Polarized-neutron-diffraction study of the microscopic magnetic structure in α'' -Fe₁₆N₂ nanoparticles

H. Hiraka,¹ K. Ohoyama,² Y. Ogata,³ T. Ogawa,⁴ R. Gallage,⁵ N. Kobayashi,^{5,6} M. Takahashi,³

B. Gillon,⁷ A. Gukasov,⁷ and K. Yamada¹

¹Institute of Materials Structure Science, High Energy Accelerator Research Organization (KEK), Tokai, Ibaraki 319-1106, Japan

²Center of Neutron Science for Advanced Materials, Institute for Materials Research (IMR), Tohoku University, Sendai 980-8577, Japan

³New Industry Creation Hatchery Center (NICHe), Tohoku University, Sendai 980-8579, Japan

⁴Graduate School of Engineering, Tohoku University, Sendai 980-8578, Japan

⁵Technical Strategy Department, Research and Development Division, TODA Kogyo Corporation, Otake, Hiroshima 739-0652, Japan

⁶T&T Innovations Inc., Otake, Hiroshima 739-0652, Japan

⁷Laboratoire Leon Brillouin (LLB), CEA/CNRS, F-91191 Gif-sur-Yvette Cedex, France

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Polarized-neutron-diffraction (PND) measurements were carried out using a pseudo-single-phase powder sample of ferromagnetic α'' -Fe₁₆N₂ nanoparticles. For the well-identified α'' -Fe₁₆N₂ phase, sizes of the magnetic moments at the three crystallographic Fe sites were determined in the absolute scale. The agreement between the magnetization value deduced from the present PND and that measured by a magnetometer (M_{VSM}) supports the hypothesis that M_{VSM} is primarily caused by the magnetization value in the target α'' -Fe₁₆N₂; thus there is no evidence for macroscopic giant saturation magnetization, at least for α'' -Fe₁₆N₂ nanoparticles. On the basis of the large magnetic moment size at one of the Fe sites, a possible coexisting state of localized spins and itinerant electron spins is inferred. Drawing a distinction between thin films and nanoparticles is currently necessary because of their divergent magnetic evolutions.

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I. INTRODUCTION

Beyond the saturation-magnetization values of α -Fe ($M_s = 217 \text{ emu/g}$) [1] and Fe₇₀Co₃₀ ($M_s \sim 250 \text{ emu/g}$) [2], a very large saturation magnetization of $M_s \sim 275 \text{ emu/g}$ and $\sim 310 \text{ emu/g}$ were reported for thin films of α'' -Fe₁₆N₂ by Kim and Takahashi [3] and Sugita *et al.* [4], respectively. Since then, whether a giant saturation magnetization in α'' -Fe₁₆N₂ is attainable or not has been a controversial question [5].

Recently, Ji *et al.* measured the depth dependence of the saturation-magnetic induction of an epitaxial α'' -Fe₁₆N₂ thin film by polarized neutron reflectometry [6]. They concluded that a giant saturation magnetization of $M_s \sim 330$ emu/g is realized locally in the near-substrate interface region over ~ 10 nm thickness, while the average value over the whole film is 258 emu/g. On the other hand, the Takahashi and Ogawa group succeeded in synthesizing *single-phase* α'' -Fe₁₆N₂ nanoparticles on the order of a few grams by ensuring that surface oxidation is avoided [7]. These premium nanoparticles result in a macroscopic magnetization of $M_s = 234$ emu/g and a coercivity of $H_c \simeq 4$ kOe at low temperatures. Their obtained M_s is 8% larger than that of α -Fe, but much smaller than the above giant-saturation-magnetization values.

From the standpoint of condensed matter physics, it is important to clarify the microscopic mechanism of such a large saturation magnetization. Most band calculations for α'' -Fe₁₆N₂ predict M_s values larger than that of α -Fe but the enhancement is limited at most to ~10% [8–11] in the band magnetism as shown in Table I; this enhanced value is nearly equal to that of Fe₇₀Co₃₀, which is the highest value in the so-called Slater-Pauling plot [2]. On the other hand, Lai *et al.* introduced strong electron correlations in the band calculation through an on-site Coulomb interaction and deduced a large M_s value of 275 emu/g [12], which shows a gain of +27% compared to α -Fe and corresponds to the value of the giant saturation magnetization in thin film.

