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Spin wave dynamics in two- and three-dimensional superlattices of nanosized ferromagnetic spheres

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The dispersion of spin wave modes which due to the dipolar interactions propagate along different directions of ordered superlattices of nanospheres is investigated. For this purpose a procedure similar to the well-known method of linear combination of atomic orbitals is applied. Different geometries of two-dimensional (triangular and square) and three-dimensional (simple cubic and hexagonal-close-packed) arrangements are considered and the influence of dimensionality on the spin wave dynamics is analyzed. A phase transition which is caused by the competition between dipolar and uniaxial anisotropy interactions is predicted by the investigation of the dispersion of the uniform Kittel mode for the superlattice of the hexagonal order. In conclusion, it is shown how the weak dipolar interaction enhances or decreases the relaxation time in the samples with a controlled direction of the easy axis. © *2008 American Institute of Physics*. [DOI: 10.1063/1.2832756]

The preparation and investigation of new nanostructured materials, especially of those which consist of superparamagnetic spheres, are of great importance due to their anticipation technological application in magnetic storage devices.[1] One of the most exigent problems is the enhancement of the storage stability, which depends on the energy barrier between the two possible ground states of uniaxial nanoparticles. This barrier primarily depends on the value of the nanoparticle anisotropy constant and there are already considerable achievements concerning the enhancement of this factor.[2] The second parameter that changes the energy barrier (and, consequently, the relaxation time and the blocking temperature) is the interparticle interaction of dipolar origin. Much attention has been paid to this question, both theoretically and experimentally.[3,4] It is known that the effect of the dipolar interaction strongly depends on the space geometrical order in nanoparticle superlattices and that it can either enhance or decrease the blocking temperature.[5]

In this article we present a theoretical investigation of spin waves (SW) which propagate through a superlattice that consists of interacting nanoscale spheres. By the application of new technologies it is possible to produce self-assembled two-dimensional (2D) and three-dimensional (3D) superlattices with various and well defined long-range translational order and with only small deviations from the average size of nanoparticles.[1,2,4] We find a considerably different effect of the dipolar interaction in 2D and 3D superlattices and calculate the specific parameters of the 3D hexagonal-close-packed (hcp) system. Due to the competition between crystalline anisotropy within every particle and the dipolar interaction between the particles, the SW dispersion suggests a phase transition from the initial ferromagnetic (FM) order into probably[6] a helicoidal order. Our interest in this topic has been stimulated by new inelastic neutron experiments on the uniform mode in small ferromagnetic particles.[7] Experimentally, the effects described here—in particular the softening of the uniform mode and the critical behavior near the phase transition point—can be probed by neutron scattering.

There are two scales at which superlattices of nanocrystals are studied: at an atomic level (i.e., the material itself) and a scale which is determined by the characteristic nanoscale length of the superlattice. A certain temptation to apply the well-developed methods of solid state physics to new artificially nanostructured materials arises naturally. In this article we apply the well-known method of linear combination of atomic orbitals (LCAO) on the analysis of propagating spin waves in 2D and 3D superlattices that consist of spherical ferromagnetic nanoparticles.[8] If the solutions of the wave functions and energies for fluctuating spins in the individual spheres are known (correspondingly, the original method LCAO works with wave functions and energies of the electron states of the atom), the solutions for the SW considered as correlated fluctuations of spins between the nanoparticles must be a correction of these individual states. Such an approach is justified if the gaps between the energies of each individual state are larger than all the characteristic values of the dipolar energies. By taking the translation invariance of the superlattice and the corresponding Bloch theorem into account, the wave function of the entire observed system can be written as a linear combination of wave functions of spin fluctuations that are centered in every sphere. By comparing the average energy scales for the intrinsic particle exchange interaction (which determines the distances between individual particle energy levels) with the characteristic dipolar energies, it can be concluded that our considerations are best applied to FM spheres with radii (R), approximately, $2\ \text{nm} < R < 10\ \text{nm}$. For much larger particles with a size of more than a few tens of nanometers the approach presented in Ref. 9 is valid. For the smaller particles

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with size of approximately 1–2 nm a consistent quantum description was already presented in Ref. 10. Based on existing experimental specimen preparation methods,[2] we choose the effective distance between particles to be 2 nm. Therefore, although the principal results only depend on the ratio R/L (with L being the distance between the centers of the spheres), we based our concrete calculations on the explicit case of nanospheres with $R=5$ nm and $L=12$ nm.

