Exchange-narrowed anisotropy contribution to the EPR width and shift in the Ag-Mn spin-glass

George Mozurkewich, J. H. Elliott, M. Hardiman,* and R. Orbach
Department of Physics, University of California at Los Angeles, Los Angeles, California 90024
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The EPR linewidth in the paramagnetic phase ($T > T_g$) of $AgMn_x$ and $AgMn_xSb_y$ contains a term which increases near T_g as a power law in $(T - T_g)/T_g$. The magnitude of the power-law contribution varies linearly with y. These results are interpreted in terms of exchange narrowing of an anisotropic, Dyaloshinski-Moriya interaction. Both width and shift depend strongly on frequency between 1 and 9.6 GHz. The shift behaves neither as a pure g-value shift nor as a frequency-independent internal field. Results are compared with the microscopic theory of Levy, Morgan-Pond, and Raghavan, and with a phenomenological approach.

I. INTRODUCTION

For the study of spin-glasses, EPR is becoming an increasingly important tool. Much current interest focusses on the complex behavior well below the susceptibility cusp temperature T_g, where the field for resonance varies with magnetic history of the sample. The experiments of Monod and Berthier[1] and of Schultz and co-workers[2] in the archetypal spin-glass Cu-Mn have inspired much theoretical work. When the sample is cooled in zero external field, the resonant field is determined principally by the anisotropy field. On the basis of magnetic hysteresis measurements,[3] Fert and Levy[4] have proposed a microscopic model for the origin of this peculiar spin-glass anisotropy, in which spin-orbit scattering plays a vital role. Indeed, the influence of spin-orbit scattering on the resonance shift below T_g was noted as long ago as 1969.[5] Anisotropy effects in spin-glasses have been reviewed by Levy, Morgan-Pond, and Fert.[6]

Corresponding experimental efforts in the paramagnetic regime $T > T_g$ have followed upon Salamon and Herman's suggestion that the divergent width in that temperature range is due to exchange narrowing of an anisotropic interaction, taken by them to be dipolar.[7,8] In Cu-Mn the linewidth was found to obey a power law in reduced temperature. The model implies that the shift in the resonance position is a g-value shift, that is, proportional to frequency, but the frequency dependence in Cu-Mn was not measured. The recent demonstration that in at least one spin-glass system[9] the shift for $T > T_g$ is better characterized as a frequency-independent internal field emphasizes the importance of frequency-dependent measurements,[10] a point which has previously been made with regard to the linewidth.[11]

In this paper we report EPR measurements in the paramagnetic phase of Ag-Mn at spectrometer frequencies between 1 and 2 GHz and at 9.6 GHz for several Mn concentrations. For one Mn concentration a series of alloys with a third element, the nonmagnetic spin-orbit scatterer Sb, was examined. For all samples we observe a divergence of the linewidth as a power law in reduced temperature. By varying the Sb concentration, we show that the predominant contribution to the linewidth involves an anisotropic spin-orbit-scattering process (as opposed to dipolar interaction) and that this same anisotropy is responsible for the low-temperature shift. The line shift for $T > T_g$ between 1 and 2 GHz is a g-value shift but fails at X band to exhibit the proportionality to frequency. In fact, over some range of temperature, the shift is smaller at X band than at lower frequencies. The shift and width are closely correlated, and therefore arise from the same mechanism. Our preliminary measurements in Cu-Mn and Cu-Mn-Pt have revealed no qualitative differences between the Cu-Mn and Ag-Mn systems.

After a description of experimental techniques in Sec. II, we present our results for width in Sec. III and for shift in Sec. IV. Section V is divided into three parts. First we examine the microscopic theory of Levy *et al.*,[12] which comes close to predicting the shape of the linewidth divergence. Next we compare the analysis in terms of exchange narrowing to phenomenology derived from Saslow[13] and Becker.[14] The values of anisotropy field extracted in this way are similar to values obtained from measurements below T_g. Although neither theoretical approach is entirely satisfactory, they are suggestive of the directions in which progress is likely to be made. Finally we briefly consider possible causes of the failure of the power-law behavior at the temperatures nearest T_g.

II. EXPERIMENTAL METHOD

The Ag-Mn spin-glass system was chosen for this investigation because it is relatively easy to produce Ag-Mn samples with minimal inhomogeneous distribution of Mn, and therefore minimal spatial variation of the magnetization cusp temperature T_g, as evidenced by the relatively weak effect of metallurgical history on the sharpness of the cusp.[15] Furthermore, the temperature of the cusp obtained from ac susceptibility shows very little dependence on measuring frequency.[11] Most experiments were performed with Mn concentration 2.6 at.%, near which the

high temperature (300—700 K) Weiss constant goes through zero,[16] indicating that the net spin-spin interaction is neither ferromagnetic nor antiferromagnetic.

Samples were prepared by arc-melting the constituent elements under an argon atmosphere. The alloys were annealed for 8 to 13 h at 835 °C, rolled into foils about 150 μm thick, and reannealed following a procedure described elsewhere.[17] Nominal concentrations are quoted in atomic percent. A superconducting quantum interference device (SQUID) was used to measure the cusp temperature in low field (9 Oe) and to obtain the dc magnetization in $H \approx 500$ Oe.

EPR at 9.6 GHz (X band) was performed with a Varian Associates E101 bridge and a TE_{011} cylindrical cavity. In the range 1—2 GHz, a low-frequency spectrometer[18] was used with either stripline[18] or split ring[19] resonators. The samples were cooled in flowing helium vapor, and the temperature of the copper sample holder was measured with a Au-Fe/copper thermocouple which was calibrated *in situ* against a calibrated silicon diode. To extract the resonance field and linewidth, the experimental derivative line shapes were fit by computer to a mixture of real and imaginary parts χ' and χ'' of the dynamic susceptibility. The best ratio of χ' and χ'' (or equivalently, the best A/B ratio[20]) was also determined by the fit. Because of the broad lines, the fitting procedure for the low-frequency spectra included the negative frequency contribution to the dynamic susceptibility. Excellent fits to the assumed Lorentzian shape were ordinarily obtained at all temperatures down to $1.1T_g$. The only exceptions were for the $AgMn_xSb_y$ samples with the largest Sb concentrations, which at the X band showed small departures from Lorentzian for $T < 1.4T_g$. Except for the largest Mn concentrations, the A/B ratios fell between 2.25 and 2.75.

