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Selective Excitation Double Mössbauer Spectroscopy

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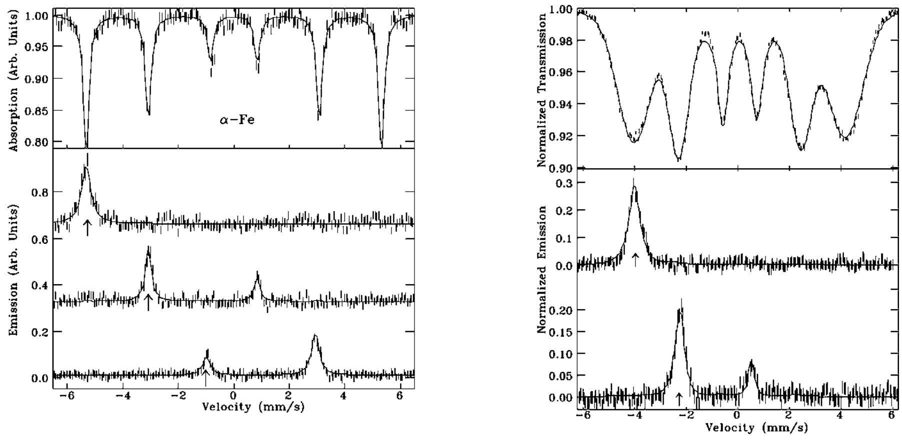
Abstract. An improved selective excitation double Mössbauer spectrometer has been used to study both static and dynamic disorder in magnetic materials. Static disorder has been identified in *a*-$Fe_{80}B_{20}$ and $Fe_{65}Ni_{35}$, while magnetic relaxation has been measured in a Fe_3O_4 ferrofluid. Simultaneous static and dynamic disorder have been established in *a*-$Fe_{92}Zr_8$.

Systems that experience magnetic relaxation undergo energy transitions not available to the Mössbauer nucleus in a static hyperfine field and show spectral line broadening. Many materials that demonstrate magnetic relaxation also suffer from chemical and structural disorder that in turn results in spectral broadening similar to that caused by relaxation. Separating these two effects in a conventional transmission Mössbauer spectrum is problematic. Selective excitation double Mössbauer (SEDM) spectroscopy, a modified Mössbauer technique (described in [1–3]) provides distinct spectral signatures from static and time-dependent hyperfine fields, so that static disorder and magnetic relaxation can be clearly separated.

In a static magnetic field, the selection rules allow only specific transitions from the excited states. This is seen most clearly in SEDM spectra of α-Fe (Figure 1) with lines corresponding to these transitions and the respective transition probabilities. In all cases, with the detector and source linewidths characterized, a complete description using a single hyperfine field (B_{hf}) is possible, as shown by the solid fitted lineshapes in Figure 1 and described in [4].

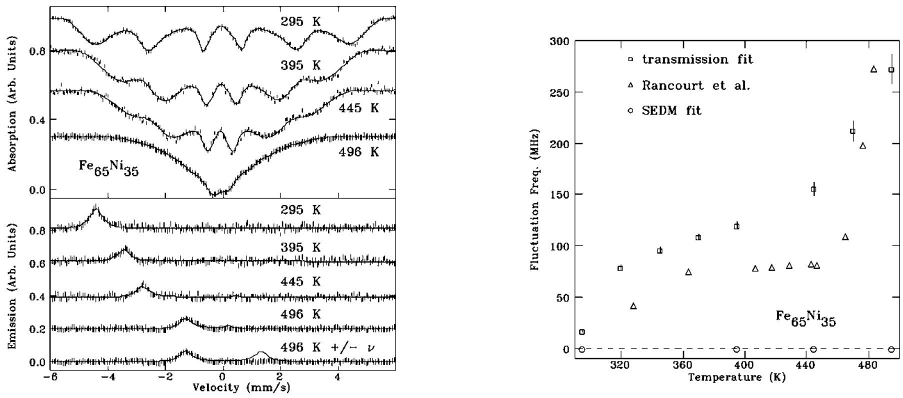
When static magnetic disorder is present in a system such as the metallic glass $Fe_{80}B_{20}$, exactly the same transitions as above are observed, shown in Figure 1. The spectra are completely described with a distribution of hyperfine fields, $P(B_{\mathrm{hf}})$, determined from the transmission spectrum, and the linewidths of detector and source [3, 4].

Transmission spectra of $Fe_{65}Ni_{35}$ (Figure 2) can be fitted equally well by either a $P(B_{\mathrm{hf}})$ or using a stochastic two state (spin-flip) magnetic relaxation model [5, 6]. A spin-flip on the timescale of the Mössbauer effect should result in additional spectral lines (described in [4, 7]) appearing in the SEDM spectrum that do no coincide with Mössbauer transitions in a static hyperfine field. SEDM spectra of



(a) (b)

Figure 1. (a) Transmission and SEDM spectra of α-Fe. (b) Transmission and SEDM spectra of $Fe_{80}B_{20}$. Pump energies indicated by the ↑.

