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Modified Brillouin function to explain the ferromagnetic behavior of surfactant-aided synthesized α -Fe₂O₃ nanostructures

Zahra Alborzi Avanaki^{1*} and Ali Hassanzadeh^{2,3}

Abstract

The α -Fe₂O₃ nanoparticles have been synthesized using some surfactants in micro-emulsion method. Magnetization, $M(H)$, of samples was measured at room temperature. Magnetic hysteresis loops were observed in all samples. A new magnetization function ($H - A$ model) was offered to explain the field dependence of the magnetization behavior of α -Fe₂O₃. In this work, the magnetization curve of all samples was analyzed based on $M_{\text{Brillouin}}$, $M_{\text{Brillouin-Akulov}}$, and $M_{\text{Brillouin-(H - A)}}$ model. The experimental data have fitted with the $M_{\text{Brillouin-(H - A)}}$ model, and the best fitting parameters were analyzed.

Keywords: α -Fe₂O₃ nanoparticles, Ferromagnetic, Akulov law, Modified Brillouin function

Correspondence/Findings

Introduction

Fine ferromagnetic particles of a nanometer-scale size have important applications in various fields of modern nanotechnology, such as magnetic recording, permanent magnet industry, biomedical applications, etc. [1]. The continuum theory of micromagnetism, which was developed in the 1930s and 1940s, was intended to bridge the gap between the phenomenological Maxwell's theory of electromagnetic fields and quantum theory based on atomic backgrounds. In Maxwell's theory, material properties are described using global permeabilities and susceptibilities valid for macroscopic dimensions. On the other hand, quantum theory allows a description of magnetic properties on the atomistic level. A modern theory of micromagnetism has been born by Brown's attempt to explain the so far unexplained $1/H$ term in the law of approach to ferromagnetic saturation. The breakthrough toward a continuum theory of magnetism is due to Landau and Lifshitz (1935), who derived a continuum expression for the exchange energy and gave a first interpretation of domain patterns [2]. Ferromagnetic materials consist of small volumes of solid called magnetic domains in which atomic magnetic dipole moments of about 10^{12} to 10^{15}

atoms align parallel to each other spontaneously. The emergence of magnetization M may be due to a change in magnetic field strength H . Hysteresis loop proceeds in three main stages: initial reversible magnetization, rapid irreversible magnetization, and the slow approach to saturation, which they related, respectively, to the reversible shifts of domain walls, irreversible rotation and shift processes, and the reversible rotation of domains [3].

These stages are being visible in the insets of Figures 1, 2, 3, and 4. Much effort is being undertaken by different scientists in order to describe all magnetization curves. Mathematical approximations are given by the multicomponent exponential or sine-series functions [4]. Some other approximations result from the general descriptions of hysteresis, such as the Preisach [5], Jiles-Atherton [6], Takacs [7], and Akulov [8] models. Moreover, it is unlikely that any one theory can be developed that will apply to all materials.

It is usually taken for granted that the Brillouin function might describe the magnetization curves of many magnetic materials. In this work, an attempt to find an approximation based on the Brillouin function is undertaken. For this purpose, the following assumptions are made:

1. The description should have a phenomenological background.
2. Its parameters should have a physical meaning.

* Correspondence: zahra_alborzi@yahoo.ca

¹Department of Physics, Faculty of Science, Urmia University, Urmia, Iran
Full list of author information is available at the end of the article

