Complete nuclear dipolar line shapes for high transverse field $\mu$SR

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Abstract

It is common in analysis of transverse field TF-$\mu$SR data to assume that the line shape contribution of the nuclear spin lattice is Gaussian. Yet, evaluation of the muon-nuclear dipolar Hamiltonian is trivial in the high field limit subject to conditions of the TF-$\mu$SR experiment. Here we clarify the experimental requirements needed to satisfy the high field limit, and point the reader to previously published calculations in this regime. We describe our calculation method and present line shapes for the tetrahedral and octahedral sites in copper for external magnetic field directions parallel to the $\langle 100 \rangle$, $\langle 110 \rangle$ and $\langle 111 \rangle$ crystal axis. Second moments of our calculated line shapes agree with second moments calculated by Camani et. al by the method of Van Vleck.

As illustrated by the calculated line shapes, dipolar broadening at the muon site is highly sensitive to the direction of the external magnetic field. Judicious choice of the external field direction can be used to minimize dipolar broadening and departures of the line shape from a gaussian character. The calculation is a valuable tool to predict field dependence for a given muon site. Alternatively, it may be used to determine the muon site from experimental data obtained at various field orientations. In situations where the line shape is not well fit by a Gaussian and/or where the muon induces lattice distortion, the calculation is a valuable tool to better fit the $\mu$SR data.

Keywords: Line shape and width, Spin Hamiltonians, Muon spin rotation, Secular limit, Local field approximation

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1. Introduction

The positively charged muon, $\mu^+$, when implanted in a solid, typically comes to rest within a few nanoseconds at an interstitial site, surrounded by a lattice of nuclei. If the nuclei posses intrinsic spin angular momentum, then magnetic dipole–dipole coupling of the muon to each nucleus contributes to the magnetic field at the muon site. Let the spin of the $j^{th}$ nucleus be $I_j$ and let its gyromagnetic ratio be $\gamma_j$. For the muon, let $S_\mu$ and $\gamma_\mu$ be the spin and gyromagnetic ratio, respectively. The Hamiltonian for dipole–dipole coupling of the muon to a lattice of $N$ nuclei is

$$H_{\text{dip}} = \sum_{j=1}^{N} \frac{\mu_0 \gamma_j \gamma_\mu}{4\pi r_j^3} \left[ I_j \cdot S - \frac{3(I_j \cdot r_j)(S \cdot r_j)}{r_j^2} \right],$$

where $r_j$, of length $r_j$, is the position vector connecting the muon site to the $j^{th}$ nucleus. In general, the spin of each nucleus interacts with other spins in the lattice, and $H_{\text{dip}}$ is a many body Hamiltonian, implicitly dependent on nuclear
spin–spin interactions, via the $I_j$ operators. In the presence of a strong external magnetic field, and for time scales typical of the $\mu$SR experiment, $H_{dip}$ may be treated as a static perturbation of a simple Zeeman Hamiltonian of non interacting magnetic dipoles. The appropriate constraints for the static perturbation approximation of $H_{dip}$ are

1. The applied field must be sufficiently strong that the quantization axis for the muon and nuclear spins can be taken to be parallel to the applied field. This ensures that Zeeman eigenstates including nuclear spin degrees of freedom are not significantly perturbed by possible quadrupolar interactions within the nuclear lattice. Also, the presence of an external field prohibits spin flip–flop interactions of the muon with the nuclei, since $\gamma_\mu \neq \gamma_j$

2. The time scale over which non-diagonal nuclear–nuclear dipolar couplings evolve must be long compared to the muon lifetime, since the calculation does not account for energy conserving spin flip–flop interactions among the nuclei. We note that this constraint may be loosened in particular scenarios where the magnetic field gradient produced by the muon magnetic moment suppresses spin flip–flop interactions among nuclei in its near vicinity. If the nuclei are asymmetrically placed with respect to the muon site, the spin flip-flop terms can no longer conserve energy and are suppressed.

Let the $\hat{z}$ direction be parallel to the external field $B_0$. The Zeeman Hamiltonian is then $H_o = \gamma_\mu S^z B_0 + \sum_{j=1}^{N} \gamma_j I_j^z B_0$. We define $H'_{dip}$ to be the appropriate form of Eq. (1) that satisfies $[H_o, H'_{dip}] = 0$ as follows:

$$H'_{dip} = \gamma_\mu S^z \cdot B_{dip}^z,$$

where

$$B_{dip}^z = \sum_{j=1}^{N} B_j^z = \sum_{j=1}^{N} \frac{\mu_0 \gamma_j}{4\pi r_j^3} (1 - 3 \cos^2(\theta_j)) I_j^z,$$

and $\theta_j$ is the angle of $\mathbf{r}_j$ with respect to $B_0$. Because it contains no spin flip–flop interactions, $H'_{dip}$ is a significant departure from the many body spin Hamiltonian treated in Van Vleck’s seminal derivation of the 2nd and 4th moments for the line shape of a crystalline spin lattice [1]. Since all operators in Eq. 3 commute with each other, the line shape or its fourier transform, the transverse field polarization function, can be evaluated in closed form [2].

In 1957, Lowe and Norberg derived the the equivalent of the TF-$\mu$SR polarization function in the high field limit and in the local field approximation for a lattice of identical spin $1/2$ nuclear dipoles [3]. Later a polarization function for identical nuclei in a cubic lattice, which allowed for any integer or half integer nuclear spin was derived by Gade and Lowe [4]. Gade and Lowe’s result was used by Cameron and Scholl to calculate the high TF-$\mu$SR polarization function for the octahedral and tetrahedral sites in a cubic crystal of identical nuclei [5]. Later, Cameron and Sholl specifically calculated the polarization function for the octahedral site in copper [6]. Unfortunately, they did not reference the empirical TF-$\mu$SR data of Camani et. al [7] and attention of the $\mu$SR community was not drawn to their work. Camani’s data also points to the significance of lattice distortions, which a practical calculation should incorporate. A particularly striking example of lattice distortion is evident in the TF-$\mu$SR data for NaF. Brewer et. al directly calculated the nearest neighbour dipolar field at the muon site in order to fit the TF-$\mu$SR data [8].

