

# Supporting Information

## Oscillatory normal forces of magnetorheological fluids

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## The relation between the field threshold and normal force

As shown in Fig. S1, the zero applied field value is negative, which means the MR fluids attract the plate come together. The value of initial normal force for each shear rate is between -0.2 N and -0.1 N, which is bigger than the systematic errors (0.03N). As soon as the magnetic field reaches the critical value, the normal force changes from negative to positive. The critical value is between 0.02T and 0.03T.

It is found that critical field strength is existed where a liquid experiences a phase transition to a solid phase<sup>1</sup>, which means chains are formed as the applied magnetic field exceeds the critical value. Below the critical value, the surface tension of the fluid and the gravity of particle adsorbed on the plate lead to a normal adsorbing effect. Then, the negative normal force is generated. Solely the critical magnetic field is reached, the normal pushing effects could overcome the attracting effect and then the normal force becomes positive.

## Normal forces under steady shear for different sample, gap distance, inverse magnetic field;

To investigate the influence of the volume fractions of the MR fluids, 10%, 20%, 40% iron based MR fluids were prepared (Fig.S2). Similar to the 30% sample, the oscillatory normal forces with a period of  $2\pi$  were found for these samples. With the increased concentration, the sinusoidal phenomenon of the normal force can be more obvious.

During the above experiments, the gap between the testing plates was kept at 1mm. To fully understand the influence of the testing condition on the normal forces, other gaps (0.3mm, 0.5mm and 0.8mm) have been measured for the 30% MR fluid. It is found that the gap distance have little effect on the result (Fig.S3). The oscillatory normal force with a period of  $2\pi$  could be observed at all these gap distances and the peak-peak oscillatory normal at 0.3mm is a little bigger than that at 0.5 mm and 0.8m.

The direction of the externally applied magnetic field was also switched by reversing the current direction through the coils. Similar to the non-reversible

magnetic field, the positive normal forces of 30% MR fluid was generated after applying the magnetic field. The oscillatory normal forces were also produced by inverting the magnetic field. This result indicated that the oscillatory normal force did not depend on the direction of the field (Fig.S4).

In a word, whatever the gap distance, particle concentration, direction of magnetic field is, the oscillatory normal force of MR fluid under steady shear will always exist.

### The linear and nonlinear region of magnetorheological fluid

With the strain sweep mode, the storage modulus of 30% MR fluid as a function of strain amplitude was measured under different magnetic fields (Fig.S5). The driven angular frequency is kept at 2 rad/s and the strain amplitude is swept logarithmically from 0.001% to 100%. When the strain amplitude is smaller than the critical value ( $\gamma_{\text{crti}}$ ), the storage modulus keeps almost constant and the dynamic behavior MR fluids belong to a linear viscoelastic region. As soon as the strain amplitude exceeds the critical value, the storage modulus decreases sharply. In this case, the changes of the storage modulus enter into a non-linear region. The critical strain values are about 0.1%-0.5%, similar to the previous reports<sup>2,3</sup>.

### Normal forces in the linear viscoelastic region

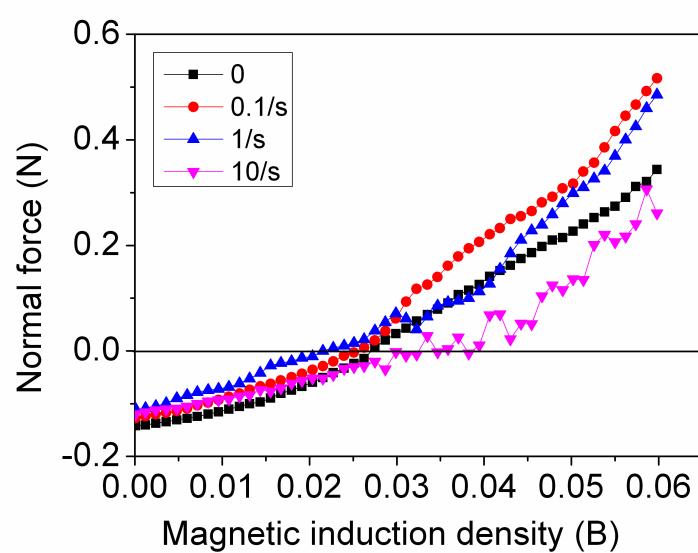
The normal forces under oscillatory shear in the linear viscoelastic region were investigated, as shown in Fig. S6. The applied strain amplitude is 0.01% under different oscillatory frequency (a)  $f=0.1\text{Hz}$  and (b)  $f=1\text{Hz}$  for the 30% MR fluid, which is smaller than the critical value ( $\gamma_{\text{crti}}$ ). The response of MR fluids is in the linear viscoelastic region. Upon application of the magnetic field, the positive normal forces generate and push the plates apart. Under a constant magnetic field, the normal forces keep a constant value and the testing time do not show any influence on them. With the increasing of the magnetic field the normal forces increase monotonously.

