Vanishing anisotropy energy of a spin glass in electron-spin resonance at $T \geq T_g$

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Our findings indicate that in ESR measurements the anisotropy energy of the spin glass *Ag*Mn vanishes at $T \geq T_g$ when the resonant field $H_{res} \rightarrow 0$. An inexplicable finite value for the anisotropy energy above T_g, mentioned in earlier reports, should be induced by the external applied field. Our data analysis gives a critical exponent for the correlation length $\nu = 1.2 \pm 0.2$.

The theories of Halperin and Saslow,[1] Saslow,[2] and Barnes[3] have predicted the existence of three modes for the spin waves in a spin glass (SG). Schultz, Gullikson, and Fredkin[4] developed a simple hydrodynamic theory for interpreting their own experimental ESR data. Their theory assumes that the anisotropy is of vector character, thus predicting two transversal modes ω^+ and ω^-. Taking into account the triad character of the Dzyaloshinsky-Moriya (DM) anisotropy[5,6] in SG, Henley, Sompolinski, and Halperin[7] extended the hydrodynamic theory of Halperin and Saslow,[1] and predicted one longitudinal mode ω_L (as denoted in Refs. 1 and 2) in addition to the ω^+ and ω^-. The ω^+ mode is investigated by a number of workers (e.g., Refs. 4 and 8–10), and is, for the case of an applied field parallel to the remanence,[4,7]

$$\frac{\omega^+}{\gamma} = \left[\frac{H_{res}}{2} - \frac{\sigma}{2\chi}\right] + \left[\left(\frac{H_{res}}{2} + \frac{\sigma}{2\chi}\right)^2 + \frac{K}{\chi}\right]^{1/2}, \quad (1)$$

where σ is the magnetic remanence, χ the susceptibility, K the anisotropy energy (density) or constant, ω^+ the resonance frequency, γ the gyromagnetic ratio, and H_{res} the resonant field. One obtains from Eq. (1) an expression for the line shift δH_{res}:

$$\delta H_{res} \equiv H_{res} - H_{res}(g=2)$$
$$= H_{res} - \omega/\gamma = \frac{-K}{\chi\omega/\gamma - \sigma}, \text{ with } \omega = \omega^+. \quad (2)$$

In Eq. (1), all variables except K can be determined experimentally. Accordingly a method has been documented for determining the value of the anisotropy energy through substitution of experimental quantities in Eq. (1).

The K (from ESR) has been found to increase with the reduction of temperature, as expected. Yet, astonishingly, K has a finite value above the glass temperature T_g at $T \lesssim 1.5 T_g$ (see inset of Fig. 1). This behavior is in opposition to the general conception that spin frustration does not occur at $T > T_g$. Consequently the DM-type anisotropy in the SG must vanish.

The task of this note is to reveal the origin of this unresolved effect. In order to have a better insight into the problem, we present the typical behavior of the ω^+ mode. Figure 1 depicts a schematic diagram of the resonance frequency of the ω^+ mode vs the resonant field, showing typical experimental data at $T = \text{const}$ above T_g (filled circles). The solid curve depicts Eq. (1) fitted to the data points with K as a fit parameter. This curve intersects the ω axis at $\omega = \gamma/2\pi(K/\chi)^{1/2}$. Assuming that the missing data points at the low frequencies (or fields) should also correspond to the theoretical curve, a finite value for K would be expected above T_g. Due to the complications in obtaining experimental data of ESR in conventional cavities at multiple frequencies, this assumption remains to be verified.

Our argument was *a priori*; if the anisotropy energy above T_g must vanish, the experimental data should behave similar to the dashed curve reaching the origin in Fig. 1, meaning that at $T > T_g$, $K = 0$, and from Eq. (2), $\delta H_{res} = 0$. To prove this we developed a new ESR technique (ESR in microcoils[11]) which enabled us to perform ESR between 0.6 and 8.5 GHz with only one microcoil.[12] Figure 2 shows experimental data of the spin glass *Ag*Mn 10.3% ($T_g = 31.2$ K) for $0.7 T_g \lesssim T \lesssim 1.3 T_g$ for a zero-field cooling (ZFC). The numerous data obtained through the microcoil technique enable a proper analysis. It can be seen at a glance that the isotherms of $T \geq T_g$ tend to meet the origin in accordance with our expectation. Figure 3 presents three ESR lines of the isotherm $T = T_g$. The narrow ESR lines are from a 11-diphenyl-2-picrylhydrazyl (DPPH) marker and represent the $g = 2.0036 \approx 2$ reso-

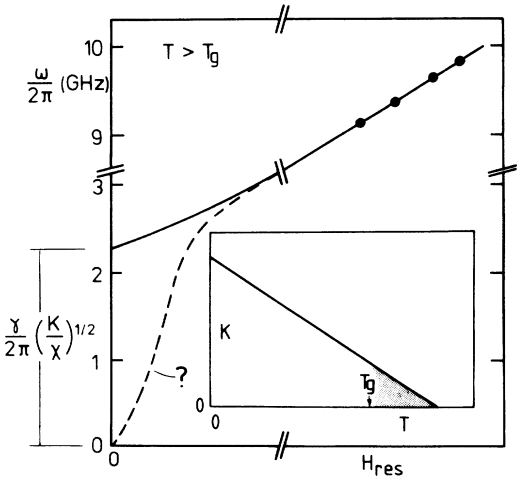

FIG. 1. Schematic behavior of $\omega^+(H_{res})$ in SG. Inset schematically shows the K obtained via the ESR. Up to now there is no explanation for a $K \neq 0$ in the shaded region.

