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The magnetic dipole-dipole interaction effect on the magnetic hysteresis at zero temperature in nanoparticles randomly dispersed within a plane

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ABSTRACT

The dipole-dipole interaction effect on the magnetic hysteresis of nanoparticles randomly dispersed within a plane is studied by micromagnetic simulation. The dependence of the coercive field with the concentration of nanoparticles varies from nonlinear and monotonic to non-monotonic dependence with a maximum at a certain concentration. It happens when the ratio of the magnetic anisotropy constant to the maximal dipole energy changes from value much larger than 1 to the value much less than unity.

1. Introduction

Magnetic nanoparticles are of increasing interest because of their current and potential applications in biomedicine, ecology, catalysis and nanoelectronics [1–12]. Material science approach to magnetic nanoparticles research involves the preparation and characterization of the nanoparticles, the design of new synthesis and research techniques and the study of correlations between the structure and properties of the material. Magnetic hysteresis loop is a key characteristic that determines the applied potential of magnetic nanoparticle-based materials (magnetic colloids, nanogranulated materials). The magnetic hysteresis of an individual magnetic nanoparticle has been studied both theoretically and experimentally. It is established that the hysteresis decreases with the increasing temperature and it completely vanishes at a blocking temperature, the value of which depends on the measurement time [1–6,10].

The effects of inter-particle interactions should be taken into account in the particle arrays used in most applications and experiments [6]. The magnetic dipole-dipole interaction is the most important interaction in the magnetic nanoparticles arrays. Its effect on the magnetization curves of nanoparticle arrays at nonzero temperatures has been studied in a number of papers. The emphasis of these studies is on the thermal stability of magnetization, the blocking temperature (T_B) [13–20], the temperature behavior of the hysteresis [20–23], the thermo-magnetic curves measured according to ZFC-FC (zero field cooling - field cooling) protocol [13–20,24]. Magnetic hysteresis at zero temperature determines the hysteresis at the final temperature along with the blocking temperature T_B. The effect of the magnetic dipole-dipole interaction on the hysteresis was studied theoretically and numerically for the ordered ensembles of nanoparticles [25–35]. Since the energy of dipole-dipole interaction decreases with inter-particle distance as $\propto r^{-3}$, the average internal field on the individual particle, and hence the magnetic hysteresis, will depend on the volume fraction of the particles (p). Authors of experimental works often refer to the Néel formula establishing a linear decrease of coercivity with increasing of particle volume fraction $H_c = H_c(0)\cdot(1-p)$ [36–38]. However, numerous experimental data on the coercive field in nanoparticles reveal qualitatively different behaviors of H_c versus p: linear [28,37,39,40], nonlinear [40–46] and even non-monotonic [40,41,43,47]. In most of materials (powders, nanogranular films), the particles are randomly scattered and additionally are randomly oriented. This causes significant fluctuations of the local magnetic field on each nanoparticle and this affects the magnetic hysteresis significantly. In particular, this effect can lead to nonlinear variations of the coercive field as a function of the nanoparticle concentration.

In this paper, hysteresis loops of nanoparticles at zero temperature randomly distributed within a plane with various values of the average surface density are studied numerically using micromagnetic approach. The calculations also take into account the random direction of the easy magnetization axes in the particle array, which is inherent in many materials (for example the particles distributed in nonmagnetic matrices). We demonstrate that the magnetic dipole-dipole interaction leads to a change in the behavior of the coercive field with concentration variations from the nonlinear and monotonic to the

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dependence with the maximum. This change is determined by the ratio of the magnetic anisotropy constant in the individual particle to the magnetic dipole-dipole interaction energy.

2. Numerical simulation

Arrays of single-domain magnetic granules were generated using random homogeneous distribution of particle coordinates within a square region. The size of the area was 4200 × 4200 × 10 nm, the cell size was 10 nm and it was equal to the particle size. Ensembles of nanoparticles with different filling density from $p = 6{\cdot}10^{-4}$ to $p = 0.999$ were generated (Fig. 1). The amount of particles varied from 10^2 to 10^5 (when the element was placed twice in the cell it was considered to be occupied by one particle). Micromagnetic simulations were carried out using the object oriented micromagnetic framework (OOMMF) [48]. The image of the ensemble was used as a mask for the micromagnetic problem, where the magnetization $M_S = 860$ G was attributed to the cells occupied by particles, while empty cells had zero magnetization. We are interested in magnetic properties in an ensemble of single-domain nanoparticles, therefore the cell size is coincided with nanoparticle size. Thus, the magnetization of each particle was set completely homogeneous.