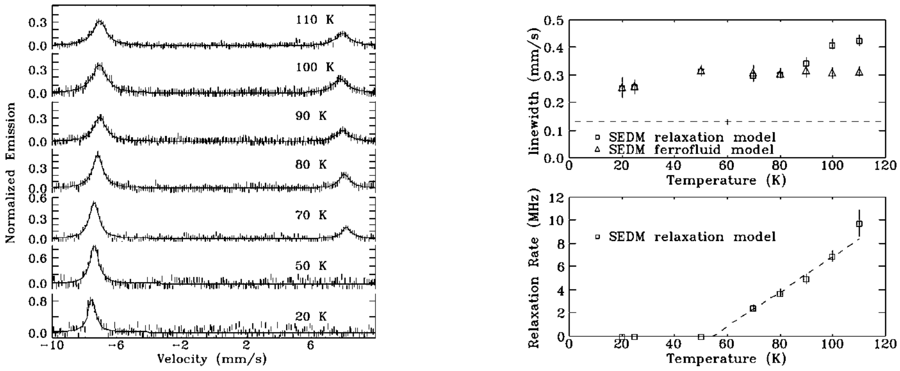


(a) (b)

Figure 2. (a) Transmission spectra of $Fe_{65}Ni_{35}$ (top) and SEDM spectra (bottom). Bottom-most SEDM spectrum shows the expected lineshape if magnetic relaxation were present. (b) Relaxation rate as a function of temperature from transmission spectra and SEDM fits. Values from Rancourt *et al.* [6] for comparison (only a single B_{hf} is necessary to fit our data while Rancourt *et al.* used a two site fit).

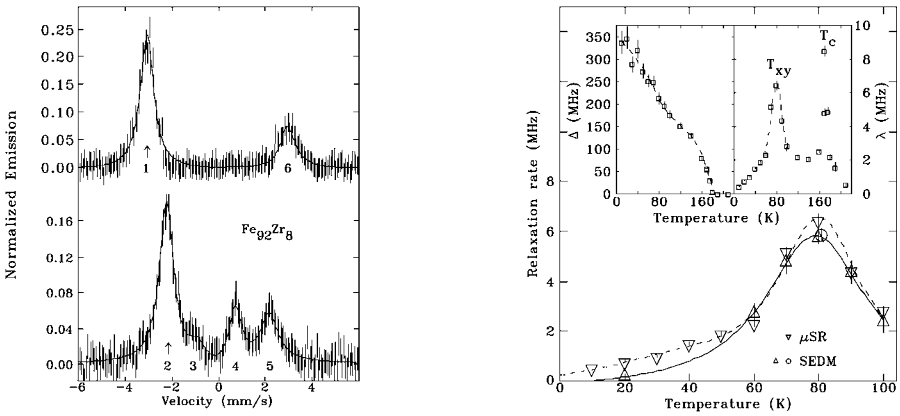
$Fe_{65}Ni_{35}$ collected at temperatures up to $0.99T_C$ (Figure 2) clearly do not show an extra line, and fitted relaxation rates (Figure 2) are zero. These SEDM results (fully described in [5]) explicitly show that no magnetic relaxation is present up to $0.99T_C$.

A magnetic fine particle system exhibits both static and dynamic disorder. Figure 3 shows SEDM spectra of a 6.0 nm Fe_3O_4 based ferrofluid. The single lines in the 20 and 25 K SEDM spectra clearly show that the moments are static. With an increase in temperature, collective magnetic excitations lead to an increase in



(a) (b)

Figure 3. (a) SEDM spectra of a Fe_3O_4 ferrofluid. Notice the appearance of the extra line above 50 K when superparamagnetic spin flips start happening. (b) Results of fits to the data using the SEDM models described in [4]. The dashed line shows the linewidths obtained by SEDM on α-Fe.



(a) (b)

Figure 4. (a) SEDM spectra at $T_{xy} = 80$ K. Pump energies are indicated by the ↑. (b) Inset: temperature dependence of the static (Δ) and dynamic (λ) relaxation rates of *a*-$Fe_{92}Zr_8$. Body: relaxation rates from ZF-μSR fits (▽) and SEDM fits (△,◯) around T_{xy}. Lines are guides to the eye.

the observed SEDM spectral linewidth (□), as described in [7]. Once above the blocking temperature, T_B, when superparamagnetic 180° moment flips happen, there are two lines. The line at $\sim +8$ mm/s is the signature of magnetic relaxation. Near T_B fitted linewidths are able to correctly describe both the drive and return lines of the SEDM spectra, a strong indication that the only dynamic behavior is due to superparamagnetic spin flips. However, once the temperature is sufficiently above T_B, the continued increase in the fitted linewidth indicates that this model is too simple (a correct description should involve no linewidth variations). A more complete description is possible when a range of relaxation rates (from transmission spectra fits described in [9]) is used to fit the SEDM spectra. These fits are shown in Figure 3 (△, SEDM ferrofluid model), and a more consistent linewidth results showing that the SEDM spectra are sensitive to range of different sized particles undergoing spin-flips.

Frustrated magnets exhibit a complex mixture of static and dynamic magnetic behavior. Zero-field muon spin relaxation (ZF-μSR) spectroscopy has been used to characterize a-Fe–Zr frustrated magnets [10]. Figure 4 shows SEDM spectra collected at the transverse spin freezing temperature, T_{xy}, in a-$Fe_{92}Zr_8$ indicating the dominant fluctuation mechanism is 180° moment flips. ZF-μSR results in Figure 4 show Δ (representing the rms static field) and λ, the dynamic relaxation rate (see [8, 10]). Since the average over all spin fluctuation modes is measured with ZF-μSR, the excellent agreement between SEDM and ZF-μSR provides strong evidence that the same magnetic relaxation phenomena is detected with the two different probes.

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