Hyperfine Spectroscopy and Characterization of Muonium in $ZnGeP_2$

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The Experiment

•Transverse field (TF) hyperfine (HF) spectroscopy (Fig. 1) performed using *HiTime* high frequency spectrometer in the M15 beamline at TRIUMF, Vancouver Canada

•Longitudinal field (LF) T_1^{-1} depolarazation measurements (Fig. 2) using EMU spectrometer at STFC ISIS Facility, UK

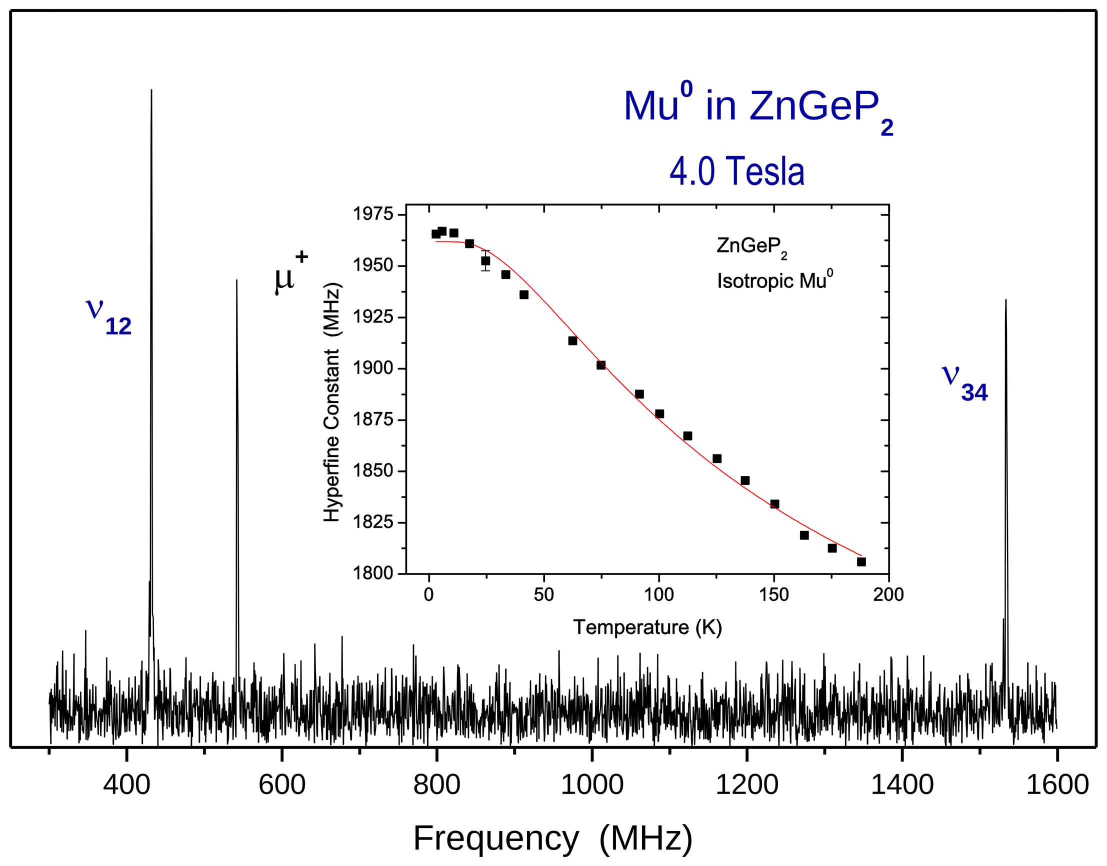

Fig. 1: Above is a TF MuSR FFT from the HF spectroscopy performed at T=5K and B=4.0T. The middle line, labeled μ^+, at 542.14 MHz is the signal from the bare muon; where ν_{12} = 432.1 MHz (left) and ν_{34} = 1534.8 MHz (right) are the HF lines. Note that ν_{12} precesses in a direction opposite of μ^+ and ν_{34}. *Inset* shows variation of Mu^0 hyperfine parameter as a function of temperature in B_{TF} = 4.0 T. The solid line is the result of the current best model fit (Einstein model) following from equation 2 [3,4].