In order to make sense of the chaos regarding the magnetization value of α'' -Fe₁₆N₂ (Table I), it is necessary to make clear whether the sample form was films or powders, whether the sample came from a single phase or a multiphase, whether the magnetization arose from the entire bulk or a local region, and which measurement tool was used. Besides, for comparison with theoretical calculations from the microscopic standpoint, it is vital to clarify the magnetic moment sizes of Fe at the three crystallographic sites: the 4*e*, 8*h*, and 4*d* positions in the *I*4/*mmm* tetragonal lattice [17,18]. Surprisingly, no experimental values are available on the magnetic moment sizes so far; hyperfine fields determined site selectively by Mössbauer [5,7,13] and NMR [14] experiments are proportional, but not directly equal, to the absolute moment sizes.

In this study we used a gram-order, well-phase-assigned α'' -Fe₁₆N₂ nanoparticle sample to investigate the strong magnetization by polarized-neutron diffraction (PND). In particular, we directly determined the sizes of the Fe magnetic moments at the three crystallographic sites; the extracted moment sizes are consistent with the macroscopic magnetization value measured by a magnetometer. Therefore, at least no giant saturation magnetization appears in α'' -Fe₁₆N₂ nanoparticles (~100 nm particle size) as a macroscopic property. We discuss the possible coexistence of localized spins and itinerant electron spins in α'' -Fe₁₆N₂ on the basis of the magnetic moment sizes determined by the present PND.

TABLE I. Observed and calculated magnetic quantities in α'' -Fe ₁₆ N ₂ . The volume fraction of the α'' -Fe ₁₆ N ₂ phase is also here	sted at $V_{\alpha''-\text{Fe}_{16}\text{N}_2}$
(%). A negative (positive) sign for $m_{\rm N}$ means $\mathbf{m}_{\rm N}\mu_{\rm B}$ is antiparallel (parallel) to H .	

Reference	Magnetic mom., $\mu_{\rm B}$ [hyperfine field, kOe]						
	m_1	m_2	<i>m</i> ₃	m _N	$M_{\rm s}~({\rm emu/g})$	$V_{\alpha''-{ m Fe}_{16}{ m N}_2}$	Comment
Kim [3]	_	_	_	_	~ 290	50-80	Film
Sugita [4]	[330]	[330]	[460]	_	~310	100	Film
Takahashi [5]	[290]	[316]	[400]	_	$\lesssim 240$	82	Film
Ji [6]	-	-	-	-	$330^{a} (258^{b})$	100	Film
Coey [13]	[301]	[312]	[395]	-	220-250	32	Bulk
Zhang [14]	[ratio 1.00]	[ratio 1.07]	[ratio 1.36]	_	285	~ 50	Bulk
Huang [15]	_	_	_	_	270-290	~ 50	Bulk
Oku [16]	_	_	_	_	~ 210	~ 30	Nanoparticles
Ogawa [7]	[297.6]	[315.4]	[403.0]	-	234	100	Nanoparticles
Present study	1.4 ± 0.2	1.8 ± 0.2	2.6 ± 0.3	(0.03 ± 0.04)	$181 \pm 14^{\circ}, 185^{d}$	>93	Nanoparticles
Min [8]	2.13	2.50	2.85	0.06	241		LMTO
Huang [9]	2.06	2.42	2.90	-0.06	236		OLCAO
Sakuma [10]	2.27	2.25	2.83	-0.07	231		LMTO
Tanaka [11]	2.17	1.95	2.74	-0.03	212		FLAPW
Lai [12]	2.36	2.75	3.53	-0.01	275		LDA+U

 ${}^{a}M_{s}$ in local determined by polarized-neutron reflectometry.

 ${}^{\mathrm{b}}M_{\mathrm{VSM}}.$

 $^{c}M_{PND}(170 \text{ K}, 5 \text{ T}).$

^d*M*_{VSM}(300 K, 1.45 T).

II. POLARIZED NEUTRON DIFFRACTION

The intensity of ferromagnetic Bragg reflection using polarized neutrons with polarization degree *P* is written as [19]

$$I^{\pm} \propto F_{\rm N}^2 + F_{\rm M}^2 \pm 2PF_{\rm N}F_{\rm M},\tag{1}$$

where the sample magnetization (**M**) is saturated by a magnetic field (**H**) applied perpendicularly to the scattering plane, and the neutron-polarization vector (**P**) is parallel/antiparallel to **M**. I^{\pm} corresponds to the intensity for the case where **P** is oriented to **H** in parallel (+) or antiparallel (-). $F_{\rm N}$ and $F_{\rm M}$ are the nuclear and magnetic structure factors, respectively, and $F_{\rm N}$ is calculated from already known crystal structure parameters. Thus one can determine $F_{\rm M}$ (i.e., the magnetic moment) in the absolute scale by measuring the flipping ratio (I^+/I^-) and solving the following equation:

$$\frac{I^{+}}{I^{-}} = \frac{F_{\rm N}^{2} + F_{\rm M}^{2} + 2PF_{\rm N}F_{\rm M}}{F_{\rm N}^{2} + F_{\rm M}^{2} - 2PF_{\rm N}F_{\rm M}}.$$
(2)