Shkel and Klingenbergs have pointed that under simple shear deformation  $\gamma$ , the

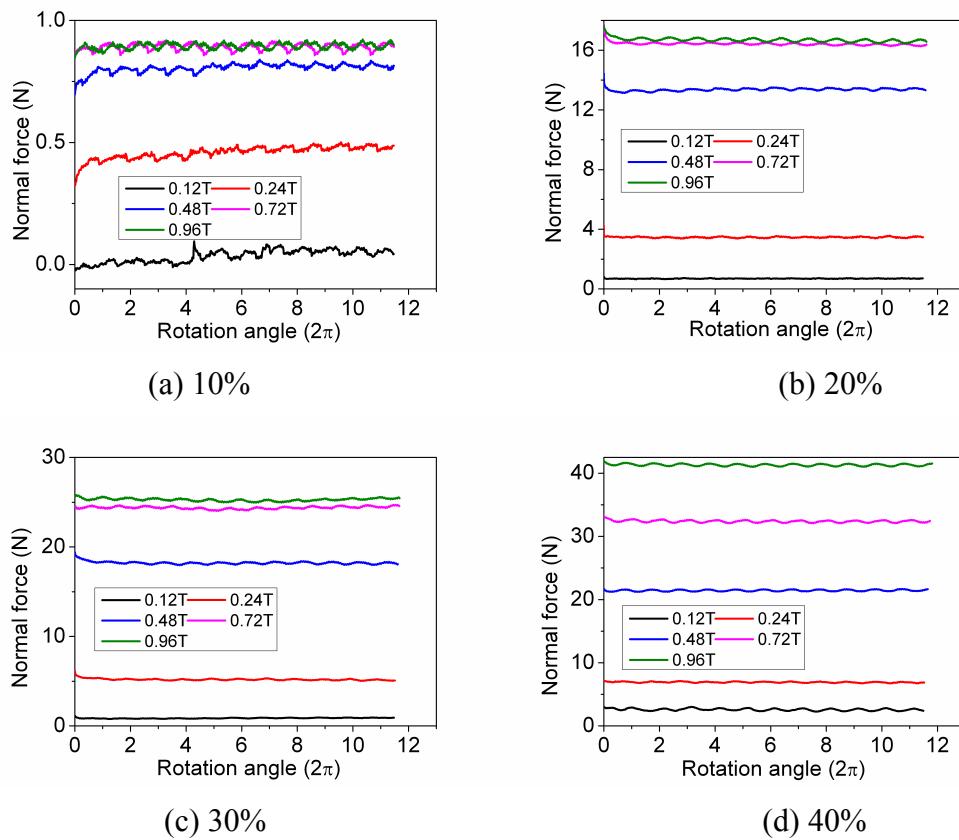
leading order  $\gamma$  for the normal force or normal stress simplifies to  $F_N \propto \sigma_{33} \propto \alpha H^2$ ,  $\alpha$  relates to the material magnetostriction coefficient and magnetic susceptibility<sup>4</sup>. That is, the normal force under small deformation (linear viscoelastic region) is independent on strain. Therefore, the normal forces measured under different testing time are almost steady values.