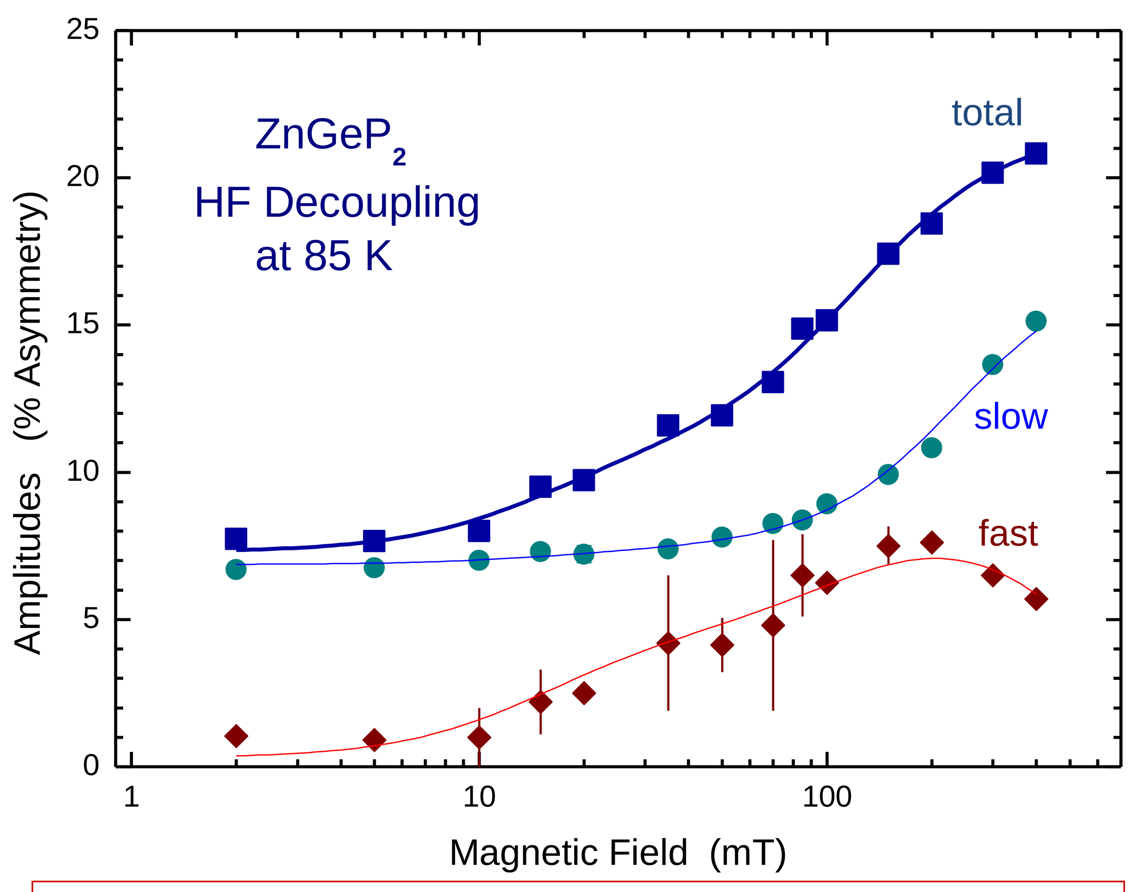

Fig. 2: Above is the HF decoupling curve from LF studies taken with fields from 0 to 400 mT. The flattening at both low and higher fields is indicative of an anisotropic hyperfine term. The isotropic piece having a value of roughly A_2 = *3185* MHz and dipolar contribution of *D = 374* MHz.

Introduction

The purpose of this study is to extend the investigation of muonium defect centers as an experimentally accessible analog of isolated hydrogen defect states in semiconductors into the chalcopyrite structured II-IV-V_2 compounds. Also, we aspire to increase the understanding of probe dynamics associated with using positive muons to characterize the atomic-scale magnetism in Mn-doped II-IV-V_2 compounds.

