

Regular article

Power-law behavior of coercivity in nanocrystalline magnetic alloys with grain-size distribution

A.S. Bolyachkin^{a,*}, S.V. Komogortsev^{b,c}

^a Ural Federal University, Yekaterinburg 620002, Russia

^b Kirensky Institute of Physics, Federal Research Center KSC SB RAS, 660036 Krasnoyarsk, Russia

^c Siberian State Aerospace University, Krasnoyarsk 660049, Russia

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ABSTRACT

Coercivity of nanocrystalline magnetic alloys depends on the grain size D according to a power law $H_c \propto D^n$ with n from 2 to 6. The law $H_c \propto D^6$ is derived based on the random magnetic anisotropy model and is clearly manifested in experimental studies of some Finemet type alloys. In this letter using computer modeling it is demonstrated that a power-law behavior with the exponent n less than 6 can be due to a grain-size distribution. An increase of grain size variance results in a decrease of the exponent from 6 to the value of about 3.

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Nanocrystalline alloys are of increasing interest as soft magnetic materials. The combination of an extremely low coercivity and a high magnetic permeability along with low eddy current losses makes them attractive for applications [1–6]. It was found that coercivity H_c in nanocrystalline alloys dramatically depends on the grain size D if $D < L_0 = \varphi\sqrt{A/K}$, where L_0 is known as a basic exchange length, φ is a dimensionless parameter of the order of one [7], A is an exchange stiffness constant and K is a magnetic anisotropy constant. A power law $H_c \propto D^6$ was derived using the random magnetic anisotropy (RMA) model [8,9]. It was confirmed on some Finemet type alloys and nanocrystalline Ni [1,3,10]. Accumulated to the present moment extensive experimental studies of nanocrystalline alloys yielded a generalized power law $H_c \propto D^n$, where the exponent n can vary from 2 to 6 [1,11]. Besides the grain size upper limit of L_0 for the $H_c \propto D^n$ applicability, there is experimentally revealed a lower one below which coercivity varies very slightly. The one order of magnitude variation in D within these limits results in the change of coercivity up to six orders according to the law $H_c \propto D^6$. For such steep dependence, a grain-size distribution should have a significant effect on the $H_c(D)$. A theoretical approach considering this effect was proposed in Ref. [12].

The fluctuations of magnetic anisotropy energy: $[(K)V_{ex}]^2 = \sum N_i (V_i K)^2$, where N_i is a number of grains of the volume V_i within the magnetic correlation volume V_{ex} , were analyzed. Assuming that $N_i = V_{ex}/V_i = (L_{ex}/D_i)^3$ and the grain size D_i is always smaller than the magnetic correlation length L_{ex} , the dependence $H_c \propto \langle D \rangle^6$ was obtained [13,14] as for the case of uniform grains. The variance of grain sizes affected only the value of the proportionality coefficient. Nevertheless, the influence of a grain-size distribution on coercivity of nanocrystalline alloys within RMA model is more complicated.

The number of grains within the magnetic correlation volume is $N = V_{ex}/V = (L_{ex}/D)^d$ for a system of exchange-coupled grains of an arbitrary dimensionality d [11,15–19]. The coercivity is finally expressed as follows [1,11,15–19]:

$$H_c \propto \frac{\langle K \rangle}{M_S} = \frac{K}{M_S} \cdot \left(\frac{D}{\sqrt{A/K}} \right)^{\frac{2d}{4-d}}, \quad (1)$$

where M_S is a saturation magnetization. Indeed, observed coercivities of nanocrystalline thin films and nanowires, which both have low-dimensional magnetization correlations, corresponded well to power-law dependencies (1) with the exponents of 2 ($d = 2$) and of 2/3 ($d = 1$) respectively [1,15,20,21]. For bulk materials ($d = 3$) the exponent of 6 is expected. Let us remind that Eq. (1) is valid if $D < L_0$. A grain-size distribution can lead to a violation of the requirement for

* Corresponding author.