The easy axes of the particles were oriented in two different ways: (a) uniformly within the plane along one side of the square plate and (b) randomly. The applied field was along one of the sides of the square plate. The only inter-particle interaction was the magnetic dipole-dipole one. The physics of this micromagnetic problem is defined by the competition between the energy of the local magnetic anisotropy and the energy of the dipole-dipole interaction between particles in the ensemble. In this case, the magnetic behavior of the material is characterized by the parameter $\varkappa = 4K/M_s^2$ [49]. At $\varkappa \gg 1$, the magnetocrystalline anisotropy prevails and at $\varkappa \ll 1$ the magnetic behavior is determined by the magnetostatic energy. For this study, the specific values of the magnetic anisotropy constant were chosen to investigate the problem in both limits.

3. Results and discussion

To interpret the shape of the hysteresis loop, we distinguish three different competing contributions to the magnetic anisotropy of the

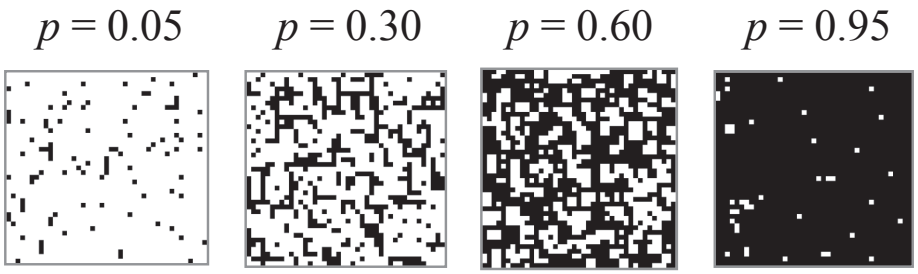

Fig. 1. Fragments of particle arrays with different filling density (p).

