

## Resonant frequency dependence of spin relaxation in concentrated metallic spin glasses

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Magnetic resonance linewidths in  $a\text{-Fe}_7\text{Ni}_{73}\text{P}_{14}\text{B}_6$  and  $a\text{-Mn}_{48}\text{B}_{52}$  spin glasses have been measured over wide ranges of frequency and temperature. For the first time, a strong "resonant" anomaly is observed at low temperatures in the relaxation rate as a function of frequency.

A thorough understanding of glassy magnetic systems which show spin-glass behavior at low temperatures requires careful studies not only at temperatures below the low-field critical temperature  $T_{SG}$ , but also at temperatures well above  $T_{SG}$ . Magnetic resonance, despite the fact that it is performed at high fields, has provided some of the most telling information involving glassy spin systems. In general, it is found<sup>1-3</sup> that there is a sizable increase in the linewidth  $\Gamma$  and a significant downshift in the resonance field  $H_R$  as the temperature is lowered towards, and through,  $T_{SG}$ . However, the details are far from fully settled, especially in the case of concentrated alloys where direct exchange, rather than Ruderman-Kittel-Kasuya-Yosida (RKKY), effects are expected to dominate. We have been studying electron paramagnetic resonance<sup>4,5</sup> in a number of concentrated glassy magnets spanning wide ranges of frequency (2–35 GHz) and temperature (4–300 K). In this spectral regime we report here the existence of a characteristic frequency  $\nu_c$ ; in each system, for which the low  $T$  rise in  $\Gamma$  is much stronger than at frequencies above and below  $\nu_c$ . Whereas there is no completely adequate theory to account for the temperature dependence of  $\Gamma$ , we suggest that the existence of this anomaly strongly points towards a high "density-of-states" local mode in the random spin system.

The samples used in this study were 20- $\mu\text{m}$ -thick ribbons of  $a\text{-Fe}_7\text{Ni}_{73}\text{P}_{14}\text{B}_6$  and  $\sim 1\text{-}\mu\text{m}$ -thick sputtered films of  $a\text{-Mn}_{48}\text{B}_{52}$  on quartz substrates. Low-field ( $\sim 10$  Oe) dc magnetization data<sup>6</sup> were used to establish that they have  $T_{SG}$  values of 20 and 17 K, respectively. The EPR measurements were done using standard techniques already described in earlier papers from this laboratory.<sup>3</sup> Data were taken in both the parallel (dc field in sample plane) and perpendicular (dc field normal to sample surface) geometries. This facilitates analysis of the line centers as discussed elsewhere.<sup>5</sup> Here, the main concern is with linewidths and we will concentrate on results obtained in the parallel configuration.

Before presenting the results we delineate a few general features: (i) the field modulation technique has been used and we noted no drastic change in line shape as the temperature was varied from  $\sim 4T_{SG}$  to  $0.2T_{SG}$ ; (ii) by making measurements on several samples and, in different mountings, we conclude that the linewidths given below are not

known to better than 10%; (iii) the line shapes are somewhat dependent upon mounting as exemplified by the observation that the asymmetry [(low-field peak height/high-field peak height) – 1] is repeatable to only a factor of 2; (iv) although data at the lowest frequency ( $\sim 2$  GHz) are limited at the low- $T$  end by the rapid downshift of  $H_R$ , this must not be construed to imply that the resonance "disappears" below any temperature. In fact, one can follow the development of the line as it "emerges" out of zero field starting at the lowest  $T$ . Thus, in principle, a line-shape calculation could be used to derive the effective linewidth at any  $T$ . However, the data described below are based on direct observation of the peak-to-peak separation and the observed resonance can be adequately represented by the

