

# Search for ALCR- $\mu^+$ SR spin polaron resonances in $\text{Cd}_2\text{Re}_2\text{O}_7$ and $\text{FeGa}_3$

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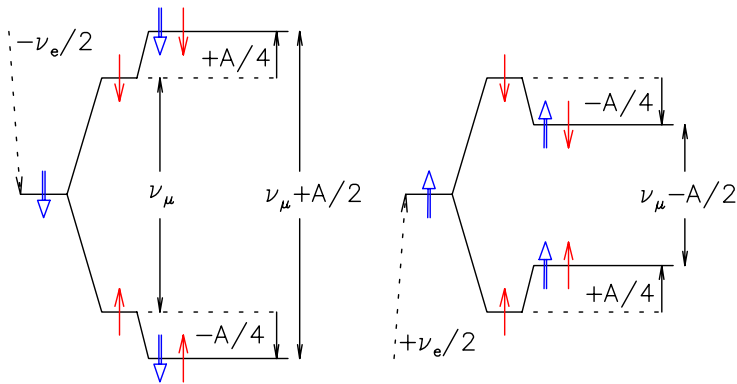
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**Abstract.** Certain magnetic materials may contain spin polarons (SP) — tiny, stable ferromagnetic “droplets” in a paramagnetic or weakly ordered “sea” of more random spins. The positive muon may become associated with SP, giving a unique window on their properties. In this experiment the  $\mu^+$ SR spectroscopy of “avoided level crossing resonance” ( $\mu$ ALCR) was used on several candidate materials to further explore this model. The results leave many questions unanswered.

## **1. Introduction**

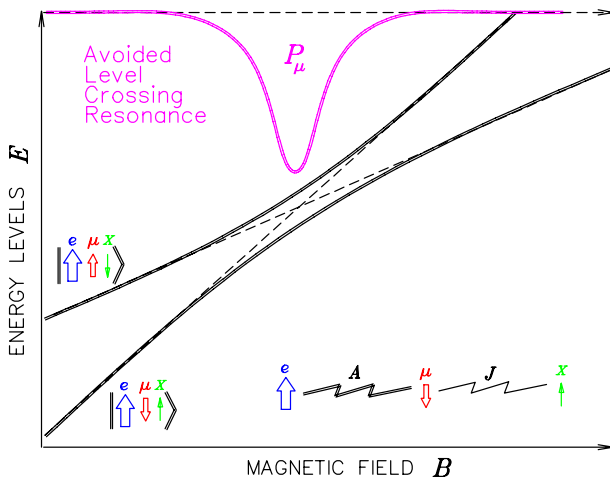
Numerous semiconducting[1, 2, 3] and metallic[4] magnetic materials have been found to exhibit characteristic two-frequency  $\mu^+$ SR precession signals in high transverse magnetic field (HTF- $\mu^+$ SR), but skepticism remains over the assignment of these spectra to muons associated with de Gennes’ legendary magnetic polarons (MP)[5] or spin polarons (SP). This is understandable, since the SP picture is a radical departure from “conventional wisdom” about both muonium and magnetism. It is therefore incumbent upon both advocates and adversaries of this interpretation to present as much spectroscopic evidence as possible in support or contradiction of the SP picture.

In studies of muonium ( $\text{Mu} = \mu^+e^-$ ) in solids and muonated radicals in liquids, the hyperfine frequency of the species containing the muon is routinely measured in high transverse magnetic



**Figure 1.** Muonium energy level diagram in the limit of high magnetic field (muon Larmor frequency  $\nu_\mu \gg A$ , the hyperfine frequency).

field (HTF- $\mu^+$ SR) as the splitting  $A$  between two precession frequencies on opposite sides of the muon Larmor frequency  $\nu_\mu$ , as shown in Fig. 1. In a SP, the electron's spin is locked to that of the local ferromagnetic (FM) "droplet" by a very strong exchange interaction, but the same sort of splitting still appears. Of course, muons in an antiferromagnetic (AFM) crystal with local fields parallel or antiparallel to the applied field will produce a similar pattern, so other considerations must help determine which scenario applies.



**Figure 2.** Avoided level crossings occur when "flipping" a third spin  $X$  that couples to the muon and/or electron causes the same change in energy as "flopping" the muon spin at a certain applied field  $B_0$ . A resonant loss of muon polarization results at  $B_0$ . This method is widely used to identify the partner  $X$ , its interaction strength  $J$  and, from that, the site and environment of the muon.

