

# Interacting magnetic nanoparticles as building blocks for miniaturized magnetic tags in anticounterfeiting strategies

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Miniaturized, unique, stable, and information-rich tags are crucial for anticounterfeiting efforts in the handling and distribution of consumable goods. Despite ongoing research into various tag technologies, a systematic approach to engineering their physical properties for optimal security is often missing. Through a numerical approach focused on in-depth modeling of the interacting magnetic nanoparticle properties, we demonstrate how their collective magnetic behavior, after suitable field conditioning, boosts the security of magnetic tags. Our miniaturized tag leverages the nonlinear response of assemblies of interacting  $\text{Fe}_3\text{O}_4$  nanoparticles to store custom information into binary keys. The magnetic properties of the nanoparticles and their spatial correlations result to be crucial to effectively decorrelate all keys from a large batch of tags produced under identical conditions. This ensures each tag encodes unique, virtually unclonable information, guaranteeing secure product identification, traceability, and quality control, enhancing consumer safety and fighting counterfeiting. The tag stability against transient disturbances of thermal and magnetic nature has been assessed. Our numerical analysis establishes magnetic nanoparticle tags as highly promising primitives characterized by high entropic information and security resilience to cloning, paving the way to better-engineered devices that harness the collective magnetic properties of nanoparticles.

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## I. INTRODUCTION

The demand for anticounterfeiting strategies that use secure, retrievable data to uniquely link information to both high- and low-value goods is rapidly increasing [1–7]. These strategies offer benefits such as enhanced traceability, improved quality control, and compliance with national regulations for food items [8,9]; they also ensure accurate dosage and transparency of pharmaceuticals [10–12], secure patient information of healthcare products [13], and robust identification, device data management, and anticounterfeiting security options for electronic and micro-electronic components [14–16]. According to the updated statistics, [17] the total impact of counterfeiting of consumer goods and commodities overcame 5 billion USD in 2024.

In today's mass production and packaging technology, a key objective is to customize each tag with unique and hard-to-clone information specific to a single item or packaging unit, thereby enhancing security and traceability.

Macroscopic variable data labels for Auto-ID [18,19] produced by advanced digital-printing techniques are an example of technical solutions to this challenge. In parallel, a variety of active and passive technical solutions have been introduced to meet the requirements posed by such a demand. They include, for instance, extensively used tools such as RFID (Radio Frequency IDentification) tags [20], one-dimensional and two-dimensional (1D and 2D) barcodes [21], cryptography algorithms based on serial numbers [22], and are characterized by different functionalities, costs, and areas of preferred application.

While macroscopic tags can be applied to large packaging, the interest in miniaturized labels is motivated by micrometric or submicrometric devices and chips that need unique identification in several fields, including connection of smart objects to the Internet [23], animal ecology [24], optical bioimaging [25], biodetection, and anticounterfeiting [26]. A variety of data-encoding methods, including physical unclonable functions [27–30], have been considered in the development of micro- and nanotags, including for instance optical [31–33], graphical [34], sequence [35], and magnetic- [36,37] encoding techniques. Various routes to obtain potentially unclonable tags using particles in the micrometer and submicrometer range have independently been explored in recent years. The common feature of these approaches is to exploit the spatial distribution of active (nonmagnetic) particles on the tag surface to

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generate a random binary key; in turn, the features of the spatial distribution of particles are inherently related to specific aspects of the process parameters involved in the fabrication of the tag. Stochastic deposition of micro-diamonds on silk fibroin films was shown to be able to produce a physically unclonable function (PUF) characterized by a unique optical response to challenge arising from a different positioning of active particles [38]. Similarly, stochastic agglomeration of approximately 100-nm zinc oxide particles in polymer films, controlled through engineered van der Waals interactions, results in the formation of random aggregates whose size critically depends on the concentration of the starting solution; such an effect was exploited to get a scalable photo-responsive PUF [39]. A different approach to producing unclonable, durable tags involves a regular distribution of 1- $\mu\text{m}$  colloidal particles on a properly indented planar surface, achieved by capillarity-assisted particle assembly; in that case the sought-after randomness emerged because the deposited particles were a random mixture of fluorescent and nonfluorescent units [40].

*Magnetic* random pattern tags exploiting the physical properties of magnetic nanoparticles are a promising alternative to the encoding of custom information linked to individual product units [41–43]. Other recently developed magnetic tags include ferromagnetic or paramagnetic macroscopic layers embedded in additively manufactured metals [44]. Generally speaking, magnetic tags offer the twofold advantage of being easily downscalable and able to guarantee uniqueness and security of the encoded information. The latter feature stems from the capability of magnetic nanoparticle assemblies to create random patterns, which cannot be replicated even knowing the encoding procedure [43]. Recently, magnetic markers containing  $\text{Fe}_3\text{O}_4$  nanoparticles have been proposed as labeling materials [45]; magnetic particles embedded in cellulose acetate films [46] or printed onto a nitrocellulose membrane [47] have been studied for tagging applications; magnetic tags obtained by self-assembling of magnetic nanoparticles combined with a hard polymer have been demonstrated [43]. Although experimental, application-oriented approaches are per se very interesting, they do not explore in full the potential of magnetic nanoparticles in this sector, so that in-deep knowledge of the factors that determine the encoding of custom data by magnetic nanoparticles is still lacking.

In this work, we demonstrate through numerical analysis and modeling how magnetic nanoparticles can be best exploited as building blocks of miniaturized tags with demonstrably individual fingerprints. Modeling has the double advantage of allowing a deeper understanding of the physical phenomena at the basis of the conceived implement and of paving the way to the fabrication of actual devices. This goal is addressed in this paper not only by studying in detail the magnetic behavior of the

particles themselves but also by analyzing the properties of the encoded information from the standpoint of encryption theory. Our aim is to demonstrate the feasibility of a miniaturized magnetic tag (MMT) with size in the micrometer range.

The proposed MMT is based on the magnetic properties of an assembly of core-shell nanoparticles comprised of a magnetite core in the magnetically blocked state surrounded by a diamagnetic shell, which are dispersed on a planar surface and whose interaction is dominated by magnetic dipolar coupling [48,49]. Magnetically blocked nanoparticles in the single-domain state are characterized by particularly high values of coercivity [50], so that they are able to store and stably retain information, which is hardly affected by temperature fluctuations and random magnetic fields.

We show that the collective magnetic properties resulting from interactions among nanoparticles ensure that, following proper binarization of the measured remanent magnetization, uncorrelated keys can be produced, even if the nanoparticle distribution on the surface of two or more tags produced by the same fabrication process happens to be correlated. In other words, encoding unique information in every single MMT is ensured by the physical effects arising from the magnetic properties of nanoparticles. In this way, a set of tags encoding custom information can be envisaged where the stored data, easily retrieved measuring an extensive magnetic property, are made resilient to malicious attacks owing to their intimate relationship with the properties of nanoparticles and their distribution on the nanometer scale.

The MMTs can be simply manufactured through a mass production process; they do not require any printed circuitry, nor electromagnetic, optical, or magneto-optical reading techniques, thereby avoiding common reading issues of RFID and optical tags [51–53]. The relevant information can be stored and retrieved using simple magnetic techniques. By leveraging the magnetic properties of nanoparticles identified in this work, miniaturized tags are expected to be particularly useful in marking small individual units produced on a large scale such as microelectronic components, and in the Internet of Small Things [54].

## II. OPERATION PRINCIPLE

The operation of the miniaturized magnetic tag (MMT) is ensured by the concurrence of three independent physical effects, which contribute to store information in a robust, reliable, and secure way:

- (a) the intrinsically nonlinear magnetic behavior of individual nanoparticles submitted to a magnetic field of sufficiently large amplitude [55,56];
- (b) an assembly of magnetic nanoparticles permanently attached to the tag's surface and randomly dispersed on

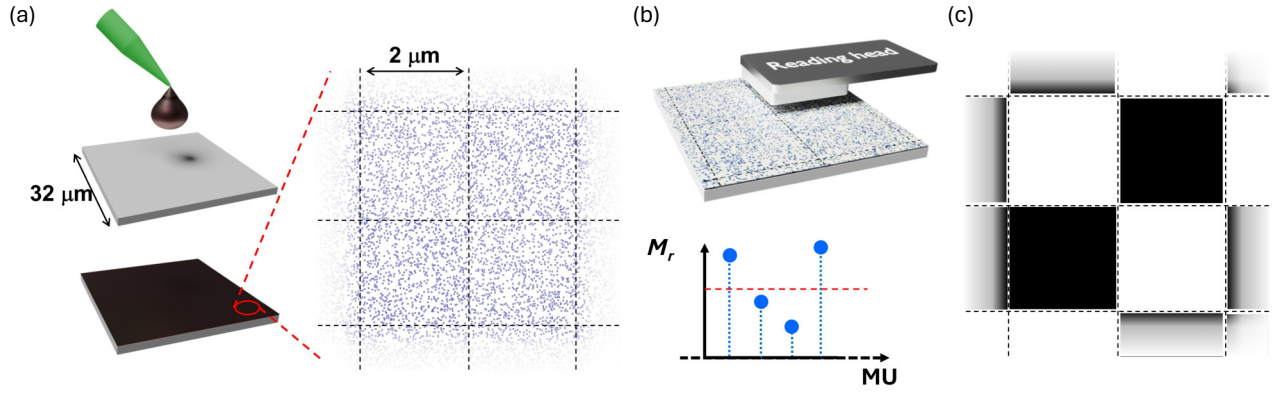


FIG. 1. (a) Principle sketch of the magnetic nanoparticle deposition on the tag surface, and magnified portion of the surface comprising four magnetic units [the blue circles represent the nanoparticles (drawing to scale)]; (b) principle sketch of the magnetic remanence reading technique, and random fluctuation of the magnetic remanence  $M_r$  in different magnetic units; (c) detail of the random checkerboard pattern obtained from magnetic data through a digitizing procedure.