In large magnetic fields, the sample's demagnetizing field significantly shifts the resonance.[21] A demagnetization correction was obtained for the X-band resonance fields by taking spectra with the applied field both perpendicular and parallel to the plane of the foil and defining $H^{\rm res} = (H_{\perp}^{\rm res} + 2H_{||}^{\rm res})/3$. At lower frequencies the difference between $H_{\perp}^{\rm res}$ and $H_{||}^{\rm res}$ was too small to be determined reliably. Therefore the spectra were measured in the perpendicular orientation only and the X-band correction was used after scaling down by the ratio of the frequencies. This correction agreed within experimental error with that obtained from SQUID magnetization measurements in $H = 500$ Oe. In principle, spatial variation of the demagnetizing field contributes to the observed linewidth.[22] However, the demagnetization distribution has been calculated for the foil geometry and Mn concentrations used in these experiments and contributes no more than 5 G to the width.

III. LINEWIDTH RESULTS

The EPR linewidth in dilute magnetic alloys increases linearly with temperature.[23] The width may be written $W = a + bT$, where a is called the residual width and b is called the thermal broadening. Departures from the linear temperature dependence are observed in more concentrated systems which undergo magnetic ordering.[23] In spin-glasses the linewidth in the paramagnetic phase ($T > T_g$) increases as T_g is approached from above, causing the overall temperature dependence to exhibit a broad minimum in the vicinity of $2T_g$. Such behavior has previously been observed in crystalline[7,8,11] and amorphous[10,24] metallic spin-glasses. A similar linewidth increase is observed in insulating spin-glasses.[25,26]

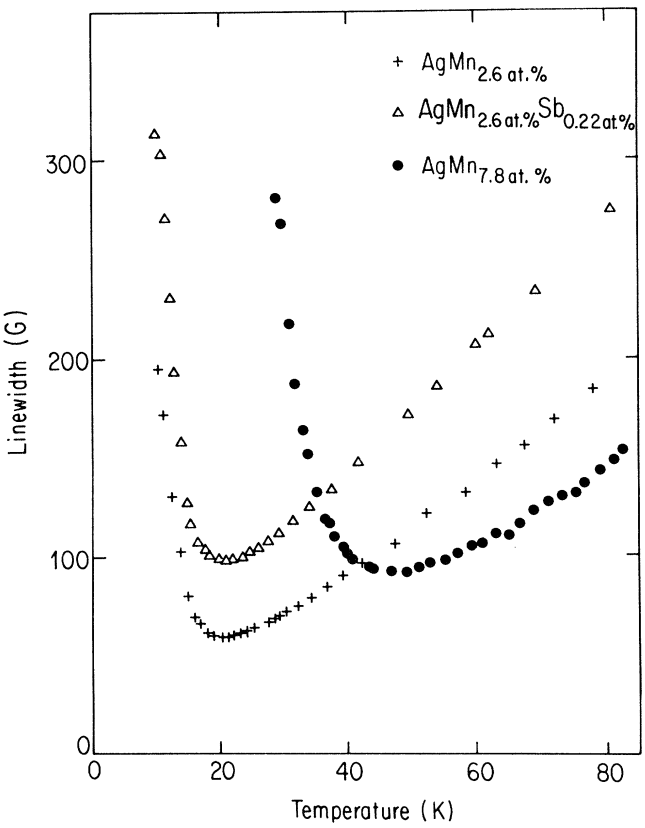

FIG. 1. Temperature dependence of the linewidth for three samples of $AgMn_xSb_y$. Spectrometer frequency was 1.9 GHz.

We have measured EPR linewidth in $AgMn_xSb_y$ with various Mn and Sb concentrations. All samples exhibit the typical behavior just described. Low-frequency (1 to 2 GHz) widths for three representative samples are shown in Fig. 1 as a function of temperature. At high temperature, the widths approach asymptotes $a + bT$, where a and b depend on concentrations x and y. As T decreases, the width departs from the linear behavior and goes through a minimum around $2T_G$, below which the low-frequency width appears to diverge as T_g is approached. The width can conveniently be expressed in the form $W = a + bT + W^{\rm ex}$, which assumes an additive combination of the high-temperature Korringa behavior plus an "excess" contribution associated with the spin-glass transition. $W^{\rm ex}$ for $AgMn_{0.026}$ is plotted in Fig. 2 against reduced temperature $t = (T - T_g)/T_g$ for two spectrometer frequencies, 1.0 and 9.6 GHz. The high- and low-frequency $W^{\rm ex}$ have the same shape above $1.6T_g$, but below this temperature the high-frequency width becomes progressively smaller than that for low frequency. Increasing either x or y increases $W^{\rm ex}$ for both frequencies at all reduced temperatures, while the temperature below

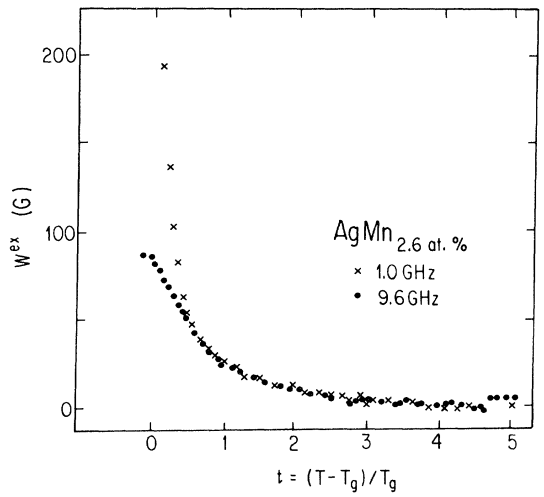

FIG. 2. Temperature dependence of the linewidth after subtraction of the high-temperature $a+bT$ contribution. Measurements at 1.0 and 9.6 GHz are compared.

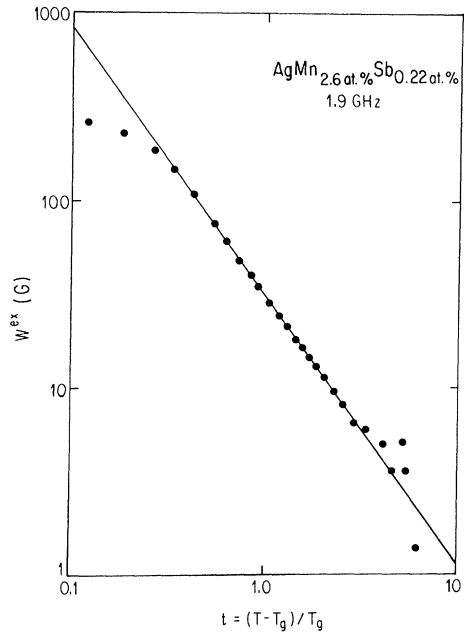

FIG. 3. Double-logarithmic plot of excess width vs reduced temperature illustrating the power-law dependence. Slope of the solid line is -1.45.

which the high- and low-frequency widths differ becomes closer to T_g as x is increased.