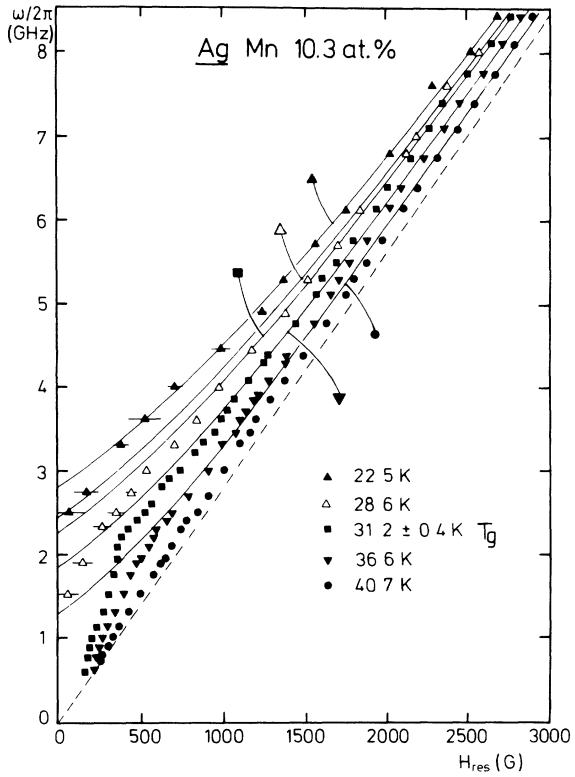

FIG. 2. $\omega/2\pi$ vs H_{res} (in ZFC experiment) at $0.7T_g < T < 1.3T_g$. The dashed line relates to $g=2$. The solid curves are the fits by Eq. (1) using K as a fit parameter.

nances. This figure shows how the line shift δH_{res} decreases at $\omega/2\pi \lesssim 2$ GHz with the reduction of ω. A vanishing δH_{res} means, from Eq. (1), a vanishing K. Figures 2 and 3 give strong evidence that the anisotropy energy in the ESR experiment approaches zero at $T \geq T_g$. Small extrapolations of the two isotherms below T_g (Fig. 2) intercept the ω axis, implying a finite value for K.[14]

The values of χ and σ were needed for fitting the data with Eq. (1), K being a fit parameter in accordance with the earlier documented method. These variables were measured in the magnetometer under the same conditions as in the ESR.[12] As seen in Fig. 2, only the data of the high frequencies $\gtrsim 6$ GHz (or fields $\gtrsim 2$ kG) can be fitted satisfactorily. The deviations of fit curves from the experimental ones are obvious at $\omega/2\pi \lesssim 4$ GHz. The intersection of a fit curve with the ω axis for any temperature is higher than the intersection of small extrapolations of low-frequency data. The difference of the two intersections diminishes with the reduction of temperature. Figure 4 shows K vs the temperature, with the triangles representing the K values extracted directly from the abovementioned extrapolation of the data. The open circles are from the fit procedure. We were curious to know the behavior of K at lower temperatures (< 22 K), but were not able to implement precise ESR under the ZFC condition at $T \lesssim 22$ K because of intensive line shift and strong line broadening. Therefore, we performed field-cooled (FC) ESR and magnetization measurements in a cooling field of 13 kG, in a 9.577 GHz cavity from $T \approx 50$ K down to 3.5 K.[12] Substituting the values of the experimental quantities into Eq. (1), we obtained a K value for each temperature. The filled circles in Fig. 4 represent the K values deduced from the FC measurements. The filled and open circles coincide with each other and give a nearly linear behavior for K (dashed line).

Our explanation for the discrepancy between the behavior of the K obtained via the earlier method, mentioned in the literature (circles, Fig. 4), and the K extracted from our analysis (triangles, Fig. 4) is primarily that the triangle data are acquired from an analysis in which $H_{res}=0$, whereas in the analysis of the round data $H_{res} \neq 0$. This is reminiscent of an induced magnetization in ferromagnets