it [a closeup of the computer-generated nanoparticle distribution in a small area of the tag's surface is shown in panel (a) of Fig. 1]. This distribution can be the result of a 2D printing process [57,58] based on, e.g., inkjet, aerosol, electrospray, drop-casting, layer-by-layer technologies [59–63]. These techniques allow to get large-area, fine-feature nanoparticle patterns on different substrates [57]; different conditions of the printing process can result in substantially different surface distributions of magnetic nanoparticles at the nanoscale [64]. A pictorial representation of the nanoparticle deposition is shown in panel (a) of Fig. 1;

(c) the presence of magnetic dipolar interactions among nanoparticles influencing the magnetic response of the nanoparticle assembly, as outlined in Sec. III and discussed in detail in Appendix A.

The magnetic signal from the deposited nanoparticles can be easily measured by interrogating the MMT through pickup techniques borrowed from the standard practice of magnetic recording (such as commercial read heads) [65,66], which typically do not require electrical or magnetic bias and whose size is in the submicrometer range [panel (b) of Fig. 1]. The magnetic signal is then converted into a binary (checkerboard) matrix, as sketched in panels (b),(c) of Fig. 1; the binarization can be done by exploiting a threshold applied to the measured remanent magnetization; such a threshold is the median of the remanent magnetization values, which ensures bit uniformity of the binary matrix [67].

### III. MODEL

In this section, the main features—both extrinsic and intrinsic—of the proposed MMT are summarized.

#### A. Geometry and size

The device is assumed to be a square of side  $L = 32 \mu\text{m}$  ( $=3.2 \times 10^4 \text{ nm}$ ). The background material of the tag must

be such that a thin layer of magnetic nanoparticles can be stably attached to the surface by the printing process; many present-day technical solutions are thoroughly reviewed in Refs. [57,58]. The surface of the tag is divided into 256 identical squares [henceforth called *magnetic units* (MUs)] of side  $B = 2 \mu\text{m}$  ( $=2 \times 10^3 \text{ nm}$ ) [see Fig. 1, panel (a)]. The choice of the number of MUs is justified by the trade-off between the need of having a sufficient long binary key to reduce the risk of key violation and the need of keeping the computational time (corresponding to the read time in a real-world scenario) as low as possible. A 256-digit binary key is compliant with the 2001 Advanced Encryption Standard (AES) [68]. The size of the MUs, corresponding to an areal information density of about  $0.16 \text{ Gbit/in}^2$ , is such that the magnetic signal originating from one MU can be easily picked up by a commercial read head, able to work up to areal densities of the order of  $1000 \text{ Gbit/in}^2$  [65,66,69]. The magnetic properties of a MU are assumed to be basically not affected by adjacent MUs, an approximation similar to that is usually done in the theory of magnetic recording [66]. The magnetic crosstalk between adjacent MUs is neglected. This approximation is justified by the fact that dipole-dipole interaction was shown to be negligible above an effective cutoff length corresponding to a particle-particle distance of about 3 times the particle diameter [70]; therefore, the effect of dipolar interaction at one MU's boundary (corresponding to an approximately 60 nm wide peripheral strip) is considered negligible with respect to the dipolar interaction present in the MU's core, where the main magnetic signal is generated.

#### B. Magnetic nanoparticles

The magnetic nanoparticles are assumed to be made of iron oxides (IONs). These nanomaterials are presently prepared in large quantities through cheap and well-established chemical techniques [71,72]. For simplicity,

TABLE I. Room-temperature magnetic properties and mean diameter of magnetite nanoparticles used in this work.

|                                         |                 |
|-----------------------------------------|-----------------|
| $\text{Fe}_3\text{O}_4$                 |                 |
| $M_s$ (emu/cm <sup>3</sup> )            | 350             |
| $K_{\text{eff}}$ (erg/cm <sup>3</sup> ) | $3 \times 10^5$ |
| $\langle D_m \rangle$ (nm)              | 22.5            |

particles made of pure magnetite ( $\text{Fe}_3\text{O}_4$ ) will be considered throughout; typical room-temperature values of their magnetic properties are given in Table I.  $M_s$  is the saturation magnetization of magnetite nanoparticles, usually lower than the bulk value because of compositional and structural defects as well as surface effects [73], and  $K_{\text{eff}}$  is an *effective* anisotropy constant comprised of a volume term related to the physical and geometric properties of the nanoparticles and a contribution from the nanoparticle surface [74].

In applications, the information stored on a tag must be preserved for a long time; therefore, superparamagnetic nanoparticles have to be discarded because they spontaneously lose magnetic information in a time of the order of 100 s [50]. The particles eligible for realizing a MMT must be in the single-domain, magnetically blocked state [75] at room temperature. In this work, the magnetic diameters  $D_m$  of magnetically blocked nanoparticles of magnetite are assumed to be narrowly distributed in size with diameters between 21 and 24 nm, according to a Gaussian law:

$$p(D_m) = \frac{1}{\sqrt{2\pi}\sigma_m} \exp\left[-\frac{(D_m - \langle D_m \rangle)^2}{2\sigma_m^2}\right] \quad (1)$$

with average value  $\langle D_m \rangle = 22.5$  nm and standard error  $\sigma_m = 0.5$  nm. The optimal choice for producing a MMT is to exploit *core-shell* particles comprised of a magnetic single-domain core surrounded by a thin diamagnetic layer (typical thickness  $t_{\text{dia}} \approx 1$  nm). The shells prevent the magnetic cores to touch, thereby avoiding the insurgence of strong contact interactions between particles, such as the exchange interaction [76]; the shell can be made of an inorganic compound such as silica or titania obtained by chemical processes compatible with the synthesis of the magnetic nanoparticles themselves ( $\text{Fe}_3\text{O}_4@\text{SiO}_2$ ,  $\text{Fe}_3\text{O}_4@\text{TiO}_2$ ) [77–79]. Core-shell magnetic nanoparticles are produced without difficulty and are available in large amounts [78,79]. The presence of a diamagnetic shell implies that magnetic dipole-dipole coupling [48,49] is the dominant residual interaction among particles. The mean value of the total particle diameter  $D = D_m + 2t_{\text{dia}}$  is therefore  $\langle D \rangle = 24.5$  nm. Such a distribution has been taken into account to better represent the real-world scenario where chemical synthesis of nanoparticles typically results in the inhomogeneity of their diameters. In the considered

range of particle sizes, the surface contribution to nanoparticle anisotropy has only minor effects on the value of  $K_{\text{eff}}$ , which is therefore considered to be constant in this work.

The particles are thought to be transferred from a ferrofluid [58,80] over the planar surface of the substrate by means of the printing process [57,58]. The thickness of the layer of deposited nanoparticles can typically be of the order of some tens of nanometers; considering that the total side of the device is  $L = 32$   $\mu\text{m}$ , the fabrication process can be tailored in such a way that the magnetic layer can be considered as a two-dimensional system. The volume concentration of nanoparticles in the ferrofluid is assumed to be such that the mean fraction of the tag surface covered by the core-shell nanoparticles is  $\langle f_s \rangle$ . This quantity is defined as the ratio of the area that results from the projection of all the deposited nanoparticles on the horizontal plane, divided by the total tag surface  $L^2$ . When the mean particle diameter is  $\langle D \rangle$ , the mean surface density  $\langle d_s \rangle$  of deposited nanoparticles is related to the surface fraction by

$$\langle d_s \rangle = \frac{4\langle f_s \rangle}{\pi \langle D \rangle^2}. \quad (2)$$

It is henceforth assumed that  $\langle f_s \rangle = 0.1$ , a value compatible with the assumption of a 2D system and ensuring that the mean number of magnetic particles per MU produces a magnetic signal detectable by commercial read heads used in magnetic recording [65,66]. In the present case, the mean surface density of nanoparticles turns out to be  $\langle d_s \rangle = 2.12 \times 10^{14} \text{ m}^{-2}$ , so that the mean number of core-shell particles per magnetic unit of the tag is expected to be of the order of one thousand; more precisely,  $\langle N \rangle = [\langle d_s \rangle B^2] \approx 848$ . In a MMT, the local number  $N$  of magnetic nanoparticles is expected to vary from MU to MU around  $\langle N \rangle$ . The random variable  $N = [d_s B^2]$  is presumed to fluctuate in accordance to the Poisson statistics [81,82] with standard error  $\sigma = \langle N \rangle^{1/2}$ , taking therefore values between  $\langle N \rangle \pm 3 \langle N \rangle^{1/2}$ . In the present case,  $\langle N \rangle^{1/2} \approx 29$ , so that the surface fraction  $f_s$  covered by nanoparticle fluctuates in different MUs around the mean value  $\langle f_s \rangle$  with standard error  $\sigma_f = \langle N \rangle^{1/2} \pi \langle D \rangle^2 / 4B^2 \approx 3.2 \times 10^{-3}$ . Therefore,  $f_s$  is expected to vary around  $\langle f_s \rangle = 0.1$  between the limits  $f_s \approx 0.09$  and  $f_s \approx 0.11$ .