Salamon and Herman have attributed the excess linewidth in Cu-Mn to exchange-narrowed dipolar interaction.[7,8] For an unspecified anisotropic interaction with variance $\langle(\delta\omega_{\text{anis}})^2\rangle$ exchange narrowing gives, for the width (in units of magnetic field),

$$W^{\text{ex}}=\frac{\hbar}{g\mu_\beta}\frac{\langle(\delta\omega_{\text{anis}})^2\rangle}{\omega_e}, \tag{1}$$

where $\omega_e=1/\tau_e$ is the reciprocal of the exchange correlation time, g is the g value, μ_β is the Bohr magneton, and $\hbar$ is Planck's constant. As the temperature is lowered, the spin fluctuations slow down and the narrowing becomes less effective. This same picture is applicable to antiferromagnetic insulators,[27] in many of which W^{ex} follows a power law in reduced temperature, describable by taking $\omega_e=\omega_0 t^{\gamma}$. We have searched for such behavior in the Ag-Mn spin-glass system and have indeed found that $W^{\text{ex}}\propto t^{-\gamma}$ represents the low-frequency data well over more than a decade in reduced temperature, as demonstrated in the log-log plot of Fig. 3. The X-band width follows the same power law over a more limited range of t. The frequency-dependent departure from the power-law behavior at temperatures near T_g is also reminiscent of EPR in antiferromagnets.[28]

To systematize the presentation of the width data, least-squares fits were made to the form

$$W=a+bT+ct^{-\gamma} \tag{2}$$

where a, b, c, and γ are adjustable. The values of these parameters giving the best fit for the low-frequency data are collected in Table I. Different exponents γ give best fit for different samples, although the observed variation is limited to 1.5 ± 0.3. Because of systematic errors during any experimental run, the best fit is not necessarily fixed by the least-squares criterion of minimum rms deviation. To reduce systematic errors, values of a, b, and c were found by the least-squares criterion for various *fixed* γ. The best γ was then selected by two methods, which gave consistent results within experimental error. In the first, the deviations between fit and experiment were plotted against T for various γ, and γ was selected to minimize

TABLE I. Parameters of the linewidth fit, Eq. (2), for $AgMn_xSb_y$. T_g is the temperature of the dc magnetization cusp, a is the residual width, b is the thermal broadening, c is the strength of the linewidth divergence, and γ is the power-law exponent.

x (at. %)	y (%)	T_g (K)	a (G)	b (G/K)	c (G)	γ
2.6	0	10.18	−8	2.48	21	1.4
2.6	0.06	9.98	−7	2.75	24	1.3
2.6	0.17	9.79	3	3.15	24	1.6
2.6	0.22	9.65	4	3.29	30	1.5
2.6	0.46	9.10	16	4.35	43	1.8
2.6	0.57	8.67	32	4.60	48	1.8
5.8	0	19.28	−43	2.53	34	1.2
7.8	0	24.56	−70	2.67	37	1.4
10.3	0	31.43	−105	2.68	40	1.4

systematic deviations from zero. In the second method, the least-squares fit was performed for only those data points taken at temperatures greater than some temperature T which we shall designate as $T_{\min}$. When the assumed functional form (including the assumed γ) is a reasonable representation of the data, then all parameters of the fit, and also the rms deviation σ, are insensitive to $T_{\min}$. On the other hand, if the form is unsatisfactory, the parameters continually readjust themselves as $T_{\min}$ is reduced.

It should be stressed that in this parametrization $t=(T-T_g)/T_g$, where T_g was determined from the low-field dc magnetization. Hence the linewidth can be analyzed consistently in the above manner with a T_g which is *independent* of measuring frequency.

The strength c of the linewidth divergence is plotted in Fig. 4 against Sb and Mn concentrations. For the 2.6 at.% Mn samples, c increases linearly at the rate 46 G/at.% Sb from an intercept of 21 G for zero Sb. The strong Sb dependence rules out dipole-dipole interactions as the major source of line breadth near T_g; any dipole-dipole broadening will be unaffected by the presence of Sb. We attribute W^{ex} to exchange narrowing of a Dzaloshinski-Moriya anisotropic interaction. Fert and Levy[4] have described a mechanism by which spin-orbit scatterers introduce into the Mn-Mn interaction a skew-symmetric Dzaloshinski-Moriya term of significant magnitude. Using Eq. (1) with[6] $\langle(\delta\omega_{\text{anis}})^2\rangle=Ax^2+Bxy$, and the assumption that $\omega_0 \propto T_g$, which goes roughly as x, one finds $W^{\text{ex}} \propto Ax+By$. The linear dependence on y is observed. That the x dependence is weaker than linear at Mn concentrations of several percent should not be too surprising. The same effects which cause T_g to increase as $x^{0.8}$ rather than linearly in x may also weaken[12] the x dependence of $\langle(\delta\omega_{\text{anis}})^2\rangle$. Furthermore, metallurgical problems such as Mn clustering may have an effect at the larger Mn concentrations.

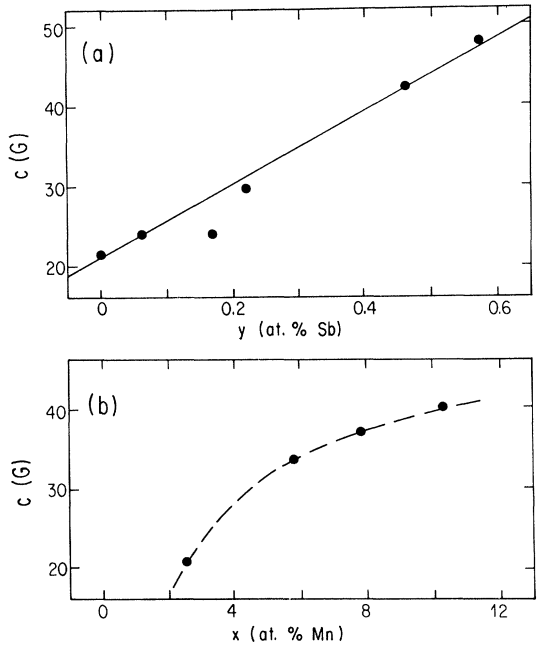

FIG. 4. Concentration dependences of the strength of the power-law term in the width (a) vs Sb concentration in $AgMn_{0.026}Sb_y$, and (b) vs Mn concentration in $AgMn_x$. Solid line has slope 46 G/at.% Sb. Dashed line is a guide for the eye.