For our own calculations, we evaluate Eq. (3) in the basis states of the Zeeman Hamiltonian [9],

$$\sum_{j=1}^{N} \langle k_j | B_j^z | k_j^\prime \rangle = \sum_{j=1}^{N} \frac{\mu_0 \gamma_j h}{4\pi r_j^3} (1 - 3 \cos^2(\theta_j)) \delta_{k_j,k_j^\prime} m_j^k,$$

where $\langle k_j \rangle$ with eigen value $\hbar m_j^k$ is one of the $2I_j + 1$ eigen states of nuclear spin operator $I_j$, chosen at random, given that the nuclear polarization is negligible at typical TF-$\mu$SR temperatures and external magnetic field values.

Each magnetic field value generated by evaluation of Eq. (4) corresponds to one possible spin configuration of the spin lattice (i.e. one set of $\langle k_j \rangle$ eigen vectors, $j=1 ..N$). To arbitrary resolution, the complete line shape may be constructed as a histogram of the magnetic field values corresponding to all possible spin lattice configurations. While each magnetic field value may be readily calculated, the complete set, for a lattice of $N$ nuclei requires a prohibitively large number of calculations ( i.e.roughly $2^N$ calculations for a set of spin $1/2$ nuclei within 2 lattice spacings of the muon site). A computational compromise is to randomly generate a subset of the lattice configurations that will converge to the exact result for increasing sampling.
2. Calculation

The calculation is performed over a rectangular volume containing $N$ nuclei, centered on the unit cell which includes the muon site. A single lattice configuration is generated by randomly selecting for the $j^{th}$ nucleus, $j \in \{1, 2, \ldots, N\}$, one of its $2I_j + 1$ eigen states. The isotopic abundance of each nucleus determines the probability with which the spin and gyromagnetic ratio corresponding to a given lattice site is selected. Then, for each lattice configuration Eq. (4) is evaluated.

We take into account the fact that the coulomb interactions, which determine the muon site, may be symmetrical about one or more crystallographic axis, so that there are a number of electronically equivalent sites where the muon may reside. The line shape we generate is a sum, over all equivalent muon sites, of the line shape histogram generated for each muon site. The code also accounts for possible lattice distortion due to the presence of the muon, by specifying the locations of the nearest neighbour nuclei with variables that are independent of the lattice constants.

3. Results and Discussion

![Unit cell of FCC copper and sites of tetrahedral (A) and octahedral (B) symmetry.](image)

We have calculated line shapes for the tetrahedral and octahedral sites in face centered cubic (FCC) copper (see Fig. 1). The line shapes are shown for each of 3 field directions in Fig. 2. The bold lines are for a finite lattice (FL) centered on the octahedral calculation site. The relatively faint lines are for nearest neighbours (NN) only. We have calculated the second moments $M_2$ for each line shape and corresponding damping rate $\sigma$ from the relation $M_2 = 2\sigma^2/\gamma^2_\mu$. The $\sigma$ values for the FL and NN line shapes agree to within 1% with those derived from 2nd moment calculations of Camani et al. [7]. Following Camani’s format, we report the $\sigma$ value for each NN line shape in brackets as a percentage of the $\sigma$ value for the corresponding FL line shape.

![Calculated line shapes in FCC copper for crystal sites with (a),(b),(c) octahedral and (d),(e),(f) tetrahedral coordination.](image)

Figure 2: Calculated line shapes in FCC copper for crystal sites with (a),(b),(c) octahedral and (d),(e),(f) tetrahedral coordination. The bold line is for the finite lattice (FL) of 729 unit cells. The lighter lines are for nearest neighbour (NN) nuclei only. Damping rates $\sigma$ and $\sigma_{NN}$, as discussed in the text, are also shown for the FL and NN line shapes respectively.
In Fig. 3(a) and 3(b), Fourier transforms of the finite lattice (FL) line shapes in Fig. 2 are compared with Gaussian curves of the form $e^{-\sigma^2 t^2}$, where each $\sigma$ value is the decay rate in $\mu$s shown to the right of an FL line shape in Fig. 2. The lowermost Fourier transform in 3(a) becomes negative beyond 4 $\mu$s, confirming the non-gaussian character of the corresponding FL line shape in Fig. 2(d). Otherwise the Gaussian approximation is reasonable and even optimal, as for the Fourier transforms of Fig. 3(a): B $\parallel$ (100) and 3(b): B $\parallel$ (111). Note that the corresponding $\sigma_{NN}$ values are zero in Fig. 2(f) and (c) respectively.

By applying the calculation to hypothetical sites of low symmetry in various crystals, we note that the line shape can be very far from Gaussian when the number of nearest neighbours is low and/or there are muon induced lattice distortions. In such cases, fit routines that can reference the complete line shape rather than a few moments are clearly advantageous. The calculation is also a valuable tool for effectively predicting the dependence of the line shape on the external field angle for a given muon site and may be used to determine the muon site from experimental data at various field orientations. At present we are working on incorporating our calculation into a generic fit routine for TF-$\mu$SR data. This will allow for a precise accounting of the nuclear contribution to the TF-$\mu$SR line shape when the muon site is known, and will aid in site identification when the muon site is not known a priori.

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References