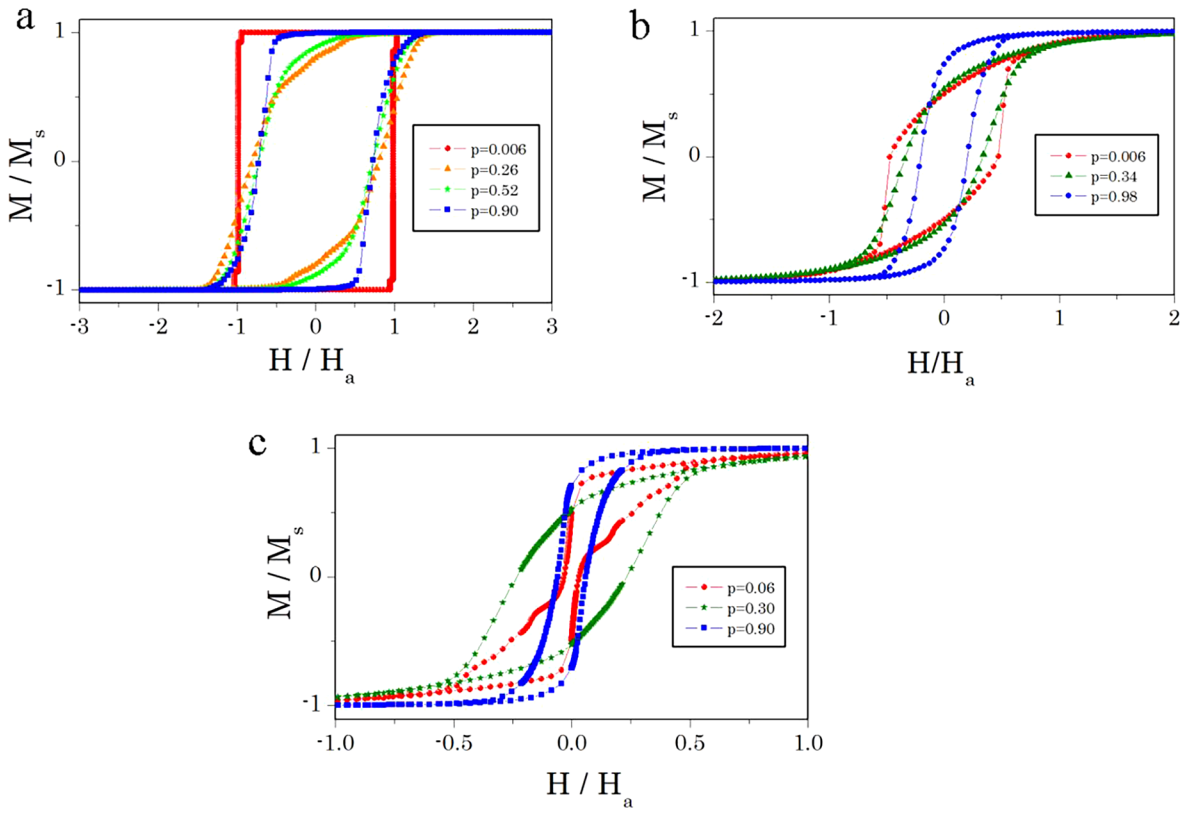

Fig. 2. Hysteresis loops of ensembles with different particle concentrations (p). Samples with uniform (a) and random (b) orientation of the easy axes for the $\varkappa = 5.4$. (c) is for$\varkappa = 0.0054$. Hysteresis loops with a uniform and random orientation of the easy axes overlap here (see text). The applied field is reduced to the field of local magnetic anisotropy of the particle$H_a = 2K/M_s$.

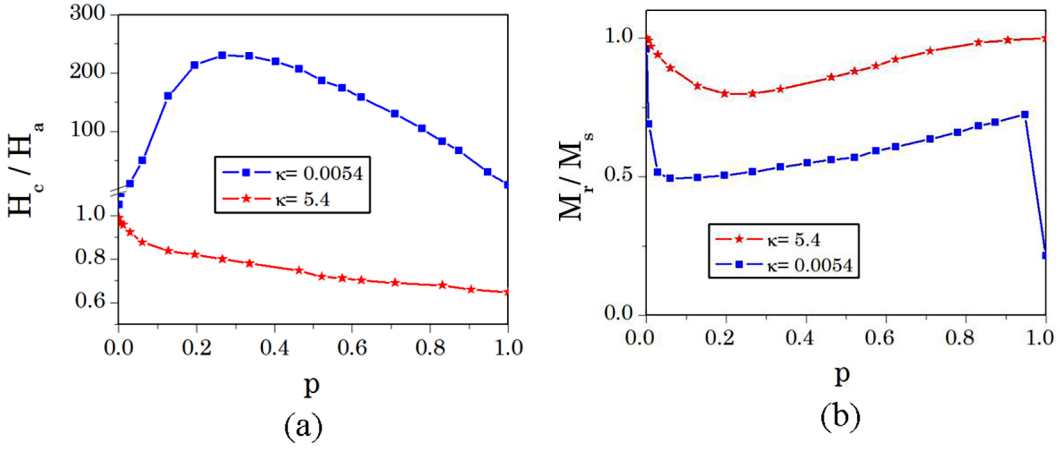

Fig. 3. Coercive field (a) and remanent magnetization (b) of particles with the uniform easy axes. The field is parallel to the sample plane and to the easy axis.

nanoparticle ensemble: the magnetic anisotropy of the individual granule with a random or uniform orientation of the easy axes, the random dipolar field on the granule produced by the entire random array of particles, and the macroscopic magnetic anisotropy, which is the magnetic shape anisotropy of the plate. The case $\kappa \gg 1$, when only the first contribution is significant, was well studied earlier [38,50]. Therefore, for the numerical experiment we have chosen the values of the constants corresponding to the parameter $\kappa = 5.4$ (the magnetic anisotropy constant in each granule is comparable with the specific magnetostatic energy) and $\kappa = 0.0054$ (the magnetostatic energy contribution is dominant).

