ROTATIONAL HYSTERESIS FOR FIELD-COOLED MAGNETIC FLUIDS NEAR MELTING POINT

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Abstract. – The magnetic torque of some magnetic fluids was investigated as a function of cooling magnetic fields. A uniaxial magnetic anisotropy in frozen magnetic fluids is induced by the formation of clustering of magnetic particles. A rotational hysteresis loss is independent of cooling field.

1. Introduction

The colloidal particles in a magnetic fluid are subject to the Brownian motion when no magnetic field is applied. In a magnetic field, however, they form chain-like clusters because of the dipole-dipole interaction between the magnetic particles. When the magnetic fluid is cooled in a magnetic field below the melting point of carrier fluid, the particles in the solvent are frozen with the formation of clustering of the particles. Therefore, a uniaxial anisotropy is induced by the field cooling [1]. In this paper, temperature and cooling field dependences of the induced magnetic anisotropy and rotational hysteresis loss observed in a torque measurement are discussed in terms of mesoscopic phase transition for viscous fluids.

2. Experimental procedure

Three kinds of magnetic fluid containing Fe_3O_4 particles whose carrier fluids are water (H_2O), heavy water (D_2O), and paraffin were prepared. The melting temperature, T_m, was determined to be about 196, 268 and 276 K for paraffin-, H_2O– and D_2O–base magnetic fluids, respectively. The torque of the specimens was measured in the temperature range from 77 to 300 K as a function of the intensity of cooling magnetic field, H_f.

3. Results and discussion

Figure 1 shows the temperature dependence of the magnetic anisotropy constant, K_u, for H_2O– and D_2O–base magnetic fluids frozen in H_f=4.0 kOe and measured in H_e=4.0 kOe. The value of K_u was almost independent of temperature below the temperature a few tens of degrees lower than T_m. The magnitude of K_u depends strongly on the H_f. For example, the value of K_u frozen without cooling field was less than one tenth of that cooled in H_f=4.0 kOe. Therefore, the anisotropy should be caused by the formation of clustering of magnetic particles in a magnetic fluid. In

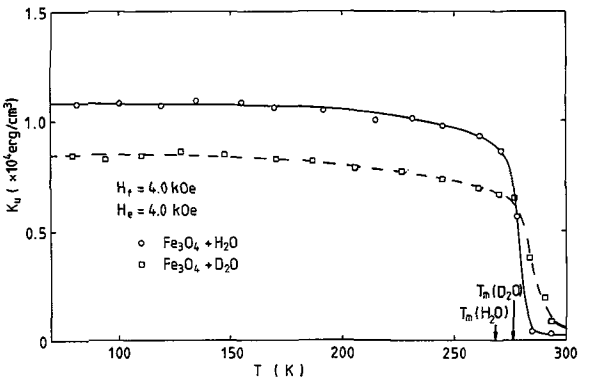

Fig. 1. – Temperature dependence of the induced magnetic anisotropy for H_2O– and D_2O–base magnetic fluids frozen in H_f=4.0 kOe. The arrows indicate the melting points of H_2O and D_2O.

the temperature range near T_m, K_u decreases with increasing temperature according to the Andrade law for viscosity [2]. Small magnetic anisotropy still exists in a liquid just above T_m but disappears with the recovery of fluidity.

The magnetic torque observed on present specimens exhibits a rotational hysteresis, E_{rot}, below T_m, whether H_f= 0 or not. This E_{rot} is independent of the intensity of H_f. Figures 2 and 3 show the temperature dependence of E_{rot} for the H_2O– and the paraffin-base magnetic fluids, respectively. Well below T_m, the value of E_{rot} decreases with increasing temperature. But near T_m, E_{rot} increases to a maximum at the temperature where $\partial K_u/\partial T$ takes maximum. At high temperatures above T_m, E_{rot} decreases gradually with the recovery of fluidity for a carrier fluid almost inversely proportional to T.

