DOES COHERENT TUNNELLING OF POSITIVE MUONS CAUSE MOTIONAL NARROWING?

A.M. STONEHAM
Theoretical Physics Division, AERE Harwell, Didcot, Oxon OX11 ORA, UK

Received 13 January 1983

Recent work by Yaouanc has suggested that coherent tunnelling has no effect on the linewidth from dipolar interactions with magnetic nuclei. We show this is not correct, ultimately because an implicit assumption is not appropriate for the short-lived muon. Thus the linewidth does depend on the extension of the muon wavefunction.

Resonance lines broadened by dipolar interactions are a common phenomenon. The best studied cases are the nuclear magnetic resonance lines, for which Van Vleck's [1] classic analysis gives expressions for the moments of the shape. Recently work on muon spin resonance [2,3] stimulated questions of whether the linewidth depended on the number n of sites over which the muon moved coherently. Yaouanc [2] argued that there was no dependence on n, i.e. that the extension of the μ^+ wavefunction did not matter. This appears to be wrong, since it ignores an important difference in the nuclear magnetic and muon resonance experiments.

Van Vleck's analysis is given most fully for like spins interacting. When one considers just a single spin A interacting with many spins B of different g-factor, it becomes clear that two types of ensemble average are involved, namely:

(a) For a given site of spin A, there is a sampling of all possible states of spins B;

(b) For a fixed configuration of spins B, the spin A can be moved to monitor all sites.

If spin A lives long enough for the configurations of spin B to be sampled properly, these two averages should be identical. This is the case in the magnetic resonance of stable nuclei. However, for a muon in a typical solid, the muon lifetime (about 2 μs) is much less than typical nuclear magnetic reorientation times (normally ms). Thus each muon A samples a frozen nuclear spin configuration B, and this leads to two distinct types of sampling.

In essence (ignoring limitations because only a few detectors are used), a muon spin rotation experiment monitors the spin reorientation angle and time delay between entry and decay for each muon. For present purposes one can regard the experiment as a measurement of the effective magnetic field ($H_{applied} + h_A$) in which muon A precesses. There are two distinct samplings of the internal field in the solid:

(c) For a given nuclear spin configuration B, h_A depends on whether the muon is localised at a single site or whether it tunnels over a group of N sites $\{i\}$, since the nuclear spin environment will differ among these sites. This is discussed quantitatively below.

(d) Successive muons are injected at different times, and may end up in different parts of the target, so that the nuclear spin configuration B may change during an experiment involving many muons. This sampling parallels that for nuclear spins [(a) and (b) above]. Indeed, if the muon localises on a single site, the muon and nuclear cases are equivalent.

Yaouanc argues that coherent tunnelling, i.e. the effect of (c) above, does not affect the observed spread in h_A. We argue now this is wrong, and that tunnelling does reduce the observed width.

The effective local field h_A for muon A is the expectation value $\langle \Phi_A | h_B(R) | \Phi_A \rangle$ of a field $h_B(R)$ which varies in space but which is constant in time for a fixed nuclear spin configuration. For simplicity, we make two working assumptions. We write the muon wavefunction Φ_A in a tight-binding form, with i labelling the interstitial sites over which coherent tunnelling occurs:

$$\Phi_A = \sum_{i=1}^{n} w_{Ai}\phi_i \, , \tag{1}$$

and assume the field $h_B(\boldsymbol{R})$ has values h_{Bi} which are essentially constant over each such interstitial site. If the normalised orbitals ϕ_i localised on each site are compact, with little overlap (so $\langle\phi_i|\Omega|\phi_j\rangle = 0$ if $i \neq j$, for any operator Ω) then we see at once

$$\langle\Phi_A|\Phi_A\rangle = \sum_{i=1}^{n} |w_{Ai}|^2 \tag{2}$$

from the normalisation. The weights w are determined by local strain, chemical inhomogeneity, etc. For the present we may assume the $|w_{Ai}|^2$ factors to be equal to $(1/n)$. The effective local field h_A now follows directly:

$$h_A = \langle\Phi_A|h_B(\boldsymbol{R})|\Phi_A\rangle = \sum_{i=1}^{n} |w_{Ai}|^2 \langle\phi_i|h_B(R_i)\,\phi_i\rangle$$

$$= \frac{1}{n}\sum_{i=1}^{n} h_{Bi} \, . \tag{3}$$

When there is a coherent tunnelling over n sites, the effective local field for muon A is the arithmetic average of the local fields h_{Bi} at these sites.

The accumulated data give a width of the distribution of h_A, not the distribution of h_{Bi}. We may use a well-known result (theorem 7.2 of ref. [4]) which demonstrates that the mean square value of h_A and that of h_{Bi} are related by

$$\langle h_A^2\rangle = n^{-1}\langle h_{Bi}^2\rangle \, . \tag{4}$$

Thus motional narrowing does occur when there is coherent tunnelling of muons. Contrary to ref. [2], but in agreement with ref. [3], the linewidth does depend on the extension of the wavefunction.

The effective value of n can be limited by several factors: spatial inhomogeneity is a common cause; interactions which destroy coherence are another factor. In general, n will itself have a distribution of values. However, it is only in special cases, e.g. when a muon is localised at a specific number of sites adjacent to a point defect, that the width $\langle h_A^2\rangle^{1/2}$ will be measurable in the presence of tunnelling. Experimental checks of (4) are thus difficult.

I am most grateful to Dr. A. Yaouanc for discussions and correspondence which provided the stimulus for this note and to Professor T. McMullen for valuable comments.

References

[1] J.H. Van Vleck, Phys. Rev. 74 (1948) 1168.
[2] A. Yaouanc, Phys. Lett. 87A (1982) 423.
[3] T. McMullen and E. Zaremba, Phys. Rev. B18 (1978) 3026;
T. McMullen, Solid State Commun. 35 (1980) 221.
[4] A.M. Mood and F.A. Graybill, Introduction to the theory of statistics (McGraw-Hill, New York, 1963).