The contribution to the energy associated with the dipole-dipole interaction is controlled by the particle concentration (p). Indeed, the shape of the magnetization curve changes significantly when the concentration of particles (p) changes (Fig. 2). The case $\kappa = 0.0054$ corresponds to the negligibly small contribution of the magnetic anisotropy of the individual granule therefore the shape of the magnetization curves here is practically independent of the orientation of the easy magnetization axes of the particles. In this regard, in Fig. 2(c) only the curves calculated for the random orientation of the easy axes are shown. For the case $\kappa = 5.4$, as shown in Fig. 2a and b, at low concentrations (p = 0.006), the interaction of the particles is negligible and the shape of the hysteresis loop is determined only by the local magnetic anisotropy constant, i.e. it follows the classical Stoner-Wohlfarth (S-W) theory [38,50].

Indeed in this limit case the loop shapes predicted in this model are observed: rectangular loop with coercivity $H_c = 2K/M_s$ and reduced remanence $M_r/M_s = 1$ for a sample with uniform orientation of the easy axes and rounded hysteresis loop with $H_c = 0.48 \times 2K/M_s$ and $M_r/M_s = 0.5$ for the sample with the random orientation of the easy axes of particles (Fig. 2a and b). In Fig. 2c hysteresis loops of different shapes are observed. For $p = 0.06$, step-like feature is on the loop, while for other particle concentrations of 0.3 and 0.9 there is no such feature. According to Neel, the contribution of the dipole-dipole interaction energy can be estimated as $2\pi \times M_s^2 \times p$. Thus, with increasing the concentration of particles, the case of negligible dipole-dipole interaction is replaced by the strong interaction regime. The case of negligible interaction corresponds to the Stoner-Wohlfarth model, where magnetostatic domains are not formed, and the uniform magnetization is localized within one nanoparticle. This corresponds to $p \sim \kappa$ condition. When ($p = 0.3$ and 0.9), the sample is multidomain, although here the magnetostatic domains are not separated by walls typical for a ferromagnet [51,52]. As the field decreases from saturation to zero, and then to negative fields, the magnetization distribution changes from uniform to multidomain. This process is inhibited by energy barriers created by the energy of the magnetic anisotropy of the particles. For the cases $p = 0.3$ and 0.9, these barriers are insignificant for the formation of a multidomain state. For the case $p = 0.06$, the dipole-dipole energy comparable with the energy barriers leads to the formation of only a small number of domains up to a minimal vortex-like configuration of the magnetization. The loop with the step (for $p = 0.06$ in Fig. 2c) resembles a loop observed in magnetic dots with a vortex magnetization [53]. This step on the loop can be regarded as a giant Barkhausen jump, while for the multidomain systems ($p = 0.3$ and 0.9) such jumps are

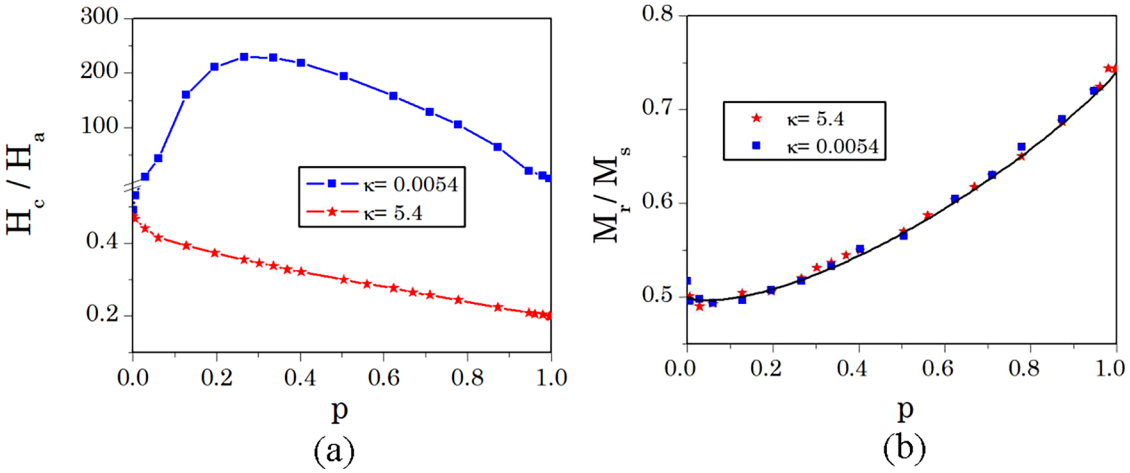

Fig. 4. Coercive field (a) and remanent magnetization (b) of the particles with a random easy axes. The field is parallel to the sample plane.

