

Dipolar interactions in arrays of iron nanowires studied by Mössbauer spectroscopy

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We have performed numerical calculations and experimental studies on the dipole-dipole interaction in the hexagonal nanowire arrays. Using the dipole approximation with a length correction, the dipolar fields in the cylindrical wire arrays were calculated. By means of Mössbauer spectroscopy, the dipolar fields in the Fe nanowire arrays fabricated by anodic porous alumina template-grown method were derived from the relationship between effective hyperfine field and applied field. Both the theoretical predictions and experimental results show there is a rather strong dipolar field with the saturation field perpendicular to the wire axis, while the dipolar interaction with magnetic moment aligned along the wire axis is too weak to be taken into account. The easy magnetization direction is determined by a competition of the dipolar interaction and the shape anisotropy. The critical ratio between the diameter of nanowire and the distance between nanowires, on which the easy axis of nanowire arrays would be changed from parallel to perpendicular to the wire axes, was calculated.

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I. INTRODUCTION

It is well known the dipole-dipole interaction is a common long-ranged interaction in magnetic materials. Although this interaction is much smaller than the exchange interaction, in modern periodically ordered magnetic nanostructures it plays an essential role, which strongly affects their magnetic properties.¹⁻⁶ The nanostructured magnetic materials, such as magnetic nanowires, nanodots, and nanotube arrays, are interesting from the potential application in ultrahigh density magnetic recording media⁷ and the fundamental research.⁸⁻¹⁰ Interdot or interwire exchange coupling is absent in this patterned magnetic nanostructures and therefore magnetic behaviors of the arrays are governed by the shape anisotropy and the dipole-dipole interaction. For magnetic nanowires, because of the high aspect ratio, the easy magnetization direction of an individual nanowire is along the wire axis, which is usually different from the easy direction of the same bulk material due to magnetocrystalline anisotropy.¹¹ It can be understood that the shape anisotropy constant is about one order of magnitude larger than that of the magnetocrystalline anisotropy constant in magnetic nanowires.¹² Once the magnetic nanowires are embedded in a periodically ordered array, the dipole-dipole interaction between nanowires should be considered to determine the easy magnetization direction of the whole array. The dipolar interaction is sensitive with the interwire distance, and thus the easy magnetization direction of nanowire arrays can be tuned parallel or perpendicular to the wire axes by changing the packed density of the arrays.^{5,13} Most of the investigations have paid much attention to the dipolar interaction in the nanostructured materials. Based on a so-called dipole approximation, the dipolar field in nanodot arrays can be calculated quantitatively by using standard textbook techniques.¹⁴ Unfortunately, it is unsuitable to use the same method to estimate the dipolar interaction in nanowire arrays, though both nanodot and nanowire arrays are periodically ordered nanostructures just with difference in the length of nanomagnets. In nanodot

arrays, the distance between the dots is far larger than the size of the dots. Therefore, the nanodot structure can be simply treated as a two-dimensional dipole array. Whereas in the arrays of nanowires, when the distance between the wires is comparable to the length of the wires, the dipole approximation are not work at all and the effect of the nanowire length should be taken into account. To our knowledge no proper model has been reported to calculate the dipolar field in nanowire systems.

Also in experiment, very few works have been successful in probing the dipolar interaction between dots or wires arranged in well-defined arrays.^{5,15-17} It is responsible for the weak coupling in the nanodot arrays with rather large distance between dots normally fabricated based on lithography techniques. Grimsditch *et al.* have estimated the dipolar field in nanodot arrays is about just several Oe.¹⁴ It is well known the nanowire arrays fabricated by the self-assembled template methods have rather small interwire distances ranged from several to hundreds nanometers.^{5,11,13} Due to the distance sensitivity of the dipole-dipole interaction, it can be expected the dipolar field in nanowire arrays is much larger than that in nanodot arrays, and which ensures this field can be measured not so difficultly. In fact, it is hard to use the traditional macro magnetometer to measure the dipolar field in nanostructured materials. In our previous works, we have proved the Mössbauer spectroscopy (MS) is a powerful technique to investigate the magnetic behavior and shape anisotropy of magnetic nanowires.^{12,18} As we know, the dipolar field results in the change of the effective hyperfine field at ⁵⁷Fe nuclei, and therefore the Mössbauer spectroscopy can be adopted to investigate the dipolar interaction in magnetic nanowire arrays. Besides Mössbauer spectroscopy, a few techniques, such as nuclear magnetic resonance (NMR),⁵ and Brillouin light scattering (BLS),^{14,15} etc., are also effective by probing the dipolar interaction in the nanostructured magnetic materials.