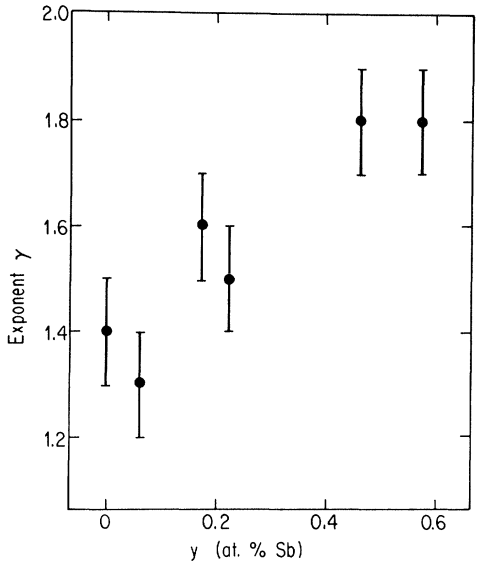

FIG. 5. Antimony concentration dependence of the power-law exponent γ in $AgMn_{0.026}Sb_y$.

The exponent γ is independent of Mn concentration but increases noticeably with Sb doping (Fig. 5). Assuming that the trend is not due to some undetected systematic error, the variation of γ could be associated with a range dependence of the exchange coupling, with consequent alteration of the spin-glass dynamics over the observed temperature range. Possible evidence for a range dependence of the exchange may also be found in the depression of T_g with Sb doping (Fig. 6), which occurs at rate -2.5 deg/at.% Sb for $x=2.6$ at.% Mn. No quantitative estimates of this effect are known to us. Furthermore, we

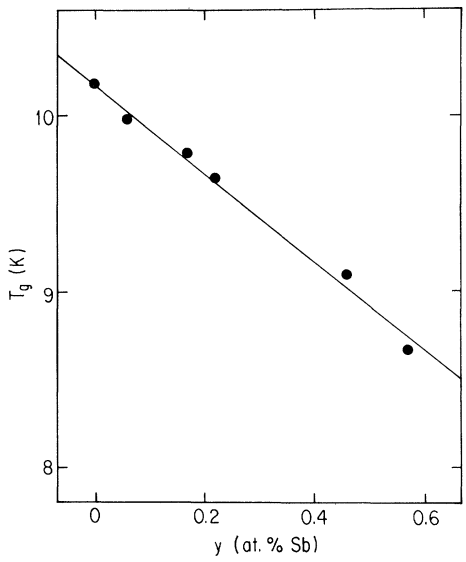

FIG. 6. Variation of the cusp temperature T_g with Sb concentration in $AgMn_{0.026}Sb_y$. T_g is depressed 2.5 K/at.% Sb.

find that T_g is *increased* with Pt doping, indicating that the introduction of spin-orbit impurities may have a more complicated effect on T_g than simply a reduction of the range of the interactions.

For the 2.6 at. % Mn samples, the thermal broadening b is linear in y with slope 3.9 G/deg at. % Sb. This behavior is a well-known consequence of the opening of the magnetic resonance bottleneck with addition of spin-orbit scatterers.[23] The slope of b vs y is determined by the Sb spin-flip scattering rate $\partial(1/T_{SL})/\partial y$. Following the analysis of Davidov *et al.*[29] and neglecting exchange enhancement of the conduction-electron susceptibility [$U\chi(0)=0$], we extract $\partial(1/T_{SL})/\partial y = 6.3\times10^{7}$ $\sec^{-1}$/ppm Sb, corresponding to a spin-flip crossection $\sigma_{Sb}=7.7\times10^{-18}$ cm^2. These values are only one-quarter as large as those of Ref. 29; however, their values of b for samples without Sb are also much different from expectations derived from Ref. 30, perhaps due to spin-glass ordering effects. The bottleneck theory also explains the thermal-broadening rate's independence of x for $x > 1\%$, where T_{SL}^{-1} is dominated by a term proportional to x. By comparing this x-independent rate with $\partial b/\partial y$, we deduce $\sigma_{Mn}=0.26\sigma_{Sb}$.

The residual width a depends linearly on both x and y, increasing at rate 65 G/at % Sb and decreasing at -13 G/at. % Mn, passing through zero for approximately 2 at. % Mn. A significantly negative residual width has been explained by Stewart,[31(a)] who argued from the Hasegawa equations that a is related to a "true" residual width a_0 via $a=a_0-b\Theta$, where Θ is the Weiss temperature. Thus a positive Θ can force the value of a deduced at high temperature to be negative. High-temperature susceptibility data from $AgMn_x$ shows that Θ increases through zero near $x=2$ at. % to 30 K for $x=10$ at. %,[16] explaining the sign of a and also a significant fraction of its magnitude. This argument has recently been made to explain the negative residual widths in $La_{1-x}Gd_xAl_2$.[9]

The $b\Theta$ term arises physically because the term proportional to T in Eq. (2) should more precisely be written as b'/M, where M is the temperature-dependent magnetization.[31(b)] If M obeys a Curie-Weiss law, a positive Θ causes an apparent negative residual width, as just noted. If M departs from a Curie-Weiss law, the temperature dependence of W^{ex} will be modified. For the magnetic fields applied in the (1—2)-GHz experiments, departure from a Curie-Weiss law occurs[17] only for $t<1.2$ in $AgMn_{0.026}$ (and at even lower t for larger x). The values of c and γ in Table I were obtained from larger t and are unaffected.

Preliminary measurements have been made in $AgMn_{0.026}Pt_y$ and in $CuMn_{0.010}Pt_y$, with results qualitatively in agreement with those of the $AgMn_{0.026}Sb_y$ series.

To conclude this section, the region nearest T_g will be considered. The excess linewidth departs from the $t^{-\gamma}$ behavior below some reduced temperature t^*, which depends on frequency ω and on Mn concentration x. The Sb concentration has no measurable effect on t^*. As x is increased from 2.6 to 10.3 at. %, t^* for the X-band width decreases from 0.6 to 0.3. At the lowest Mn concentration, for which the effect of frequency is most conspicuous, t^* for the (1—2)-GHz width is approximately 0.3. Thus the range over which the power-law dependence of W^{ex} is obeyed increases as either the frequency (i.e., resonant field) is reduced or the Mn concentration is increased. It should be noted that the departure from power-law behavior near T_g is not a consequence of our using the simple form bT in Eq. (2), for use of the more precise form b'/M would *increase* that departure.

