (dimension 64×64) over that spin degree of freedom and then re-inflating the density matrix with a tensor product with the identity matrix, taking care to ensure that the identity matrix appears in the correct position. The results shown in Fig. 4(d) show that the fast-fluctuation limit is recovered, and the simulations look superficially like those of the Markov model in Fig. 4(a). However, although qualitatively similar, they are quantitatively different, with dynamics having a weaker effect. A Markov hop is a much more drastic event than a semi-coherent hop in which only half the coherences are lost in a hop. Although that loss is still irretrievable, the most important coherence (between the muon and the nearest-neighbour fluorine which is invariant in the hopping process) is retained. The clear implication of this is that, where coherence is retained in the physical system, a Markovian model will overestimate the effect of hopping and thus imply a hop rate that is *lower* than the actual hop rate.

4 Conclusion

In conclusion, the standard Markovian model of hopping ignores quantum coherent effects which the present analysis shows can be significant. The coherent model presented here captures the insight that the muon injects quantum information into a system of nuclear spins, and quantum coherences that are subsequently built up over time affect the muon polarization, even when the muon hops because entanglements persist. A further feature that can be captured by these models is that orientation of a F–μ–F bond can rotate by 90° during a hopping event. It is also possible to mix Markovian and non-Markovian hopping into these types of model. These ideas can be straightforwardly introduced into even more realistic models in which the muon hopping is considered over a real lattice and a subset of coherences are retained, rather than carelessly discarded. Although coherent effects are most obvious in fluorides, the principle should apply to all nuclear spins and therefore the implications of considering these coherent phenomena may be of much wider relevance in muon diffusion problems.

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