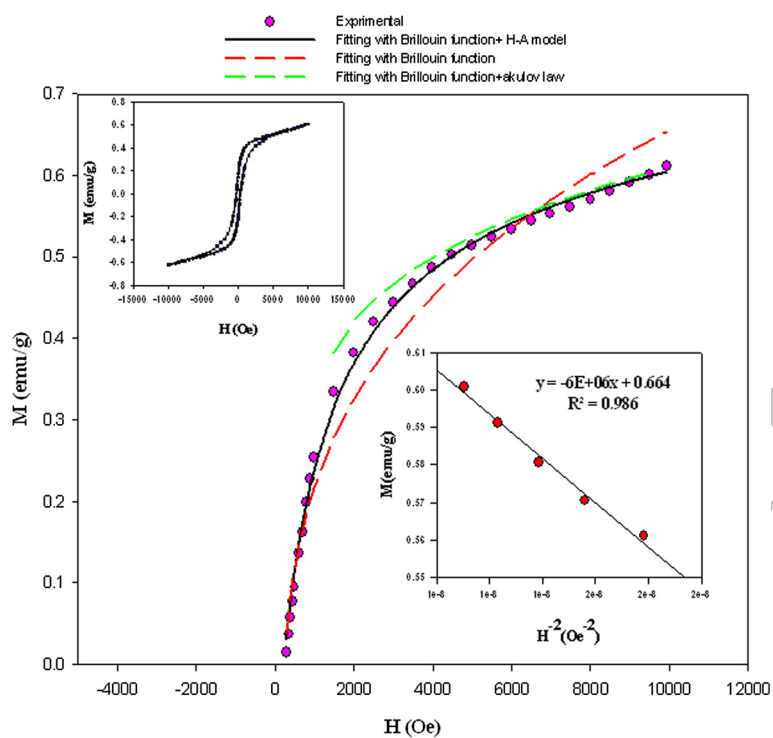


Figure 1 Fitting of experimental data using different equations for Fe_2O_3 (Merck).

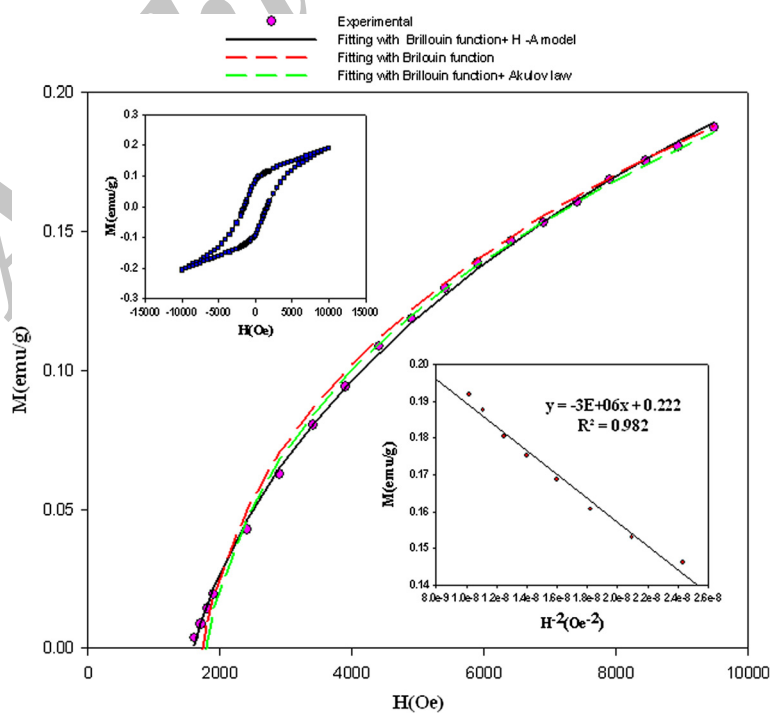


Figure 2 Fitting of experimental data using different equations for Fe_2O_3 (CTAB**).

