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Anisotropic Temperature-Dependent Interaction of Ferromagnetic Nanoparticles Embedded Inside CNT

S. Prischepa[*,†,§] and A. Danilyuk[‡,¶]
[*]*Department of Information Security*
Belarusian State University of Informatics and Radioelectronics
P. Browka 6, 220013 Minsk, Belarus
[†]*Department of Condensed Matter Physics*
National Research Nuclear University MEPhI
Kashirskoe Highway 31, 115409 Moscow, Russia
[‡]*Department of Micro- and Nanoelectronics*
Belarusian State University of Informatics
and Radioelectronics
P. Browka 6, 220013 Minsk, Belarus
[§]*prischepa@bsuir.by*
[¶]*danilyuk@nano-center.org*

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We analyze the magnetization versus magnetic field curves of Fe-based nanoparticles embedded inside CNT. Measurements were performed at different temperatures and orientations of the magnetic field. We demonstrate that, for the parallel field the magnetic anisotropy dominates and the coherent anisotropy is of great importance at low temperatures. At high temperatures, the exchange coupling becomes stronger, but the coherent anisotropy still occurs. For the perpendicular field, the coherence anisotropy is absent, and the dimensionality of the system reduces to 2D. The results are discussed in the framework of the correlation functions of the magnetic anisotropy axes.

Keywords: Exchange interaction, magnetic anisotropy, magnetic nanoparticles, carbon nanotubes.

1. Introduction

Interaction of ferromagnetic (FM) nanoparticles (NPs) is determined mainly by the interplay between the exchange coupling and magnetic anisotropy. The matrix materials, in which NPs are embedded, can have a great impact on the overall properties of the nanocomposite. It has been established that carbon nanotubes (CNTs) can enhance both the exchange interaction and magnetic anisotropy depending on the sample's morphology and thus leading to the possibility to adjust and control the magnetic properties.[1–3]

In this paper, we analyze magnetization M versus magnetic field H, $M(H)$ dependencies, in a wide temperature range, 2–350 K, and both for parallel and perpendicular with respect to the CNT axis

orientations of the external magnetic field. Investigating the law of the approach to saturation (LAS), one can get useful information about a nature of magnetic interaction in a nanocomposite as well as extract the dimensionality d and correlation function of the magnetic anisotropy axes, $C(r)$.

2. Results and Discussion

Here we analyzed properties of vertically aligned arrays of CNTs with ferromagnetic NPs embedded inside inner CNT channels. The samples have been fabricated by chemical vapor deposition (CVD) using ferrocene $Fe(C_5H_5)_2$ as a source of iron-based NPs.[1]

The exchange coupling between FM NPs occurs when their average size R_c is less than the exchange correlation length R_f, $R_c < R_f$. The exchange correlations depend on the dimensionality of the system. This, in turn, reflects in LAS.[4] The phenomenological LAS can be estimated as:

$$\frac{\delta M}{M_S} = \frac{\{M(H) - M_S\}}{M_S \sim H^{-\alpha}}, \qquad (1)$$

where M_S is the saturation magnetization and exponent $\alpha = (4-d)/2$.[4] Measured $M(H)$ curves were analyzed according to Eq. (1) for $\alpha = 1/2$, 1, 3/2 and 2. It was obtained that such analysis does not provide an unambiguous answer about the dimensionality d. What is important, the field range $\{H_{\min}, H_{\max}\}$ of the validity of a certain exponent α depends on the temperature and orientation of the magnetic field. In Fig. 1 we plot both $H_{\min}$ and $H_{\max}$ for each exponent α in Eq. (1), versus temperature T for both orientations of the magnetic field.

From this result, it follows that, for parallel orientation, within LAS one can find an appropriate magnetic field range of the validity for each exponent α. There is general tendency of a crossover from $d = 3$ to $d = 0$ with the field growth exists, but it occurs via the series of crossovers with the intermediate dimensionalities $d = 1$ and 2. At $T < 100$ K, the region with $\alpha = 2$ dominates. This corresponds to the dominant role of the anisotropy.[4] At $T > 200$ K, the main region for LAS is described by $\alpha = 1/2$, which corresponds to strong exchange coupling and $d = 3$. For the perpendicular orientation $d = 0$, the dominant role of the magnetic anisotropy was not detected. At $T < 50$ K, the main region for LAS corresponds to $\alpha = 1/2$, whereas for $T > 50$ K $\alpha = 1$ ($d = 2$). Thus, we may conclude that the interaction between FM NPs inside inner CNT channels depends significantly on the orientation of the magnetic field and temperature.