## Reference

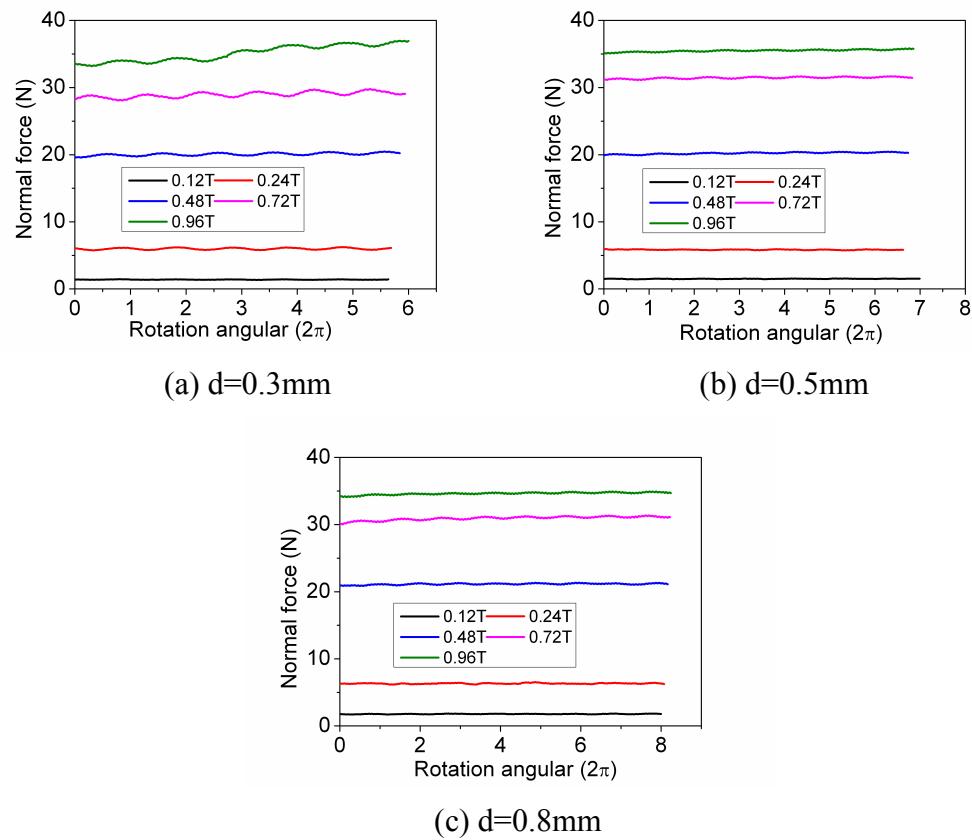
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- 2 W.H. Li, H.J. Du, G. Chen, S.H. Yeo and N.Q. Guo, *Rheol. Acta*, 2003, **42**, 280-286.
- 3 J. Claracq, J. Sarrazin and J.P. Montfort, *Rheol. Acta*, 2004, **43**, 38-49.
- 4 Y.M. Shkel and D.J. Klingenberg, In: *Tao R (Ed) Proceedings of the 7th International Conference on Electro-rheological Fluids, Magnetorheological Suspensions*, 1999,



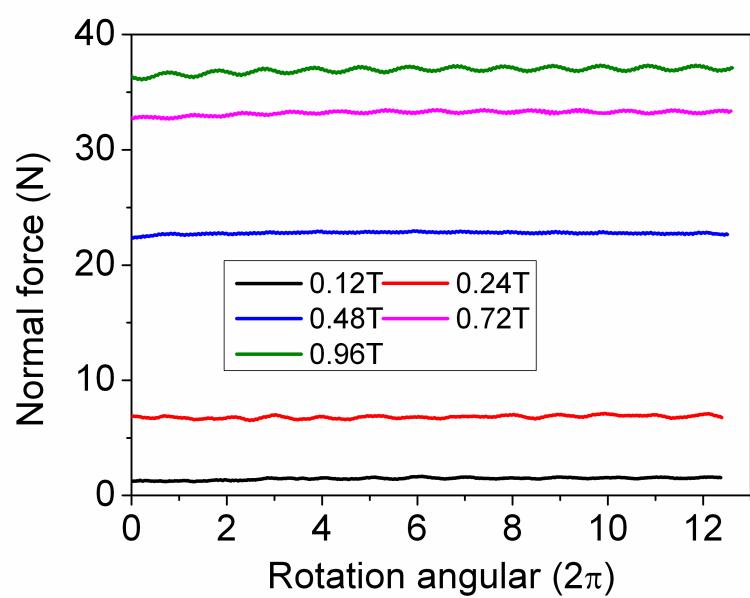
**Fig. S1** Normal forces of 30% MR fluid with magnetic field under different shear rates