IV. LINE-SHIFT RESULTS

At high temperature, $T \gg T_g$, all samples exhibit a resonant field corresponding to $g\approx2$, in agreement with previous investigations.[30] As T approaches T_g from above, the resonance shifts towards lower field. The nature of the shift for the 2.6 at. % Mn sample with no Sb doping is illustrated in Fig. 7 for several spectrometer frequencies. The shift is plotted as a g value obeying $h\omega=g\beta H^{res}$. Clearly the shift can be properly described neither as a pure g-value shift nor as a simple, frequency-*independent* internal field. Within the narrow range 1—2 GHz, the shift is adequately representable as a g-value shift; at higher frequency and below t^* it may be described as a frequency-dependent internal field. The magnitude of the low-frequency shift at any given reduced temperature is rather insensitive to Mn concentration but increases with Sb doping. These behaviors are shown in Figs. 8 and 9.

Within the low-frequency limit $\omega \ll \omega_e$ the shift corresponding to Eq. (1) is

$$\Delta H^{res}=\frac{\hbar}{g\mu_\beta}\frac{\omega\langle(\delta\omega_{anis})^2\rangle}{\omega_e}, \tag{3}$$

implying a g-value shift, as observed for $\omega/2\pi \lesssim 2$ GHz, and a power-law dependence $\Delta H^{res}\propto t^{-2\gamma}$. This power-law prediction is difficult to test directly, partly because the lines become broad, making determination of H^{res} im-

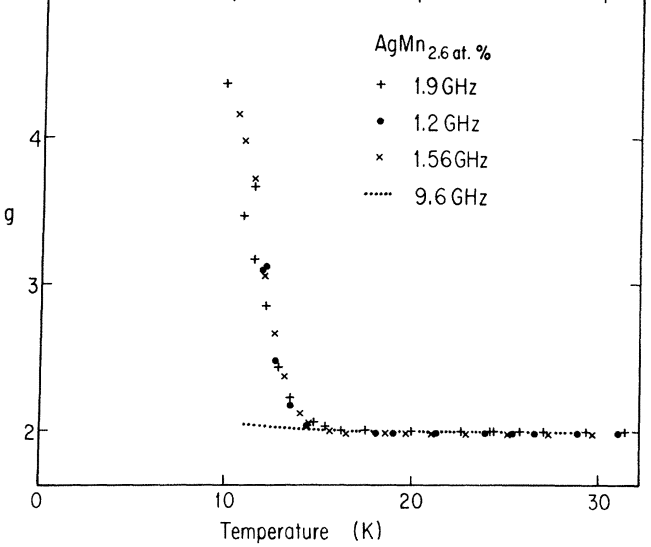

FIG. 7. Temperature dependence of the line position in $AgMn_{0.026}$ for four spectrometer frequencies. Line position is presented as an effective g value.

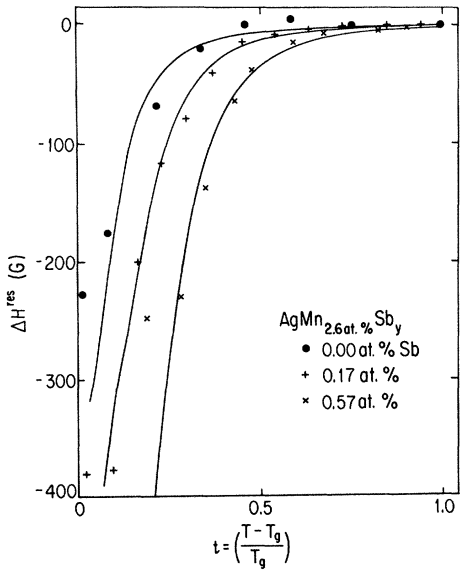

FIG. 8. Line shift vs reduced temperature in $AgMn_{0.026}Sb_y$ for three values of y at spectrometer frequency 1.9 GHz. Solid lines are the respective predictions of Eq. (3) with parameters chosen as described in the text.

precise, and partly because the shift data is useful for fitting only over the very narrow temperature range $t^* \lesssim t \lesssim 0.5$. At temperatures $t > 0.5$, the shift is not measureably different from zero, and at $t < t^*$, the low-frequency limit no longer applies. For our best data set, $AgMn_{0.026}Sb_{0.0022}$ at 1.88 GHz, we obtain an exponent 3.0, exactly twice the exponent which fits the corresponding W^{ex}; for other data sets, the exponents for the shifts scatter widely.

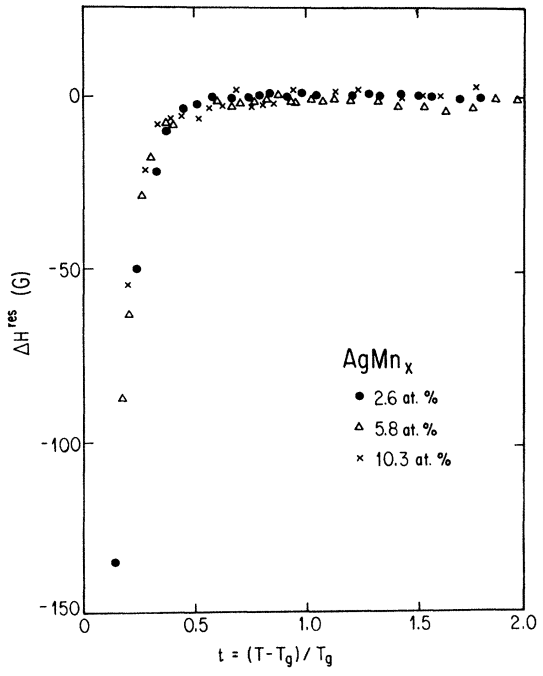

FIG. 9. Line shift vs reduced temperature in $AgMn_x$ for three values of x at spectrometer frequency 1.0 GHz.

The $t^{-2\gamma}$ form for the shift can be tested indirectly in the following way. The values $\langle(\delta\omega_{anis})^2\rangle^{1/2}/2\pi = 1.6$ GHz, $\omega_0/2\pi = 29.2$ GHz, and $\gamma = 1.5$ reproduce both width and shift in $AgMn_{0.026}Sb_{0.0022}$ at low frequency. The shifts for other Sb concentrations can then be predicted from Eq. (3) by taking $\langle(\delta\omega_{anis})^2\rangle \propto c$ (consistent with the width data) and ω_0 independent of y. In Fig. 8 the resulting predictions are compared to the measured shifts for three Sb concentrations using values of c and γ from Table I. The $t^{-2\gamma}$ behavior and the absolute magnitudes are successfully predicted.