Herein, we have investigated the dipolar interaction in the hexagonal-close-packed nanowire arrays by using the dipole

approximation with a length correction. Mössbauer spectroscopy with applied fields was adopted to probe the dipole-dipole interaction in the Fe nanowire arrays. When the saturation field was perpendicular to the wire axis, a dipolar field of 2.8 kOe, which is much stronger than that of nanodot arrays,¹⁴ was observed. The experimental results agree well with the numerical calculations.

II. EXPERIMENT

The arrays of Fe nanowires were fabricated by electrodepositing Fe into porous anodic aluminum oxide (AAO) templates. The detailed procedures for preparing AAO templates and electrodepositing Fe nanowires have been described in our previous works.^{11,18} The magnetic properties of Fe nanowire arrays were measured by a superconducting quantum interference device magnetometer (Quantum Design). For Mössbauer measurements, the AAO films were flaked off from the aluminum substrates after completely dissolving the Al foils in an HgCl_2 saturated solution, and a stack (eight pieces) of as-prepared AAO film instead of a single piece was used to improve the absorptive signals. The ^{57}Fe Mössbauer spectra were collected at 10 K and RT using a conventional constant-acceleration spectrometer with a $^{57}\text{Co}(\text{Pd})$ source, with the γ beam parallel to the nanowire axes. The low temperature and the various magnetic fields up to 50 kOe both parallel and perpendicular to the nanowire axes are provided by an Oxford Spectromag SM4000-9 superconducting split pair, horizontal field magnet system. The values of velocities were calibrated using a α -Fe foil at room temperature.

III. RESULTS AND DISCUSSION

Previously, the dipolar field in nanodot arrays has been calculated base on the dipole approximation. However, this method is unsuitable for nanowire system due to the length of the nanowires. Herein, we developed a length correction to estimate the dipolar field in the nanowire arrays, based on actual structures that can be simply synthesized by using the above-mentioned methods. The magnetic nanowire arrays we studied are ordered in a two-dimensional hexagonal-close-packed array, typically of diameter about 20 nm, lattice constant 60 nm, and length about 10 μm . Because of the shape anisotropy resulted from the high aspect ratio of the nanowires, the magnetization is oriented along the axes of the nanowires, giving rise to square hysteresis loops.¹² Prior to the investigation of the dipolar interaction, three typical states of magnetic nanowire arrays are defined firstly. They are the demagnetized state (state I), the saturation field parallel to the wire axes (state II), and the saturation field perpendicular to the wire axes (state III).

In state II, considering the length of nanowire, one nanowire is not same as a nanodot to be viewed as a dipole. The suitable way to treat the long nanowire is to put monopole with magnetic charge $m=MS$ on the end of each wire, where M is the magnetization of the nanowire, S is the area of the nanowire end [Fig. 1(a)]. For a point P on the nearest wire but far away the end of nanowire, that is to say

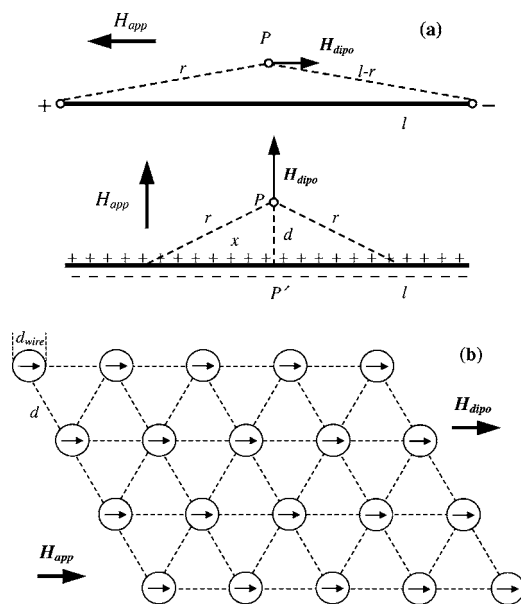


FIG. 1. (a) Magnetic charge distribution in a magnetic nanowire and dipolar field on point P produced by the wire, with the magnetic field applied parallel and perpendicular to the wire axis. (b) Schematic drawing of a top view of hexagonal dipolar arrays of nanowires with a lattice spacing d , when the magnetization is perpendicular to the wire axis. The dipolar interaction in the nanowire arrays gives rise to an additional field collinear to the applied field.