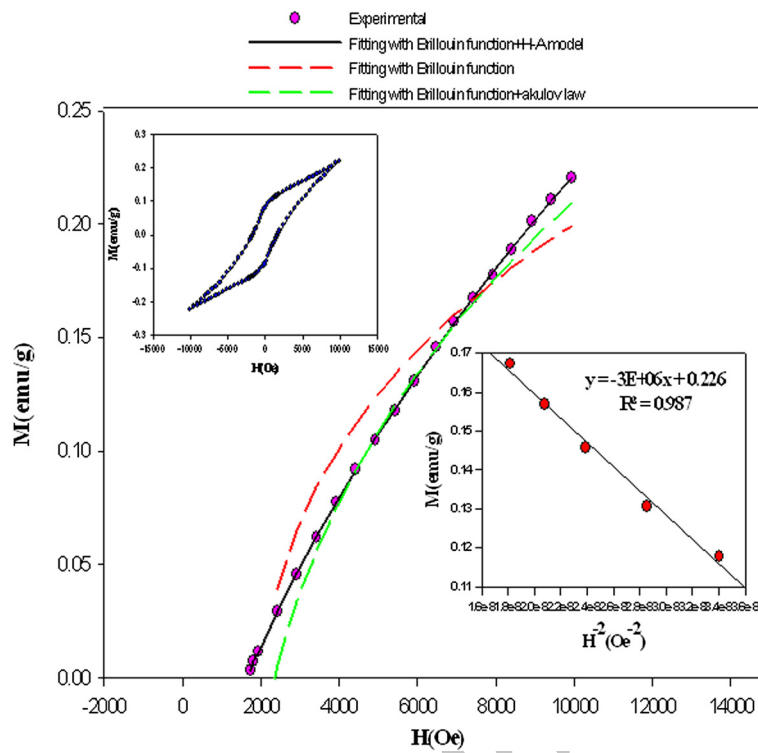


Figure 3 Fitting of experimental data using different equations for Fe_2O_3 (CPC).

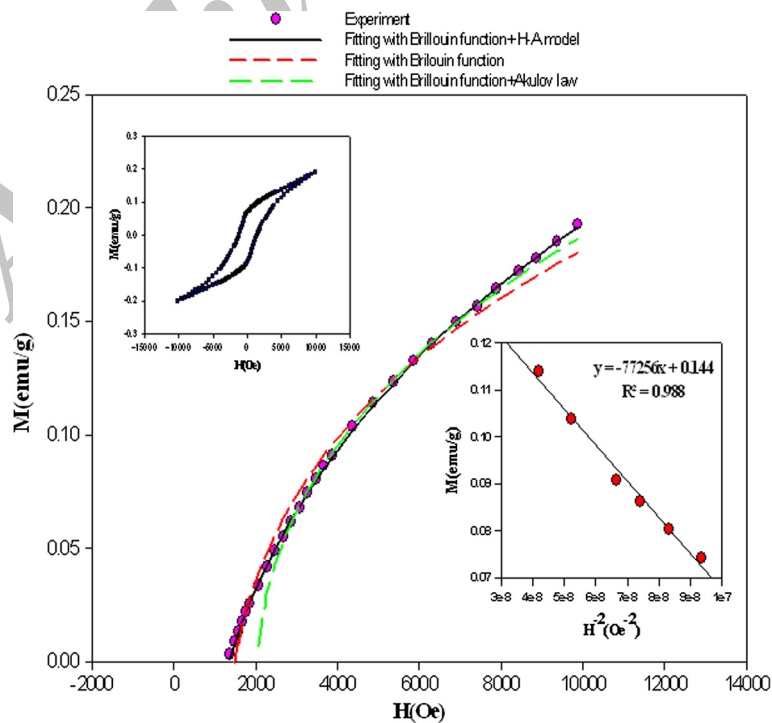


Figure 4 Fitting of experimental data using different equations for Fe_2O_3 (CTAB*).

3. The modified Brillouin function, chosen for modeling, should be verified using experimental data.
4. It is expected to achieve high accuracy in the region of wide range of magnetization changes.

For these purposes, the experimental data of α -Fe₂O₃ nanoparticles have been utilized such that their magnetization results were measured by vibrating sample magnetization (VSM).