The deposition process—as accurately controlled as it can be—introduces by its own nature a nondeterministic distribution of magnetic nanoparticles on the tag surface. The printing process is influenced by various factors—both of physical and chemical nature—so that an intrinsic randomness of their positions on the tag’s surface is naturally created. Directly measuring the position of each particle on the surface would result in a potentially unclonable key, as recently demonstrated in an analogous case by point-to-point identification through computer vision concept of randomly dispersed nanofeatures on a complex artificial

structure [83]. In the present case, point-to-point identification of individual nanoparticles with diameters of the order of 20 nm would require advanced imaging techniques based, e.g., on surface and transmission electron microscopy (SEM and TEM). The difficulty of generating a random key by direct particle observation can be overcome by properly exploiting the magnetic properties of the assembly of nanoparticles.

### C. Magnetic behavior of nanoparticles

The magnetic nanoparticles, once deposited on the tag's surface from the ferrofluid, lose all translational and rotational degrees of freedom. As a consequence, Brownian relaxation [84] is suppressed and the magnetization of the system evolves via Néel relaxation only [84,85]. In these conditions the nanoparticles are suitably described as double-well systems (DWSs) where the populations of each potential well changes with time according to magnetic rate equations [86,87]. The magnetic behavior of a set of nanoparticles is explained in terms of the changes in the populations of each potential well by effect of temperature and magnetic field [85]. A detailed treatment of the rate-equation approach applied to DWSs can be found in Appendices A 2 and A 3.

Each nanoparticle is characterized by a single easy axis of magnetization, corresponding to the minimum of the uniaxial magnetic anisotropy energy; in zero magnetic field the magnetic moment  $\mu$  of the particle is along one of the two directions defined by the anisotropy axis, corresponding to one of the two energy minima [see Fig. 2, panel (a)]. The direction of each magnetic moment is characterized by the Euler angles  $(\theta, \phi)$  with  $0 \leq \theta \leq \pi$  and  $0 \leq \phi \leq 2\pi$ . In the literature on magnetic nanoparticles, the effective magnetic anisotropy is usually considered to have uniaxial symmetry [88]. Although this is an approximation, it has proven to account for many experimental observations and provides an adequate picture of the main features of magnetic nanoparticles of interest in applications [89]. The uniaxial symmetry has been largely used even in the case of cubic-symmetry nanoparticles, implicitly assuming that effects related to nanoparticle surface and/or shape give a uniaxial contribution to  $K_{\text{eff}}$ , which dominates over the cubic term [74,89,90].

In general, the magnetic moment is reversed by an activated jump over the anisotropy energy barrier of the DWS. However, for magnetically blocked nanoparticles of magnetite with size in the above-defined interval of values, the relaxation time for spontaneous reversal of the magnetic moment at room temperature is very long, being of the order of hundreds of years for a particle of magnetic diameter  $\langle D_m \rangle$ ; therefore, jumps above the energy barrier are forbidden [panel (b.1) of Fig. 2].

The height of the barrier of a DWS is determined not only by the intrinsic parameters of the nanoparticle (such

as the effective anisotropy constant and the particle size), but also by the dipolar interactions among all nanoparticles in a MU. In our magnetic model, such an interaction brings about an increase of the barrier between energy minima of all nanoparticles [70,91] (see Appendix A 1). Dipolar interaction will be shown to play a central role in the operation of the tag because it is very sensitive to the degree of inhomogeneity of the distribution of a set of nanoparticles on a plane.

After nanoparticle deposition, the magnetic moments in all MUs of the tag are expected to point with equal probability along all directions on the unit sphere, because the printing process is not done under a magnetic field nor is able to induce an anisotropic distribution of the easy axes. The resulting magnetization of the whole tag as well as the one of each MU is therefore equal to zero.

### D. Conditioning process

The information is stored in the tag by submitting the as-prepared device to a simple, easy *conditioning process* at room temperature. A constant, uniform magnetic field of suitable amplitude is applied to the tag for a suitable conditioning time  $t_c$ . In this work, the conditioning time and field are assumed to be  $t_c = 300$  s and  $H_c = 300$  Oe. The latter value is easily obtained in practice by using, e.g., a simple solenoid or a permanent magnet; the choice of  $t_c$  is explained by the result of the calculation shown in Fig. 8. The direction of  $\mathbf{H}_c$  defines the  $z$  axis in panel (a) of Fig. 2. Selecting other  $t_c$  and  $H_c$  values in the same range would not change the main conclusions of this paper.

The conditioning field modifies the energy landscape of all DWS in a MU. In particular, the energy barrier decreases for the nanoparticles whose magnetic moment  $\mu$  forms with  $\mathbf{H}_c$  an angle  $\theta$  higher than  $\pi/2$  [panel (b.2) of Fig. 2], so that the activated jump becomes much easier and the relaxation time for magnetic-moment reversal drops until it becomes comparable or less than the conditioning time. The  $\phi$  angle does not play any role in the process. As a consequence, the magnetic moments initially directed with an antiparallel component with respect to the field have a finite probability of reversing in order to reach the energy minimum characterized by lower energy [Fig. 2, panel (b.2)], corresponding to the lowest angle between  $\mu$  and  $\mathbf{H}_c$ . However, the effect is influenced by the fact that the probability of magnetic-moment reversal strongly depends on the height of the energy barrier of the DWS, which is proportional to the cube of the nanoparticle diameter [Eq. (A7) of Appendix A 1]. Therefore, the relaxation time governing magnetic-moment reversal,  $\tau_{\text{eff}}$  [Eq. (A11) of Appendix A 2], can be either longer or shorter than  $t_c$ , depending on the size of the considered nanoparticle [see panel (c) of Fig. 2 for a pictorial representation of the effect].

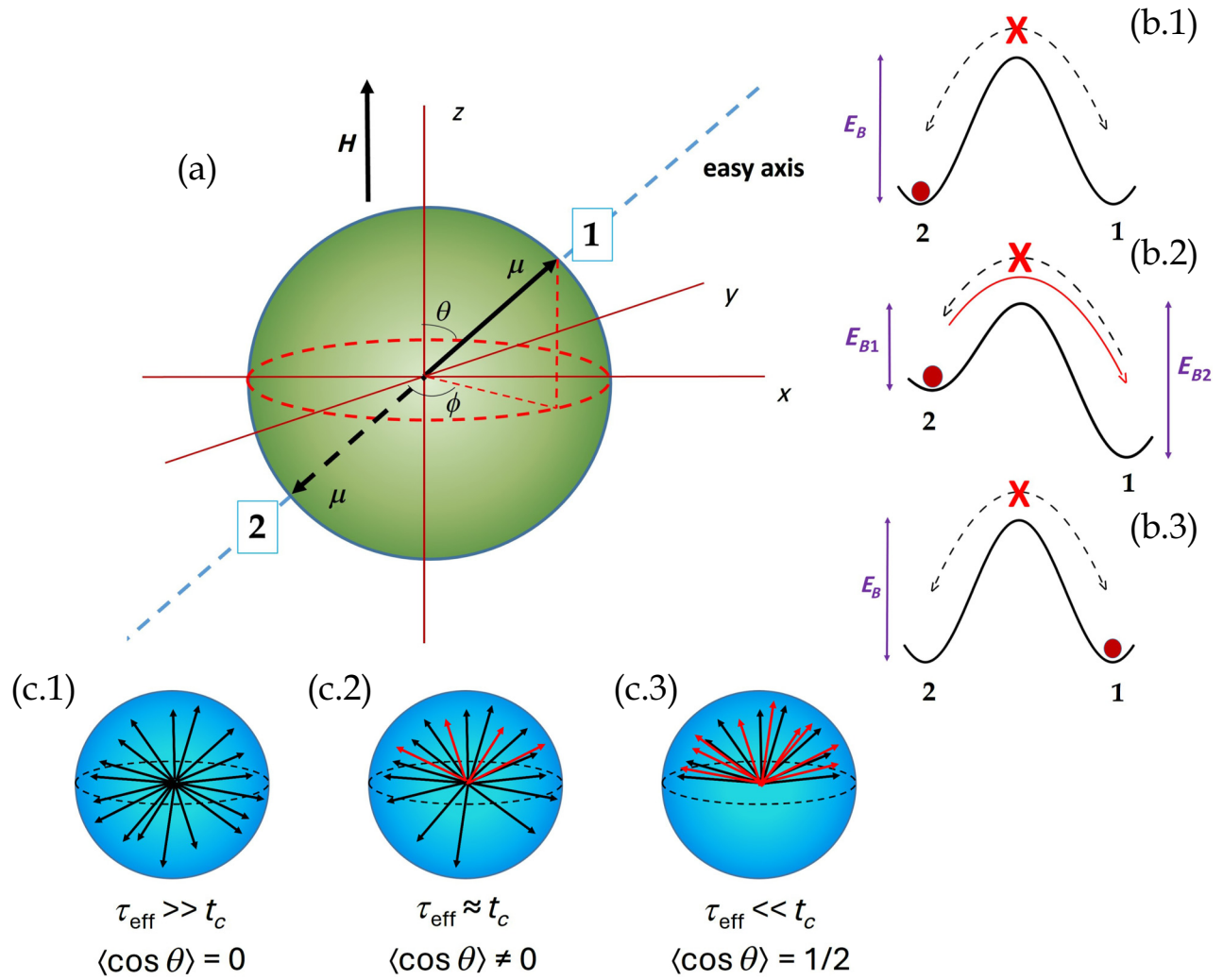


FIG. 2. (a) Euler angles defining the direction of a nanoparticle's easy axis on the unit sphere; at zero applied field, the magnetic moment  $\mu$  is along one of the two equivalent positions labeled 1 and 2; (b) sketch of the energy landscape of the DWS and of the allowed and forbidden activated jumps [(b.1) and (b.3), zero applied field; (b.2), field along the positive  $z$  axis]; (b) pictorial representation of the distribution of magnetic-moment directions during and after the conditioning treatment of duration  $t_c$ .