$r \approx l/2$ with r the distance between this point and the monopole, l the length of nanowire, the magnetic potential is $\varphi = \varphi_+ + \varphi_- = (m/r) + [m/(l-r)] \approx 2m/r$. Consequently, the dipolar field on point P produced by the wire is $H_{\text{dipo}} = -\nabla\varphi = 2m/r^2 \approx 2\pi M(d_{\text{wire}}/l)^2$, where d_{wire} is the diameter of nanowire. For a nanowire with high aspect ratio d_{wire}/l , both the magnetic potential and the dipolar field in the middle part of nanowire arrays are about zero. Thus, the total dipolar field acting on P produced by all the surrounding magnetic nanowires is also very weak. In the demagnetized state (state I), the magnetization of nanowire is still along the wire axis due to the shape anisotropy, but the direction alternates up and down in an ordered zig-zag pattern to reach no net magnetization.^{8,13} For an individual nanowire, state I is the same as state II. Therefore, the sum of the dipolar field produced by all the wires in the ground state is also about zero. Actually, for the arrays of nanowires with finite length, the dipolar field is quite inhomogeneous along the wire axes. But the magnitude of the dipolar field in the state I and state II is too small (lower than 1 Oe) to be measured by conventional technology including the Mössbauer spectroscopy.

In state III, when the saturation field is applied perpendicular to the wire axes, the nanowire arrays can be viewed as two-dimensional dipole arrays with a length correction. The dipole moment of each dot is $p = p_x \mathbf{x} + p_y \mathbf{y}$. The position of each dipole dot can be defined as $\mathbf{r} = i\mathbf{a} + j\mathbf{b}$, where i and j are integers, \mathbf{a} and \mathbf{b} are basic vectors of two-dimensional hexagonal-close-packed lattice. In the rectangular coordinate, $\mathbf{a} = \frac{1}{2}d\mathbf{x} + (\sqrt{3}/2)d\mathbf{y}$ and $\mathbf{b} = \frac{1}{2}d\mathbf{x} - (\sqrt{3}/2)d\mathbf{y}$, where d is the distance between nanowires. The component of the dipolar field at a wire located at

the origin, generated by all other wires, can be written in the form.^{14,19}

$$H_x = \sum_{ij} \frac{1}{d^3} \frac{(10ij - i^2 - j^2)p_x + \sqrt{3}(i^2 - j^2)p_y}{4(i^2 + j^2 - ij)^{5/2}}, \quad (1)$$

$$H_y = \sum_{ij} \frac{1}{d^3} \frac{3\sqrt{3}(i^2 - j^2)p_x + (5i^2 + 5j^2 - 14ij)p_y}{4(i^2 + j^2 - ij)^{5/2}}. \quad (2)$$

Considering a large two-dimensional array, for example 2000×2000 array, the dipolar field is calculated to be $H_{\text{dipo}} = 5.517p/d^3$. The dipolar field is collinear to the direction of the applied field and the dipole moment, which is shown in Fig. 1(b). The dipolar field is isotropic in the x - y plane no matter what direction is the applied field. However, for a finite array, a sixfold anisotropy of the dipolar field in plane is resulted from a so-called edge effect.¹⁴ For the nanowire arrays in state III, the dipole moment cannot be simply written as $p = MV$, just like nanodot arrays, where V is the volume of a dipole. A length correction should be carried out in a nanowire system. In state III, each nanowire can be considered as equivalent to two infinite lines with the magnetic charge line density $\sigma_m = MS/d_{\text{wire}}$ [Fig. 1(a)]. The magnetic potential on point P produced by positive magnetic charge and the component perpendicular to the wire axis is $\varphi_{\perp} = \int_{-l/2}^{l/2} [\sigma_m d / (x^2 + d^2)] dx = 2\sigma_m \arctan(l/2d)$, the component parallel to the wire axis is zero for the symmetric distribution of magnetic charge. The magnetic potential produced by a long line with magnetic charge is equivalent to the effect of a monopole on the point P' . The magnetic charge of the equivalent monopole is $\sigma_m l_{\text{eff}} = \varphi_{\perp} d = 2\sigma_m d \arctan l/2d$, and the dipole moment in two-dimensional lattice is $p = \sigma_m l_{\text{eff}} d_{\text{wire}} = MS l_{\text{eff}}$, where we define $l_{\text{eff}} = 2d \arctan l/2d$ as the effective length of nanowire, that is to say, in state III the dipole moment of a nanowire in the dipole approximation is just equivalent to the total moment of a nanowire with the length of l_{eff} . For an infinite long nanowire, the effective length is πd . Therefore, the dipolar field is $H_{\text{dipo}} = 1.379\pi^2 M (d_{\text{wire}}/d)^2$. For a typical nanowire array, using $d_{\text{wire}} = 20$ nm, $d = 60$ nm, and $4\pi M \approx 20$ kG (slightly lower than bulk Fe but typical for Fe nanomagnetic materials),¹⁴ the dipolar field of the Fe nanowire arrays is about 2.4 kOe in state III.