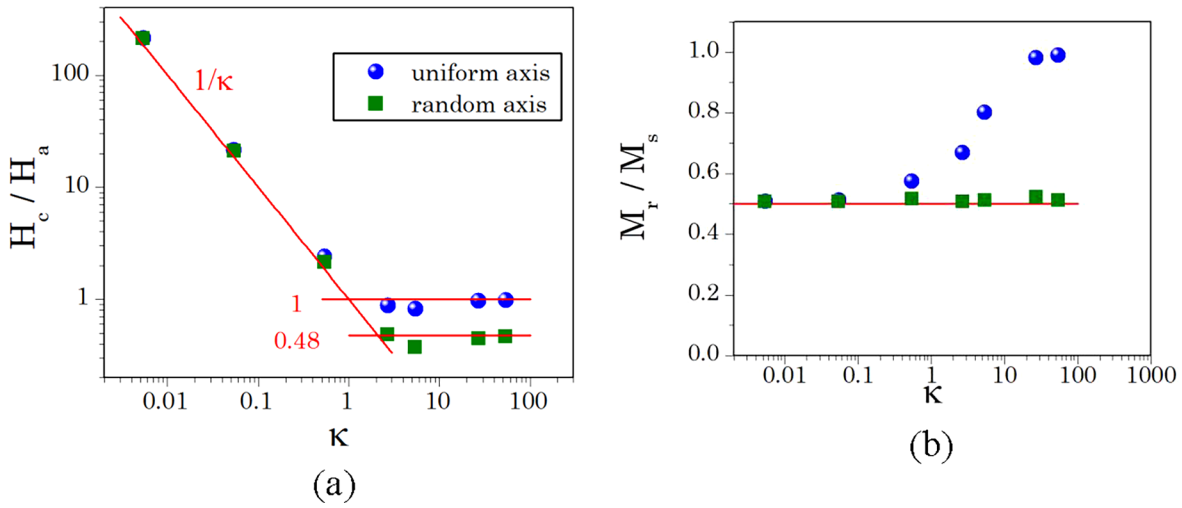

Fig. 5. The coercive field (a) and the remanent magnetization (b) for the particle concentration $p = 0.196$ and different ratios of the local magnetic anisotropy constant to the energy of the dipole-dipole interaction. Blue circles show data for particles with uniform easy axes, while green squares correspond to random easy axes. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

numerous and small in magnitude, leading to a visible lack of singularities on the loop.

As shown in Figs. 3a and 4a, different qualitative behaviors of the $H_c(p)$ dependence are observed depending on the value of κ for both aligned and randomly oriented easy-axes samples. In the case $\kappa = 5.4$, we observe a monotonic decrease of H_c with increasing concentration with a linear dependence in a wide range (Figs. 3a, 4a). However, for $\kappa = 0.0054$ there is a non-monotonic behavior of $H_c(p)$ with a maximum at $p \approx 0.3$ (Figs. 3a, 4a).

In the case of uniform easy axes, the remanent magnetization $M_r(p)/M_s$ has a minimum at $p \approx 0.25 \div 0.3$ for $\kappa = 5.4$, and at $p \approx 0.05 \div 0.1$ for $\kappa = 0.0054$ (Fig. 3b). The minimum value of M_r/M_s in Fig. 3b is close to 0.5 for $\kappa = 0.0054$ and reaches 0.8 for $\kappa = 5.4$. In Fig. 3b, for low $\kappa = 0.0054$ we have the completely random local magnetic anisotropy at all particle concentrations. For ensembles with $p \approx 0.25 \div 0.3$ and $\kappa = 5.4$ (Fig. 3b, asterisk-symbols), the contribution of the random anisotropy energy induced by dipole-dipole interaction is apparently comparable with the contribution from the uniform anisotropy. That leads here to the maximum deviation of data on M_r/M_s from the prediction of the S-W model (Fig. 3b, asterisk-symbols).