E-mail address: anton.bolyachkin@urfu.ru (A.S. Bolyachkin).

some grains within the V_{ex} that results in $N = V_{ex}/V < (L_{ex}/\langle D \rangle)^3$ for the 3D case. This discrepancy can be resolved by the assumption of $N = (L_{ex}/D)^d$ with effective $d < 3$ if supposing the scaling approach is still applicable. Thus, in bulk nanocrystalline alloys the power-law behavior $H_c \propto D^n$ with $n < 6$ can be caused by a grain-size distribution. This work is focused on the study of this opportunity using compute modeling of major hysteresis loops of exchange-coupled polydisperse ensembles of grains with the random magnetic anisotropy.

An ensemble of close packed polyhedra was considered as a model of a polycrystalline alloy. It was being constructed in two stages. At the first one, a gradual pouring of spherical particles with a lognormal distribution of diameters into a container was performed using the molecular dynamic package LAMMPS [22]. After that, in order to eliminate edge effects of pouring, the central part of the ensemble was cut out and used further. It was close to a cubic shape and consisted of at least 62,000 particles. At the second stage, the radical Voronoi tessellation of the ensemble was done using diameters of particles as weights and taking into account periodic boundary conditions. This procedure was realized based on the VORO++ library [23] which allowed to obtain all required statistics for polyhedra (volumes, lists of neighbors, lists of contact areas, etc.). A cross-section of the ensemble created as described above is shown in Fig. 1 (a). It was able to tune polyhedra size distribution, which satisfied well the lognormal one (Fig. 1 (b)), varying the spheres size distribution at the first stage. Hereafter the quantity $D = \sqrt[3]{V}$ was chosen as a characteristic size of a polyhedron. Besides polydisperse ensembles of polyhedra, an arranged monodisperse ensemble of 64,000 rhombic dodecahedra ($40 \times 40 \times 40$) was considered [24].

For polydisperse ensembles the following statistics was obtained: a mean grain size $\langle D \rangle = \frac{1}{N} \sum_i^N D_i$, a volume-weighted mean grain size $\langle D \rangle_V = \frac{\sum_i^N \nu_i D_i}{\sum_i^N \nu_i}$, a standard size deviation $\sigma = \sqrt{\frac{1}{N} \sum_i^N (D_i - \langle D \rangle)^2}$ and a volume-weighted one $\sigma_V = \sqrt{\frac{\sum_i^N \nu_i (D_i - \langle D \rangle_V)^2}{\sum_i^N \nu_i}}$, where N is a number of particles and $\nu_i = V_i / \sum_i^N V_i$. The different types of averaging are relevant for both a theoretical analysis of modeling and its potential comparison with experiments (e.g. TEM and XRD provide $\langle D \rangle$ and $\langle D \rangle_V$ respectively).

It was assumed that each polyhedron has a uniform magnetization changing by the coherent rotation. This assumption, that leads us to the single-spin approximation [25,26], is valid if the interatomic exchange within small grains is sufficiently strong to ensure parallel spin alignment [27]. Exchange interaction between neighboring grains i and j was simulated as a direct Heisenberg-like one $E_{ex} = -J_{int} S_{ij} (\vec{\mu}_i \cdot \vec{\mu}_j)$ [24–26], where J_{int} is an intergrain interaction constant, S_{ij} is a contact area between adjacent grains and $\vec{\mu}_{ij}$ are unit vectors of magnetizations (the inset of Fig. 1 (a)). For the case of ideal grain interface and the simple cubic crystal lattice the following equalities can be established: $J_{int} = J/a^2 = A/a$, where J is an exchange constant and a is an atomic lattice constant. Actually, it is often assumed that $J_{int} \ll J/a^2$ [24–29]. The uniaxial magnetic anisotropy with uniform constant K and randomly oriented easy magnetization axes (EA) of grains was used in our modeling. Magnetostatic interaction between grains was neglected. Hence, the normalized energy of a grain i was calculated by the following:

$$\epsilon_i = -(\vec{\mu}_i \cdot \vec{n}_i)^2 - 2\vec{\mu}_i \cdot \vec{h} - \sum_{j=1}^{N_i} \frac{J_{int} S_{ij}}{KV_i} \vec{\mu}_i \cdot \vec{\mu}_j, \quad (2)$$

where \vec{n}_i is a unit vector, that is collinear to EA, $\vec{h} = \vec{H}/H_a$ is an external magnetic field reduced to the anisotropy one $H_a = 2K/M_s$, N_i is a number of neighboring grains and V_i is the grain volume. The list of constants for modeling was the following: $J_{int} = 1$ erg/cm², $M_s = 10^3$ emu/cm³ and $K = 10^6$ erg/cm³. Mean grain sizes $\langle D \rangle$ varied from 10 to 60 nm.

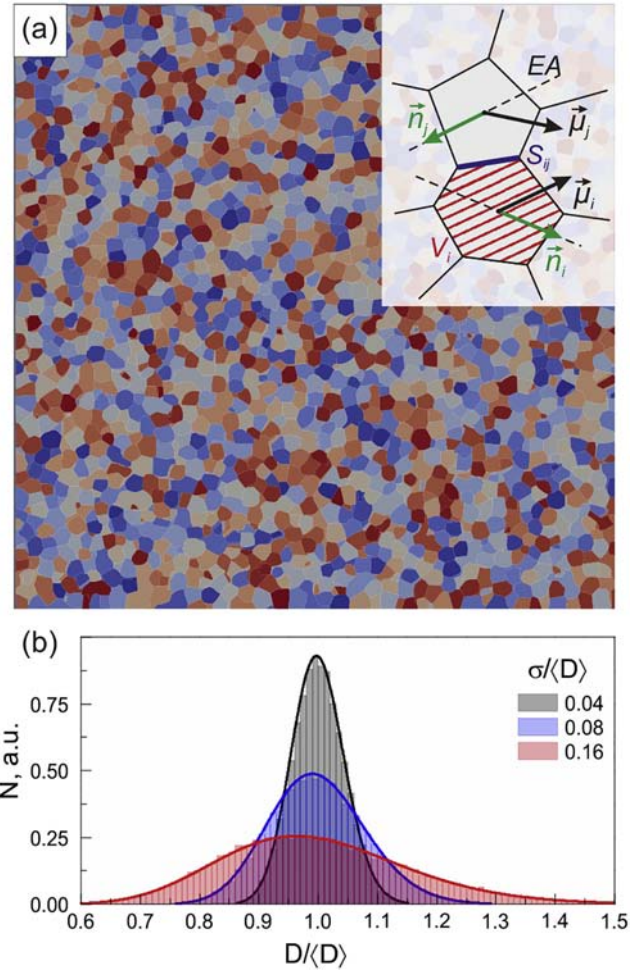


Fig. 1. A cross-section of a model polydisperse ensemble of grains (a). Its color pallet corresponds to projections of unit vectors \vec{n}_{ij} , that are collinear to easy magnetization axes (EA), on a normal of the cross-section plane. In the inset there is a 2D diagram illustrating quantities used for energy calculations (2): $\vec{\mu}_{ij}$ are unit vectors of magnetizations, V_i is a volume of grain i , S_{ij} is a contact area of adjacent grains i and j . Histograms of grain sizes D , determined as $\sqrt[3]{V}$, are presented for ensembles with different standard size deviations σ (b). Fits of lognormal distribution functions to histograms are given with solid lines.

Equilibrium micromagnetic structures of ensembles were obtained by reiterative subsequent energy (2) minimization for all grains at each magnitude of magnetic field [24,30]. Since in our model only adjacent grains interact with each other, the minimization was realized taking into account periodic boundary conditions for nearest neighbors only. During the procedure coordinates of vectors $\vec{\mu}_{ij}$ were determined with uncertainty of 10^{-9} ; the number of iterations was up to 10^3 . These parameters along with the number of grains provided the compromise between a computing time and an accuracy of results.