We now consider an assembly of spherical ferromagnetic nanoparticles with easy axis anisotropy and all the easy axes of the individual nanoparticles aligned parallel to the z axis. Preparation of such oriented nanoparticles is of great technological importance[11] which is reflected by reports on new technological methods that allow to grow highly oriented nanoparticle structures.[12] We assume that an external magnetic field (which can be removed after this action) aligned the magnetic moments of all particles in one direction and arranged them in one of two possible minima of easy-axis energy states before the external magnetic field is switched off again. Below the blocking temperature magnetic moments precess around the easy axis and can be described by $\mathbf{M}=\mathbf{M}_0+\mathbf{m}$, where $\mathbf{M}_0$ is the saturation magnetization of the ferromagnetic material and $\mathbf{m}$ is a vector that describes small fluctuations. In linear SW approximation the vector $\mathbf{m}$ is perpendicular to $\mathbf{M}_0$. In accordance with the LCAO method we write the two components of $\mathbf{m}$ as

$$m_i = \frac{a_i}{\sqrt{N}} \sum_j \psi(\mathbf{r}-\mathbf{r}_j) e^{i\mathbf{k}\mathbf{r}_j}, \tag{1}$$

where $\mathbf{k}$ is a wave vector of the SW propagating through the nanocrystal superlattice due to the long-range dipole interaction between the nanoparticles, N is the total number of the nanoparticles, $\mathbf{r}_j$ is a coordinate vector giving the center of each nanoparticle sphere, $\psi(\mathbf{r}-\mathbf{r}_j)$ is a function describing the SW of chosen symmetry centered at the individual sphere with index j, and the sum is taken over all the nanoparticles in the superlattice. This approach also allows one to take the specific shape of nanoparticles into account. The corresponding collective modes with energy $E(\mathbf{k})$ feel the influence of the boundary conditions due to the surface of the nanoparticles. The dependence of $E(\mathbf{k})$ on the superlattice Bloch wave vector $\mathbf{k}$ can be considered as a correction of the energy levels of the individual spheres.

Intrinsic exchange forces and the boundary conditions on the surface of the sphere determine the profile of standing SW for an individual nanoparticle. We remind that the non pinning condition is $\partial\mathbf{m}/\partial n=0$ on the surface of a nanoparticle (n is the normal to the surface), in contrast to strong pinning, when $\mathbf{m}=0$ on the surface. In the case of a small surface anisotropy for the nanoparticles of spherical (ellipsoidal) shape—i.e., when the particle size is small compared to the exchange length—the nonpinning case occurs.[13] Consequently, the lowest mode is the uniform Kittel mode.[14] Recent experimental observations of the uniform mode in maghemite nanoparticles prove a validity of such an aproach.[7] In small particles the energy level of the Kittel mode lies much lower than other (exchange) SW levels. For a zero external field and for typical values of the anisotropy simple estimation gives $E_{\text{Kittel}} \propto 50-100$ μeV. In the case of weak pinning, i.e., for small particles, corrections to the Kittel mode should be considered. However, in the case of strong pinning i.e., for particles with a size of approximately 50–100 nm, the angular symmetric mode of the shape $j_0(K\rho)$ takes its place (compare with the case of cylindrical dots[15]). Here we only consider the case of uniform Kittel mode.

For the uniform mode the wave function in Eq. (1) is

$$\psi(\mathbf{r}-\mathbf{r}_j) = X(|r|) = \begin{cases} 1, & |r| \le R \\ 0, & |r| > R \end{cases}.$$