For ensembles with $p = 0.3$, the hysteresis loops for different values of κ (Fig. 5) were calculated for both random and uniform easy axes. The results confirm that for $\kappa > 1$ the coercive field and the remanent magnetization follow the predictions of the S-W model. Moreover, for a coercive field the transition to the S-W regime requires only a twofold exceeding of the threshold $\kappa = 1$, whereas for M_r/M_s, such an exceeding must be at least an order of magnitude. For $\kappa < 1$, we have $M_r/M_s \approx 0.5$ and the coercive field doesn't vary with κ, with $H_c \approx 0.5M_s$. Fig. 4a shows that the used values of $\kappa = 0.0054$ and $\kappa = 5.4$ are in good agreement with the modes, when the hysteresis is determined only by the dipole–dipole interaction between the particles or their local magnetic anisotropy respectively.

The position of the minimum on the dependence $M_r(p)/M_s$ and the minimum value as well are sensitive to the value of κ for the case of the uniform easy axes. This can be used in experimental researches to estimate the weight fraction of particles with known κ or for the κ estimation if the particle concentration is known. For the random easy axes, the shape of $M_r(p)/M_s$ curve does not depend on the value of κ. The sharp decrease in the remanent magnetization $M_r(p)/M_s$ for $p \to 1$, observed in Fig. 3b for the case $\kappa = 0.0054$, is due to the fact that the energy of the demagnetizing field with the chosen measurement geometry and the used plate size (4200 × 4200 × 10 nm) is larger than the local magnetic anisotropy energy. This leads to an increase in slope of the hysteresis loop and, as a consequence, to a sharp decrease in M_r/M_s.

To study the macroscopic magnetic anisotropy the hysteresis loops of flat ensembles of particles were calculated for two directions of the external field: perpendicular and parallel to the plane (the inset to Fig. 6). The effective magnetic anisotropy constant was calculated as:

$$K_{eff} = \int_0^\infty (M_\perp(H) - M_\parallel(H))dH$$

The upper part of the major loop $M(H)$ corresponding to demagnetization from an infinite field to zero field was used for calculation, and the summing interrupted when the difference between $M_\perp(H)$ and $M_\parallel(H)$ was less than 0.1%. The main source of macroscopic magnetic anisotropy in this case is the shape anisotropy of the plate containing the nanoparticles. According to mean-field theory, the constant of this anisotropy is $K_{sh} = 2\pi \times M_s^2 \times p$ (see for example [54–58]). The blue solid line in Fig. 6 corresponds to this equation. The calculated data are in a good agreement with the predictions of the mean-field theory.

The fact that the composite plate behaves like a uniform magnetic film means that the results obtained in our work do not depend on the sizes of the plate and on the absolute number of particles, i.e. these results can be considered as universal for all granular films.

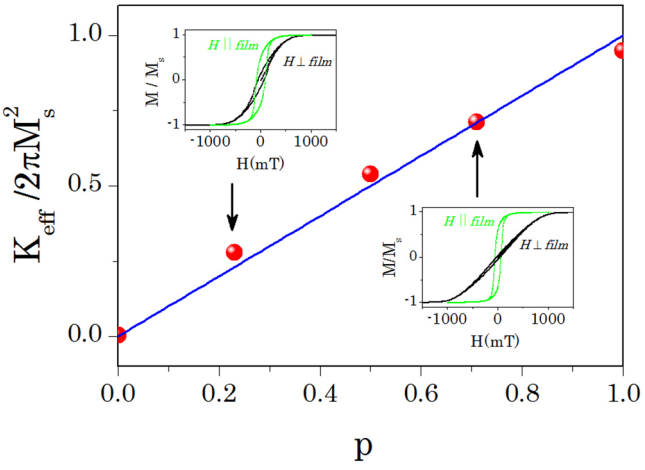

Fig. 6. The macroscopic magnetic anisotropy constant of ensembles with different particle concentrations (p) and with a random distribution of easy axes.

4. Conclusion

Hysteresis loops of nanoparticles randomly distributed in the plane for various surface densities of particles are numerically investigated. Uniform or random easy axes of the particles were set. It is shown that the dipole-dipole interaction leads to a change in the dependence of the coercive field on the particles concentration from the nonlinear monotonic to the dependence with the maximum. This behavior differs from linear Néel's dependence of the coercive field on the particles concentration. The observed change in coercive field behavior is found to be determined by the ratio of the magnetic anisotropy constant of the individual particle and the specific dipole energy.

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