Complete nuclear dipolar line shapes for TF-μSR in the limit of high magnetic fields

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Characterizing the magnetic field line shape of a nuclear lattice by TF-μSR

Transverse field muon spin resonance spectroscopy, **TF-μSR**, is a very sensitive magnetic probe of fields that are *internal* to a sample. The analysis of TF-μSR data requires that contributions to the local field be identified and properly characterized so that the magnetism of interest may be isolated.

Much of our research involves **crystalline solids**, and we must account for the magnetic field distribution, or ***line shape***, at the muon site due to the surrounding lattice of nuclear magnetic dipoles. **Calculation** of this line shape is a many body problem for which an exact solution is generally intractable. The many body problem includes so called "flip-flop" terms which are *energy conserving* exchanges of spin direction between dipoles (*i.e.* a "down" spin flips up and an "up" spin flops down) .

However, unique properties of the **TF-μSR** experiment allow one to **drop flip-flop terms** when calculating, to a very good approximation, the nuclear dipolar fields at the muon site.

Firstly, the dipole moment of the muon, and therefore its Zeeman energy in a given field, is more than 3 times that of any nucleus, so that flip-flop interactions of the muon with neighbouring nuclei do not conserve energy, and may not occur in the presence of an external field. **Secondly**, the dipolar field of the muon creates a magnetic field gradient in its vicinity which suppresses energy exchange amongst nuclei near the muon. **Finally**, the rate at which the nuclei exchange energy is slow compared to the muon lifetime, so that the nuclear field at the muon site is essentially static.

Further simplifications are relevant when a sufficiently strong external field is applied such that the quantization axis for both the muon and the nuclei is approximately parallel to the external field. In practical terms, the **external field** must be **large** compared to the nuclear dipole field at the muon site. Also, the Zeeman energy of the nuclei in the external field must be large compared to any nuclear-nuclear or nuclear quadrupolar interactions. Typically, an external field of a few hundred gauss is required.

Given a sufficiently strong external field, the nuclear dipolar contribution to the local field at the muon site, as measured in a TF-μSR experiment is given by:

$$\sum_i \frac{\mu_0 \mu_i}{4\pi} \frac{[3(\hat{r}_i \bullet \hat{z})^2 - 1]}{|\vec{r}_i|^3} \hat{z}$$

- $\hat{z}$ is the direction of the external magnetic field
- μ_i is the nuclear magnetic moment at the i th lattice site
- $\vec{r}_i$ is the vector displacement of the i th lattice site from the muon site
- $\hat{r}_i$ is a unit vector parallel to $\vec{r}_i$

Determining the muon site by TF-μSR

Note that the numerator of the i th term of the sum above is zero for $(\hat{r}_i \bullet \hat{z}) = \cos(54.7^o) = \sqrt{3}^{-1}$

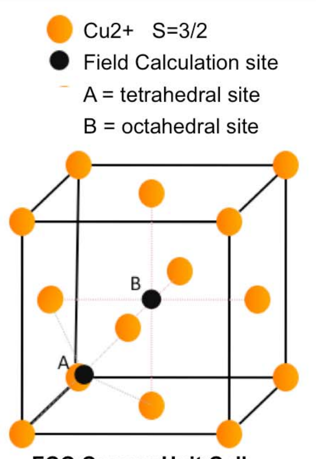
In cases where the muon has a **single** nearest neighbour, as for several proposed muon sites in $La_{2-x}Sr_xCuO_4$ (LSCO), the nuclear line shape depends ***very sensitively*** on the **external field direction.** The magnetic field direction can be chosen to **negate** the dipolar field contribution of the muon's nearest neighbour. If this external field direction is approached in a series of TF-μSR experiments**,** significant narrowing of the field distribution should be apparent, if the muon is at the proposed site . Alternatively, a single μSR experiment may be sufficient to **rule out** several sites as possible hosts for the muon.

In the calculations shown below for Cu and LSCO the external field direction is varied for select muon sites and the sensitivity of the field distribution on the external field angle is clearly demonstrated.