To get deeper inside into the problem, we apply more general approach for the LAS analysis based on the conception of the real space correlation function of the magnetic anisotropy axes.[4] This approach allows obtaining important information about the mechanism of magnetic interaction between NPs intercalated inside CNTs: interplay between exchange coupling and magnetic anisotropy, role of the coherent anisotropy (CA) and its relationship with the random anisotropy and, finally, indication of the impact of the CNTs medium on the interparticle interaction.[4] The CA is introduced by substituting the external field H by $H + H_{CA}$ (H_{CA} being the CA field) in the case when the H_{CA} is aligned along the external one.[5] The explicit expression for LAS under these conditions depends

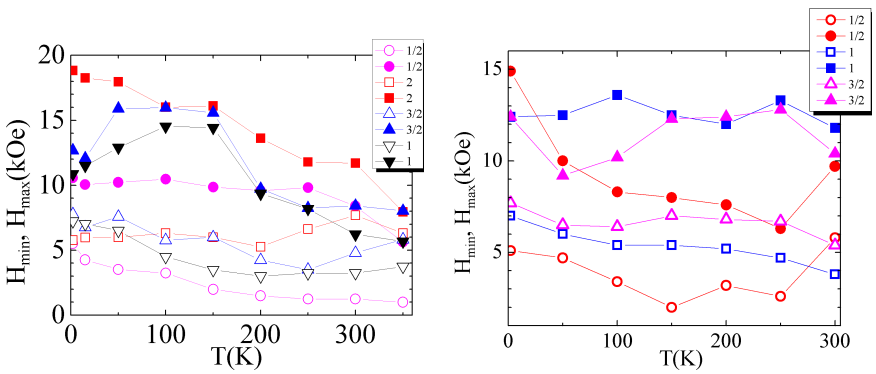

Fig. 1. $H_{\min}$ (open symbols) and $H_{\max}$ (closed symbols) versus temperature for parallel (a) and perpendicular (b) magnetic field. Numbers in the legend mean α values.

on the dimensionality of the sample. In particular, for $d = 2$ the magnetization approaches saturations as[4]

$$\frac{\delta M}{M_S} = \frac{1}{32}\left(\frac{H_{\rm ra}}{H_{\rm ex}}\right)^2\left(\frac{H_{\rm ex}}{H}\right)^{\frac{1}{2}} \times \int_0^\infty d^3x C(x)x^2 K_1\left[x\left(\frac{H}{H_{\rm ex}}\right)^{1/2}\right], \quad (2)$$

where $H_{\rm ra}$ is the random anisotropy field, $H_{\rm ex}$ is the exchange field, the coordinate x is normalized to $R_{\rm a}$, the length over which the magnetic anisotropy axes are correlated and K_1 is the modified Henkel function. For other dimensionalities, the general expression for LAS is[6]

$$\frac{\delta M}{M_S} = \frac{1}{30}\left(\frac{H_{\rm ra}}{H_{\rm ex}}\right)^2\left(\frac{H_{\rm ex}}{H}\right)^{\frac{1}{2}} \times \int_0^\infty d^3x C(x)x^2 \exp\left[-x\left(\frac{H}{H_{\rm ex}}\right)^{1/2}\right]. \quad (3)$$

Analyzing the experimental data with (2) and (3) it is possible to get the explicit form of the correlation function $C(x)$. The performed analysis revealed that for $\alpha = 2$ the correlation function (CF) is step-like:

$$C(x) = \frac{1}{\{1 + \exp[(x - x_{1/2})/2]\}}, \quad (4)$$

where $x_{1/2}$ is a coordinate at which the amplitude of the correlation function reduces to 1/2.

For $\alpha = 1/2$ the experimental $M(H)$ dependence close to saturation is better explained applying[2]

$$C(x) = b^{-1/2}x^{(\nu/2)-2}J_\nu[2b^{1/2}(x + x_0)^{1/2}], \quad (5)$$

where J_ν is the νth order Bessel function of the first kind, b and x_0 are constants.

Finally, for $\alpha = 1$, the CF for CNT-based magnetic nanocomposites is described by the following

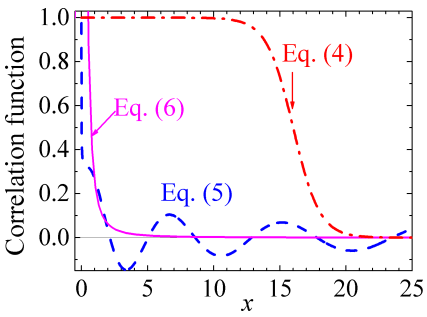

Fig. 2. Correlation functions as obtained analyzing the experimental data.[2]

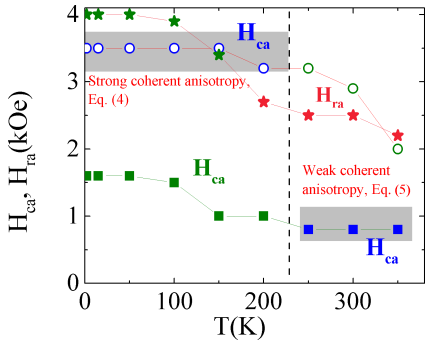

Fig. 3. $H_{\rm CA}$ versus T for CF calculated according to Eqs. (2) (upper curve) and (3) (lower curve). $H_{\rm ra}$ versus T is the same for both functions.

expression[3]:

$$C(x) = \frac{1}{x^2}\frac{1}{(x^2 - a^2)^{1/2}} \times \{[x + (x^2 - a^2)^{1/2}]^{2m} + [x - (x^2 - a^2)^{1/2}]^{2m}\}, \quad (6)$$

with constants $a = 0.01$ and $m = 0.07$. It should be noted that Eq. (6) is well approximated by simple $1/x^2$ dependence. The correlation functions obtained according to Eqs. (2)–(6) are depicted in Fig. 2.