V. DISCUSSION

A. Microscopic theory

The first attempt to calculate microscopically the EPR linewidth and shift in the paramagnetic phase of spin-glasses was made by Salamon.[8] In essence he attributed the excess linewidth to exchange-narrowed dipolar interaction. The calculation was based on the Mori-Kawasaki formalism, which has long been applied near ordinary critical points.[27] In this case the spins were expanded in the basis λ of the isotropic Ruderman-Kittel-Kasuya-Yosida (RKKY) exchange interaction[32] instead of Fourier components. The resulting four-spin correlation functions were factored in the random-phase approximation, retaining only the terms diagonal in λ. The resulting divergences, as $(T/T_g - 1)^{-3}$ for the width and as $(T/T_g - 1)^{-5}$ for the shift, were stronger than observed in Cu-Mn by Salamon[7,8] and in Ag-Mn and Cu-Mn in the present work.

Levy, Morgan-Pond, and Raghavan[12] have recently presented a microscopic theory which differs from Salamon's in two important respects. Firstly, the dominant anisotropy contributing to the exchange-narrowed width is the Dzaloshinski-Moriya interaction, rather than dipolar anisotropy. Secondly, while decoupling the four-spin correlation functions, attention is focussed on the off-diagonal ($\lambda \neq \lambda'$) contributions, compared to which the diagonal terms are of order N^{-1}. Thus Salamon's integral over a single Kinzel-Fischer correlation function is replaced by a double integral over a product of two correlation functions. The linewidth and line shift are given by the real and imaginary parts of

$$\Gamma(\omega,T) = \frac{2N\langle D^2\rangle_c S(S+1)}{9\hbar^2\Delta} I(\omega,T) = A_0 I(\omega,T) , \qquad (4)$$

where N is the density of spins S, $\langle D^2\rangle_c$ is an average over a distribution of Dzaloshinski-Moriya couplings, and Δ is the relaxation parameter in a Langevin equation of motion of the spin. In the zero-frequency limit,

$$I(0,T) = \left[\frac{T}{T_g}\right]^2 \left\{\frac{2a}{\pi}\left[\frac{T}{T_g}\right]\left[K\left[\frac{1}{a}\right] - E\left[\frac{1}{a}\right]\right] - 1\right\} , \qquad (5)$$

where K and E are elliptic functions and $a = (T^2 + T_g^2)/(2TT_g)$. At finite frequencies, $I(\omega,T)$ can be cal-

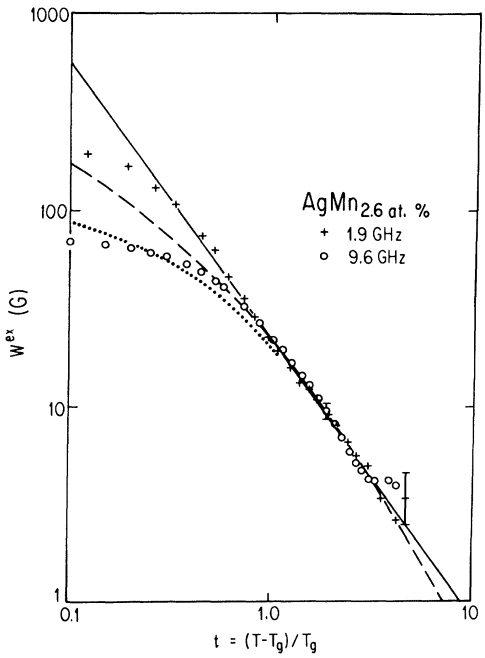

FIG. 10. Comparison of experimental linewidths in $AgMn_{0.026}$ at two frequencies (symbols) with predictions of the microscopic theory, Eqs. (4) and (5), using $A_0=280$ G. Dashed line: $\omega=0$. Dotted line: $\omega/2\pi=50$ GHz. Solid line shows the power-law fit with $\gamma=1.4$.

culated numerically. Levy *et al.* estimate $A_0=660$ G for $AgMn_{0.026}$. The linewidth prediction of Eq. (4) is qualitatively in agreement with our data (see Fig. 1 in Ref. 12) with $A_0=312$ G, but the predicted shift is much smaller than observed.

As a more stringent test of the theory, we have plotted the temperature-dependent part of the width from Eq. (4) as a dashed line on the log-log plot of Fig. 10. The slope for $t>1$ agrees well with the measurements, but the detailed shape of the zero-frequency prediction falls below the data on approaching T_g. This discrepancy, which begins at higher temperature than that at which the power-law behavior (solid line) breaks down, is worsened when the frequency dependence of $I(\omega,T)$ is taken into account. Adjustment of the residual width a and Korringa rate b does not appreciably improve the agreement. Furthermore, the predicted frequency dependence is weaker than that observed. For example, the dotted line shown with the X-band data in Fig. 10 was calculated for $\omega/2\pi=50$ GHz using $\Delta=kT_g$ as in Ref. 12. It should be noted that the theory also predicts a temperature-independent width of magnitude $A_0/2$, which would appear as a large positive contribution to the residual width. The data in Table I show no evidence for such a term.

Linear concentration dependences of A_0 on x and y are predicted by the theory within the same approximation considered in Sec. III. As noted, the predicted y dependence is observed, but the actual x dependence is weaker than linear. The relative strengths of the x and y dependences are difficult to estimate because $N\langle D^2\rangle_c$ is uncertain. In particular, $N\langle D^2\rangle_c$ depends on Mn spin S as $(\frac{5}{2}-S)^2$ and should be adjusted rapidly downward if S is closer to $\frac{5}{2}$ than supposed in Ref. 12. An alternate evaluation[33] using the zero-temperature anisotropy energy gives a value of $N\langle D^2\rangle_c$ which is smaller by a factor of 20. If we should simultaneously reduce $N\langle D^2\rangle_c$ and Δ by a factor of 5 from the values used for the comparison of Fig. 10, A_0 would remain unchanged while agreement with the frequency dependence of the width and the magnitude of the shift would be improved. Although we have no theoretical justification for this at present, adjustments to mean-field theory may result in such a reduction.

To summarize, the theory of Levy, Morgan-Pond, and Raghavan successfully predicts the power-law temperature dependence of W^{ex} for $T\gtrsim 2T_g$, and, with readjustment of $N\langle D^2\rangle_c$ and Δ, may fit the shift also. Thus the agreement is potentially quantitative. The outstanding theoretical question is why does the predicted W^{ex} for $\omega=0$ falls so much below the low-frequency data for $T<2T_g$, or from a different perspective, why do we see a better power-law behavior than that anticipated by the theory?