The chalcopyrite structured II-IV-V_2 compounds are well developed and optimized as infrared active non-linear optical materials [1]. Several members of this class of compounds have shown room temperature ferromagnetism when doped with Mn, allowing these compounds to be possible candidates for future spintronics applications [2].

Results and Interpretation

• μ^+ follows $f = \gamma_\mu B$ where γ_μ = 135.54 MHz/T which is the diamagnetic frequency [4]

• ν_{12} and ν_{34} show behavior characteristic of hyperfine splitting from a muonium atom, Mu^0 with varying temperature and applied field [5]

•HF interaction for promptly formed Mu^0 state (visible in TF) is isotropic since variations in sample orientation produced identical results.

• T_1^{-1} depolarization data in LF, fig. 2, showed state with anisotropic hyperfine interaction [6]

• T_1^{-1} depolarization rates as a function of field using single state model assuming dipolar fluctuating piece of the hyperfine parameter responsible for depolarization yields hop rates ($1/\tau_c$), shown in Fig. 4, extracted from [7]

$$T_1^{-1} = \left(1 - x/\sqrt{1+x^2}\right)\frac{D^2\tau_c}{1+\omega_{12}^2\tau_c^2} \quad (1)$$

where $x = B_{LF}/B_2$ and B_2 = 0.1131 T (decoupling field)

Further Discussion

• Sites where Mu^0 may reside:
  - T-site with Phosphorous near neighbors; where isotropic Mu^0 formed
  - T-site associated with II-IV sublattice; where anisotropic Mu^0 formed

• Variation in HF parameter, fig. 1 inset, as function of temperature fits local-mode vibrational model (Einstein model) [8,9] ie.

$$A(T) = A_0 + \frac{C_1}{\exp(h\nu/k_BT)} + \frac{C_2}{\left[\exp(k\nu/k_BT)\right]^2} \quad (2)$$

A_0 = isotropic zero-temperature HF parameter

C_1 & C_2 = coupling constants (temperature independent)

ν = single vibrational frequency

• For this particular experiment, $h\nu$ = 7.20 meV, similar to Mu_{II} in structurally similar ZnSe [10]

• Hop rates and $2\pi A_0$ having similar order of magnitude confirm the isotropic characteristic of the 1962 MHz HF term in T_P-site and exclude the possibility of motional averaging

Conclusions

• Upon muon implantation, Mu^0, with isotropic hyperfine parameter of 1962 MHz, promptly formed in a phosphorous T-site

• Mu^0 rapidly hops, visiting other sites with different hyperfine parameters where there is an abrupt change in isotropic to anisotropic hyperfine parameter occurring at each subsequent hop

• While occupying the T-site associated with the II-IV sublattice, Mu^0 has anisotropic hyperfine parameter (with axial symmetry) of A_2 = 3185 MHz and D = 374 MHz

• Since hop rates increase with decreasing temperature, motion below 80 K can be assigned to quantum tunneling; however, the slower decrease at lowest temperatures is commonly assigned to interactions with defects or impurities

• Motion between 100 K and 200 K is commonly assigned to thermally activated hopping

•Rapid increase in the relaxation rate (fig 3) above 200 K yields an ionization energy near 200 meV

•Further study necessary to develop a model for the relaxation due to motion involving two distinct Mu^0 centers

References:

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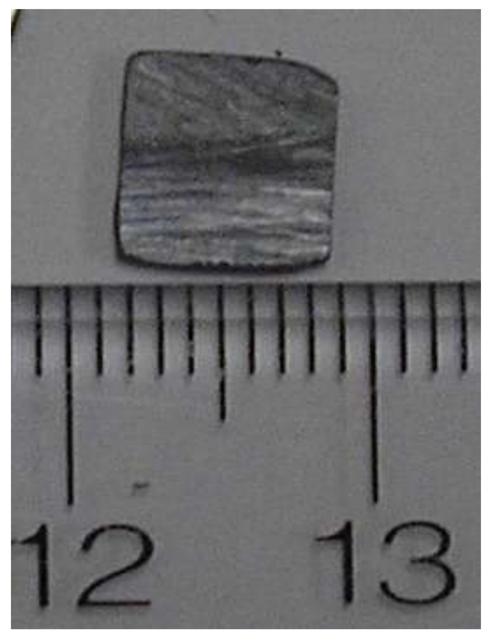
The Sample