The basic mechanism of muon avoided level crossing resonance ( $\mu$ ALCR) is depicted in Fig. 2. Usually it is assumed that  $J \ll A$ . The opposite is the case in a SP, if  $J$  is the exchange interaction between the SP electron and the paramagnetic ions comprising the SP. However, if  $J$  is the *nuclear* hyperfine (NHF) coupling between the SP electron and a nearby *nuclear* spin, the same situation may apply as in ordinary semiconductors[6] or radicals [7], for which  $\mu$ ALCR spectroscopy [6, 7, 8] has yielded a plethora of information about the location, structure and effective spin hamiltonian of the paramagnetic center.

It is therefore natural (even mandatory) to explore the  $\mu$ ALCR spectroscopy of candidate SP systems in the same way. We performed an initial study of several nominally dissimilar materials with nearly identical SP-like HTF- $\mu^+$ SR spectra.

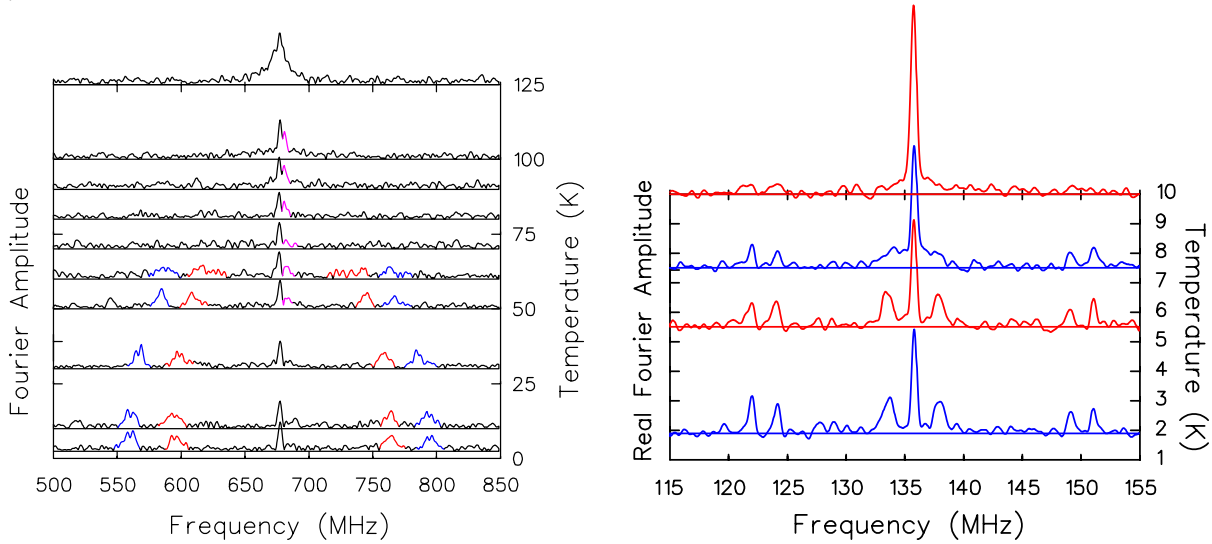
## 2. SP $\mu$ Spectroscopy

The MP or SP is formed when a free electron in a paramagnetic or weakly magnetic host has a huge exchange interaction  $J$  with neighboring magnetic ions, causing their spins to be locked together (along with that of said electron) into a small ( $\sim 1$  unit cell) ferromagnetic (FM) "droplet" — the SP — widely believed to be the root cause of various important phenomena

such as metal-insulator transitions in magnetic semiconductors (MS)[9]. The energy decrease due to this local FM ordering compensates for the increased kinetic energy of the SP electron caused by its localization. In some cases the additional electrostatic binding energy of said electron to the  $\mu^+$  assists in the localization; in other cases a SP population exists independent of the muon but the negatively charged SP attracts (and is bound to) the  $\mu^+$  in a neutral “ $\text{SP}\mu$ ” complex, analogous to muonium or muonated radicals.

The ability of HTF- $\mu^+$ SR to probe the SP depends upon the muon’s incorporation into the SP in a consistent way; thus the system in question is often described as a *bound* magnetic polaron (BMP). However, this designation is usually taken to imply a SP that is not free to move through the lattice, which may be a poor picture of the  $\text{SP}\mu$  system in cases where the muon is “bound” to the SP only by the latter’s negative charge, providing only a small extra impedance to the mobility of this exotic charge carrier.

The two SP candidate materials chosen for this  $\mu$ ALCR study are the metallic, magnetically frustrated pyrochlore  $\text{Cd}_2\text{Re}_2\text{O}_7$  and the magnetic semiconductor  $\text{FeGa}_3$ , both of whose HTF- $\mu^+$ SR frequency spectra are shown in Fig. 3.



**Figure 3.** LEFT: Frequency spectra of HTF- $\mu^+$ SR signals in  $\text{Cd}_2\text{Re}_2\text{O}_7$  at  $H = 1$  T[4]. The splittings do not change appreciably with  $H$  (not shown). RIGHT: HTF- $\mu^+$ SR frequency spectra at  $H = 1$  T in the magnetic semiconductor  $\text{FeGa}_3$ . Again the splittings do not change significantly with  $H$ .