B. Phenomenological theories

A number of phenomenological theories applicable to EPR in the low-temperature spin-glass phase have been developed in response to experimental work.[1,2] Schultz and co-workers[2] presented a model free energy containing both remanence and anisotropy which explained the slope of their linear field-frequency relationship and which predicted the existence of a second mode, which was observed.[2] Their free energy was put on a firmer theoretical basis by Henley, Sompolinsky, and Halperin[34] who calculated the field-frequency relationship for general orientation of H relative to the equilibrium remanent magnetization. No damping was included in either calculation. Saslow[13] extended the work of Halperin and Saslow[35] to include damping and anisotropy in the macroscopic equations of motion. More recently, Becker[14] has derived the spin dynamics, including relaxation, using a projection operator approach.

One may attempt to extend Saslow's theory to $T>T_g$ by assuming that the remanent magnetization becomes zero in the paramagnetic phase while the anisotropy triad $(\hat{N},\hat{P},\hat{Q})$ continues to persist. Any additional applied field (such as the microwave H_1) changes both the magnetization and the local, metastable spin triad $(\hat{n},\hat{p},\hat{q})$, but the spin triad relaxes back towards $(\hat{N},\hat{P},\hat{Q})$ at rate u (equal to U^{-1}). For narrow lines, the line shift is given by[36]

$$\Delta H^{\rm res}=\left[\frac{\hbar}{g\mu_\beta}\right]\omega-H^{\rm res}=\left[\frac{g\mu_\beta}{\hbar}\right]\frac{(K/\chi)\omega}{\omega^2+u^2}\,, \tag{6a}$$

and the increase of the width over the high temperature $a+bT$ behavior is, in field units,

$$W^{\rm ex}=\left[\frac{g\mu_\beta}{\hbar}\right]\frac{(K/\chi)u}{\omega^2+u^2}\,, \tag{6b}$$

with K the anisotropy constant and χ the magnetic susceptibility. The theory of Becker[14] gives the same forms for these equations with the identification $u\equiv M_2/$

(Kk_BT).

As pointed out by Becker[14] these phenomenological equations concur qualitatively with experiment in several important respects. The shift is proportional to frequency in the low-frequency limit, but for high frequency, $\omega > u$, the shift becomes smaller. W^{ex} is independent of frequency for low frequency but is reduced when $\omega > u$. That these frequency dependences are observed in our data for $t \lesssim 0.5$ indicates that the order of magnitude of u is $2\pi \times 10^{10}$ sec^{-1}. In the low-frequency limit, $\omega \ll u$, ΔH^{res} and W^{ex} are proportional to $(K/\chi)u^{-2}$ and $(K/\chi)u^{-1}$, respectively, which are identical in form to the expressions of the exchange-narrowing model[8] if one arbitrarily identifies u with $\omega_e = \tau_e^{-1}$. In fact, Eqs. (6) then have the more general forms expected for line narrowing when the condition $\omega \ll \omega_e$ cannot be assumed.[37]

Equations (6) may be treated as phenomenological forms in which the empirical behavior of the parameters must be deduced. In this spirit, we compare the low-frequency limits of Eqs. (6) to Eqs. (1) and (3), which have already been fit to the low-frequency widths and shifts. That comparison forces the identifications $(K/\chi) = (\hbar/g\mu_\beta)^2 \langle (\delta\omega_{anis})^2 \rangle$ and $u = \omega_e$. Thus we immediately learn from the results of Sec. IV that $u = u_0 t^\gamma$ with u_0 independent of y, that K/χ is independent of temperature[38] and that $K/\chi \propto c$. It is interesting to note that the magnitudes of K obtained from these identifications are in fact similar to those determined from EPR below T_g. Recent measurements within the low-temperature phase[39] give, for $x = 6.3$ and 4.9 at.%, $K(T_g) = 0.22\text{K}(0) = 65$ Oe^2 emu/g and 45 Oe^2 emu/g, respectively. Assuming a quadratic dependence of D on x, one expects $K(T_g) = 12$ Oe^2 emu/g for $x = 2.6$ at. %. With $K/\chi \propto c$, we obtain for $AgMn_{0.026}$, $K/\chi = 2.3 \times 10^5$ Oe^2, and using our experimental static susceptibility at the cusp, $K(T_g) = 28$ Oe^2 emu/g, in disagreement by less than a factor of 3.

According to Eqs. (6) the power-law dependence of W^{ex} is obtained only for temperatures high enough that $u \gg \omega$. Thus t^* is the frequency-dependent temperature below which the low-frequency limit begins to fail. As ω increases, t^* also increases, reducing the range over which W^{ex} obeys the power law. The experimental fact that t^* decreases with increasing Mn concentration can be explained if the additional Mn enhances u_0. Furthermore, the low-frequency shift is essentially independent of Mn concentration, as shown in Fig. 9. Since $K/\chi \propto c$ and c increases with x (Fig. 4), Eq. (6a) requires that u_0 increase with x. This result seems natural if one keeps in mind the identification between u and the reciprocal of the exchange correlation time.

Outside the low-frequency limit, the frequency dependences contained in Eqs. (6) are too strong. Figure 11 shows W^{ex} at two frequencies along with the predictions. In the high-frequency limit, $\Delta H^{res} \propto \omega^{-1}$, predicting an X-band shift which is much smaller than observed. If Eqs. (6) still apply at the X band, then both K/χ and u_0 must themselves depend on frequency. Unfortunately there is too much scatter in the present shift data to allow a confident point by point extraction of K/χ and u at different temperatures. However, it should be noted that the low-frequency data show no hint of frequency dependences in the parameters. The existence of a g-value shift and the frequency independence of W^{ex} require that $(K/\chi)u_0^{-2}$ and $(K/\chi)u_0^{-1}$ be independent of ω, in turn requiring that K/χ and u_0 be individually independent of ω, at least at small ω.

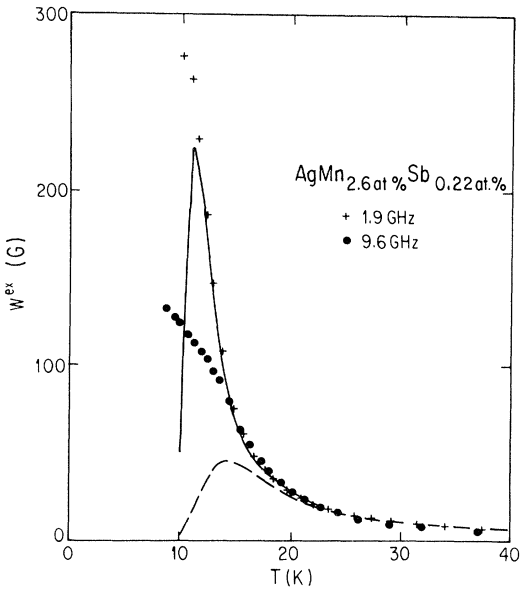

FIG. 11. Comparison of experimental linewidths in $AgMn_{0.026}Sb_{0.0022}$ at two frequencies (symbols) with predictions of Eq. (6b). Solid line: $\omega/2\pi = 1.9$ GHz. Dashed line: $\omega/2\pi = 9.6$ GHz. In these curves, $K/\chi = 3.1 \times 10^5$ Oe^2 and $u_0/2\pi = 29.2$ GHz.