At the end of the conditioning process, the field  $\mathbf{H}_c$  is rapidly removed and each magnetic moment stays in the energy minimum where it was at  $t = t_c$ , because the nanoparticles are again magnetically blocked [panel (b.3) of Fig. 2]. As a result, the conditioning process induces a stable imbalance of the directions of the magnetic moments, with an increase of the number of moments with a component parallel to the  $z$  axis. The magnetization of each MU at zero applied field is now different from zero, i.e., each magnetic unit of the tag displays a magnetic remanence ( $M_r$ ). The remanent magnetization turns out to be simply expressed by  $M_r = M_s \langle \cos \theta \rangle$  (see Appendix A 5), where  $M_s$  is the saturation magnetization of magnetite and the  $\langle \cos \theta \rangle$  average is a quantity no longer equal to zero (see Appendix A 4). Actually, this average can be shown to take values between 0 and 1/2 [Eq. (A18) of Appendix A 5], so that the magnetic remanence in

every MU is between 0 and  $M_s/2$  (approximately equal to 175 emu/cm<sup>3</sup> in magnetite). The magnetic remanence of each MU can be measured by means of a read head [65,66].

It is worth noting that the conditioning step, characterized by a constant and uniform field applied for a short time, can be easily integrated with the tag-printing process in order to achieve the mass production of macroscopically identical MMTs by means of a single controllable procedure.

### E. Microstates of different homogeneity

Each MU of a typical tag is characterized by a local number  $N$  of nanoparticles, which can be considered as a random quantity described by Poisson statistics and fluctuating around  $\langle N \rangle$ . It is useful to figure out how these  $N$  particles are actually placed inside the MU.

In the absence of interactions among particles during the deposition process, it is expected that the MU will be populated by the nanoparticles according to the laws of complete spatial randomness (homogeneous spatial Poisson distribution) [92,93]. However, attractive or repulsive [94] interactions of chemical and physical nature among nanoparticles could favor the tendency to form uneven patterns in the MU, no longer fulfilling the requisites for complete spatial randomness.

In the present model, it is assumed that the actual nanoparticle positions satisfy two requirements: (a) the nanoparticles have a finite size, so they are not to be considered as ideally pointlike; (b) no superposition, even partial, of the projection of particles on the plane of the MU is possible. In other words, the nanoparticles can be viewed as hard-core disks in the plane, in line with 2D statistical approaches where particles are rigorously assumed to be randomly distributed or clustered on a plane (planar random processes) and where particle superposition is formally excluded [95,96].

A typical example of a computer-generated distribution of  $N$  particles inside a MU is given in panel (a) of Fig. 3 (top left). In this particular case, each particle was placed completely at random inside the square of side  $B$ , with the sole constraint of respecting the requirements of the previous paragraph; in the example, the particle diameters are distributed around the mean value  $\langle D \rangle = 24.5$  nm according to Eq. (1).

It can be shown that a particle pattern generated in this way complies with the conditions of complete spatial randomness on the plane. To this aim, a set of 25 independent, random patterns containing the same number  $N$  of particles were generated by the computer, and the distances between all  $(i,j)$  pairs of particles in the set were measured and counted, thereby obtaining a frequency density function  $\tilde{f}_L(d_{ij})$ . The curve drawn in black symbols in panel (b) of Fig. 3 is the average of this function obtained from the 25 independently generated patterns; the total number of points is  $[N \times (N - 1)/2]$ . Such a function takes values between the minimum and maximum distances between particle centers in the MU (i.e., 21 nm and  $\sqrt{2}B^2 \approx 2828$  nm, respectively), and shows a broad maximum around 960 nm. The full red line in the same panel represents the theoretical prediction for the probability density function (PDF)  $f_L(d_{ij})$  for a homogeneous point Poisson process on a square of side  $B$  [97], whose explicit analytical expression is given in Appendix B. The excellent agreement between the calculated frequency density function and the theoretical PDF shows that complete spatial randomness is indeed achieved by instructing the computer to place all particles at random in the MU.

In the case of weak interactions during the printing process, the final patterns of the deposited nanoparticles are assumed not to be characterized by particularly strong clustering effects (such as the ones described in terms of

Matern or Thomas clusters [96]). They are rather described in terms of an alteration of the pattern produced by a homogeneous spatial Poisson process; in other words, the pattern is assumed to be described in terms of an inhomogeneous Poisson process, characterized by a spatially varying intensity [96].

Different degrees of spatial inhomogeneity in a MU have been mimicked by generating patterns of hard-core nanoparticles distributed in size according to the usual rule. The variation with respect to a homogeneous Poisson process is obtained by building the pattern in a sequential way, making use of a single dimensionless *positional parameter*  $P$ , which takes positive values starting from  $P = 1$ . The nanoparticle pattern is generated as follows:

(a) in a void MU, a first nanoparticle ( $n = 1$ ) is placed by the computer in a completely random position; the diameter is chosen at random according to the prescribed distribution law;

(b) the diameter of particle 2 is chosen at random as before; the center of the second particle is then placed at a center-to-center distance  $d_{12}$  from particle 1 according to a radial Gaussian probability law centered on particle 1 with standard error  $\sigma_p = [P \times \langle D \rangle]$ ; therefore, the choice of  $P$  strongly influences the probability of placing the second particle at a shorter and larger distance  $d_{12}$  from particle 1; once this distance has been selected, particle 2 is placed around particle 1 with uniform probability with respect to the round angle on the plane, as sketched in Fig. S1 of the Supplemental Material [98]. Of course, the condition of nonsuperposition of particles has to be respected; otherwise, particle 2 will be discarded and another attempt will be made;

(c) the process is iterated every time a new particle is added to the pattern; that is, the center of the  $n$ th particle is placed according to the same law around the  $(n - 1)$ th particle. The process proceeds sequentially until particle  $N$  is successfully placed.

A small value of  $P$  favors the probability that the pattern obtained by the sequential algorithm is characterized by a higher unevenness, with regions where the particles are closer to each other with respect to the homogeneous case. This effect is shown by some examples obtained at decreasing values of  $P$ , displayed in panel (a) of Fig. 3. The frequency density functions for the patterns generated by the sequential process according to three values of  $P$  are shown in panel (b) of Fig. 3 (full symbols in color). For small values of  $P$  the pattern generated by the sequential algorithm is substantially different from the one obtained without imposing any particular constraints on the positions of the particles; with increasing  $P$ , the pattern becomes increasingly similar to the one characterized by complete spatial randomness; in fact, it can be shown that the frequency density function of the Poisson process