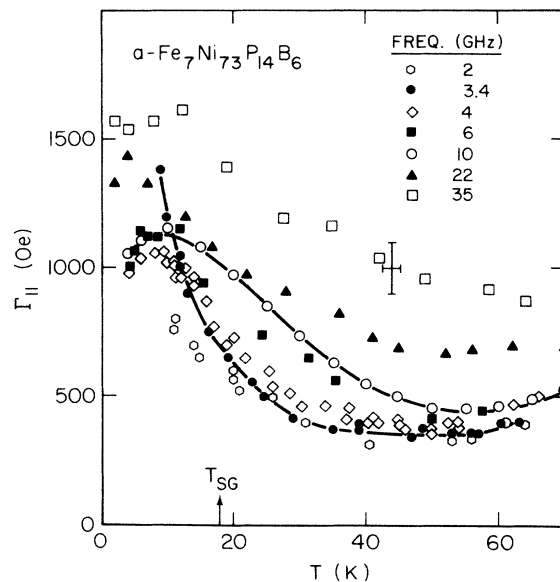


FIG. 1. Temperature dependence of the peak-to-peak linewidth of electron paramagnetic resonance in  $a\text{-Fe}_7\text{Ni}_{73}\text{P}_{14}\text{B}_6$  observed in the parallel geometry for several frequencies between 2 and 35 GHz. Note that the low- $T$  increase in  $\Gamma_{||}$  is sharper at 3.4 GHz than at any other frequency. The lines are guides to the eye.

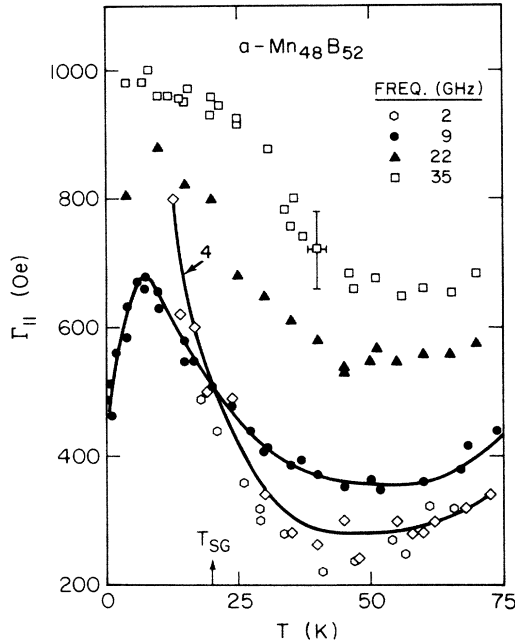


FIG. 2. Parallel linewidth vs temperature for  $a\text{-Mn}_{48}\text{B}_{52}$ . Here the characteristic frequency is very close to 4 GHz.

usual phenomenological equations.<sup>7</sup>

In Figs. 1 and 2 the linewidths are shown as functions of  $T$  for  $4 < T \leq 70$  K. Note that the 2-GHz data do not go down to 4 K. The behavior at higher  $T$  is discussed in Ref. 5, and, as further described there, the low-temperature rise is well represented by

$$\Delta\Gamma = (\Gamma - \Gamma_0) = \Gamma_1 \frac{T}{T_0} \exp(-T/T_0) \quad (1)$$

where  $\Gamma_0(\nu)$  represents a temperature-independent background. This can be seen also in Fig. 3 where the "scaled" low- $T$  increase ( $\Delta\Gamma/\Gamma_1$ ) is plotted as a function of  $(T/T_0) \exp(-T/T_0)$  and one notes that, within limits of experimental error, the data follow the 45° line in accord with Eq. (1). However, an anomaly becomes clear when we focus attention on data in the neighborhood of 3 to 4 GHz [see the data for 3.4 (4 GHz) in Fig. 1 (2)] and find a low- $T$

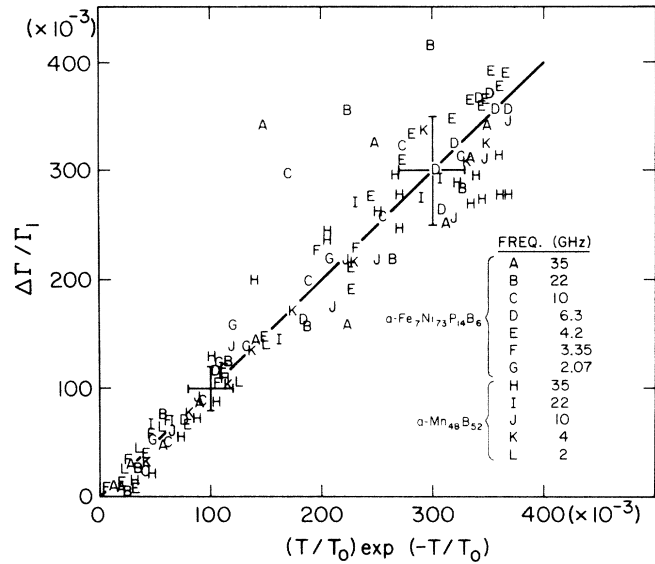


FIG. 3. Scaled plot of the low-temperature increase in  $\Gamma$  (Figs. 1 and 2) in terms of Eq. (1). The relevant parameters are in Table I.