Material and methods

The α -Fe₂O₃ nanoparticles have been prepared by micro-emulsion method in the system of water/toluene. The experimental procedure details of the synthesis of nanoparticles have been reported in [9]. In this method, there are two methods for synthesis of nanoparticles. In the first method, 25 ml of 0.1 M aqueous FeCl₃ and 160 ml of toluene were poured into a beaker. The solution was stirred, and cetyltrimethylammonium bromide (CTAB) was added as a surfactant. Then, maintaining stirring, 70 ml of 0.1 mol/l aqueous NaOH was added after the mixed system was stirred for 6 h. The first method was denoted for CTAB*. In the second method, 2 g of CTAB as surfactant was dissolved into 240 ml of CHCl₃ and 160 ml of CH₃CH₂CHOH. The mixture system was stirred for 30 min. Then, the transparent solution formed was separated into two parts. One was added with a certain volume of 0.1 mol/l FeCl₃ aqueous. The other part was added a certain volume of 0.1 mol/l NaOH aqueous. The two mixtures were stirred until they became transparent solutions; then, they were mixed by ultrasonic wave and stirred for 1 h. The second method was denoted for CTAB**. In the two methods, the α -Fe₂O₃ nanoparticles capped with surfactants were obtained by evaporating the solvent at a 350°C heat treatment under the protection of nitrogen. For cetylpyridinium chloride (CPC), as a surfactant instead of CTAB, just the second method was used.

Theory/calculation

A vibrating sample magnetometer (Meghnatis Daghigh Kavir Co., Kashan Kavir, Iran) with a sensitivity of 10⁻³ emu and magnetic field up to 10 kOe was utilized to evaluate the magnetic saturation and hysteresis loops of NPs at room temperature with field sweeping from -10 to +10 kOe. The magnetic field was changed uniformly at a rate of 20 Oe/s

(shown in Figures 1, 2, 3, and 4). Their VSM results are shown in Table 1.

The magnetic domain's spontaneous magnetization is due to the so-called Weiss mean (or molecular) field that aligns all of its atomic dipole moments along the same direction. This spontaneous magnetization appears essentially because of the exchange interaction processes between magnetic atoms.

In a real crystal, the spins of these electrons interact with each other, and the main contributions are the Coulomb interaction and the exchange symmetry for two electrons instead of the dipole-dipole magnetic interaction [10]. Modern quantum mechanics indicates that the existence of such fields is due to quantum mechanical exchange interactions among neighbor electrons with overlapping wave functions. The magnetic properties of certain ferromagnetic materials depend upon the direction in which they are measured. This phenomenon is called magnetic anisotropy as a result of which the magnetization of a ferromagnetic material is much easier accomplished along a particular crystal axis defined as the material's easy axis of magnetization [11].

Generally because the value of anisotropy energy is very small, it is difficult to find the real reason about its behavior. However, there are cases in which the anisotropy of the magnetization should be appreciable, and also, these values can be observed in Table 1. The closely associated problem of the anisotropy energy is at least qualitatively understood. Akulov [8], Keffer [12], Van-Vleck [13], and others have shown that the first anisotropy constant model of cubic materials falls from its zero-temperature value as the tenth power of the magnetization.

The contribution of both phases has been used to analyze magnetization curves. An ideal ferromagnetic sample would show a Brillouin function behavior, but a number of factors can influence the shape of a curve. Unfortunately, the effect of the distribution of particle size, demagnetizing effects, magneto-crystalline anisotropy, and magnetostriction must have caused the flattening of the ideal Brillouin curve and cannot easily have separated from the similar effect of a diamagnetic contribution. Recently, a theoretical investigation was presented, in which the correlation between magnetostriction and antiferromagnetic domain formation in the bulk and at surfaces was studied. The authors found

Table 1 VSM results for the synthesized magnetic nanoparticles

Sample	Surfactant	M_r (emu/g)	M_s (emu/g)	H_{c-} (Oe)	H_{c+} (Oe)	Size (nm)	M_r/M_s (dimensionless)	ρ (g/cm ³)	K_1 (erg/g)
Fe ₂ O ₃ (Merck)	-	0.177	0.548	-260	289	97.2	0.322	5.27	1,770.827
α -Fe ₂ O ₃	CPC	0.078	0.22	-1,683	1,726.5	54.4	0.224	4.98	1,091.2
α -Fe ₂ O ₃	CTAB*	0.073	0.192	-1,388.6	1,369.1	40.7	0.38	5.20	972.195
α -Fe ₂ O ₃	CTAB**	0.084	0.188	-1,591.5	1,508.8	35.3	0.446	5.26	892.53

that, similar to magnetic charges at ferromagnetic surfaces, 'elastic charges' at antiferromagnetic surfaces cause an 'elastic stray field,' which determines the domain structure [14].