Shown (Right)

Nominally undoped single crystal of $ZnGeP_2$ with dimensions of 8x8x1 mm^3 and an [001] orientation



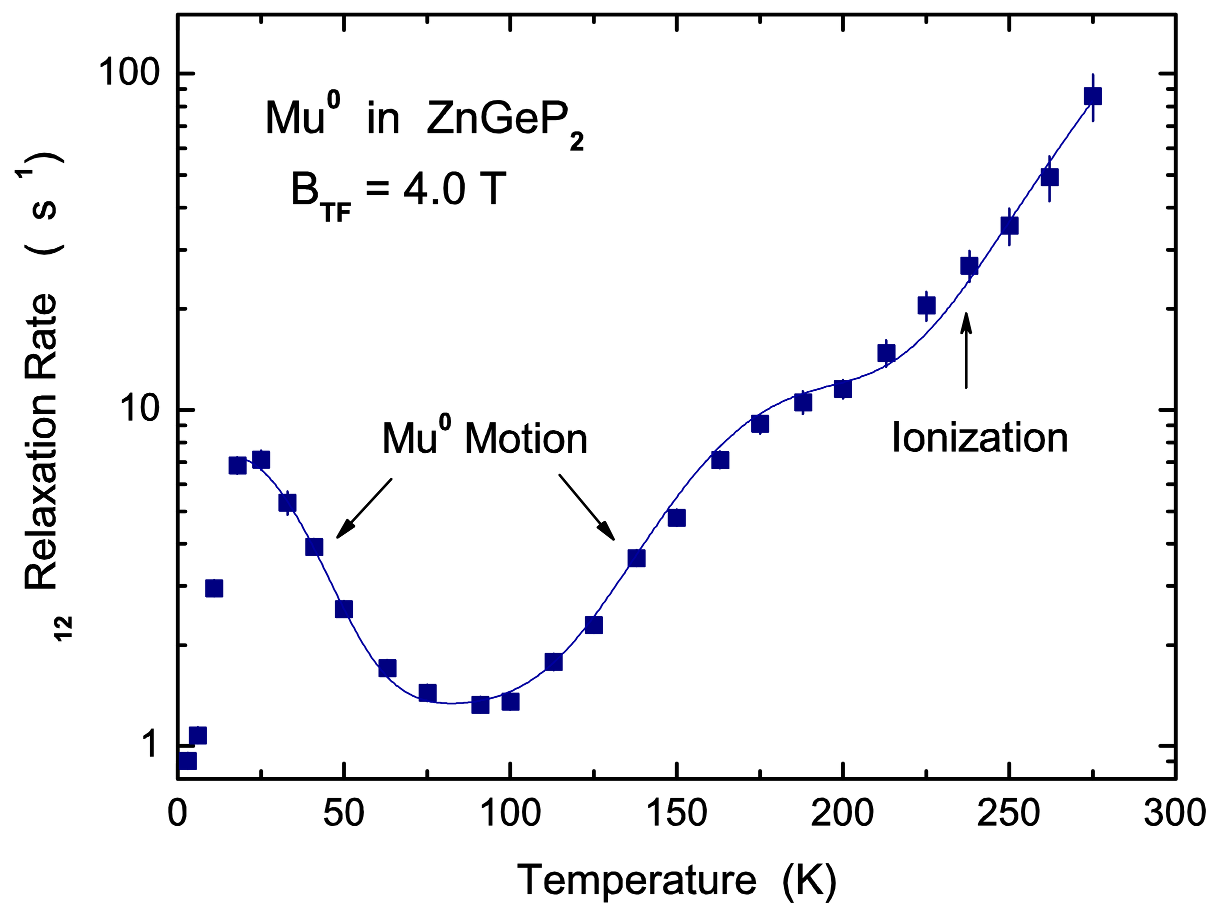


Fig. 3: Above is the relaxation rate of the ν_{12} from TF measurements (LF shows similar results). The high relaxation rates at low temperatures (T<50K) have been assigned to quantum tunneling. Increasing relaxation rates from 100 to 200 K are assigned to thermally activated hops.

