Comment on "Quenching of the Nonlinear Susceptibility at a $T = 0$ Spin Glass Transition"

In a recent Letter [1] Wu, Bitko, Rosenbaum, and Aeppli (WBRA) report a study of the quantum critical behavior of the Ising spin glass $LiHo_{0.167}Y_{0.833}F_4$. The spin glass (SG) phase undergoes a transition to the paramagnetic phase on increasing the strength of the transverse magnetic field H_t, increasing the quantum fluctuations. Experimentally this phase transition has been most extensively studied by WBRA and co-workers in Ref. [1], using the nonlinear susceptibility χ_3 and in two previous studies using the dynamic susceptibility [2,3].

The purpose of this Comment is to point out the following. In the work by WBRA and co-workers the SG transition temperature $T_g(H_t)$ is determined from dynamic susceptibility measurements (i) (Ref. [2], and compared to in Ref. [1]) and from nonlinear susceptibility measurements (ii) (Ref. [1]). However, in both cases incorrect methods to determine $T_g(H_t)$ have been employed. Consequently, the critical analysis of Ref. [1] is incorrect.

(i) The SG relaxation begins at the typical spin flip time $t_{\min}$ and ends at the maximum relaxation time $t_{\max}$ (in the SG phase $t_{\max}$ is infinite) [4]. In the time interval $t_{\min} \ll t \ll t_{\max}$ the equilibrium relaxation is rather close to logarithmic. In order to determine T_g from dynamic measurements, WBRA chose the temperature at which the relaxation becomes nearly logarithmic, $\alpha \approx 0$ (notation from Ref. [2]), in the experimental frequency window. However, this criterion does not correspond to the divergence of $t_{\max}$ and, therefore, does not correspond to T_g but rather to what is commonly called the freezing temperature $T_f(\omega)$ of the system (since on decreasing the temperature the criterion corresponds to the temperature where the maximum realization time just extends out of the experimental frequency window). $T_f(\omega)$ is only an upper bound on T_g and $T_g = T_f(\omega \to 0)$ [4]. As an example of how misleading this criterion can be, consider a classical 2D SG which has $T_g = 0$ K but finite freezing temperatures. The criterion $\alpha \approx 0$ in the experimental frequency window would incorrectly suggest a finite T_g for a 2D SG.

(ii) The nonlinear susceptibility χ_3 diverges at an ordinary SG transition. However, when χ_3 is measured by a finite probing frequency the response falls out of equilibrium before the transition temperature (at the temperature where the out-of-phase component first appears) and does not diverge at T_g. $\chi_3(\omega)$ instead shows a maximum at $T \approx T_f(\omega)$. The behavior is illustrated in Fig. 1 for the classical Ising spin glass $Fe_{0.5}Mn_{0.5}TiO_3$. In Ref. [1] (Fig. 3) $\chi_3(\omega)$ measured at a higher temperature and lower H_t has a larger maximum then $\chi_3(\omega)$ measured at a lower temperature and larger H_t. This does not imply that one of the curves diverges or that one of them would not diverge if zero frequency could have been used. It is, therefore, not correct to claim that the transition appears first order at one of the temperatures. In Ref. [1] the transition at lower H_t is analyzed by a static scaling analysis of the nonlinear susceptibility. The data analyzed are, however, in an (H_t, T) regime where the measured $\chi_3(\omega)$ does not correspond to its equilibrium value (there is an essential out-of-phase component in the regime used), e.g., at $T = 0.098$ K only data for $H_t \gtrsim 10$ kOe are equilibrium data (cf. Fig. 3, Ref. [1]). The claim of Ref. [1] that $\chi_3(\omega)$ is independent of frequency for $\omega < 2$ Hz and $H_t < 10$ kOe is most probably due to measurement uncertainty in a limited frequency interval (cf. Fig. 2, Ref. [1]). Since the static scaling (cf. Fig. 4, Ref. [1]) is made with dynamic data, the values deduced for "$T_g(H_t)$" and "γ" in Ref. [1] are incorrect [one expects that the measured dynamical $\gamma \to 0$ as $T \to T_f(\omega)$ since $\chi_3(\omega)$ does not diverge but shows only a maximum at $T_f(\omega)$].

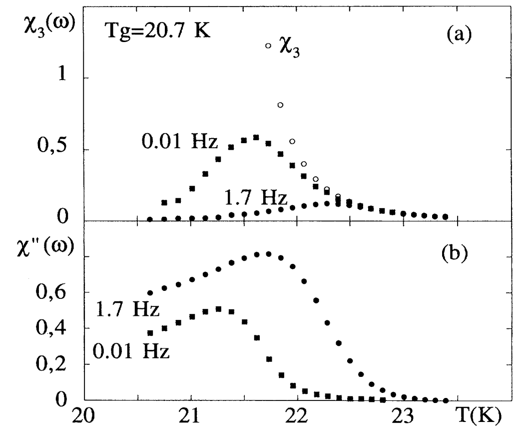

FIG. 1. (a) $\chi_3(\omega)$ and (b) $\chi''(\omega)$ vs temperature for $\omega = 0.01$ and 1.7 Hz measured on the classical Ising SG $Fe_{0.5}Mn_{0.5}TiO_3$ [5] $T_g = 20.7$ K. The open circles are χ_3 as found from static scaling.

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[2] W. Wu *et al.*, Phys. Rev. Lett. **67**, 2076 (1991).
[3] D. H. Reich *et al.*, Phys. Rev. B **42**, 4631 (1990).
[4] In the current system $t_{\min}$ is in the order of the experimental time scales, in some cases even within the experimental frequency window. (This is in contrast to many classical SG's where $t_{\min}$ is of the order of 10^{-12} s.) Experimental time scales, therefore, correspond to relatively short time scales in this system.
[5] Data from K. Gunnarsson *et al.*, Phys. Rev. B **43**, 8199 (1991).

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