

Thermodynamics of magnetic suspensions in correlation with temperature dependence of magnetic susceptibility

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In a thermodynamic circumstance, single-domain magnetic nanoparticles behave uniquely as they can exhibit superparamagnetism at room temperature, followed by spin-glass-like transition at low temperature. Experimentally, these two behaviors can be easily identified from the absence of hysteresis in the field strength-dependent magnetization curve, and the presence of thermo-remanent magnetization in the temperature-dependent magnetization curve, respectively. Maxwell relation additionally describes that the total entropy of such ferromagnetic system decreases with increasing the applied magnetic field (for a given temperature), and it increases with increasing temperature (for a given magnetic field). Referring to this magneto-caloric effect¹⁾, the change of magnetic entropy due to a magnetic field fluctuation is thermodynamically contributable to the temperature rise in magnetic suspension observed during the irradiation of alternating magnetic field. Therefore, understanding thermodynamic processes of dynamically-magnetized magnetic suspension and the corresponding measurable-parameters are of importance in optimizing the heat generation of magnetic nanoparticles used for practical application of magnetic hyperthermia.

The particle movement in fluidic medium, as well as spin fluctuation, is strongly affected by thermal energy in addition to the availability of external magnetic field. For constant magnetic field, increasing temperature will amplify both the random Brownian motions and the spontaneous flipping of magnetic moments, giving rise to a lower magnetization value. A sufficiently-high temperature will completely randomize the magnetic moments so that paramagnetism appears with respect to Bloch's law. Hence, the change of magnetic entropy (i.e. spin entropy) is associable with the temperature-dependent magnetic susceptibility. To investigate the entropy change due to alternating magnetic field, we recorded complex magnetic susceptibility of two types of magnetic suspension containing: sodium α -olefin sulfonate-coated iron oxide nanoparticles (Specimen 1) and carboxydextran-coated iron oxide nanoparticles (Specimen 2) at 30°C to 50°C under 6.5 Oe rms magnetic fields with frequency of 1 kHz, 10 kHz, 100 kHz, and 1000 kHz. We found in both specimens that the decrease of magnetic susceptibility χ in higher temperature was clearly observable at 1 kHz, but increasing frequency of the applied field to 1 MHz resulted in a less temperature-dependence of χ (Fig. 1a). Meanwhile, the gradient of magnetic susceptibility upon temperature $d\chi/dT$ estimated from the linear regression of Fig. 1a further convinces that temperature has a small contribution to the high-frequency relaxation-dynamics in the respective specimens (Fig. 1b). Here, Fig. 1b also means that the change of magnetic entropy is negligible at higher frequency.

Reference

- 1) N. A. de Oliveira and P. J. von Ranke, Phys. Rep., **489** (2010) 89-159.

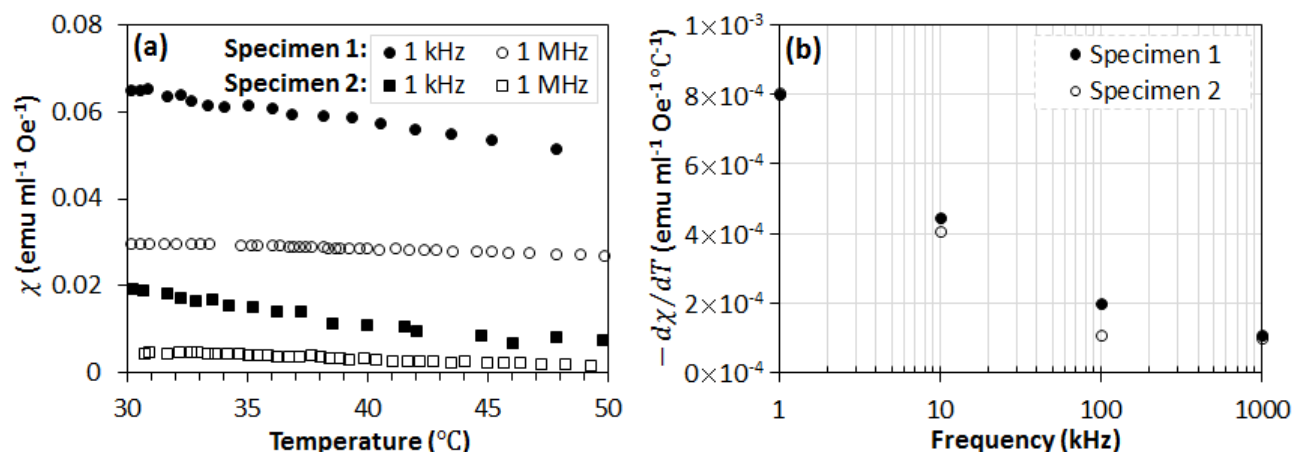


Fig. 1 Measurement result of (a) temperature-dependent magnetic susceptibility and (b) the respective gradient of magnetic susceptibility upon temperature