FCC Cu : Calculations compared to published 2nd moments

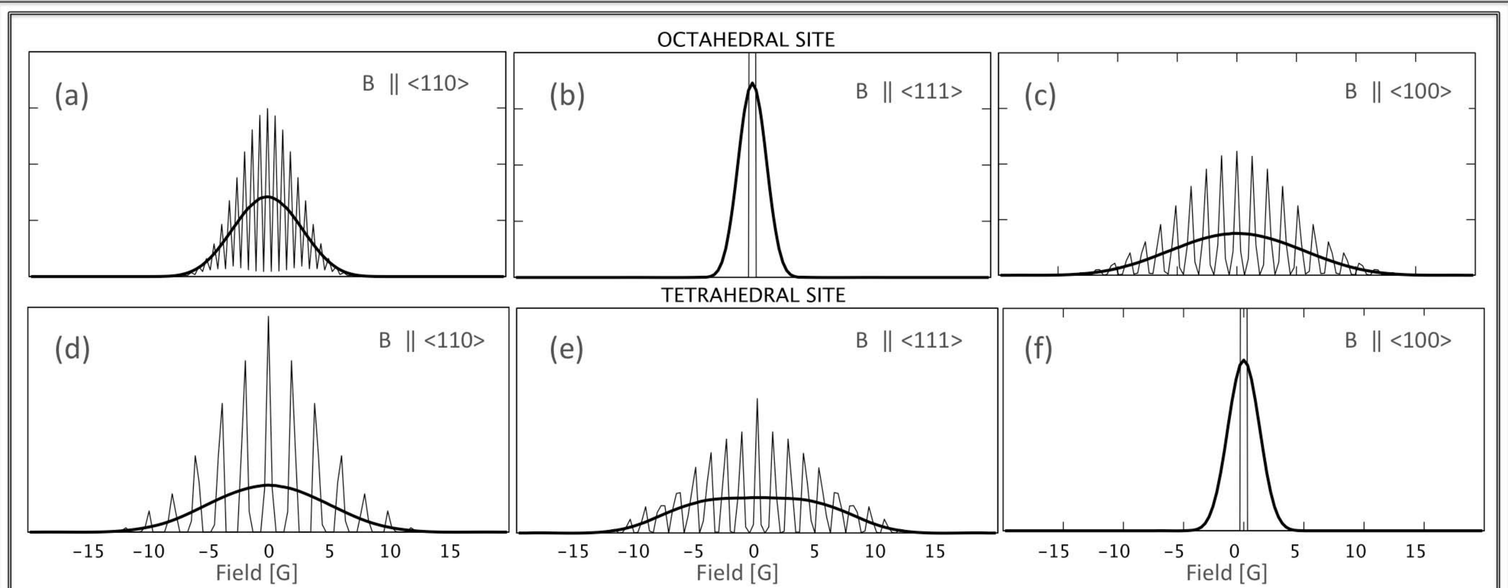
Initial testing of the line shape calculation

For the sake of trouble shooting numerical errors, and to validate the expression used for the line shape calculation, published data for nuclear dipolar fields as measured or calculated for the **TF-μSR** experiment was compared to calculated line shapes for **Cu** (see right) and for **NaF** (below)



FCC Copper Unit Cell

The value of the calculation, for characterizing nuclear fields, can be appreciated in those line shapes (*see right* "(e)") that are markedly **non-gaussian**, since a gaussian line shape is the approximate form most commonly used in fitting TF-μSR data



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 a cubic lattice of 729 unit cells. The lighter lines are for nearest neighbour nuclei only.