Whenever methods of solid state physics are applied to artificial structures, it is important to make use of the similarities between systems with discrete energy levels even though these levels are of different physical origin and nature. It allows applying the same approach for both, quantum mechanics and phenomenological equations. In our case the role of the equation of motion is taken by the Landau–Lifschitz equation (LL) for the dynamic components of the magnetization, instead of the Schroedinger equation which is found in the usual LCAO. By substituting Eq. (1) into LL, one obtains the linear system for the unknown coefficients a_i. The determinant of this system has to be equal to zero. The SW energy $E(\mathbf{k})$, is then expressed in a usual form as

$$E(\mathbf{k}) = g\mu B(\{H_{\text{eff}} - [h_z - h_x(\mathbf{k})]\}\{H_{\text{eff}} - [h_z - h_y(\mathbf{k})]\})^{1/2}, \tag{2}$$

where $H_{\text{eff}}=H_{\text{ext}}+H_{\text{anisotropy}}$ (remember that we assume that all the anisotropy axes are parallel to the external field and therefore also parallel to $\mathbf{M}_0$). The terms h_z, $h_x(\mathbf{k})$, and $h_y(\mathbf{k})$ are effective dipolar matrix elements given by

$$h_z = \frac{M_0}{V} \sum_{i\neq j} \int_{V_i} \int_{V_j} d\mathbf{r} d\mathbf{r}' \frac{\partial}{\partial z} \frac{\partial}{\partial z'} \frac{1}{|\mathbf{r}-\mathbf{r}'|},$$

$$h_l(\mathbf{k}) = \frac{M_0}{V} \sum_{i\neq j} e^{i\mathbf{k}(\mathbf{r}_i-\mathbf{r}_j)} \int_{V_i} \int_{V_j} d\mathbf{r} d\mathbf{r}' \frac{\partial}{\partial l} \frac{\partial}{\partial l'} \frac{1}{|\mathbf{r}-\mathbf{r}'|}.$$

Here we denote $l=x$ or $l=y$, the summation is over the vectors of the centers of the spheres $\mathbf{r}_{i,j}$ and integration runs over the volumes V_i, V_j of each of the spheres, V is an average volume of the nanoparticles.

As it is known, the value of the effective dipolar fields strongly depends on the geometry of the superlattice. This is also closely linked with theoretical investigations of ground states and dynamical behavior of dipole lattices which derive from the classical works of Luttinger and Tisza (see Refs. 16–18, and references therein) and is also connected with the resent analysis of collective excitations in nanodots and nanospheres.[9,19] Here we calculate the SW dispersion in the most important arrangements which are 2D triangular and 3D hexagonal structures. We assume that the 2D structure is a triangular monolayer (consisting of spheres) with the rhombic angle of $\pi/3$. The 3D hexagonal structure consists then of such monolayers. The distance between them corresponds to the ideal hcp structure with a ratio of $\sqrt{3}/2$ between interplane and in-plane lattice parameters.[20] We also assume the z and the x axis are in the plane of the monolayer,

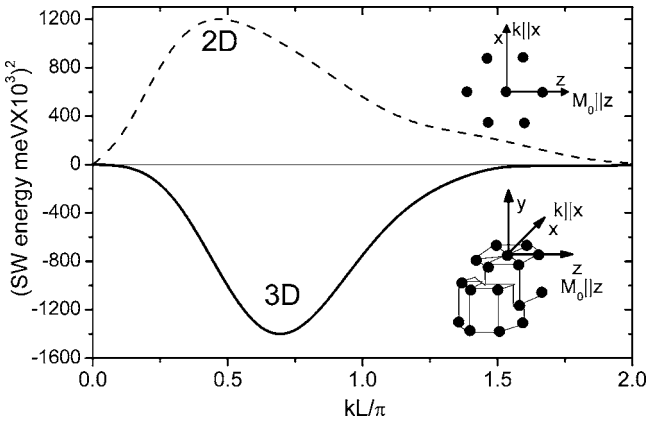

FIG. 1. The square of the SW dispersion of the uniform mode ($H_{\text{eff}}=0$) as calculated for the dipolar interaction between Co spheres for 2D triangular and 3D hexagonal superlattices with $\mathbf{M}_0 \| z$, $k \| x$, $|\mathbf{M}_0|=0.14$ T (bulk Co), and $g=2$. The rhombic angle in the xz plane in both 2D and 3D structures is assumed equal to $\pi/3$. Correspondingly, the 3D hexagonal structure is an ideal hexagonal (hcp) structure with a ratio of $\sqrt{3}/2$ between interplane and in-plane lattice parameters The insets give the corresponding structures and vectors, respectively. The ratio between the radius of nanospheres R and the distance between their centers L is $R/L=5/12$ (as it was used for all the calculations throughout the article).

and that $\mathbf{M}_0 \| z$. In order to compare our approach with previous investigations, we also present results for square and simple cubic (SC) structures.