On the other hand, the interference term in Eq. (1) is obtained by subtracting I^- from I^+ :

$$I^+ - I^- \propto 4P F_{\rm N} F_{\rm M}.\tag{3}$$

During the current data analysis, we examine the d-spacing (D) integrated intensity of a polycrystalline sample. Thus the following formula applies:

$$\int dD (I^+ - I^-)_{hkl} = C \frac{m_{hkl}}{\sin\theta \sin 2\theta} P(F_{\rm N}F_{\rm M})_{hkl}, \quad (4)$$

where C, m_{hkl} , and $1/(\sin\theta \sin 2\theta)$ represent the scale factor, the multiplicity of the Bragg reflection (hkl), and the Lorentz factor, respectively. The integrated intensity of the left-hand side in Eq. (4) is experimentally quantified from diffraction data in order to determine $F_{\rm M}$ in a relative scale.

III. EXPERIMENTS

A powder sample of α'' -Fe₁₆N₂ nanoparticles was prepared as previously described [7]. The x-ray diffraction pattern guarantees a main phase of α'' -Fe₁₆N₂, residues of α -Fe ($\leq 5\%$) and Fe₄N (<1%), and a paramagnetic oxide Fe₂O₃ ($\leq 1\%$). Accordingly, the α'' -Fe₁₆N₂ phase volume accounts for over 93% of the volume fraction; this value is considerably larger than the volume fractions in bulk powders [13-15] and coated nanoparticles [16] reported previously. The particle size used here was ~ 100 nm and the nanoparticles were uncoated so that the magnetism of pristine α'' -Fe₁₆N₂ could be examined. Magnetization measurement at room temperature and at H =14.5 kOe using a vibrating-sample magnetometer (VSM) showed $M_{\rm VSM} = 185$ emu/g for the current slightly impurityphase-admixed sample. On the other hand, the optimized α'' -Fe₁₆N₂ single-phase nanoparticles exhibited $M_{\rm VSM}^{\rm opt} =$ 208 emu/g under the same condition of H and T [7]. Hence the magnetization value in the current sample is smaller than $M_{\rm VSM}^{\rm opt}$ by ~11%.

PND measurements were carried out on a polarized hotneutron 5C1 diffractometer [20] at the Laboratoire Leon Brillouin, France. A polarized neutron beam with P = 0.85and $\lambda = 0.84$ Å was used. A vertical field of 5 T, which is much higher than H_c , was applied to the sample to ensure saturation in the sample magnetization. The details of the PND experimental configuration are described in the Supplemental Material [21].

IV. RESULTS AND ANALYSES

Figure 1(a) shows the raw data of I^+ and I^- as a function of the *d* spacing, *D*. The finite deviation between the two corresponds to $F_M \neq 0$ in Eq. (3) or the presence of magnetic



FIG. 1. (Color online) (a) Raw *D* profiles of I^+ and I^- . The series of bars at the bottom indicate D_{hkl} positions calculated from the crystal parameters of α'' -Fe₁₆N₂, α -Fe, and Al. The non-negligible Al scattering comes not from the sample itself but possibly from an Al attachment above the sample holder. (b) Difference plot of $I^+ - I^-$. The inset shows a magnification of the large *D* region. Thirteen Bragg reflections, denoted by green indices, are used for analysis (see Supplemental Material [22]), and the resultant fit is shown by the green curve.

scattering from the sample, as expected for ferromagnets. However, the background level is relatively high and the nuclear Bragg peaks from α -Fe and Al are located close to those of α'' -Fe₁₆N₂. Because of the use of a powder sample and medium collimation, it is difficult to separate and assign Bragg reflections to the broad peaks, particularly in the small *D* region (<2 Å). This is why a full flipping-ratio analysis using Eq. (2) is unsuitable for the current case.