Major hysteresis loops of the monodisperse ensemble and a series of polydisperse ones with fixed $\sigma/\langle D \rangle$ were calculated using the described above technique. In Fig. 2 only segments of the hysteresis loops near coercivity are presented. The coercivity decreased and remanence increased with the decreasing $\langle D \rangle$. Demagnetization curves of polydisperse ensembles (Fig. 2, bottom; $\sigma/\langle D \rangle = 0.16$) were flatter than the ones of the monodisperse ensemble at large grain sizes (e.g. 60 nm).

Coercivities of both monodisperse $H_c(D)$ and polydisperse ensembles $H_c(\langle D \rangle)$ had close values approaching a limit (Fig. 3), which is the coercivity of the Stoner-Wohlfarth ensemble $H_c^{SW} = 0.479 \cdot H_a$

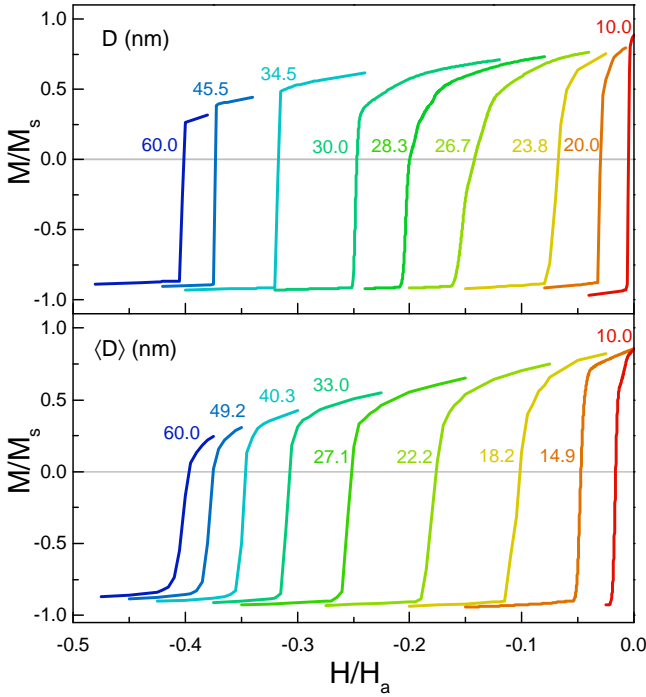


Fig. 2. Segments of major hysteresis loops of the monodisperse ensemble (top) and the polydisperse one (bottom; $\sigma/\langle D \rangle = 0.16$) calculated for different mean grain sizes $\langle D \rangle$ that are noted in nm near corresponding curves.

[31,32], at large grain sizes ($D > 30$ nm). The sharp power-law decrease of coercivity $H_c \propto D^6$ of the monodisperse ensemble was observed in the narrow grain size range from 21 to 30 nm. An intersection of the H_c^{SW} and the D^6 trend was defined as the basic exchange length L_0 in our discretized model since its continuous-based form $\varphi\sqrt{A/K}$ was nontrivial to adapt. This L_0 value actually determines the grain size below which average exchange energy prevails over anisotropy one that provides the power-law coercivity regime of the monodisperse ensemble. Further changes of $H_c(D)$ with decreasing grain size ($D < 21$ nm) were smaller than ones suggested by the power law (Fig. 3). This region of small grain sizes, which typically has higher relative uncertainties [25,26], deviates

due to the effect of the modeling area finite size. With increasing number of grains in the monodisperse ensemble, D^6 coercivity trend was extended to lower grain size values.