In order to validate our model, the Fe nanowire arrays with the distance between wires d about 60 nm, the diameter of nanowires d_{wire} is about 20 nm, the length of the nanowires l is about $7 \mu\text{m}$, have been fabricated. The sizes of the templates and nanowires were checked by the transmission electron microscope, which have been reported in our previous works.¹¹ Figure 2 shows the hysteresis loops of Fe nanowire arrays in AAO films with the applied field parallel and perpendicular to the wire axes at 10 K and 300 K, respectively. The hysteresis loops of Fe nanowire arrays reveal that the saturation fields (H_S) perpendicular to the wire axes are about 13 kOe and 11 kOe at 10 K and 300 K, respectively. The saturation field is usually very close to the anisotropy field (H_A) in nanowires. But the value of saturation field is larger than the theoretical value ($2\pi M = 10$ kOe) of the shape

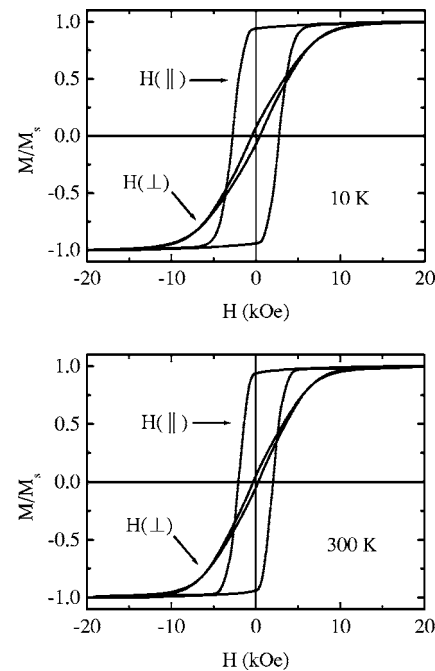


FIG. 2. Normalized hysteresis loops of Fe nanowire arrays with typical sizes at 10 K and room temperature, $H(\parallel)$ indicates the applied field parallel to the wire axes, $H(\perp)$ denotes the magnetic field perpendicular to the wire axes.

anisotropy of Fe nanowire arrays. Considering the additional dipolar field in state III, the actual saturation field is even larger than that just obtained from hysteresis loops. The large saturation field is perhaps attributed to the surface spins of Fe nanowires.²⁰ Due to the coordination and the interface between the Fe wire and the template, surface spins are normally very difficult to be aligned, which gives rise to a very larger saturation field.

It was proved that the applied field Mössbauer spectroscopy is a powerful technique to probe the internal field at ^{57}Fe nuclei.^{12,21} The dipolar field in nanowire arrays could be probed directly by comparing the changes of the internal field between the above-mentioned states. Figure 3 shows some typical Mössbauer spectra of Fe nanowire arrays in all states. Figures 3(a)–3(c) are Mössbauer spectra with zero-field, the applied field of 20 kOe parallel and perpendicular to wire axes at 10 K, respectively. It is well known that, in magnetically split spectra, the relative intensities of the second and fifth absorption peaks (corresponding to the $\Delta m = 0$ nuclear transitions) are given by $I_{2,5} = 4 \sin^2 \theta / (1 + \cos^2 \theta)$, where θ is the angle between the Fe spin and the γ beam direction. In the case of all Fe spins are collinear to the γ beam direction $I_{2,5} = 0$, while all magnetic moments are perpendicular to the γ beam $I_{2,5} = 4$. In Figs. 3(a) and 3(b), the 2–5 lines in the sextets almost disappear, which indicates all Fe spin orient parallel to the nanowire axes. For the magnetic field is far larger than the saturation field in the direction of parallel to the wire axes, the magnetization vectors of Fe nanowires should be parallel with each other in the case of Fig. 3(b), which corresponds to state II. For the zero-field Mössbauer spectrum, the sample is in demagnetizing state, i.e., state I. The magnetization vectors of Fe nanowires might