For superlattices consisting of point dipoles it has been shown that dipole forces stabilize the ferromagnetic 2D structure in 2D triangular lattices.[18] As it follows from properties of the dipolar forces the magnetic moment lies in the plane of the dipoles. For our case with the rhombic angle of equal to $\pi/3$ this plane is isotropic. From this point of view the properties of the 2D superstructure both of point dipoles or spheres are similar to those of easy plane thin films[21] and even a small disorder in the anisotropy can destroy the long-range order. It is also similar to the case, when—in accordance with Mermin–Wagner theorem—thermal fluctuations can destroy the long-range order in 2D system. However, if we consider the ordered assembly (the entire easy axes are aligned and the system is ordered ferromagnetically), the SW energy should be real valued (meaning that, its square is positive) for any direction of the wave vector $\mathbf{k}$. Figure 1 shows the square of the SW energy as it was obtained for zero external and zero anisotropy fields for both 2D and 3D lattices of identical nanospheres. For these calculations it is convenient to locate the origin of the coordinate system at the center of one selected sphere. Because of the long-range order of dipolar interaction one has to take a quite large cluster of the superlattice[9] into account. The strongest dispersion of collective SW arises if the wave vector is parallel to the x axis (compare with Fig. 4 in Ref. 17). For a 2D triangular lattice the square of the energy is positive, but that of a 3D close packed lattice is negative and, hence, the energy of the SW is imaginary. This suggests instability of the FM order in the system. It is important to note that the instability arises even if the amount of layers of nanospheres is small. Three layers are already sufficient to obtain an imaginary energy for a SW that propagates along the x direction.

The reason for this can be understood in the following way. It follows from the Eq. (2), that if H_{ext} is zero, the sign of $E(\mathbf{k})^2$ directly depends on the signs of the terms $[h_z - h_x(\mathbf{k})]$ and $[h_z - h_y(\mathbf{k})]$. In the triangular lattice (2D case) both are negative. Furthermore, $[h_z - h_x(0)]$ becomes zero which suggests the absence of the anisotropy of dipolar origin in the plane, and $[h_z - h_y(0)]$ is negative which indicates the easy plane anisotropy. If we move to the 3D lattice, the term $[h_z - h_x(\mathbf{k})]$ does not change its sign because the geometry of the plane remains the same. However $[h_z - h_y(\mathbf{k})]$ does change its sign and becomes positive. From this follows that if the SW energy is real for some direction of dispersion in a 2D structure, it becomes imaginary in the corresponding 3D superlattice and vise versa.

It is interesting to illustrate this observation by some results on SW dispersion in square and cubic lattices. It is already known that the FM state with a magnetization along the (1,0) direction is unstable in square lattices and the FM state with a magnetization direction along the diagonal (1,1) is metastable (Ref. 17, and references therein). The ground state for the corresponding 3D SC lattice is antiferromagnetic.[16] Concequently, the FM order must be unstable. For the unstable FM order in a 2D square lattice $[\mathbf{M}_0 \| (1,0)]$ we obtain that the energy of SW, which propagates parallel to $\mathbf{M}_0$, is imaginary. However, for SW propagating perpendicularly to $\mathbf{M}_0$, it is real valued. This is just the opposite in the 3D SC structure. Their corresponding curves are presented in Figs. 2(a) and 2(b). The real-valued SW energy for metastable 2D squares becomes imaginary in the 3D SC lattice, if the magnetization preserves the same direction along the in-plane diagonal (not shown).

We can now address the question how these properties of dipolar interaction influence the observable properties of ordered superlattices of ferromagnetic nanospheres in presence of an easy-axis anisotropy. For this purpose we consider the magnetic moment of the nanoparticles aligned in a FM order as described earlier. Figure 3(a) shows that the SW energy becomes zero for a 3D hexagonal superlattice with $R/L=0.417$ (the distance between nanoparticles is $L-2R=2$ nm, $R=5$ nm, $L=12$ nm) for a certain value of H_{eff}, and the corresponding wave vector of $\mathbf{k}=(k_{0x},0,0)$ in the considered geometry. The value $k_{0x} \approx 0.7\pi/L$ can be estimated numerically. Due to the infiniteness of the dipolar sums, k_{0x} can be incommensurable with the principal vectors of the Brillouin zone. For comparison, the SW energy is presented in Fig. 3(b) for the wave propagating along the y direction (perpendicular to the original monolayer).