We conclude that the phenomenological theories are adequate to represent the low-frequency data to exactly the same extent as Eqs. (1) and (3) with the implications that $u = u_0 t^\gamma$ and that K/χ is independent of temperature. These theories also describe one manner in which the low-frequency limit begins to break down. However, comparing the 9.6-GHz data to the (1—2)-GHz data, the predictions give too strong frequency dependences for the width and shift, implying that K/χ and u_0 themselves depend on frequency (or applied field). We have not yet discovered the functional forms for such dependences. Further experiments at frequencies between 2 and 9.5 GHz are in progress. In the next section we turn to a more general discussion of the breakdown of the low-frequency limit.

C. Behavior near T_g

Throughout this paper we have ascribed the EPR linewidth in the paramagnetic phase to exchange narrowing of an anisotropic interaction, as expressed in Eq. (1). The form $\omega_e \propto t^\gamma$ predicts a power-law behavior for W^{ex} which is observed only for $t > t^*$. The eventual failure of the power law for each frequency at lower temperatures admits two broad categories of explanation, which may be termed intrinsic and extrinsic[27]: Either the slowing down of ω_e does not continue as a power law because of some intrinsic behavior of spin-glasses in applied fields, or the nature of the EPR probe precludes continuing observation of that power law.

The suggestion that spin-glass dynamics may be changed by an applied magnetic field has been made by us in an earlier Ag-Mn EPR investigation.[11] As evidenced by susceptibility and magnetization measurements,[40,15] static spin-glass properties near and below T_g depend strongly on field. The strength of such dependences is characterized by the ratio $g\mu_\beta H/kT_g$, indicating that the temperature regime over which field effects can be felt decreases with T_g. Thus we expect t^* to increase with applied field, decrease with Mn concentration, and remain relatively insensitive to Sb concentration. These expectations agree with observation but are not in themselves sufficient evidence for the intrinsic nature of the effect.

The alternate viewpoint, advanced by Salamon,[8] attributes the failure of the power law at low t to breakdown of extreme motional narrowing.[41] The simple formulas (1) and (3) apply only so long as $\omega_e \gg \langle(\delta\omega_{\text{anis}})^2\rangle$. For $AgMn_{0.026}$, $\langle(\delta\omega_{\text{dipolar}})^2\rangle^{1/2}/2\pi \approx 0.4$ GHz from the method of moments, while from Sec. IV, $\langle(\delta\omega_{\text{anis}})^2\rangle^{1/2}/2\pi \approx 1.3$ GHz. Because the predominant anisotropy comes from the spin-orbit mechanism, the failure of this usual narrowing condition requires that t^* depend on y as well as x, and not on ω, and therefore can be ruled out immediately.

The simple narrowing formula can, however, fail in another sense. In the short-correlation-time limit defined by the Kawasaki condition $\omega_e \gg \omega$, both secular and nonsecular terms contribute to the average $\langle(\delta\omega_{\text{anis}})^2\rangle$, while in the long-correlation-time limit, which contains the rigid-lattice case, only the secular terms are involved.[27,37] Thus as the temperature is reduced while remaining entirely within the extreme motional-narrowing regime, exchange might no longer sufficiently dominate the Larmor frequency and the line might become narrower than otherwise expected as the nonsecular terms drop out. In this case t^* should increase with ω, decrease with x, and be insensitive to y, as observed. Furthermore for $AgMn_{0.026}$, $\omega_0 t^\gamma = 2\pi(9.5\text{ GHz})$ for $t \approx 0.5$, which is very nearly the measured t^*. As for the shift, when the Kawasaki condition has broken down, at least part of the system is transiently "condensed" into a spin-glass state for times longer than a Larmor period. This leads to the establishment of an internal anisotropy field whose value depends on the magnitude of the "cooling" field, or, in our case, the field (and hence, frequency) for resonance.

At present there is insufficient evidence to distinguish between the intrinsic and Kawasaki mechanisms for the rounding near T_g, especially if both are operative simultaneously. From the standpoint of the phenomenological theories, what seems to be required is a careful extraction of K/χ and u as functions of frequency. The necessary experiments are underway. Once u is characterized in greater detail, the question of the effect of magnetic field on spin-glass dynamics may be readdressed.

VI. CONCLUSION

The principal experimental conclusions of this investigation are the following.

(1) The linewidth diverges as $T \to T_g$ from above as a power law in reduced temperature $t = (T - T_g)/T_g$. This divergence is interpreted in terms of exchange-narrowed anisotropic interaction. The divergence ceases below a frequency and Mn concentration-dependent temperature t^* due either to the effect of the applied field on spin-glass dynamics or to failure of the Kawasaki condition.

(2) The strength of the linewidth divergence increases linearly with Sb concentration, supporting the view that spin-orbit scattering rather than dipolar interaction is responsible for the anisotropy. The line shift correlates with the width as Sb concentration is changed, demonstrating that width and shift arise from the same mechanism.

(3) The line shift is proportional to frequency at low frequency, but this proportionality does not continue to the X band. The shift can be properly described neither as a simple g-value shift nor as a frequency-independent internal field.

(4) Preliminary measurements in Cu-Mn and Cu-Mn-Pt have revealed no qualitative differences between the Cu-Mn and Ag-Mn systems.

Comparison with theory is, at present, inadequate. The phenomenological theories[13,14] fit the width and shift in the low-frequency limit, where they have the same form as the standard exchange-narrowing model. The fits imply that K/χ is independent of temperature and that $u = u_0 t^\gamma$. The deduced values of K agree with EPR and other measurements within the low-temperature phase. But the predicted frequency dependences are too strong. If the forms of the phenomenological equations are to be preserved, then K/χ and u_0 must depend on frequency or applied field.

The microscopic theory[12] fits the magnitude of the low-frequency width and, for $T \gtrsim 2T_g$, its temperature dependence. From the standpoint of this theory, it is particularly mystifying why we observe power-law behavior over such a large range of temperature. The predicted shift is entirely too small, unless perhaps if $N\langle D^2\rangle_c$ and Δ are changed in tandem. In any case, the temperature-independent contribution to the width remains a problem.

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*Present address: Physics Building, University of Sussex, Brighton BN1 9QH, U.K.

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