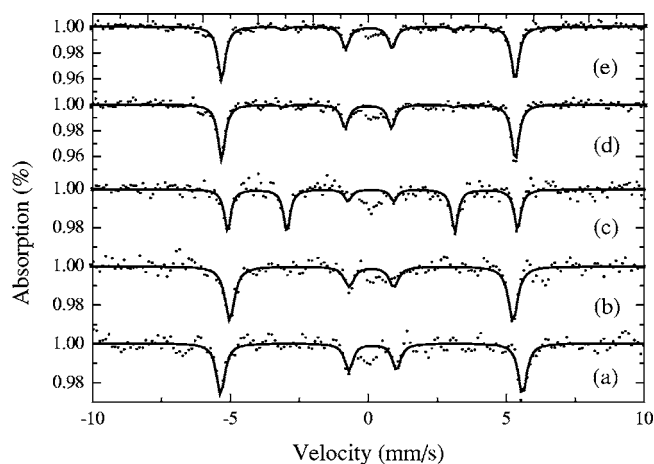


FIG. 3. Typical Mössbauer spectra of Fe nanowire arrays in the zero-field (a), the applied field of 20 kOe parallel (b) and perpendicular (c) to wire axes at 10 K, the demagnetized state (e) and remanent state (f) at RT, respectively.

be antiparallel with the adjacent nanowires to achieve no net magnetization. When the applied field is larger than the saturation field in the direction of perpendicular to the wire axes, the Mössbauer spectrum with $I_{2,5}=4$ in Fig. 3(c) indicates all magnetic moments align completely perpendicular to the wire axes, that is to say the Fe nanowire arrays have reach saturation, which satisfies state III we have described.

The effective hyperfine field H_{eff} at ^{57}Fe nuclei in the nanowire arrays can be expressed as

$$H_{\text{eff}} = H_{\text{hf}} - H_{\text{app}} + H_{\text{dem}} - H_{\text{dipo}}, \quad (3)$$

where H_{hf} is the hyperfine field, H_{app} is the applied field, H_{dem} is the self-demagnetizing field of the nanowires, and H_{dipo} is the dipolar field produced by the surrounding Fe nanowires. It should be noted that the hyperfine field at ^{57}Fe nuclei is antiparallel to the magnetic moment. The demagnetizing field is given by $H_{\text{dem}} = -NM$, where N is the demagnetization factor that depends on the direction of magnetization, and M is the magnetization vector. As we known, the demagnetization factors of an individual infinite wire are 0 and 2π in the direction of parallel and perpendicular to the wire axis, respectively. Due to the difficulty in measuring the weight of nanowires in AAO films, the saturation magnetization of Fe nanowires cannot be directly obtained from the magnetization measurement. Herein, we also use $4\pi M \approx 20 \text{ kG}$ as the saturation magnetization of Fe nanowire arrays, which leads to $H_{\text{dem}\perp} = 10 \text{ kOe}$ in state III and $H_{\text{dem}\parallel} = 0$ in state I and state II. Because the dipolar field is collinear to the applied field in state III, those fields are all in a line for all the states we defined. Only under this condition, the vector sum of those field can be expressed as Eq. (3).

Figure 4 illustrates the effective field H_{eff} for Fe nanowire arrays as a function of the applied field H_{app} . Except the zero-field Mössbauer spectrum, the fields applied in others spectra are far larger than the saturation field obtained by magnetization measurement, which ensures the nanowire arrays have reached saturation. In the state I, the hyperfine field of Fe nanowire arrays matches perfectly with the value of

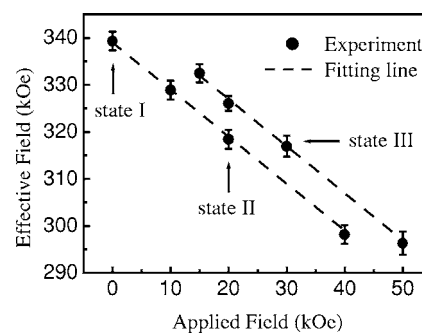


FIG. 4. The effective field H_{eff} for Fe nanowire arrays as a function of the applied field H_{app} at 10 K. The dots and dashed lines are experimental and linear fitting results, respectively. State I is the demagnetized state. State II and state III denote the nanowire arrays reach saturation in the direction of parallel and perpendicular to the wire axes, respectively.