The results presented in Figs. 3(a) and 3(b) suggest a phase transition in the vicinity of $H_{\text{eff}} \propto 0.25$ T for the superlattice with the chosen parameters of the magnetization ($|\mathbf{M}_0|=0.14$ T) and the L/R ratio. If $H_{\text{eff}} > 0.25$ T, the FM order stabilizes due to the influence of the anisotropy and external fields. If the L/R ratio is smaller H_{eff} is larger. The questions of the order of the magnetic structure which forms after the transition and the question of the kind of the corresponding phase transition are both complicated to answer. In any case, the new magnetic order forms due to the competition between anisotropy and external field on the one side and the dipolar field on the other side. Both questions are

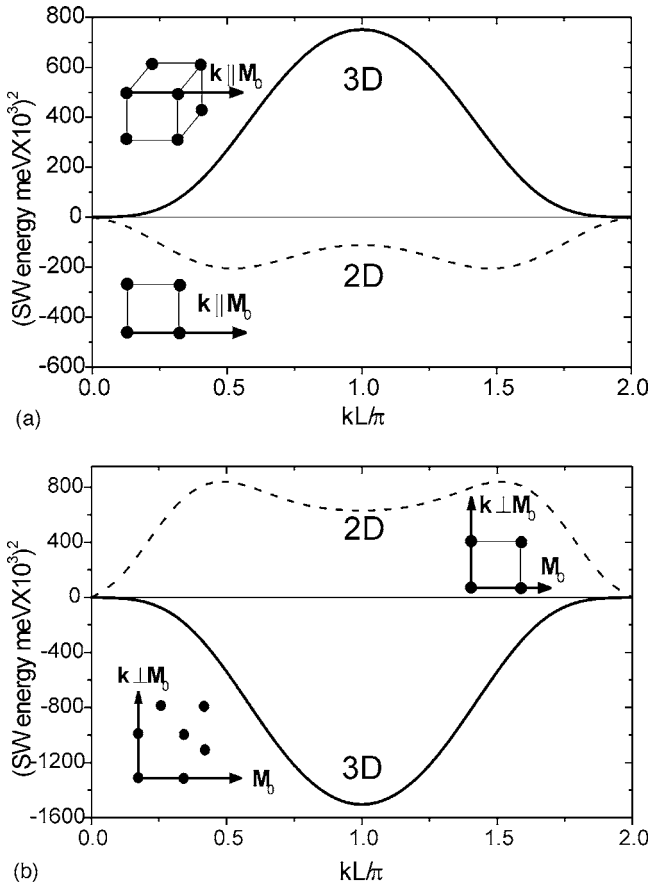

FIG. 2. (a) and (b) The square of SW dispersion of the uniform mode ($H_{\text{eff}}=0$) for square (2D) and cubic (3D) lattices with $\mathbf{M}_0\|(1,0,0)$ for (a) $k\|\mathbf{M}_0$ and for (b) $k\perp\mathbf{M}_0$ (but within the same plain).