In fact, better quality information is derived from the difference data between I^+ and I^- , as shown in Fig. 1(b); nonmagnetic background signals are completely removed so that purely magnetic components are accurately extracted, as formulated in Eq. (3). The peak positions in the difference plot agree well with the D_{hkl} values that accompany the large $|F_N|$ of α'' -Fe₁₆N₂ (see Supplemental Material [22]). Therefore, the ferromagnetic cross section of this nanoparticle sample has been confirmed to originate most dominantly from α'' -Fe₁₆N₂; this fact is consistent with the prevailing structural volume fraction of α'' -Fe₁₆N₂, as characterized by the x-ray diffraction.



FIG. 2. (Color online) Prefactor-corrected net integrated intensity A_{hkl}^{obs} versus the calculated interference term ($\propto F_N F_M$). In this plot, $m_1 = 0.8m_2, m_3 = 1.5m_2, B_{Fe} = 4.8 \text{ Å}^2$, and $B_N = 2.7 \text{ Å}^2$ are used. The lower inset magnifies the crowd portion around the origin. The upper inset shows a unit cell of α'' -Fe₁₆N₂, drawn by VESTA [23].

To quantify the integrated net intensity of each ferromagnetic Bragg peak from α'' -Fe₁₆N₂, $A_{hkl}^{obs} \equiv \int dD (I^+ - I^-)_{hkl}$, the difference profile was fitted to a Gaussian complex by floating the peak height and peak width but holding the peak position fixed to the D_{hkl} values calculated for α'' -Fe₁₆N₂. Figure 2 plots the prefactor-corrected A_{hkl}^{obs} against the calculation of $(F_N F_M)_{hkl}$ (see Supplemental Material [22]). For calculating $(F_N F_M)_{hkl}$, five parameters were used: m_1, m_2, m_3 , $B_{\rm Fe}$, and $B_{\rm N}$. The first three parameters represent the magnetic moment sizes of Fe at the $Fe_1(4e)$, $Fe_2(8h)$, and $Fe_3(4d)$ sites, respectively (upper inset of Fig. 2); here, the Fe magnetic moments of $\mathbf{m}_n \mu_{\rm B}$ (*n* = 1,2,3) were arranged collinearly in the calculation. The last two parameters, $B_{\rm Fe}$ and $B_{\rm N}$, are the isotropic atomic-displacement parameters for Fe and N, respectively (see Supplemental Material [22]). To establish a linear relationship between the observation and calculation data, we computed the sum of squared residuals (χ^2) and attempted to minimize χ^2 by changing the five parameters one by one. After several iterations, the magnetic moment ratios converged to $(m_1/m_2) = 0.8 \pm 0.1$ and $(m_3/m_2) = 1.5 \pm 0.1$. Here, the error was evaluated by allowing a deviation of $3\sqrt{\chi^2}$ in χ^2 . The resultant linearity was found to be excellent over a wide range (including the negative region), as shown in Fig. 2. Indeed, the green curve in Fig. 1(b) using the final parameters reproduces the observations very well.

Next, in order to determine the absolute values of m_n , we tried to evaluate a flipping ratio between I^+ and I^- .

Fortunately, the scattering on the tail at $D \sim 1.6$ Å, which is mainly composed of (004) and (213) Bragg reflections, is less contaminated by α -Fe and Al, as seen in Fig. 1(a). Because of the small splitting in D_{hkl} between (213) and (004) reflections (see Supplemental Material [22]), as compared with the *D* resolution, we employed the summation $(I^{\pm})_{213} + (I^{\pm})_{004}$ for I^{\pm} in Eq. (2). Then, the flipping ratio was experimentally determined as $(I^+/I^-)_{213+004} = 2.0 \pm 0.2$. By solving Eq. (2), we finally obtained the following values: $m_1 = 1.4 \pm 0.2$, $m_2 = 1.8 \pm 0.2$, and $m_3 = 2.6 \pm 0.3$.

Now, we describe a possible magnetic moment induced at the N 2*a* site ($\mathbf{m}_N \mu_B$). By introducing a sixth parameter of m_N , we repeated the above simulation and estimated the order of magnitude of $m_N = 0.03 \pm 0.04$ at this stage. In order to further investigate the moment m_N , however, another high-resolution PND experiment is required to reduce the uncertainty.