Coercivities of all polydisperse ensembles exceeded the monodisperse one at $\langle D \rangle < 30$ nm and $H_c(\langle D \rangle)$ followed the power law $\langle D \rangle^n$ with exponents n lower than 6 in extended ranges of $\langle D \rangle$. Data for ensembles with $\sigma/\langle D \rangle = 0.02$ and $\sigma/\langle D \rangle = 0.16$ are shown in Fig. 3. For them power laws $H_c \propto \langle D \rangle^n$ with $n = 4.5 \pm 0.1$ and $n = 3.2 \pm 0.1$ were obtained respectively. Let us note that even a very small grain size standard deviation resulted in a significant decrease of the exponent from 6 and the bigger $\sigma/\langle D \rangle$ was the smaller n was. These power laws $\langle D \rangle^n$ located mainly in the intervals of grain sizes where all ones were lower than L_0 (color filled regions in Fig. 3). It was important also that observed power-law behavior was the same for both $H_c(\langle D \rangle)$ and $H_c(\langle D \rangle_V)$; there was just a parallel shift of coercivity toward the larger grain sizes if volume-weighted ones $\langle D \rangle_V$ were used instead of $\langle D \rangle$ (it is demonstrated in Fig. 3 (b) with triangle symbols). Based on this a recommendation of not mixing methods of grain size estimation can be given for experiments.

Described results cannot be explained using the mentioned analytical approaches [12–14]. To demonstrate it we put in Fig. 3 dashed curves that were calculated based on the expression following from Ref. [13]:

$$H_c(\langle D \rangle) = \left(\int_0^\infty \sqrt{H_c(D)} f(D) dD \right)^2 \quad (3)$$

where a grain-size distribution function $f(D)$ was chosen as a lognormal one with appropriate $\sigma/\langle D \rangle$ and $H_c(D)$ was the spline interpolation of the simulated result for the monodisperse ensemble. Let us note that Eq. (3) is valid only if $\forall D_i < L_0$, so corresponding regions were considered. Within them the dashed curves slightly deviate from the $H_c(D)$ but they are far from the corresponding numeric data. The exponent of $H_c \propto D^n$ for polydisperse ensembles is shown in Fig. 4 versus the reduced grain size standard deviation. It sharply decreased first and then it changed gradually reaching values of about $n = 3$ with increasing $\sigma/\langle D \rangle$. The dependence $n(\sigma_V/\langle D \rangle_V)$ based on volume-weighted quantities varied slightly within uncertainties in respect to $n(\sigma/\langle D \rangle)$.

Power-law behavior of coercivity $H_c \propto \langle D \rangle^n$ with the exponent n less than 6 was observed in experiments [1,33]. The most extensive discussions were dedicated to the frequent case of $H_c \propto \langle D \rangle^3$ [1,3,34,35]. A competition between random magnetic anisotropy and induced one, in which easy axis is uniform within volumes much