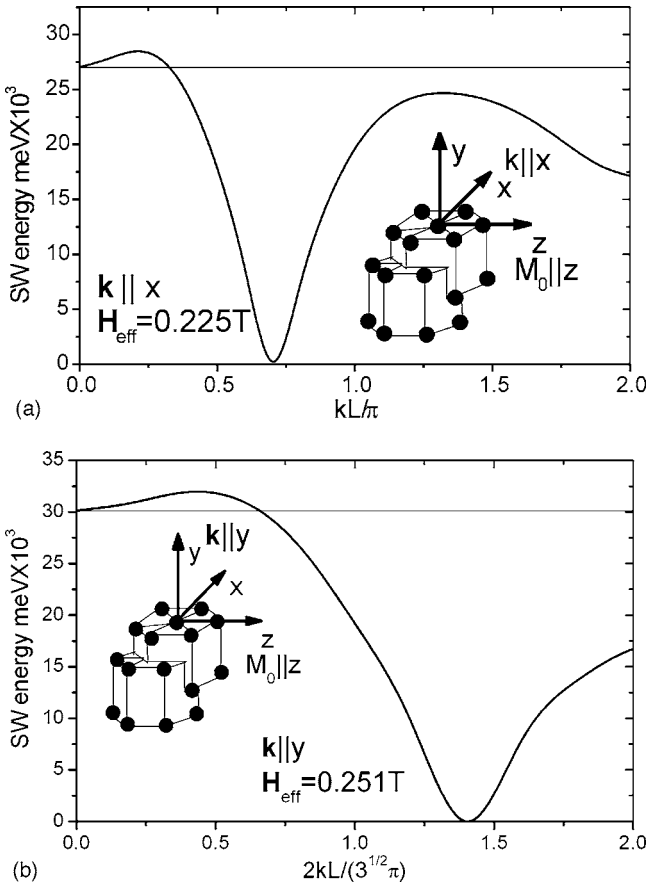

FIG. 3. (a) and (b) The uniform mode energy dispersion in a hcp superlattice of FM spheres for $\mathbf{M}_0\|z$, with (a) $k\|x$ and (b) $k\|y$ [Fig. 3(b)]. The horizontal lines indicate the energy of the uniform mode as obtained for the same case of $H_{\text{eff}}=0$, but without dipolar intersphere interaction.

also closely linked to the problem of how to determine the ground state in hexagonal 3D dipolar lattices. To our knowledge, extensive investigations of the dipolar order were reported for cubic lattices (SC, body-centered-cubic, and face-centered-cubic[16,22]), but for hexagonal 3D lattice the question is still unanswered at the present time. The SW theory presented here suggests that this magnetic order should be rather helicoidal than FM or AFM. However, this question demands an additional investigation and is out of the scope of this article.

From numerical results, it follows that the influence of dipolar interaction is rather small as it is known that dipolar FM are "soft." Hence, it obviously is negligible for samples with a large crystallographic anisotropy constant (as it was shown[2] in experiments with $CoPt_3$). However, it would be quite considerable in the materials with decreased anisotropy as it was observed in experiments with ε-Co (the first article in Ref. 4 and the second article in Ref. 11).

We emphasize that the FM ordered models considered here are attractive for investigations of the relaxation time in such systems. The influence on the superlattice ordering is simple and definite due to the known direction of the crystallographic anisotropy field (one can find the influence of arbitrary direction external field in Ref. 23). Evidently, if both the anisotropy and the dipolar field enhance the ordering effect of each other, the energy barrier increases. On the other hand, a competition between these fields leads to a reduction of the energy barrier. For the geometry considered here, the energy of the system with anisotropy and dipolar interaction can be written as a sum of two terms, $E=KV\sin^2\theta+C\sin^2\theta\sin^2\varphi$, where $K>0$ is the easy-axis anisotropy energy constant, θ is the angle between the magnetization and the easy axis, and $C\propto-[h_z-h_y(\mathbf{0})]$ corresponds to the dipolar interaction. Assuming that $C/kT\ll 1$, one obtains the expression $\tau=\tau_0\exp((KV+C/2)/kT)$ for the relaxation time.[7] As shown earlier, for the 2D triangular lattice C becomes positive. Hence, the relaxation time increases for the 2D case. In comparison, in the 3D case of hexagonal lattices C is negative, and thus the relaxation time decreases for the 3D case.[24]

In conclusion, in this article we considered 2D (square and triangular) and corresponding 3D (SC and hcp) superlattices of dipolar interacting nanospheres with ferromagnetically ordered moments. The well established method of LCAO used in solid state physics and modified here for ascertained spin wave modes in selected structures of nanospheres can be easily generalized for any kind of mode in a variety of regular superlattices of nanoparticles. We showed that by SW analysis an evidence for a reorientation phase transition is given. For the first time, we considered in detail how these properties are connected with a change in the dimensionality (from 2D to the corresponding 3D ordered systems). Our computational results are in a good agreement

with a theory of ordered lattices of point dipoles (see Refs. 15–17, and references therein) which has been developed over many decades. The applied approach allows one to estimate the relaxation time in ordered systems which are in accordance with experimental results and are of great technological importance.

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[24]Note, that the same effect (decrease of relaxation time) could be achieved also in the case of 2D triangular ordering, where the spheres are magnetized perpendicularly to the plane.