The demagnetizing and Lorentz (dipole-dipole) field are omitted since their effect is small compared to the molecular field. The magnetization of the ferromagnetic materials in the spontaneous magnetization region is given by

$$M_{\text{Brillouin}} = N J g \mu_B F(J, y'), \quad (1)$$

where the $F(J, y')$ function is called the Brillouin function and is denoted by

$$F(J, y') = \left(1 + \frac{1}{2J}\right) \coth \left[\left(1 + \frac{1}{2J}\right) y' \right] - \frac{1}{2J} \coth \left[\frac{y'}{2J} \right]. \quad (2)$$

Also, the value of y' for ferromagnetic materials is given by

$$y' = \frac{J g \mu_B (H + \gamma M)}{k_B T}, \quad (3)$$

where J is the total electronic angular momentum, g is the Lande g value of the magnetic ion, μ_B is the Bohr magneton constant, N is the number of cation sites per gram, k_B is the Boltzmann constant, γ is the Weiss molecular field coefficient, and T is the temperature of ambient.

Equation 1 is in need of extension of $\coth(x)$ as follows: $\coth(x) = \frac{1}{x} + \frac{x}{3} - \frac{x^3}{45}$, in addition to that $x = \left(1 + \frac{1}{2J}\right) y'$ and $x = \frac{y'}{2J}$. The relation between the magnetization M and applied field H can be calculated by the solution of third-degree Equation 1. It is thought that by the computation of Equation 1, the form of Brillouin function is equaled to

$$M_{\text{Brillouin}} = \left[\frac{H}{\alpha \beta \gamma} + \sqrt{\frac{H^2}{4(\alpha \beta \gamma)^2} + \frac{1}{27} \left(\frac{1}{\alpha \beta \gamma} - \frac{1}{\beta} \right)^3} \right]^{\frac{1}{3}} - \frac{\left(\frac{1}{\alpha \beta \gamma} - \frac{1}{\beta} \right)}{\left[3 \left[\frac{H}{\alpha \beta \gamma} + \sqrt{\frac{H^2}{4(\alpha \beta \gamma)^2} + \frac{1}{27} \left(\frac{1}{\alpha \beta \gamma} - \frac{1}{\beta} \right)^3} \right]^{\frac{1}{3}} \right]}, \quad (4)$$

where $\alpha = \frac{N(g\mu_B)^2(J^2+J)}{3k_B T}$ and $\beta = \frac{(g\mu_B)^2(J^2+J+\frac{1}{2})}{9(k_B T)^2}$.

Equation 4 is fit by allowing the value for N , J , and γ as free parameters. It can be seen in Figures 1, 2, 3, and 4 that the magnetic data have not fitted well with the $M_{\text{Brillouin}}$ function. It is written that the Akulov law

corresponds to the magnetic saturation for poly-crystals and nanoparticles without exchange interaction between them. In the system of exchange-coupled particles, the magnetization curve is described by another power law [15]. Also, this law could be used for single-domain noninteracting nanoparticles [16]. It seems that to approach the best fitting of magnetic saturation curves of the investigated nanoparticles, we should use the Akulov law:

$$M_{\text{Akulov}}(H) = M_s \left[1 - \frac{1}{15} \left(\frac{H_a}{H} \right)^2 \right], \quad (5)$$

whereas H_a is the critical field related to nucleation processes. As the value of the critical field is reached, the spontaneous magnetization reverses. In the case of high-density magnetic materials, H_a is related to anisotropy constant, saturation magnetization, etc. [17], that has been followed by

$$H_a = \frac{2K_1}{M_s}, \quad (6)$$

where K_1 is the anisotropy energy constant and M_s is the saturation magnetization [1]. The K_1 value of samples was calculated, and it is shown in Table 1.

