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Prof. Andreas Honecker
Editor-in-Charge
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re: *ms* # 202504_00035v2:

"Muonium reaction in MgO: A showcase for the final steps of ion implantation"

by Rui C. Vilão, Ali Roonkiani, Apostolos G. Marinopoulos, Helena V. Alberto, João M. Gil, Ricardo B.L. Vieira, Robert Scheuermann and Alois Weidinger

Dear Prof. Honecker,

This substantial work combines detailed μSR measurements with sophisticated DFT calculations to construct a model of the behavior of positive muons stopping in an MgO crystal, from the high-energy charge-exchange process to the epithermal diffusion process to the response of the host lattice to the muon (Mu^+) or muonium (Mu^0) at nearly-thermal energies to their abrupt thermalization in an inelastic process, forming the metastable "doorway state" in which the muon is only loosely bound to an electron, to the stable final Mu^+ or Mu^0 state.

The paper begins with a list of assorted applications of energetic ions (especially protons) in which (the authors declare) the final stages of the stopping process are poorly understood, despite being essential to effective utilization. Since no description is given of what is unknown in the several cases, the reader is left to either read all the references or just take the authors' word for it.

This is followed by a succinct summary of the proposed mechanisms for energy loss of positive muons in matter and efforts to infer how the muons lose their last few eV from the subsequently observable states of the μ^+ or the muonium ($Mu^0 = \mu^+ e^-$) atom in its various forms. The authors describe their previous work as firmly establishing their model in which the two possible final states (a μ^+ bound to an oxygen ion or an interstitial Mu^0 atom) are governed by a "transition state" that lives long enough to be observed as a fast relaxing signal with a frequency shift.

The experimental section is thorough but not excessively detailed, clearly showing the subtle but essential point that the "diamagnetic-like" precession signal is both anomalously broadened and (especially at low temperature) shifted in frequency well below that of the free muon frequency (or a μ^+ bound to an oxygen ion). These features are shown as a function of temperature. The temperature dependence of the muonium signal is also displayed clearly and that of the Mu^0 hyperfine frequency is convincingly explained in terms of thermal vibration of the Mu^0 atom in its iterstitial well. This is nice work!

They then argue convincingly that the rapid relaxation of Mu^0 at high temperature cannot be due to trapping and must be due to thermal ionization of Mu^0 or its thermal conversion into the weakly-bound "doorway state" which is later proposed to be the precursor of the "diamagnetic-like" state Mu^+.

In the Theory section they describe DFT calculations of the locations and properties of the Mu^0 and "diamagnetic-like" Mu^+ states, culminating in a detailed model of the Mu^0 diffusion path over the potential barrier between two sites in the MgO lattice. This model is then adapted to include the effects of lattice relaxation and Mu^0 zero point motion as it passes the top of the barrier.

The picture so far is that the Mu^0 is still carrying kinetic energy from its original source – the muon beam. The earlier charge-exchange regime left some muons in the energetic Mu^0 state and others in a similarly energetic Mu^+ state.[1]

They then explain that the Mu^0 hops more freely through the lattice while it has high energy and moves too fast for the lattice to react to its presence; but as it slows down the lattice is able to relax in reponse to its passage, opening up the lower barrier as a result. It seems to me that this would *ease* the motion of Mu^0, but instead they say it makes possible the important "doorway state" which forms in an inelastic process that leaves the surrounding host vibrating until that energy disspiates to leave the Mu^0 strongly coupled to its environment for times "up to hundreds of nanoseconds". In this long-lived "doorway state" the muon has only a tiny hyperfine interaction with its electron, which is later lost to form Mu^+ (along with the prompt Mu^+ fraction). It is the long-lived "doorway state" that is proposed to produce the fast-relaxing and frequency-shifted early component of the "diamagnetic-like" Mu^+ signal.[2]

The proposed "doorway state" is modeled in great detail based upon measurements of its broadened and shifted "diamagnetic-like" precession frequency. The picture proposed is that the epithermal Mu^0 state, in which the electron is strongly bound to the muon, has an inelastic interaction with the lattice when its velocity is low enough for the lattice to relax during its passage over the barrier between two sites, and this inelastic process releases the electron sufficiently for it to have only a very weak hyperfine interaction with the muon. Only because the electron spin is polarized (by the strong magnetic field in thermal equilibrium) is its influence on the muon observable.

I have some problems with this picture.

First, the "doorway state" is proposed to last for "hundreds of nanoseconds" before losing its electron completely, during which time it has a slightly shifted precession frequency due to the loosely associated, thermally-polarized electron, after which it precesses at the free muon Larmor frequency ω_D. If so, then a Fourier transform omitting the first few hundred ns should yield a sharp line at ω_D. Was this done? If not, it should be. If so, and it yielded the same broadened and shifted line as the full time spectrum, then this picture cannot be correct.

Second, the energy scales seem inconsistent. A fully-bound, compact Mu^0 state is bound by some substantial fraction of a Rydberg unless its electron is already shared with the lattice. (Weakly bound Mu^0 states are frequently observed in solids, but they always involve the lattice intimately.) Therefore the picture of a epithermal weakly-bound Mu^0 state diffusing rapidly through the lattice is self-contradictory. Such diffusion is only possible for a compact, strongly-

[1] For some reason they refer to the 1947 Fermi and Teller paper concerning μ^- capture.

[2] At the bottom of p. 11 they say, "The inelastic reaction is caused by the force exerted by the squeezed muonium on the surrounding atoms." If the Mu^0 is "squeezed" rather than "expanded", then why is the hyperfine interaction *weaker* than in vacuum?

bound Mu^0 state, which therefore arrives at the crucial inelastic collision with a significant fraction of a Rydberg of energy. This energy is supposedly released in said inelastic process, contributing to the local "thermal spike" that raises the effective temperature of the local lattice (which must dramatically reduce the thermal equilibrium polarization of the weakly-bound electron in the "doorway state"). How is it then possible that the resultant frequency shift is so strongly temperature dependent?

It may be that my difficulty in following the logic of this paper is due to the somewhat artificial separation of experimental observations from the theory that explains their interpretation; this is probably structurally mandatory, but I get confused when flipping back and forth between assertions and their separate justifications.

Despite my reservations about some details of the conclusions, I strongly recommend publishing this impressive work.

Sincerely,

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