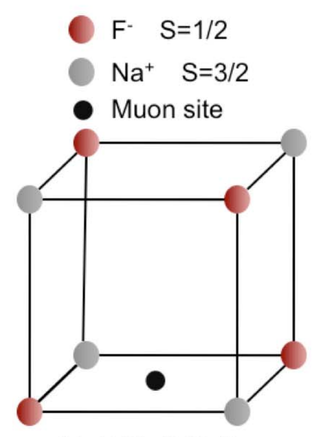
Line widths at select sites in FCC copper

SITE	External field parallel to (100) axis	(110) axis	(111) axis
Octahedral site	**0.308 μs⁻¹** 5.11 Gauss	**0.165 μs⁻¹** 2.74 Gauss	**0.068 μs⁻¹** 1.13 Gauss
Tetrahedral site	**0.077 μs⁻¹** 1.28 Gauss	**0.279 μs⁻¹** 4.63 Gauss	**0.318 μs⁻¹** 5.28 Gauss

The above line widths, derived form the 2nd moments of our calculated line shapes are in complete agreement with previously published values. See :

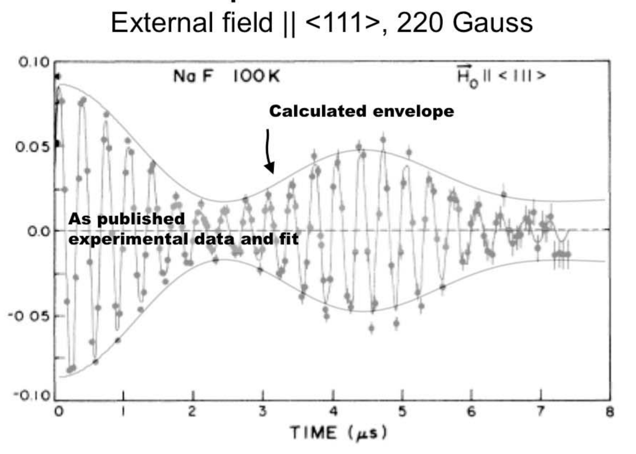
"Positive Muons in Copper…" Camani PRL 39 (1977) 836

NaF: Calculations compared with TF-μSR experimental data



NaF Unit Cell

TF-μSR data for NaF
External field || <111>, 220 Gauss

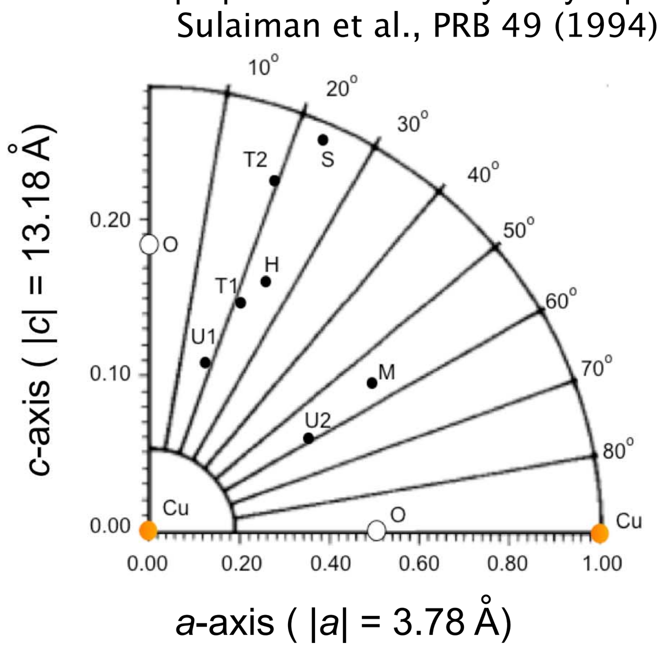


On the left, we see experimental TF-μSR data. The relatively high frequency oscillations visible in the data are due to Larmor precession of the muon in an applied field of 220 G. The relatively low frequency beat is characteristic of the line shape at the muon site. The distance of the **μ⁺** to its nearest neighbour **F⁻** ions determines the beat frequency. This **μ⁺ - F⁻** distance is greatly contracted due to coulomb attraction.