In a further study on finding the most appropriate formulation for fitting the experimental data, a combination of the Brillouin function and Akulov law was used:

$$M_{\text{Brillouin-Akulov}} = M_{\text{Brillouin}} + M_{\text{Akulov}}. \quad (7)$$

The H^{-2} term is probably the result of domain and imperfection effects. This dependence of power was observed for all magnetization curves. As you know, the behavior of ferromagnetic samples matched with Akulov law where $M \rightarrow H^{-2}$ was linear (shown in the right insets of Figures 1, 2, 3, and 4). In order to obtain a good correspondence between the theoretical and experimental hysteresis, loops are necessary to introduce new parameters in the magnetization equation. In this work, the following equation is considered:

$$M_{H-A \text{ model}} = \frac{AH}{B + H}. \quad (8)$$

One of the considered parameters is A , whereas the other one is named B . In Equation 8, which shall be referred to as the $H - A$ model, this situation can occur when the magnetic ions are distributed as sparsely dispersed large clusters and the magnetic moment density associated with an ion is of short range [18]:

$$M_{\text{Brillouin-(H-A) model}} = M_{\text{Brillouin}} + M_{H-A \text{ model}}. \quad (9)$$

The VSM results and fitting of their curves by Equations 4, 7, and 9 for the synthesized magnetic nanoparticles are shown in Figures 1, 2, 3, and 4.

Since the exchange interaction is of short range, the value of J is largest for nearest-neighbor spins. This tendency to align the nearest-neighbor spins parallel (for positive J) causes complete parallel alignment of the entire spin system, which results in ferromagnetism. This is equivalent to the assumption that the value of J is the same for all spin pairs, not just nearest-neighbor pairs [19]. The values of the parameters of Equation 9 are shown in Table 2.

It is thought that by comparing the results of Tables 1 and 2, we can almost consider the A parameter as M_S . With a short calculation, it is clear that this term is proportional to $\frac{1}{B+H}$. We know that the dimension of B parameter is similar to the applied field (Oe). It is thought that this parameter B is used for high magnetic field. The presence of this parameter B in Equation 8 is under investigation. According to the fitted value of A and B parameters (shown in Table 2), it is thought that this function behaves linearly with H , in the range of the applied field H in <1 to >150 Oe, and this function can be related to the Curie-Weiss law.

Findings and discussion

The hysteresis loop of the samples modified using CPC, CTAB*, and CTAB** evidences the ferromagnetic character of the materials, which can be categorized in the hard magnetic material group. The coercive force of powder ferromagnetic materials depends on the particle sizes and forms, but the dimensional dependence of H_c is very unusual [11].

In systems with very big multidomain particles, the coercive force increases with a decrease in particle size [20]; this case was not observed for the samples considered in this work because these samples were synthesized by several surfactants. The effects of the type of surfactant on coercive force were observed in many works [21,22], and its effect must be larger than the size effect. Furthermore, the hysteresis loops of the samples show a small asymmetry with a shift toward the negative field. This is due to the exchange interaction between the ferromagnetic and antiferromagnetic phase at their interface [23].

It is thought that the value of anisotropy energy constant K_1 is reduced by decreasing saturation magnetization M_S and increasing nanoparticles size [24].

According to the fitting of the experimental data with Equation 7, it was seen that there is not a good fitting

approximately in scope with $H < 4,000$ Oe. Thus, it is believed that the exchange interaction between the α -Fe₂O₃ nanoparticles was not observed in the range of the applied field H in <4 to >10 kOe.

Recently, a new approximation introduced by Zhang et al. rather than Akulov's approximation, which is more accurate, has been obtained to approximate analytical solutions for the nonlinear equations of magnetization, which were in good agreement with the numerical results [25], but they did not fit the experimental data with a theoretical curve similar to the one presented in this work.