$\text{Cd}_2\text{Re}_2\text{O}_7$  is a weakly metallic pyrochlore with geometrically frustrated magnetism, a heavy electron effective mass and a superconducting transition at  $\sim 1$  K. In the region below  $\sim 60$  K, where its resistivity follows a  $T^2$  dependence (characteristic of a Fermi liquid regime), the HTF- $\mu^+$ SR frequency spectrum at  $H = 5$  T shows the splittings characteristic of a SP with a large hyperfine coupling[4].

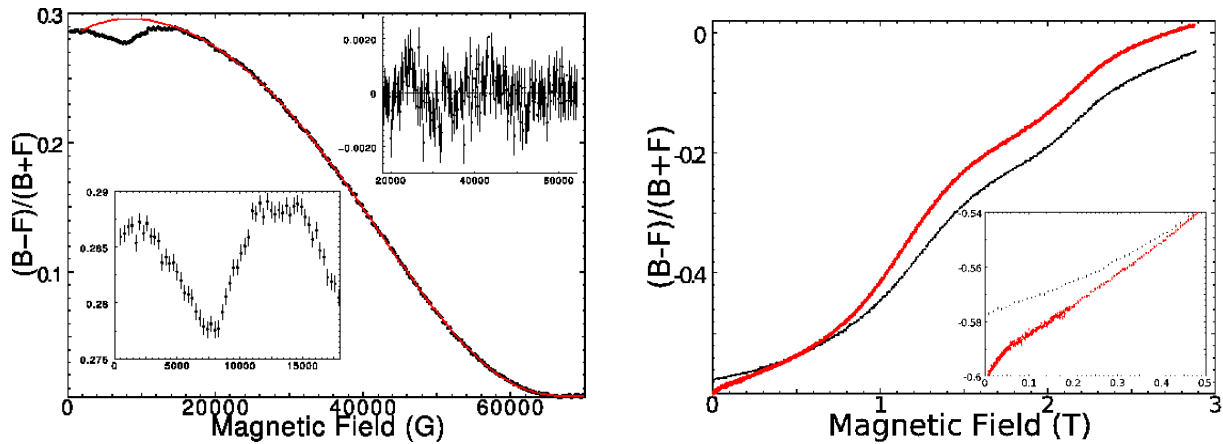
$\text{FeGa}_3$  is a narrow-gap diamagnetic semiconductor for which the gap formation is attributed to strong electron correlations within a narrow  $3d$  band. Below about 10 K an extremely narrow SP band is thought to form; in the same range, characteristic SP splittings are observed in the HTF- $\mu^+$ SR frequency spectra, with splittings independent of field up to 5 T[3].

Both materials have plentiful nuclear spins which might couple to the SP electron, possibly producing a rich  $\mu$ ALCR spectrum.

The HTF- $\mu^+$ SR “signature” of  $\text{SP}\mu$ , like that of Mu or a muonated radical, is a pair of frequencies split about the free muon Larmor frequency ( $\nu_\mu[\text{MHz}] = 135.54B_\mu[\text{T}]$ , where  $B_\mu$  is the local magnetic field at the muon site) by  $+\Delta\nu_+$  and  $-\Delta\nu_-$  with  $\Delta\nu_+ - \Delta\nu_- = A$ , the

effective frequency of the  $\mu$ - $e$  HF interaction. This similarity, plus the fact that SP $\mu$  spectra were first discovered in MS, may tempt one to attribute such spectra to normal Mu atoms; but this cannot be the case, first because (as in Cd<sub>2</sub>Re<sub>2</sub>O<sub>7</sub>, shown in Fig. 3[left]) SP $\mu$  is seen in *metallic* hosts where the Coulomb attraction of the  $\mu^+$  is screened by conduction electrons, and second because the fields of large nearby moments in the paramagnetic phase would instantly relax the Mu electron if it were not tightly locked to the spins in the FM “droplet”. Very similar spectra are seen in the MS FeGa<sub>3</sub> (Fig. 2) at low temperature. It is remarkable that the signature of SP $\mu$  should be observed in such different magnetic systems; usually one does not expect a hitherto undiscovered phenomenon to be quite so ubiquitous. In each instance one can often find alternative explanations for the observed pairs of lines — two sites with different Knight shifts, or one site in an antiferromagnetic environment, or other more exotic interpretations invoking modification by the  $\mu^+$  of the local magnetic properties of the host. However, such alternative-hunting fails to explain the remarkable consistency of the HTF- $\mu^+$ SR spectra observed in such diverse magnetic metals, semiconductors and insulators.