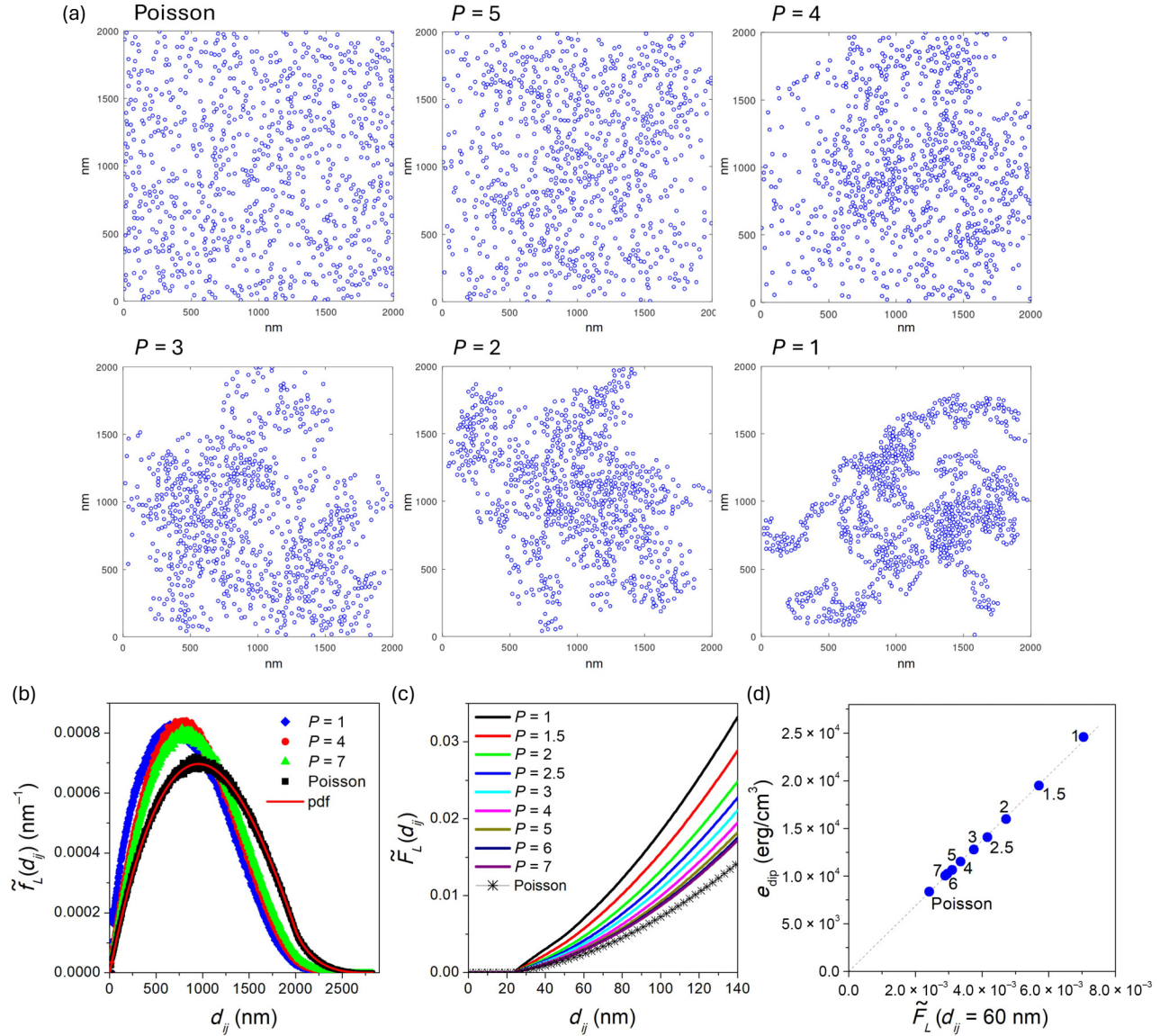


FIG. 3. (a) Examples of simulated nanoparticle patterns in a MU for different values of the positional parameter  $P$ ; the Poisson point process (top left) corresponds to the limit  $P \rightarrow \infty$ ; (b) frequency density functions of the interparticle distances  $d_{ij}$  in the MU for different values of  $P$  (full symbols) and theoretical PDF of the Poisson point process (red line); (c) CDF of the interparticle distances  $d_{ij}$  in the MU for various finite values of  $P$  (full lines) and for the Poisson point process (symbols); (d) linear dependence of the dipolar energy density of a MU as a function of the CDF [ $d_{ij} \leq 60 \text{ nm}$ ] for different values of  $P$  in the 1–7 range (full symbols) and for the Poisson point process.

corresponds to the limit  $P \rightarrow \infty$  (see Fig. S2 of the Supplemental Material [98]). In practice, a value of  $P$  of the order of 30 produces a nanoparticle pattern barely distinguishable from the case of complete spatial randomness. A comparison with the frequency density function of the Poisson process [black symbols in panel (b) of Fig. 3] shows how the average interparticle distance is affected by the positional parameter. In particular, the probability of finding low values of the distance  $d_{ij}$  increases with decreasing  $P$ , as demonstrated by panel (c), where the cumulative distribution function (CDF)  $\tilde{F}_L(d_{ij})$ —i.e., the

integrated occurrence frequency of interparticle distances shorter than a given value  $d_{ij}$ —is reported for low values of  $d_{ij}$ .

A key consequence of such a behavior, and the key effect for producing an efficient MMT, is the remarkable increase of the mean dipolar energy stored in a MU in dependence of the decrease of the positional parameter. As discussed in Appendix A 1 [Eqs. (A6) and (A7)], the energy density of dipolar nature  $e_{\text{dip}}$  is proportional to the quantity  $\langle 1/d_{ij}^3 \rangle$ , which is markedly sensitive to the actual distribution of interparticle distances. In fact,  $e_{\text{dip}}$  linearly increases with

increasing the fraction of short interparticle distances in the MU, as shown in panel (d) where the CDF for distances  $d_{ij} \leq 60$  nm is displayed on the horizontal axis. The points corresponding to all investigated values of  $P$ , including the one associated to the Poisson process, stay on the same straight line.

Therefore, a stronger tendency towards an inhomogeneous nanoparticle pattern (as parametrized by the positional parameter  $P$ ) brings about a linear increase of the energy barrier  $E_B$ , which can be written as

$$E_B = (K_{\text{eff}} + e_{\text{dip}})\langle V_m \rangle - \mu \cdot \mathbf{H}, \quad (3)$$

where  $K_{\text{eff}}$  is the effective magnetic anisotropy,  $\langle V_m \rangle$  is the mean magnetic volume of the particle,  $\mu$  is the particle's magnetic moment [see Eq. (A7) of Appendix A 1 for the complete expression of  $E_B$ ]. Such an increase modifies the response of the nanoparticles to the conditioning field, because the relaxation time for magnetic-moment reversal increases as well, affecting the value of the  $\langle \cos \theta \rangle$  average and the magnetic remanence  $M_r$ . A closely similar approximate treatment of dipolar interaction in the rate-equation framework was introduced and discussed elsewhere [70]. As a matter of fact, dipole-dipole interactions among magnetic nanoparticles are expected to give rise to local inhomogeneous arrangements of magnetic moments, which are in general very complex and inherently dependent on the angles between pairs of magnetic moments and with the line connecting any two moments. This type of effect is not taken into account by a change of the height of the DWS energy barrier experienced by any single nanoparticle, as assumed here [Eq. (3)]; therefore, the present approximation is to be viewed as a mean field approach to an intrinsic many-body problem, useful to reduce complexity and calculation time. A mean field theory is still a suitable way to describe a set of interacting magnetic nanoparticles when their number is as large as in the present case.

In particular,  $M_r$  increases with increasing  $P$ , i.e., with increasing the local *homogeneity* of the nanoparticle distribution, as shown in Fig. 4 for  $f_s = \langle f_s \rangle = 0.1$  (thick black line). In other words, the more the nanoparticle pattern is irregular, the lower will be the magnetic remanence.

As a matter of fact, the magnetic remanence of a MU depends on *two* independent parameters: the local fraction  $f_s$  of surface actually occupied by nanoparticles, which is a random variable taking values between 0.09 and 0.11 (as discussed in Sec. III B) and the degree of local inhomogeneity of the pattern, expressed by the positional parameter  $P$ , which is investigated here between the values 1 and 7 (for higher  $P$  values the magnetic remanence is almost not influenced by the positional parameter, as shown in Fig. 4). Therefore, the magnetic remanence of a MU is expected to take all possible values, which are inside the red dashed region of Fig. 4.

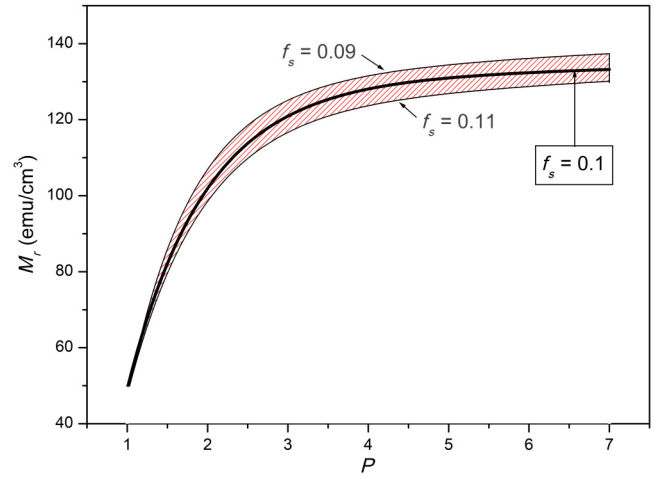


FIG. 4. Behavior of the magnetic remanence of a magnetic unit,  $M_r$ , as a function of the positional parameter  $P$  for three values of the surface fraction occupied by the nanoparticles,  $f_s$ ; the thick line is for  $f_s \equiv \langle f_s \rangle = 0.1$ , while the thin lines correspond to the assumed limits of the variability interval of the local  $f_s$ . The dashed red region encompasses all possible pairs of  $(f_s, P)$  values used in the simulations.

A key feature of this behavior is that two MUs with the same value of  $f_s$  can be characterized by significantly different values of the magnetic remanence by effect of a different degree of unevenness of the respective nanoparticle patterns.

#### IV. FEATURES OF THE MINIATURIZED MAGNETIC TAG

In applications where custom information is needed, the data stored by a single, randomly selected tag must be fully uncorrelated from the ones stored by any other tag of the same set. The aim is to deal with a set of tags in each of which the stored information is unique, i.e., where every tag has individual fingerprints.