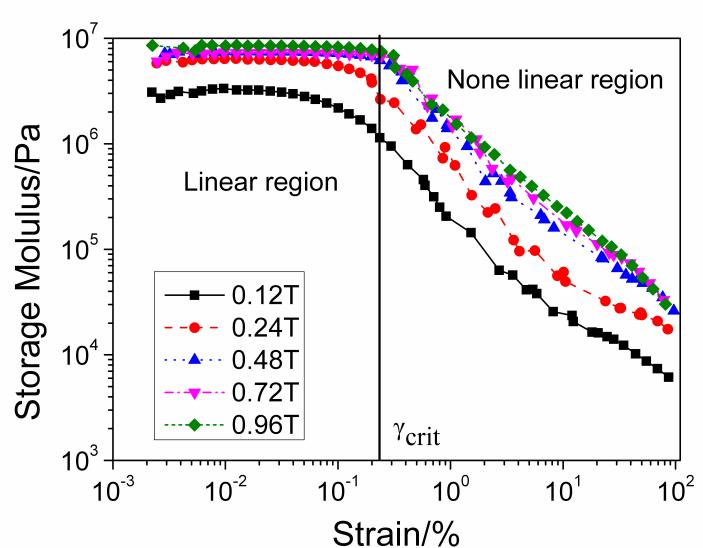
**Fig. S2** Normal force under steady shear for different concentration samples



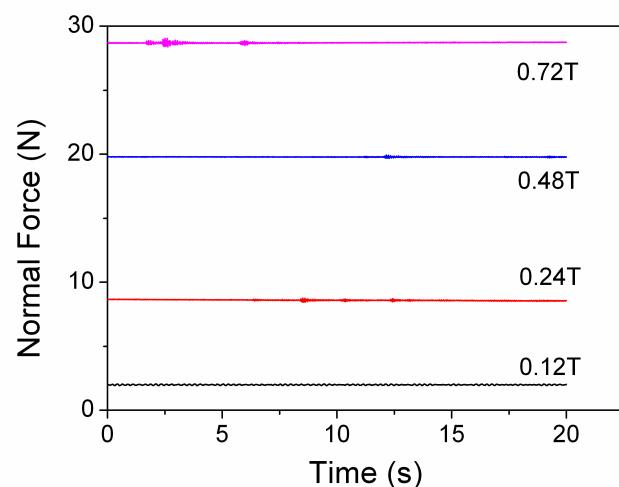
**Fig. S3** Normal force of 30% MR fluid under steady shear for different gaps



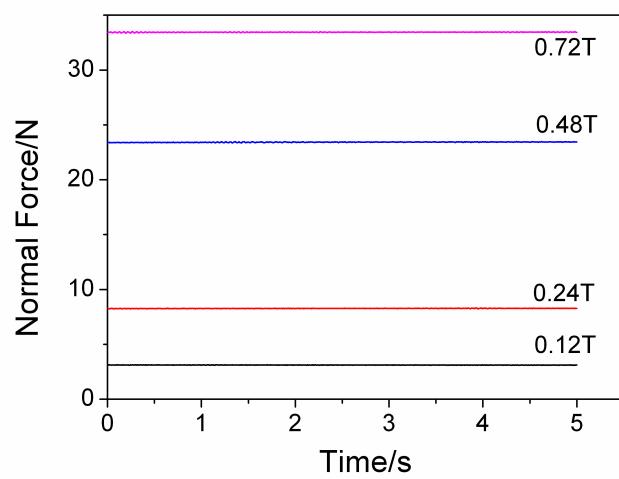
**Fig. S4** Normal force of 30% MR fluid under steady shear for inverse magnetic field



**Fig. S5** Storage modulus as a function of strain amplitude for 30% MR fluid



(a)  $f=0.1\text{Hz}$ ,  $\gamma = 0.01\%$



(b)  $f=1\text{Hz}$ ,  $\gamma = 0.01\%$

**Fig. S6** Normal force of 30% MR fluid under oscillatory shear with time in the linear region