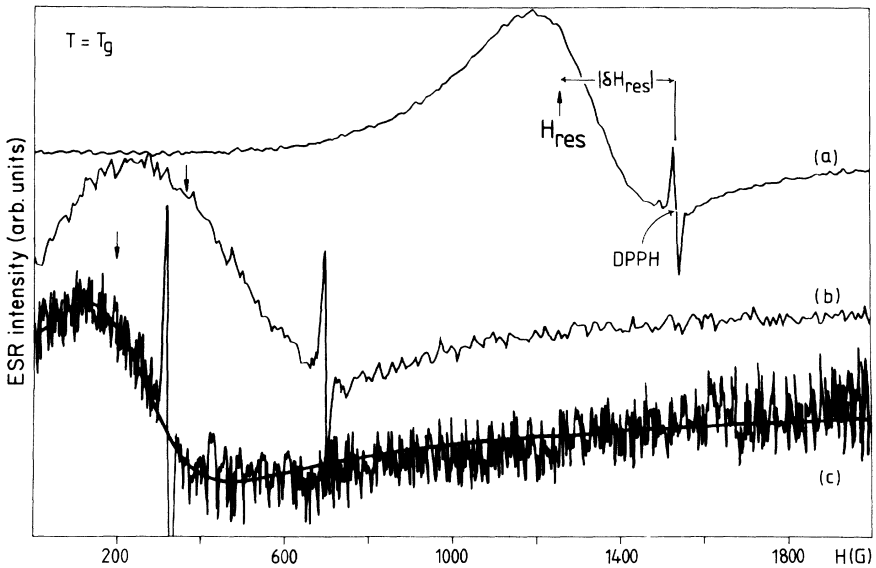

FIG. 3. Three ESR lines (see text): (a) $\omega/2\pi=4.3$ GHz, (b) $\omega/2\pi=1.95$ GHz, (c) $\omega/2\pi=0.88$ GHz. The vertical arrows show the resonance positions. Spectrum (c) includes also the Lorentzian fit curve (see Ref. 13 for our fit procedure).

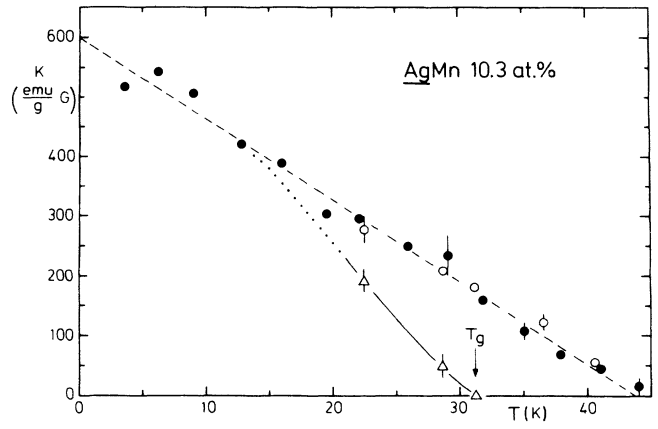

FIG. 4. The anisotropy constant K, determined under different conditions vs the temperature. Open circles denote K extracted from the fits of Fig. 2. Filled circles assigned to K resulted from the FC measurements. The triangles represent K, which is deduced by a reasonable extrapolation of the data of Fig. 2 to $H=0$.

when the external field is finite. If T_g is a critical temperature, the anisotropy energy is coupled to the order parameter q in SG.[15] (It is believed that the nonlinear susceptibility in SG behaves as q for $T>T_g$.[16,17]) A finite external field can influence the critical fluctuations, and induce an order parameter and consequently induce an anisotropy energy in the SG. An anisotropy energy intrinsic just to the spin system must be measured in $H\rightarrow 0$.[18]

Assuming a phase transition at T_g, we obtain the critical exponent of K ($H=0$), $\mu=1.2\pm0.2$, via fitting the triangles with a power law in the reduced temperature $t\equiv(T-T_g)/T_g$ (solid curve Fig. 4),

$$K(H=0)\propto|t|^{\mu}\propto(T-T_g)^{\mu}. \tag{3}$$

The study of Sompolinsky, Kotliar, and Zippelius[15] predicts an anisotropy constant for the Heisenberg SG below T_g, which vanishes at T_g in the mean-field limit as $(T-T_g)^{\mu}$, with $\mu=3$ in dimension $d=6$. If we use the Josephson relation $\mu=\nu(d-2)$,[19] where ν is the critical exponent of the correlation length for $d=3$, we get $\mu=\nu=1.2\pm0.2$. A Monte Carlo simulation of the Ising SG in $d=3$ has estimated $\nu=1.2\pm0.1$,[20] and the measurements of the nonlinear susceptibility give $\nu=1.3$,[17] both of these being in agreement with the ν of this work.

In Fig. 4 the optional dotted line matches the solid curve with the dashed line at $T\approx 0.5T_g$, meaning that for $T\lesssim 0.5T_g$, K should be field independent.

In summary, the static field can account for a $K\neq 0$ above T_g. An anisotropy intrinsic to merely the spin system can be measured in $H\rightarrow 0$. Assuming a phase transition at T_g, we deduced the exponent value of the correlation length $\nu=1.2\pm0.2$, this being in agreement with the results of the computer simulation and the magnetization measurements. In this investigation we did not observe the ω^- and ω_L modes, their having been detected only in CuMn 8% Ni 0.37%.[4,21] Additionally, the ω_L mode can be observed only in a frequency scanning ESR,[21] which does not apply here.

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