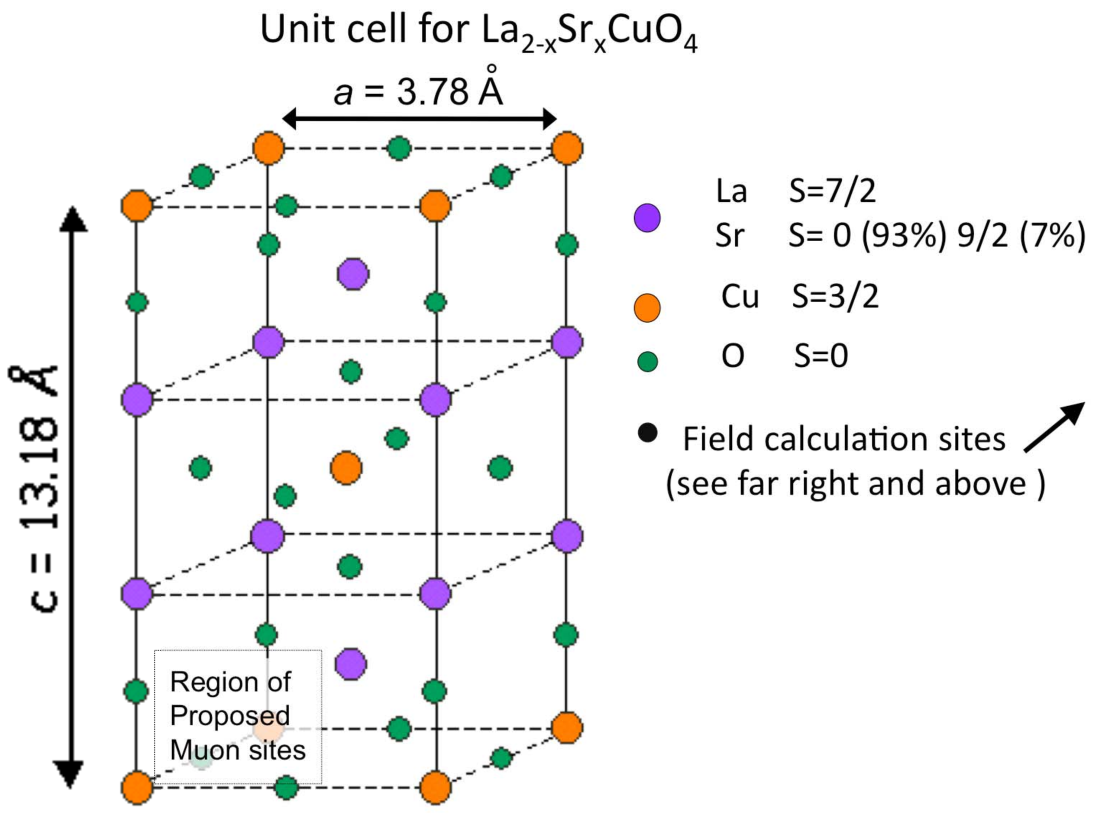
Our calculation accounts for lattice distortion due to the presence of the muon. In **NaF,** the distance of the **μ⁺** to its nearest neighbour **F⁻** ions is rougly 70% of the distance in the unperturbed lattice. Taking this lattice disortion into account, we have calculated the line shape for the muon site in **NaF**. See "**Calculated envelope**" on the left, which is the Fourier transform of the calculated line shape.