bulk iron. In the state II, the effective field can be fitted in the formula of $H_{\text{eff}} = H_{\text{hf}}(\text{state I}) - H_{\text{app}}$ within the experimental error. In the state III, the effective field can be fitted as $H_{\text{eff}} = H_{\text{hf}}(\text{state I}) - H_{\text{app}} + H_{\text{dem}\perp} - 2.8 \text{ kOe}$. Therefore, the dipolar fields are 0, 0, and 2.8 kOe for state I, state II, and state III, respectively. The results are in good agreement with the calculated value according to our model. The deviation of the dipolar fields between experimental results and theoretical predictions might be arisen from the error of the size parameters of Fe nanowire arrays and the line widths of Mössbauer spectra.

Furthermore, we have measured the room-temperature Mössbauer spectra of the Fe nanowire arrays in the demagnetized state and the remanent state [Figs. 3(d) and 3(e)]. The remanent state is for the sample having been saturated with the 20 kOe magnetic field along the wire axes. This state is a special case of state II without the applied field. The magnetizations of Fe nanowires are all in the same direction on account of the hysteresis loop with high squareness. No deviation in the internal field between the two cases is found and the internal fields agree very well with the hyperfine field of the bulk iron, which also demonstrates that the dipolar field in the state I and state II is about zero in the nanowire system. We also found in state I and state II the nanodot arrays are quite different from the nanowire arrays. We believed that if using Mössbauer spectra to study the nanodot arrays, the deviation between the demagnetized state and the remanent state would be found, and there would be a relationship of $H_{\text{dipo}}(\text{state II}) = -2H_{\text{dipo}}(\text{state III})$.^{5,22,23}

From our model we can understand that a competition between dipolar interaction and demagnetization energy can lead to a preferential direction of magnetization parallel or perpendicular to the wires. When all the spins are aligned along the wires, the effective applied field acting on one wire is the sum of the dipolar field and the demagnetizing field $H_{\parallel} = H_{\text{dem}\parallel} + H_{\text{dipo}\parallel} = 0$. When all the moments are perpendicular to the wires, the total fields are $H_{\perp} = H_{\text{dem}\perp} - H_{\text{dipo}\perp} = 2\pi M - 1.379\pi^2 M(d_{\text{wire}}/d)^2$, where the positive sign corresponds to the fields of directions opposite to the magnetization vector. When $H_{\perp} > H_{\parallel}$, the preferential direction of magnetization is along the wires. While $H_{\perp} < H_{\parallel}$, the preferential

direction of magnetization is perpendicular to the wire axis, and $H_{\perp}=H_{\parallel}$ corresponds to the critical ratio between the diameter of nanowire and the distance between nanowires, so that $(d_{\text{wire}}/d)_{\text{critical}}=0.68$. Therefore the easy magnetization direction can be tuned parallel or perpendicular to the wire axes by change the ratio porosity of templates. For our Fe nanowire arrays, the ratio of $(d_{\text{wire}}/d)\approx 0.33$ is far lower than the critical ratio, so that the magnetic easy axis is along the wires and the hysteresis loops is quite square. For an actual nanowire system, the magnetocrystalline anisotropy should be taken into account to correct the critical ratio, especially for nanowires with high magnetocrystalline anisotropy such as Co nanowire.

IV. CONCLUSION

In this work we have investigated the dipole-dipole interaction of two-dimensional arrays of parallel ferromagnetic Fe nanowires embedded in nanoporous alumina templates. By combining the dipole approximate with a length correction, we have predicted the dipolar field with the saturation field applied perpendicular to the wire axes is rather strong

while the dipolar interaction with magnetization aligned along the wire axes is quite weak. By means of Mössbauer spectroscopy, we have measured the effective field as a function of the applied field in saturated magnetization states. The Mössbauer results show the dipolar fields are 0, 0, and 2.8 kOe for the demagnetized state, the saturation field parallel and perpendicular to the wire axis, respectively. The experimental results of dipolar fields are in good agreement with our theoretical model. By comparing the dipolar interaction with the shape anisotropy, the critical ratio between the diameter of nanowire and the distance between nanowires, which is a key parameter to determine the easy magnetization direction parallel or perpendicular to the wire axes, was calculated.

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