V. DISCUSSION

We identified the α'' -Fe₁₆N₂ composition as the nearly magnetic single phase and succeeded in determining m_1 , m_2 , and m_3 in the absolute scale with a collinear ferromagnetic model. From the relative abundance of 1:2:1 among the three Fe sites, $M_{PND} = 181 \pm 14$ emu/g is derived from the absolute values of m_n for the α'' -Fe₁₆N₂ phase, which agrees well with M_{VSM} . This good agreement confirms that M_{VSM} , which measures magnetization in the whole volume, emerges mostly from α'' -Fe₁₆N₂. In other words, the possible surface matrices, such as iron oxides, should be less significant in the present surface-uncoated nanoparticle sample. From this fact, we inductively conclude that the M_s (=234 emu/g) of the optimally magnetized sample [7] is governed by the intrinsic magnetic property of α'' -Fe₁₆N₂.

Next, we discuss the sizes of the Fe magnetic moments, which give rise to the large magnetization values. Table I lists the extracted Fe moment sizes together with those from several band calculations [8-10] and hyperfine fields from Mössbauer [5,7,13] and NMR [14] experiments. Some common features can be identified. First, m_3 is greater than the moment size of α -Fe (2.2 μ _B) and fairly consistent with the band calculation results. Second, the magnitude relation $m_1 \lesssim$ $m_2 < m_3$, which was anticipated from the local hyperfine fields and the band calculations, is directly confirmed by the present PND study. At first glance, these results support the effectiveness of the band picture for Fe magnetic moments in this system. Third, the inspection of m_n between the present data and the band calculations indicates that the degradation of M in the present sample is synchronized with the large shrinkage of m_1 and m_2 . Because a considerable hybridization of wave functions is expected in N-Fe1 and N-Fe2 electron bondings [9], the decrease in m_1 and m_2 is possibly related to unexpected electronic states at the N site, such as a N deficiency in a putative α'' -Fe₁₆(N_{1- δ})₂ phase. (Ohoyama *et al.* recently conducted high-resolution, unpolarized-neutron powder diffraction measurements using α'' -Fe₁₆N₂ nanoparticles synthesized in the same way and quantitatively determined a percentage-order N deficiency [24].)

These results remind us that the magnetic moment in α'' -Fe₁₆N₂ comprises two types of spins: itinerant electron

spins near N atoms (small m_1 and m_2) and localized spins far from N atoms (large m_3). Such a qualitative difference is parallel to band calculations of the partial density of state [11]: an intermixed field of bcc Fe and fcc Fe at the Fe3 site, instead of the bcc-Fe like field at the Fe1 and Fe2 sites. Besides, the coexisting state of itinerant electron spins and localized spins is consistent with a recent band calculation based on the LDA+U method [25,26], which leads to a partially localized electron configuration with a long-range ferromagnetic order. Indeed, the magnetic moment at the Fe₃ site reaches almost $3\mu_{\rm B}$ so that a high-spin state or an intermediate-spin state of localized spins would be the appropriate explanation. The generation of the localized spins can (i) contribute to the enhancement of H_c of α'' -Fe₁₆N₂ [7] via an increase in the single-ion anisotropy of Fe spins and (ii) additionally produce magnetic interaction with itinerant electron spins, such as the RKKY interaction. Therefore, the simple band theory may be inadequate to understand and predict the magnetism of α'' -Fe₁₆N₂.

Finally, we mention the difference in M_s values between the current nanoparticles and thin films. According to Ji et al. [6], the giant saturation magnetization in films is thought to be an outcome of the spatial distribution in magnetization and it appears not entirely in a sample but partially in a mesoscopic scale of ~ 10 nm thickness because of the strong strain force near the interface region. Hence the absence of giant saturation magnetization or the smaller M_s values is a natural consequence from strain-free regions: not only inside films [6] but also the most volume in nanoparticles. Present results are consistent with their findings from polarized neutron reflectometry and support their proposal. Indeed, as seen in Table I, M_s values of nanoparticles are mostly smaller than those of thin films. To dispel further confusion on the controversy about magnetization in α'' -Fe₁₆N₂, it would be better to discuss thin films and nanoparticles separately.

VI. SUMMARY

PND measurements were carried out to determine the sizes of Fe magnetic moments in α'' -Fe₁₆N₂ nanoparticles. On the basis of the good agreement between M_{PND} and M_{VSM} , it is concluded that M_{VSM} primarily originates from α'' -Fe₁₆N₂ so that a giant saturation magnetization is unlikely to occur at least for α'' -Fe₁₆N₂ nanoparticles as a macroscopic quantity. The possible microscopic coexistence of two types of spins, i.e., localized spins and itinerant electron spins, may be a more suitable hypothesis for the microscopic magnetism in α'' -Fe₁₆N₂ than simple band magnetism. We propose to discuss thin films and nanoparticles separately because of the difference in magnetism evolution.

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