To this end, in principle one could directly exploit the local surface density  $f_s$  of nanoparticles in each MU, which is indeed a random variable. However, this is not an optimal choice for at least two reasons: (a) the tag would work only by detecting the actual positions of all nanoparticles on its surface, a hard task unless advanced and expensive imaging techniques (SEM and TEM) are exploited; (b) even more relevant, the mass production of several tags through a serial printing technique (which should be basically operated under the same conditions of the physical and chemical process parameters) can result in a set of devices that retain a certain degree of unwanted positional correlation of the deposited nanoparticles (in other words, the values of  $f_s$  measured in the same MU of two randomly selected tags can be at least partially correlated).

We are able to show that when two or more MMTs are interrogated by reading the magnetic remanence of

their MUs, it becomes possible to associate to any tag a binary key demonstrably uncorrelated from the corresponding key of any other tag, allowing us to conclude that the information stored in each device of the set is really unique; this happens also when the values of the local surface density are partially correlated.

### A. Testing the operation of a MMT

The performance of the proposed MMT has been tested considering a set of  $N_D = 100$  devices characterized by the same  $\langle f_s \rangle$ . In each computer-generated tag the local  $f_s$  is a random variable following, in the  $Z = 256$  MUs, a Gaussian distribution law peaked at  $\langle f_s \rangle$  with standard error  $\sigma_{f_s} = 3.2 \times 10^{-3}$  (see Sec. III B). The probability that the positional parameter  $P$  takes a given value in a MU is assumed to be uniformly distributed within a preset interval of variation [in particular, the regions  $1 \leq P \leq 3$  (strong inhomogeneity of particle patterns) and  $3 \leq P \leq 6$  (mild inhomogeneity) have been explored].

Once the nanoparticle pattern of each MU has been generated using a randomly selected pair of values  $(f_s, P)$ , the magnetic remanence resulting from the conditioning treatment is obtained from Eqs. (A10), (A17), (A19) of Appendix A (see Fig. 4 also).

The  $M_r$  values corresponding to every MU of a single tag are shown in panels (a),(b) of Fig. 5 for two preset intervals of variation of the positional parameter  $P$ . In both panels, the median of the  $Z$  values is also shown (red dashed line). The fluctuation of  $M_r$  with respect to the median is much stronger when the positional parameter takes small values ( $1 \leq P \leq 3$ ) than in the case of mild unevenness of patterns ( $3 \leq P \leq 6$ ). This is expected because a marked inhomogeneity of the nanoparticle pattern enhances the fluctuation of  $M_r$ . In any case, even the weaker fluctuation of  $M_r$  observed in panel (b) [corresponding to a median absolute deviation (MAD) of  $2.70 \text{ emu/cm}^3$ ] can be easily measured by a standard pickup equipment or a commercial high-sensitivity magnetic head [99].

The key to the performance of the device is the measure of the magnetic remanence in all the MUs and the concomitant evaluation of the median of the set of the measured values. A *random checkerboard* representation of the magnetic remanence states in the MMT [see a sketch in Fig. 1, panel (c)] is obtained starting from the values of  $M_r$  measured in the  $Z$  magnetic units of the tag. The analog values of the magnetic remanence are digitized evaluating whether the value of  $M_r$  in the  $i$ th MU is above or below the median. In the first case, the value  $n_i = 1$  is attributed to the MU; otherwise, the value  $n_i = 0$  is taken. The checkerboard patterns, respectively, derived from the data shown in panels (a),(b) are shown in panels (c),(d) of Fig. 5; each MU is drawn white or black depending on the local value of the binary digit  $n_i$ . Comparing the remanence measured in each magnetic unit to the median ensures that

the checkerboard pattern is comprised of the same number of white and black squares.

A *binary key* can now be associated to every tag of the set. The  $i$ th element of the binary key of the  $m$ th tag,  $n_i^{(m)}$ , is equal to the digitized value of the magnetic remanence of the corresponding MU. In the present case,  $N_D$  binary keys are generated.

The proposed digitizing technique always results in binary keys containing an even number of elements; each key contains exactly the same number ( $Z/2$ ) of ones and zeros. Therefore, the maximum number of binary keys  $N_{BK}$ , which can be ideally generated is

$$N_{BK} = \frac{Z!}{(\frac{Z}{2})! (\frac{Z}{2})!} \approx \frac{2^Z}{\sqrt{\frac{\pi}{2}Z}}, \quad (4)$$

the second equality deriving from Stirling's approximation. When  $Z = 256$ , the ideal number of binary keys would be as large as  $N_{BK} \approx 6 \times 10^{75}$  (in the absence of correlations).

It is worth noting that the information encoded in the checkerboard pattern of a MMT by means of the conditioning process (see Sec. III D) is very stable with respect to temperature variations. A measure of the stability of the information stored in a MMT is the average value of the relaxation time for magnetization reversal in zero applied field,  $\tau_{\text{eff}}^0(D_m, T)$ , defined in Appendix A 2, Eq. (A12). The average is done at fixed temperature on the particle diameters with weighing function  $p(D_m)$  given by Eq. (1):

$$\overline{\tau_{\text{eff}}^0(T)} = \int_0^\infty \tau_{\text{eff}}^0(D_m, T) p(D_m) dD_m. \quad (5)$$

The behavior of  $\overline{\tau_{\text{eff}}^0(T)}$  as a function of absolute temperature is shown in Fig. 5, panel (e). It can be concluded that the stability of the information stored by the nanoparticles around room temperature is much longer than the expected lifetime of the magnetic tag.

We now study in detail the correlation degree between the random values taken by the digitized magnetic remanence  $M_r$  in magnetic units, both in the absence and in the presence of correlations of the local surface fraction occupied by nanoparticles,  $f_s$ . The latter quantity is a continuous random variable, so that the degree of correlation between local surface densities  $f_s$  of any two devices of the computer-generated set is studied using Pearson's linear correlation coefficient, as discussed in detail in Appendix C, where the possible correlation degree of  $f_s$  is parametrized by introducing the quantity  $\rho_{f_s}$ .

### B. Fractional Hamming distance between keys derived from magnetic remanence

We consider the set of  $N_D$  binary keys whose elements were generated starting from the values of the magnetic

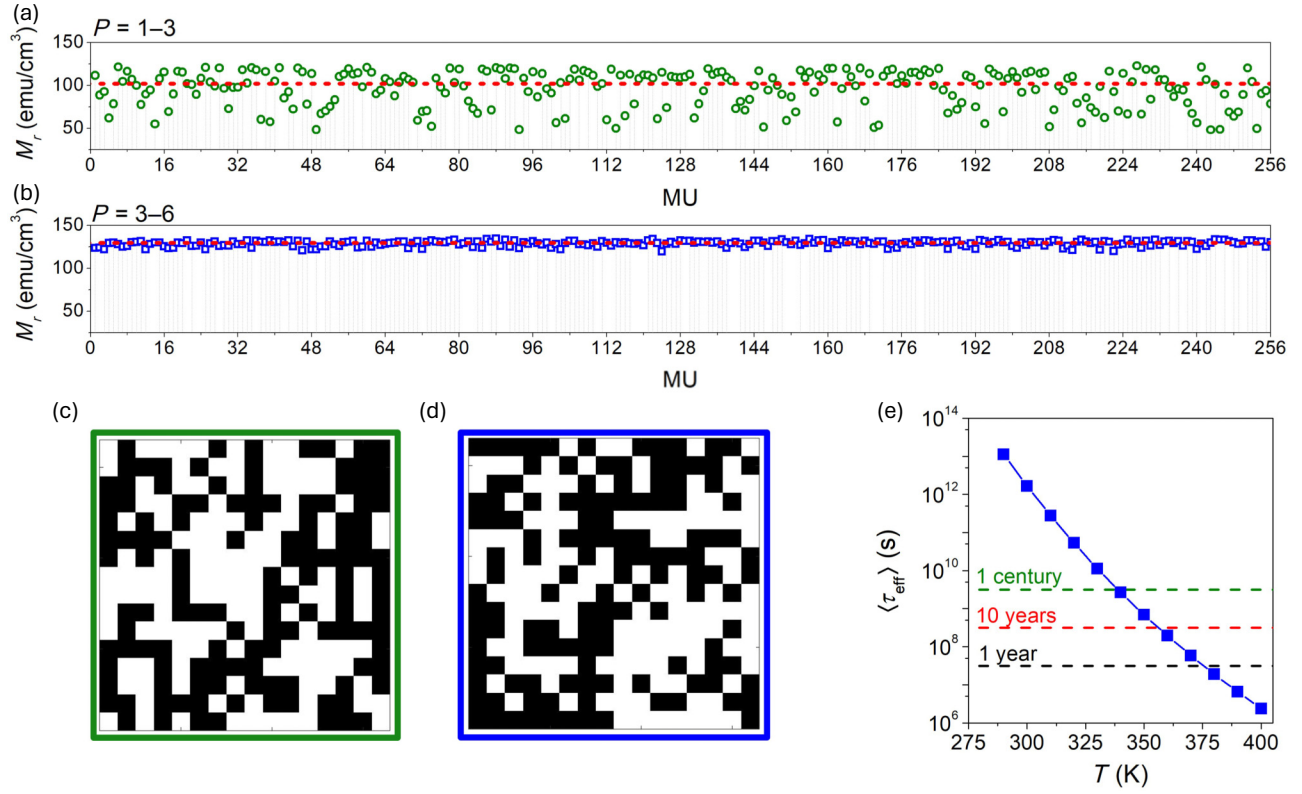


FIG. 5. (a),(b) Typical values of the magnetic remanence  $M_r$  found in the  $Z$  MUs of a randomly selected tag, for two intervals of variability of the positional parameter  $P$ ; (c),(d) random checkerboard patterns, respectively, resulting from the magnetic remanence data of panels (a),(b), according to the procedure outlined in Sec. IV A; (e) time stability of the information stored in the MMT around room temperature.