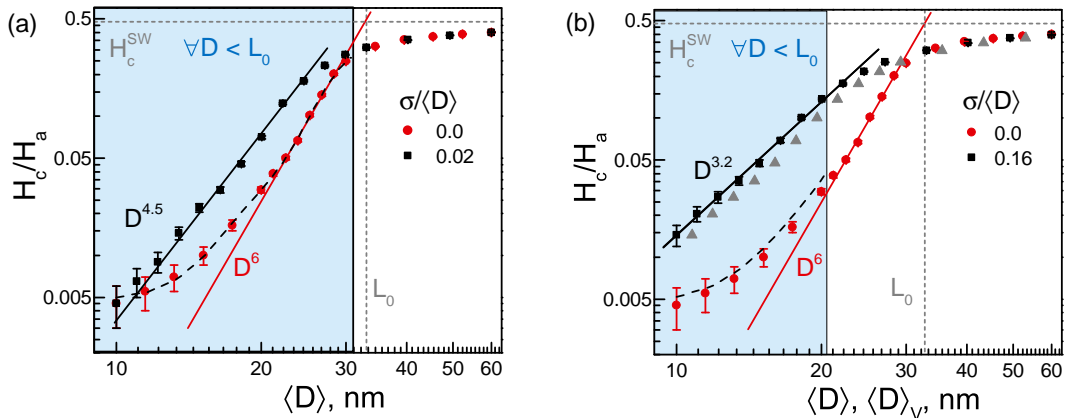


Fig. 3. Dependencies of reduced coercivity on mean grain size of the monodisperse ensemble ($\sigma = 0.0$) and the polydisperse ones with $\sigma/\langle D \rangle = 0.02$ (a) and $\sigma/\langle D \rangle = 0.16$ (b). Power laws D^n describing the dependencies are shown with solid lines. Triangle symbols (b) demonstrate the same coercivity dependence ($\sigma/\langle D \rangle = 0.16$) plotted vs. volume-weighted mean grain size $\langle D \rangle_V$. The level H_c^{SW} is a coercivity of the Stoner-Wohlfarth ensemble. L_0 is the basic exchange length. The color filled regions correspond to ones where all grain sizes are smaller than L_0 . Dash curves were calculated using numerically obtained $H_c(D)$ of the monodisperse ensemble in Eq. (3) with appropriate $\sigma/\langle D \rangle$.

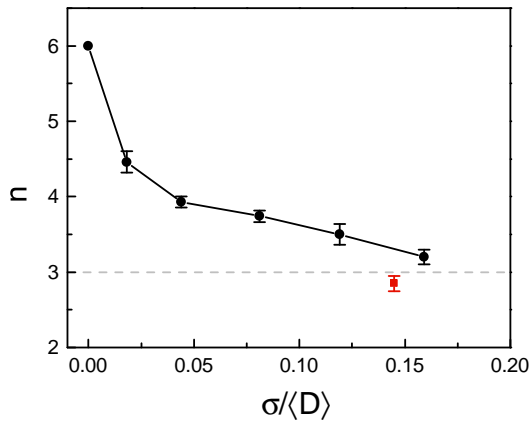


Fig. 4. The exponent n in coercivity dependence $H_c \propto \langle D \rangle^n$ of nanocrystalline ensembles with different grain size standard deviations (solid line is a guide for the eye). The result obtained for the ensemble of polyhedra generated based on random points in a cubic volume is marked with the square symbol.

larger than V_{ex} , was proposed as a reason for such behavior [34,35]. Indeed, an induced magnetic anisotropy appears frequently in real nanocrystalline alloys and therefore the explanation should be recognized as reasonable [1,3,34–36]. Presented numeric results bear that a grain-size distribution can also cause the behavior $H_c \propto \langle D \rangle^n$ with $n < 6$. The problem statement for computer modeling did not consider an induced anisotropy, so it did not contribute to the results. According to Fig. 4 the exponent $n \approx 3$ in nanocrystalline alloys can be due to the wide grain-size distribution with $\sigma/\langle D \rangle > 0.1$.

In addition, the exponent n can be influenced by the type of grain-size distribution. It was tested using hysteresis loop calculations for an ensemble of polyhedra, that was generated by a different algorithm than the described one, were performed. The ensemble was created using Voronoi tessellation based on an array of random points in a cubic volume. The grain-size histogram of this ensemble was described well by the normal distribution function. A $H_c(\langle D \rangle)$ was found to follow the power-law $H_c \propto \langle D \rangle^n$ with $n = 2.9 \pm 0.1$ (it is marked by the square symbol in Fig. 4). The result slightly but definitely deviated from the main series of $n(\sigma/\langle D \rangle)$ data obtained for ensembles with the lognormal grain-size distribution.

In summary, the major hysteresis loops of the monodisperse and polydisperse ensembles of exchange-coupled single-domain polyhedra with the random magnetic anisotropy was studied using micromagnetic modeling. Different mean grain sizes $\langle D \rangle$ and standard deviations $\sigma/\langle D \rangle$ of ones were used in the calculations. The power-law behavior of coercivity $H_c \propto \langle D \rangle^n$ was observed in certain grain-size ranges for all ensembles. There was a sharp decrease of the exponent n from 6 to about 3 with increasing $\sigma/\langle D \rangle$. A deeper understanding of reasons for such power-law behavior of coercivity would become an issue for further studies. We hope this work contributed to the problem of power-law behavior of coercivity in nanocrystalline alloys. It demonstrates that it is not only control of mean grain sizes that is essential for development of new nanocrystalline materials with the best soft magnetic properties but grain-size distributions in general.

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