### 3. SP- $\mu$ ALCR Experiments



**Figure 4.** LEFT:  $\mu$ ALCR spectrum from 0 to 7 T in metallic Cd<sub>2</sub>Re<sub>2</sub>O<sub>7</sub> at 30 K, taken on the M15 muon channel at TRIUMF with the *HiTime*  $\mu$ SR spectrometer. Upper right inset: residuals from a polynomial fit (red line) to the systematic decrease at high field due to curling up of electron orbits. Lower left inset: detail of a broad resonance at  $\sim 0.75$  T ( $\nu_\mu \approx 100$  MHz  $\approx A_{SP}/2$ ) and a hint of a smaller resonance at  $\sim 1.3$  T. RIGHT:  $\mu$ ALCR spectra from 0 to 3 T at 5 K in FeGa<sub>3</sub> (upper curve, red circles) and Ag (lower curve, black squares), taken on the new M20 muon channel at TRIUMF with the *Helios*  $\mu$ SR spectrometer. If there is any resonant “dip” in the FeGa<sub>3</sub> polarization curve, it is either extremely broad or extremely narrow. The decrease in the FeGa<sub>3</sub> curve at very low field (see inset, lower right) is presumably just decoupling from local dipolar fields.

We therefore measured  $\mu$ ALCR spectra over a very large field range in the correlated metallic pyrochlore Cd<sub>2</sub>Re<sub>2</sub>O<sub>7</sub> at 30 K and in the semiconductor FeGa<sub>3</sub> at 10 K. The results are shown in Fig. 4. In each case the measurements were made (as usual) by scaling the Forward (F) and backward (B) raw positron count rates and calculating the asymmetry as  $(B - F)/(B + F)$ . As is often the case, this “raw” asymmetry is susceptible to huge systematic longitudinal field (LF) dependences as positron orbits “curl up” in high fields. Another common problem is stochastic variations due to instabilities in beamline power supplies or the proton beam drifting on the production target; that effect requires use of differential method where the applied LF is rapidly “toggled” between  $H - \delta H$  and  $H + \delta H$ . This latter method was not required for

these experiments on M15 and the newly-rebuilt M20, whose power supplies are very stable. Therefore the huge systematic changes in the “baseline” asymmetry are evident in Fig. 4.

The large, broad resonance in  $\text{Cd}_2\text{Re}_2\text{O}_7$  at  $\sim 0.75$  T (corresponding to a frequency  $\nu_\mu \approx 100$  MHz) is consistent with a zero-crossing resonance (ZCR) for muons at a site with a local field of that magnitude antiparallel to the applied field, as in  $\text{MnF}_2$  [10]. This would imply that the SP hyperfine splitting  $A_{SP}$  in  $\text{Cd}_2\text{Re}_2\text{O}_7$  at low  $T$  is not the large ( $\Delta\nu_{\text{big}} \approx 200$  MHz) splitting shown in Fig. 3 but the smaller ( $A_{SP} = \Delta\nu_{\text{small}} \sim 30$  MHz) splitting between blue and red peaks in each of the groups on either side of  $\nu_\mu$ , while  $\Delta\nu_{\text{big}}$  comes from local magnetic ordering at the muon site. If there is actually a smaller resonance at  $\sim 1.3$  T, it might be a true  $\mu\text{ALCR}$ , presumably with  $X = \text{Re}$  nuclear moment.

Presumably the  $H$  dependence of the asymmetry in Ag is due entirely to systematics and represents a calibration for full asymmetry, except for a possible offset due to slight geometrical differences. It is therefore unsurprising that the  $\text{FeGa}_3$  asymmetry has the same shape at high LF (above about 1.6 T) except for a geometrical offset. At lower fields, however, considerable asymmetry is lost in  $\text{FeGa}_3$  (relative to Ag), in a monotonic drop of about 0.066, nearly half of a typical full asymmetry in such LF measurements. This is consistent with Mu decoupling in a semiconductor with nearly 100% Mu formation and a nearly vacuum-like Mu HF coupling. The additional drop-off as  $H \rightarrow 0$  is presumably due to decoupling from local nuclear dipolar fields. The HTF- $\mu^+$ SR splittings observed for  $\text{FeGa}_3$  in Fig. 3 are certainly not consistent with a HF coupling of close to  $A_0 = 4463$  MHz for a majority of muons forming vacuum-like Mu atoms.

#### 4. Conclusions

Our search for rich  $\mu\text{ALCR}$  structure in these SP candidate materials has been generally unsuccessful. The NHF couplings between the SP electron and neighboring nuclei may be either too weak or too anisotropic to engender resonances. There might be extremely narrow resonances that would require scanning the appropriate field range with much smaller step sizes. (We used steps of 4, 10, 50 and 100 G in low, medium and high field regions.) Such a survey would benefit enormously from a good guess of where such resonances might be expected to appear.

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