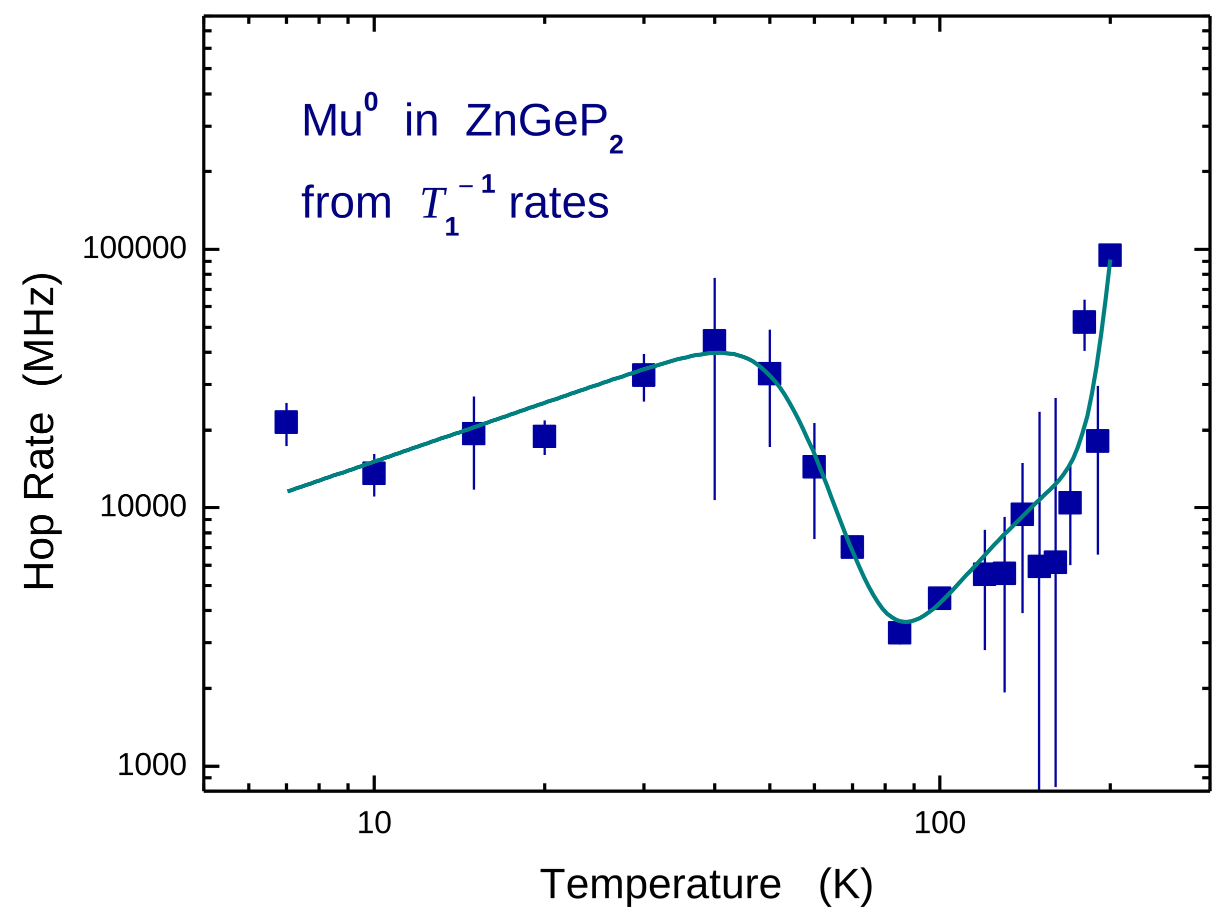


Fig. 4: Hop rates for anisotropic Mu^0 in $ZnGeP_2$ as extracted from longitudinal depolarization data using equation 1.

Acknowledgements:

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