remanence of the MUs of each tag according to the procedure outlined in Sec. IV A. Taking any two keys  $(m, n)$  of the set a column vector is created, whose elements  $n_k^{mn}$  are determined by the condition:

$$n_k^{(mn)} = n_k^{(m)} \oplus n_k^{(n)}. \quad (6)$$

In other words, the  $k$ th element of the vector is in turn equal to 0 or 1 when the two corresponding elements of keys  $(m, n)$  are equal or different, respectively. The correlation coefficient  $h_{mn}$  is calculated according to the expression:

$$h_{mn} = \frac{1}{Z} \sum_{k=1}^Z n_k^{(mn)}, \quad (7)$$

the summation in this equation being the *Hamming distance* between the two vectors [100]. The quantity  $h \equiv h_{mn}$  (fractional Hamming distance, FHD) takes  $[N_D \times (N_D - 1)/2]$  discrete rational values between the limits 0 (corresponding to identical, i.e., perfectly correlated keys) and +1 (corresponding to perfectly anticorrelated keys).

For completely uncorrelated binary vectors, the quantity  $h$  is expected to be distributed around the probability  $p = \frac{1}{2}$  according to the binomial law:

$$\tilde{p}(h) = \binom{Z}{Zh} p^{Zh} (1-p)^{Z(1-h)} \quad \left( h = 0, \frac{1}{Z}, \frac{2}{Z}, \dots, 1 \right) \quad (8)$$

with standard error:

$$\left\langle \left( h - \frac{1}{2} \right)^2 \right\rangle^{\frac{1}{2}} = \frac{1}{2\sqrt{Z}}. \quad (9)$$

The normalized histograms representing the actual distribution of the FHD in the considered set are shown in blue color in panels (a),(b) of Fig. 6. When the positional parameter  $P$  is small ( $1 \leq P \leq 3$ ), i.e., in the presence of markedly inhomogeneous nanoparticle patterns, the histograms of the correlation coefficients of *all* computer-generated binary keys derived from magnetic remanence are superimposed, independently of the value of  $\rho_{fs}$ , i.e., of the degree of correlation of the local surface density, and well described in position and shape by Eqs. (8) and

(9) [red full line with dots in panel (a)], indicating that the binary keys derived from the proposed procedure are fully uncorrelated even in the presence of significant correlation between the values of the random variable  $f_s$ . Therefore, measuring the magnetic remanence is the key to deal with completely uncorrelated devices also in the case of tags characterized by a partial correlation between the local surface density of nanoparticles.

On the other hand, weaker inhomogeneities of the nanoparticle pattern, characterized by larger values of the positional parameter ( $3 \leq P \leq 6$ ), make the decorrelation operated by magnetism somewhat less effective, as checked in panel (b) of Fig. 6, where the cases corresponding to uncorrelated ( $\rho_{f_s} = 0$ ) and markedly correlated ( $\rho_{f_s} = 0.62$ ) surface densities  $f_s$  are compared. It can be noted that even when  $\rho_{f_s} \neq 0$  the histogram of the correlation coefficients  $h$  is well fitted both in position and width by the binomial distribution [Eq. (8)] with a suitable  $p < \frac{1}{2}$ .

In summary, in the case of stronger inhomogeneities of the nanoparticle pattern the binary keys turn out to be fully uncorrelated, as expected, while for mild inhomogeneity the mean  $p$  of the  $\tilde{p}(h)$  distribution is slightly displaced towards zero with respect to  $h = \frac{1}{2}$ , indicating that in the presence of surface-density correlation  $\rho_{f_s}$  a residual positive correlation between binary keys survives, although magnetism greatly reduces the degree of correlation. Such a behavior is observed for all positive  $\rho_{f_s}$  values in the explored range.

The effect of the surface-density correlation parameter  $\rho_{f_s}$  on the mean  $p$  of the  $\tilde{p}(h)$  distribution is shown in panel (c) of Fig. 6. In the case  $1 < P < 3$  the mean of the distribution is not affected by  $\rho_{f_s}$  and is always equal to  $\frac{1}{2}$ , whereas in the case  $3 < P < 6$  the mean  $p$  decreases almost linearly with  $\rho_{f_s}$ ; the observed reduction is however rather small.

Since the  $\tilde{p}(h)$  distribution derived from the computer-generated set of tags is always distributed according to a binomial law independently of the degree of surface-density correlation, the number of degrees of freedom associated to each binary key [101] turns out to be always equal to  $Z = 256$ , i.e., to the number of magnetic units in a tag. Such a result is intrinsically related to the method of extraction of the binary key exploited in this work, which does not take into account possible spatial correlations in the  $f_s$  distribution across adjacent MUs in the same tag.

The present results support the idea that significant local fluctuations of the aggregation degree of nanoparticles should be looked for in order to produce a set of MMTs characterized by demonstrably individual fingerprints.

### C. Stability of the MMT

Although a MMT is expected to work at room temperature in the absence of substantial external magnetic fields,

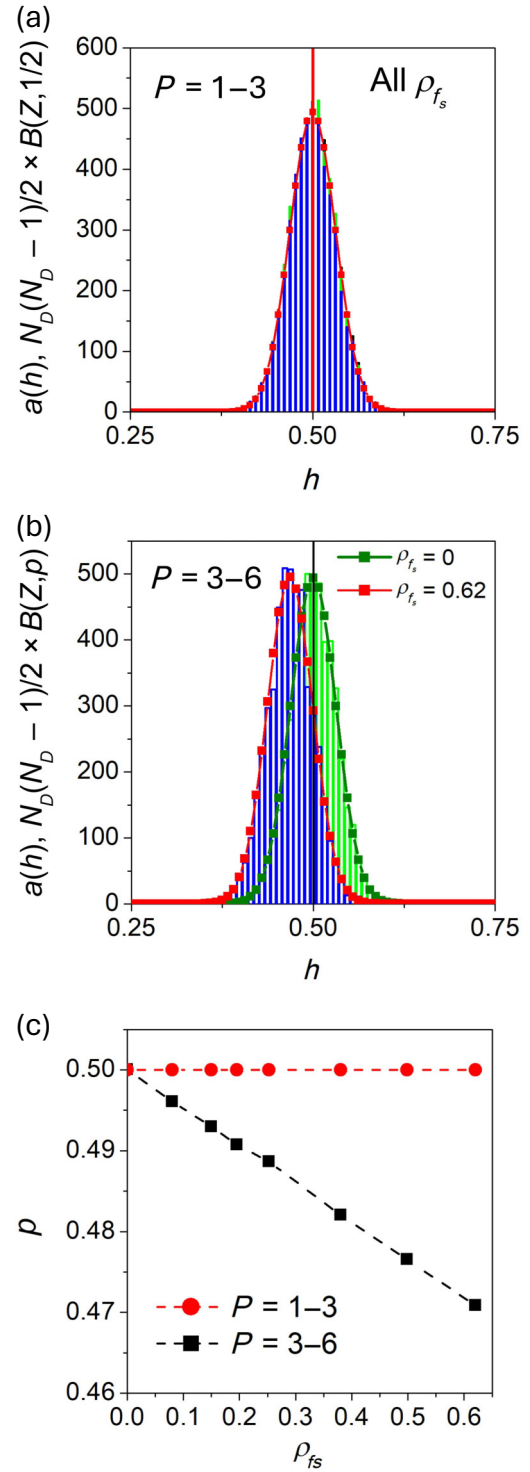


FIG. 6. (a) Distribution of the FHD for the digitized magnetic remanence, obtained from the pairwise comparison of the computer-generated tag set for all explored values of  $\rho_{f_s}$ , in the case of strong inhomogeneity of the nanoparticle patterns; blue bars, normalized histograms; full red line with dots, theoretical prediction. (b) Same as in panel (a) for  $\rho_{f_s} = 0$  (green bars) and  $\rho_{f_s} = 0.62$  (blue bars) for mild inhomogeneity. (c) Effect of  $\rho_{f_s}$  on the mean  $p$  of the distribution of the binary correlation coefficients for strong and mild inhomogeneity.