As a result of the fitting procedure, the $M(H,T)$ dependences were explained. For the parallel field data,[2] the coherent anisotropy was important. Without the CA, it was impossible to fit these data. At low temperatures, at which $\alpha = 2$, the CF obeys Eq. (2). Whereas at high temperatures ($\alpha = 1/2$), the correlation function is described by Eq. (3). In Fig. 3, we plot the evaluated fields versus temperature for the parallel orientation.

Shadow areas in Fig. 3 mark regions where the mentioned correlation functions are valid. Despite higher values of the coherent anisotropy field for the correlation function described by Eq. (2), the better agreement is reached in the calculations with (3) at high temperatures. This means that the crossover from low dimensionality to 3D occurs at temperatures around 200 K. The real values of the coherent anisotropy magnetic fields drop significantly due to the thermal energy.

For the perpendicular field, the CA is absent and the $M(H)$ dependencies are described by two correlation functions depending on the field. For low magnetic fields, the $M(H)$ curve is better described by the CF according to Eq. (3), whereas for high fields the CF was calculated with Eq. (6), i.e., we observed the 3D→2D crossover with the field (Fig. 4).

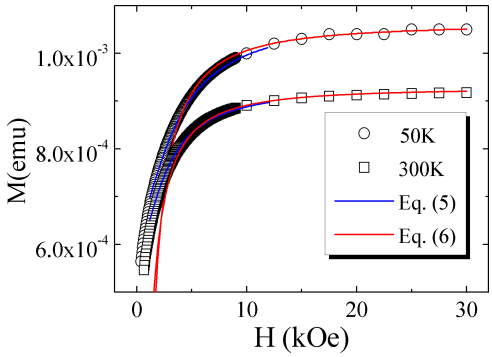

Fig. 4. Measured (symbols) and calculated (lines) $M(H)$ dependencies at two temperatures.

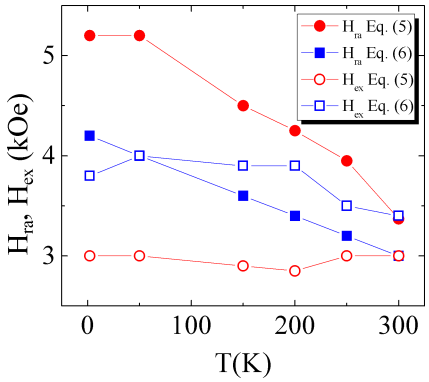

Fig. 5. $H_{\rm ra}$ (closed symbols) and $H_{\rm ex}$ (open symbols) versus T extracted from the fitting procedure.

In Fig. 5, we present the temperature dependencies of $H_{\rm ra}$ and $H_{\rm ex}$ as obtained applying these two correlation functions. It follows that at low temperatures the anisotropy dominates over the exchange interaction between NPs for both CFs. At high T, anisotropy still dominates for 3D case, whereas for 2D the exchange interaction is stronger. The latter could be caused by short range character of the correlation function determined by Eq. (6).

3. Conclusion

We have performed investigation of the magnetic properties of CNT-based nanocomposite in the temperature range, from 2 K to 350 K and for both parallel and perpendicular orientations of the external magnetic field. It has been demonstrated that analysis of the law of the approach to magnetic saturation is a very powerful technique for determining the interplay between exchange interaction and magnetic anisotropy. In particular, at low temperatures and parallel orientation of the external magnetic field, the magnetic anisotropy dominates. Moreover, the contribution of the coherent magnetic anisotropy is evident. The correlation function of the magnetic anisotropy axes is a step-like one with the range of correlations of tens of nanometers. At high temperatures, the coherence is still present, but the contribution of the exchange interaction dominates. The correlation function becomes a Bessel-like one with traces of a long-range magnetic anisotropy. The most likely reason of the serious loss of the coherence is thermal fluctuations, which destroy the long-range magnetic order.

For perpendicular fields, the contribution of the magnetic anisotropy becomes negligible in the whole investigated temperature range. As a result, the exchange interaction dominates. Thus, the ensemble of magnetic nanoparticles inside CNTs is described by two correlation functions, depending on the magnetic field. At low fields, the Bessel-like correlation function is valid, whereas at high fields, close to magnetic saturation, the CF is described by the $1/x^2$ dependence. In other words, the field-induced crossover from $d = 3$ to $d = 2$ is a significant feature of the perpendicular filed orientation.

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