The temperature variation of E_{rot} can be explained by the following two processes:

(1) $T < T_m$; the colloidal particles frozen below T_m can be allowed no relative motion. When the particle has a uniaxial magnetic anisotropy K_p, the magnetic moment in the particles, whose volume V is smaller

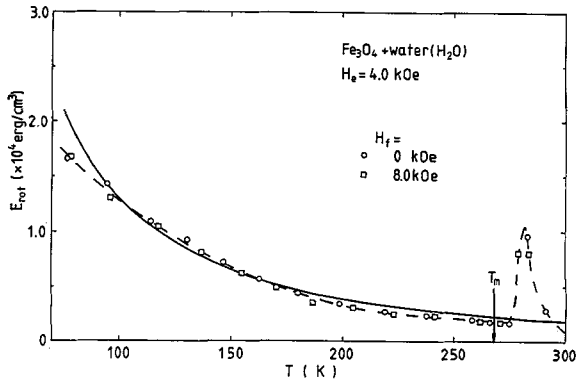

Fig. 2. – Temperature dependence of the rotational hysteresis loss for H_2O–base magnetic fluid frozen in H_f= 0 or 8.0 kOe and measured in H_e=4.0 kOe. The solid line represents the E_{rot} calculated by the method described in the text.

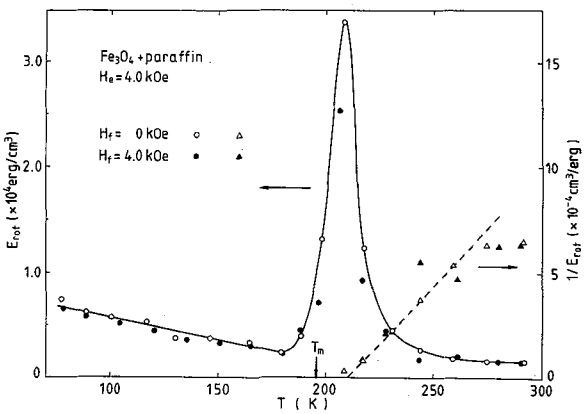

Fig. 3. – Temperature dependence of the rotational hysteresis loss for paraffin-base magnetic fluid frozen in H_f= 0 or 4.0 kOe and measured in H_e=4.0 kOe.

than 25 kT/K_p, exhibits superparamagnetic and can overcome the potential barrier of the anisotropy energy K_pV. In an external magnetic field H_e, the potential barrier E_pV is approximately expressed by $(K_p - M_sH_e + M_s^2H_e^2/4K_p)V$ [3], where M_s is a saturation magnetization. In this case, the magnetic moment in the particles with the volume smaller than 25 kT/E_p overwhelms the potential barrier, while the ferromagnetic moment in the particles with $E_pV > 25\ kT$ is hard to align along the direction of H_e and, therefore, causes a rotational hysteresis. We assume that E_{rot} is given by the following integral;

$$E_{rot} = NE_0 \int_{25\ kT/E_p}^{\infty} Vf(V)\,dV, \qquad (1)$$

where $f(V)$ is the size distribution function of the particle [4], E_0 the rotational hysteresis loss per unit volume of the particle, N the number of the particles per unit volume of the magnetic fluid. These quantities are evaluated by using the relationships between the magnetization curve and the size distribution [5]. As a result, the average volume of the particle, the standard deviation of the distribution and N for water-base magnetic fluid are estimated to be 2.1×10^{-20} cm^3, 1.8 and 5.1×10^{17} particles/cm^3, respectively. By using these parameters, the temperature dependence of E_{rot} is obtained. With least squares fitting of experimental results to equation (1), we estimate the magnitudes of E_0 and K_p as 5.7×10^7 erg/cm^3 and 5.0×10^5 erg/cm^3, respectively. The evaluated value of K_p agrees well with other result obtained from the cooling field dependence of K_u reported in a previous paper [2].

(2) $T > T_m$; in this temperature range, the magnetic fluid recovers fluidity and the particles in the solvent can exhibit relative motions including a pure rotation. The ferromagnetic moment in the particles with larger volume than 25 kT/K_p is pinned to the direction of the magnetic easy axis. Therefore, these particles themselves are rotated so as to reduce the magneto-static energy when H_e is applied. The free rotation of the particles is obstructed in the viscous fluid by a frictional torque given by $8\pi\eta r^3\omega$, where η is the viscosity of a fluid, r the radious of the particle, ω the rotating angular speed. The torque also causes the rotational hysteresis which is proportional to η. When temperature dependence of η is given by the Andrade law, E_{rot} depends on the temperature as follows;

$$E_{rot} \propto \eta_0 \exp(\Delta E/kT). \qquad (2)$$

At high temperatures, E_{rot} is approximated to a function of $A + B/T$, where A and B are constant. The magnitude of ΔE for paraffin-base magnetic fluid is estimated to be 2.9×10^{-13} erg from the temperature dependence of E_{rot}, while ΔE is also evaluated to be 0.9×10^{-13} erg [2] by the temperature variation of the viscosity for paraffin. The magnitude of activation energy estimated by both methods agrees well each other.

Acknowledgments

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