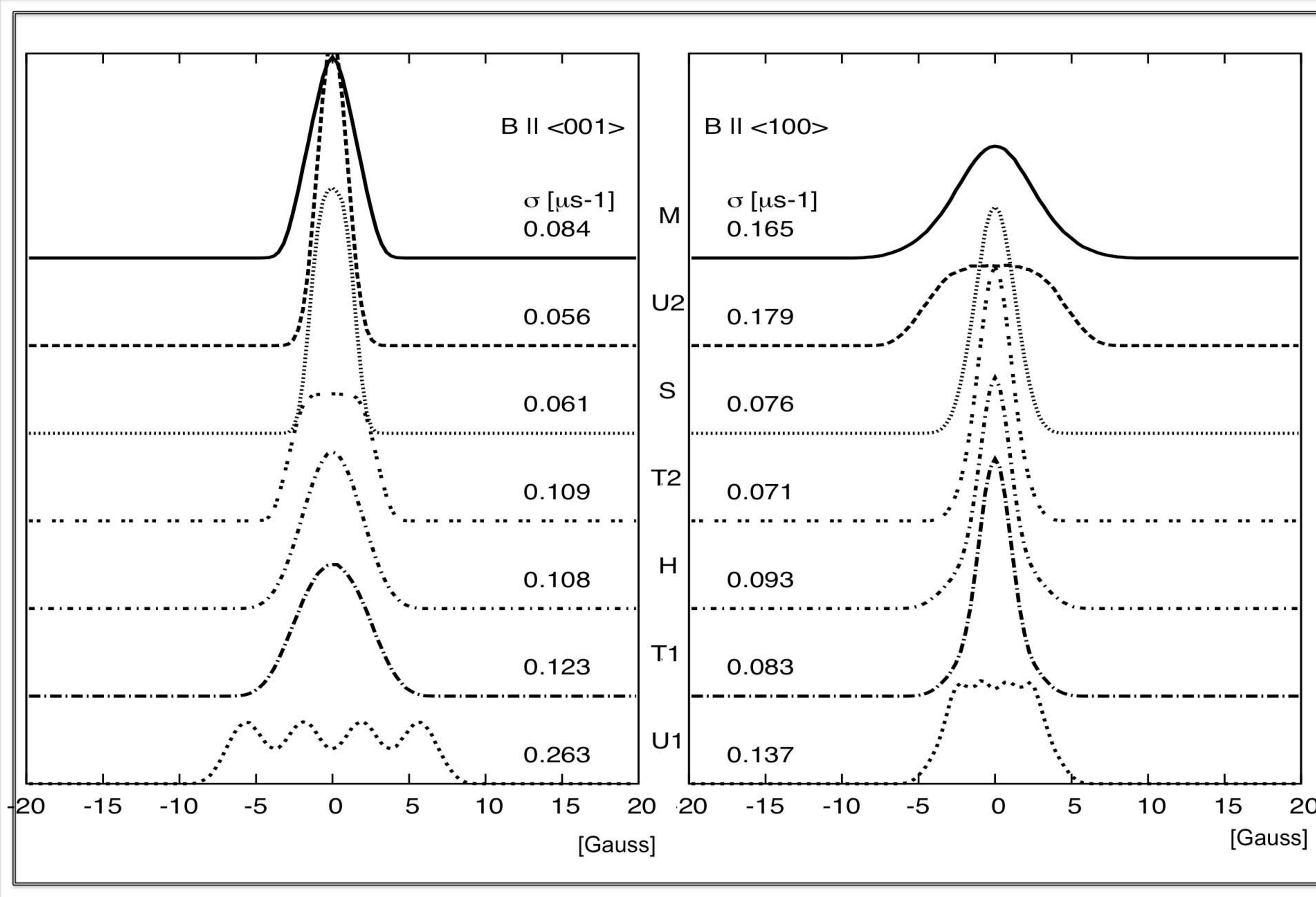
Experimental data source:
Brewer, J. H. *et al*, PRB 33 (1986) 7813 (R)

Proposed muon sites (•) in the *a*-*c* plane of the **LSCO** unit cell as proposed theoretically or by experiment
Sulaiman et al., PRB 49 (1994) 9879



LSCO: Calculation of the nuclear field distribution for proposed muon sites.





Lines shapes at proposed muon sites in **$La_{2-x}Sr_xCuO_4$**

On the left, calculated line shapes are shown for the sites displayed above, for two external field orientations: field parallel to the *c*-axis (left) and *a*-axis (right). The width "σ" of each line shape is also shown.

By comparison of line widths in the left and right panels for a given site, we see that the magnetic field dependence of the TF-μSR line shape is unique for each muon site, and thus in principle can be used to determine the precise location of the stopped muon in the sample.

The field dependence of the line width that we extract from fits to TF-μSR data most nearly agrees with calculated line widths for the U1 site. However, the magnitude of the experimentally derived line width suggests a muon site between T1 and U1.

Currently we are incorporating our calculation into a generic fit routine for TF-μSR data, to aid in our search for the muon site in **$La_{2-x}Sr_xCuO_4$** and to allow, in general, a precise account of the nuclear dipole contribution to the TF-μSR line shape in single crystals.