increase which is far more rapid than for other frequencies. The "resonant" character is particularly evident from the parameter values listed in Table I, where  $\Gamma_1$  is roughly constant except near a characteristic frequency (underlined), where it is higher by 50% to 100%. The "width" of the resonant anomaly is roughly 2 GHz. In Table I, the temperature regimes used for obtaining  $\Gamma_1$  are also listed and one notes that the anomaly is reflected in the entire range of increase of  $\Gamma$  and not merely by the lowest-temperature data.

As mentioned earlier there is no theory to quantitatively account for Eq. (1). Formally, a similar result has been obtained by Continentino<sup>8</sup> by considering coupling to two-level systems (TLS), but his results are truly applicable only at very low  $T$ . It is also not particularly fruitful to try and connect the present results to earlier work on spin relaxation in Sierpinski gasket (SG) alloys, since most of the previous studies were not done on such highly concentrated materials as  $\text{Fe}_7\text{Ni}_{73}\text{P}_{14}\text{B}_6$ . (It is useful to recall<sup>6</sup> that in the  $\text{Fe}_x\text{Ni}_{80-x}\text{P}_{14}\text{B}_6$  system the critical concentration for the on-

TABLE I. Parameter values for Eq. (1).

Temp. range (K)	$a\text{-Fe}_7\text{Ni}_{73}\text{P}_{14}\text{B}_6$ ( $T_{SG} = 17$ K)			Temp. range (K)	$a\text{-Mn}_{48}\text{B}_{52}$ ( $T_{SG} = 20$ K)		
	Frequency (GHz)	$\Gamma_1$ (Oe)	$T_0$ (K)		Frequency (GHz)	$\Gamma_1$ (Oe)	$T_0$ (K)
4-55	35	2000	11	4-40	35	1000	10
4-40	22	1800	8	4-40	23	1000	9
4-40	10	2000	10	1.5-35	10	900	7
4-40	6.3	2000	9	5-35	5	1000	9
8-40	4.2	1700	7	9-35	4	1900	7
8-35	3.8	2200	7	16-35	3.3	1500	7
12-35	3.4	2800	5	18-35	2.7	1700	6
17-35	<u>2.7</u>	2900	6	18-35	2	1500	6
16-35	2.5	2600	6			(± 10%)	(± 1)
16-35	2	1600	6				
		(± 10%)	(± 1)				

set of ferromagnetism is  $x_c = 7.7$ ). Note that the anomaly reported here is observed only when the concentration is rather high. For instance, preliminary data on  $\alpha$ - $\text{Fe}_{32}\text{B}_{68}$  exhibit a similar anomaly at  $\nu_c \approx 2.8$  GHz. While the present paper was in review, we learned<sup>9</sup> that the concentrated spin glass  $\text{AuFe}_{15}$  may also show a comparable effect. On the other hand, no unusual behavior is discovered when the linewidths in reentrant alloys ( $x \geq x_c$ ) are examined over wide ranges of frequency and temperature.<sup>10</sup>

The existence of the present anomaly in the relaxation rate is clear without appeal to any specific model. It is highly likely that it arises from coupling to a local mode (TLS?) in the magnetic network, which has a high density of states and a frequency "matching" the applied radio frequency,

thereby giving rise to an enhanced relaxation rate. Alternatively, it may be a reflection of the fact that in the neighborhood of a "percolation" concentration, there are additional relaxation channels due to the presence of large spin clusters. Further measurements are in progress to study the dependence of the anomaly on concentration and other parameters.

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<sup>7</sup>A detailed discussion will be given in a forthcoming paper; see also Ref. 5.

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