A modified Brillouin function was used for explaining the ferromagnetic behavior of α -Fe₂O₃ nanoparticles. A good agreement was found between the experimental magnetization data and fitting curves by the $H - A$ model that it is given in Equation 9. According to the behavior of the samples fitting in the equations, the existence of crystalline phase, amorphous phase, and dislocation of elastic dipole is visible, and finally, the $H - A$ model term characterizes the presence of defects in materials by attention to Brown's theory. The continuum theory of micromagnetism was intended to bridge the gap between the phenomenological Maxwell's theory of electromagnetic fields and quantum theory based on atomic backgrounds; the theory of micromagnetism is an efficient tool to describe magnetization processes; it is thought that by comparing between the results of Tables 1 and 2, we can almost consider the A parameter as saturation magnetization M_S . With a short calculation, it is clear that this term is proportional to $\frac{1}{B+H}$. We know that the dimension of B parameter is similar to the applied field (Oe). It is thought that this parameter B is used for a high magnetic field. The presence of this parameter B in Equation 8 is under investigation. Approximately, this $1/H$ term results from small linearly extended elastic dipoles due to the agglomeration of vacancy type so-called free volumes [26], or on the basis of Brown's theory, it may have its origin in the presence of defects in samples: inclusions, voids, or simply inhomogeneities or lattice irregularities [27]. The molecular field coefficient γ of these samples takes reasonable value. In Weiss' original theory, the molecular field coefficient γ has a positive value, and it is perfectly well understood that the molecular field, having a finite and positive value when the atomic moments are all parallel and in the same direction, can cause this ordered state [28]. It seems that the high value of the quantum number J must be equivalent to the

Table 2 Fitting parameters using Equation 9

Sample	Surfactant	N (number/g)	J (quantum number)	γ (Oe/(emu/g))	A (emu/g)	B (Oe)	Accuracy of fitting
Fe ₂ O ₃	Merck	3×10^{15}	5×10^7	2.8	0.54	1,065.1	2×10^{-3}
α -Fe ₂ O ₃	CPC	4×10^{15}	7×10^7	0.17	1.04	42,043	6.5×10^{-6}
α -Fe ₂ O ₃	CTAB*	2×10^{16}	5×10^7	0.48	0.23	16,091.5	2×10^{-5}
α -Fe ₂ O ₃	CTAB**	5×10^{15}	4×10^7	1.9	0.17	12,133.9	6×10^{-5}

assumption that the value of J is the same for all spin pairs, not just nearest-neighbor pairs [19].

Conclusions

This paper considered the possibility of using the modified Brillouin function to explain the ferromagnetic behavior of surfactant-aided synthesized α -Fe₂O₃ nanoparticles. It was started with a magnetization measurement of samples with VSM. A theoretical framework based on modified Brillouin function which considers the effects of exchange interactions, ferromagnetic domains, elastic dipoles, dispersed particles, and defect within the sample was elaborated. On the basis of calculations, it can be stated that the Brillouin function combined with the rational fraction relationship might yield an appropriate description of magnetization curves. It follows from the analysis that the effects of elastic dipoles in the considered α -Fe₂O₃ nanoparticles are noticeable. As per this description, it may be useful both for practical and teaching purposes, and it has been used for other ferromagnetic materials. Also, researchers have been researching on the effect of the magnetic phases of magnetic materials on X-ray diffraction patterns.

Competing interests

The authors declare that they have no competing interests.

Authors' contributions

ZAA has carried out the experimental part, set up the experimental setup, analyzed the data, prepared the manuscript, and drafted the manuscript. AH has participated in the design of the experimental setup, analysis of the data, and manuscript preparation. Both authors have read and approved the final manuscript.

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Author details

- ¹Department of Physics, Faculty of Science, Urmia University, Urmia, Iran.
²Department of Chemistry, Faculty of Science, Urmia University, Urmia, Iran.
³Nanotechnology Research Center, Urmia University, Urmia, Iran.

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