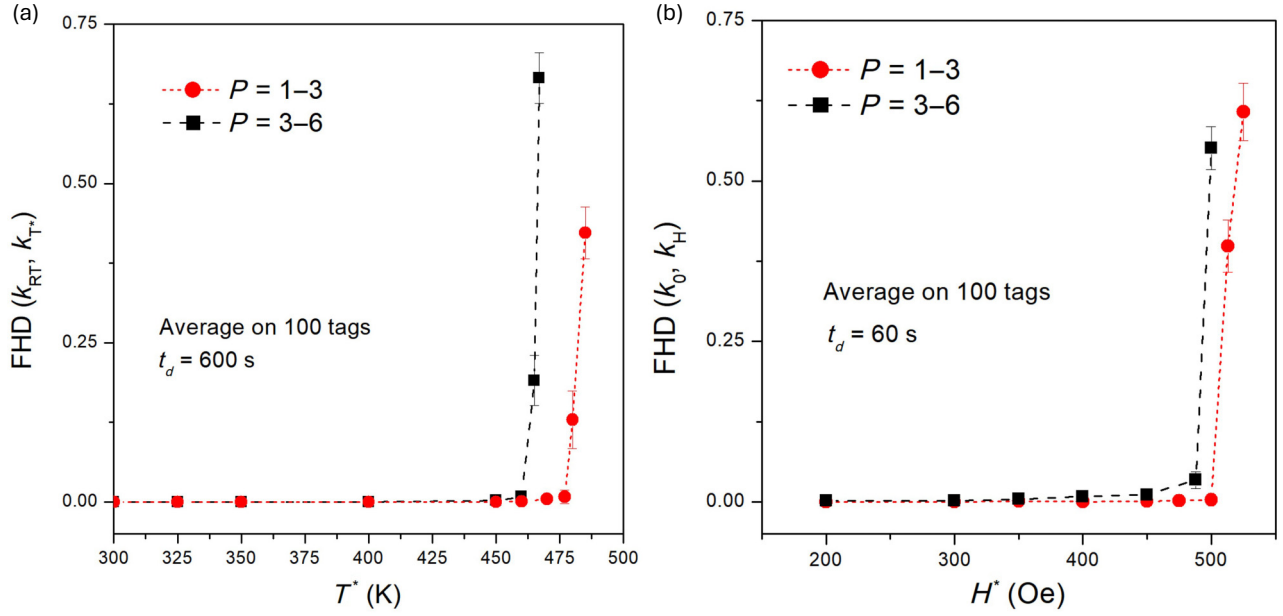


FIG. 7. (a) Fractional Hamming distance (FHD) between room-temperature binary keys associated to a MMT before ( $k_{RT}$ ) and after ( $k_{T^*}$ ) a transient thermal disturbance at the temperature  $T^*$  for  $t_d = 600$  s. (b) The same as in panel (a) for room-temperature binary keys associated to a MMT before ( $k_0$ ) and after ( $k_{H^*}$ ) a transient magnetic disturbance induced by a field  $H^*$  acting for  $t_d = 60$  s. In both panels, the vertical bars show the standard deviation of the data obtained on a set of 100 computer-generated tags.

during its lifetime it can be accidentally exposed to the effects of either high temperatures or magnetic-field disturbances. In both cases, the magnetic remanence of every MU is changed, so that the binary key associated to the tag can be in principle modified with an ensuing loss of information [the stability in time of the MMT at room temperature was already discussed in Sec. IV A; see panel (e) of Fig. 5].

The thermal stability of the MMT has been tested by simulating the effect of a transient thermal disturbance. It is assumed here that an as-conditioned tag characterized at room temperature (RT = 300 K) by a specific binary key ( $k_{RT}$ ) is suddenly brought to some high temperature  $T^* > RT$  for a time  $t_d = 600$  s (without applying a magnetic field) and quickly brought back to RT. The treatment at  $T^*$  acts to reduce the magnetic remanence of every MU because temperature enhances the tendency towards equilibrium of all DWS in the MU (corresponding to  $M_r = 0$  when no applied field is present; see Appendix A); as a consequence, the  $M_r$  value of every MU is irreversibly reduced when the tag is brought back to RT, the amount of the effect depending on the height of the local energy barrier for the DWS, which in turns depends on the local values of  $f_s$  and  $P$  (Sec. III E).

The stability of the information stored in a MMT submitted to such a treatment is shown in panel (a) of Fig. 7 where the FHD between the binary keys associated to the tag before ( $k_{RT}$ ) and after ( $k_{T^*}$ ) heating is plotted.

The binary tag is not modified by the thermal transient up to a temperature  $T_M^* > 450$  K (the computed FHD being exactly equal to zero in this temperature range). The key begins to be irreversibly modified at higher temperatures, as indicated by the quick increase displayed by the FHD. It should be noticed that  $T_M^*$  is slightly influenced by the degree of inhomogeneity of the particles, being larger when  $P$  is small ( $1 \leq P \leq 3$ ). The vertical bars in the figure represent the standard deviation of the FHD data obtained on 100 different computer-generated tags. The high value of  $T_M^*$  resulting from this simulation ensures that the information stored in a tag is indeed a robust quantity, hardly affected by the temperature history during a device lifetime.

In a similar way, the stability of the MMT against random magnetic disturbances has been tested by simulating the effect of a transient magnetic field acting at room temperature on an as-conditioned tag characterized by a specific binary key ( $k_0$ ). In the simulation, a magnetic field of amplitude  $H^*$  is applied for a time  $t_d = 60$  s and then quickly removed (owing to the isotropic distribution of the nanoparticle easy axes, the magnetic-field direction plays no role here). The transient magnetic disturbance acts to modify the magnetic remanence of every MU (see Appendix A); as a consequence, the  $M_r$  value of every MU is irreversibly changed when the field is removed, and the extent of the change again depends on the height of the local energy barrier for the DWS.

The stability of the device against magnetic disturbances is shown in panel (b) of Fig. 7 where the FHD between the binary keys associated to the tag before ( $k_0$ ) and after ( $k_{H^*}$ ) the application of the magnetic field is plotted as a function of  $H^*$ . In the considered case, the binary tag is not modified for magnetic-field amplitudes up to  $H_M^* \gtrsim 375$  Oe  $\simeq 29.8$  kAm $^{-1}$  (the FHD being exactly equal to zero in this field range), whereas the key begins to be irreversibly modified for larger fields, as indicated by the quick increase displayed by the FHD. Even in this case,  $H_M^*$  is influenced by the degree of inhomogeneity of the particles, being larger when  $P$  is small ( $1 \leq P \leq 3$ ). The vertical bars in Fig. 7(b) indicate the standard deviation of the FHD data obtained on 100 different computer-generated tags. The value of  $H_M^*$  obtained in the simulation ensures that the information stored in a tag is a robust quantity even with respect to the exposition to accidental magnetic disturbances during a device lifetime.

As a final comment, it should be stressed that even if the room-temperature binary key of a tag was irreversibly modified by a thermal or magnetic disturbance, the device is still fully reusable; in fact, the lost original key can be easily restored by demagnetizing the tag and applying again to it the same conditioning procedure as the first time. Therefore, the desired information can again be stored in a MMT whose binary key was accidentally modified.

## V. CONCLUSION

Miniaturized magnetic tags with size in the micrometer range are able to encode custom information, which can be uniquely attached to every single unit of a mass production process. In this context, magnetic nanoparticles dispersed on a surface provide random, miniaturized, cheap and easy to fabricate tags. We have developed a numerical model to thoroughly analyze in detail the magnetic behavior of an assembly of interacting magnetic nanoparticles by studying the routes to decorrelation from the standpoint of encryption theory. This numerical study lights up the role of magnetic interaction among nanoparticles to assess the randomness of the binary keys that can be extracted from a tag of  $32 \times 32$  squared microns. Interestingly, we show that the decorrelation of geometric origin stemming from the random patterns of magnetic nanoparticles on the local scale is substantially reinforced by dipolar interactions among nanoparticles. Spatial inhomogeneities at short distances in the nanoparticle pattern markedly enhance the fluctuations and favor the decorrelation. The proposed model could be straightforwardly upgraded to describe a three-dimensional distribution of nanoparticles in order to make it more adherent to a real-world scenario where fabrication techniques may not be able to produce a perfect single layer of nanoparticles. On the other hand, the physical effect at the basis of the proposed detection

method, i.e., the random variation of magnetic remanence from MU to MU, would not be modified by the presence of a non-negligible thickness of the deposited nanoparticles.

The results of this study show that a custom piece of information can be stored in a MMT by means of a simple conditioning process done at room temperature, not requiring the use of ac fields and easily integrable in the tag-printing process; the information can be subsequently retrieved by interrogating the tag by means of a commercial read head. Using magnetic nanoparticles in the magnetically blocked state ensures the stability of the information at room temperature for times much longer than a tag's lifetime. The information stored in a MMT has been shown to be a robust quantity against random thermal or magnetic disturbances.

The proposed MMT is microscopically sized, so that it can be used to mark single microcomponents of a complex system; it is sustainable, being made of nontoxic and easily disposable component materials; it is versatile, reliable, and easily readable. The inherent stability ensures that the encoded information is not degraded even if the tag is applied to a local source of heat such as a power electronic component. In spite of the reduced size of the proposed device, the magnetic signal can be easily detected using a standard magnetic recording head.

From the viewpoint of security, the MMT can be hardly replicated, even knowing the printing and conditioning processes, so that the encoded information is virtually unclonable. From a technical viewpoint, the MMT needs no added circuitry, it can be printed on several substrate materials, and can be fully reconditioned and reprogrammed when submitted to magnetic or electromagnetic environmental alterations. Finally, it should be emphasized that the practical feasibility of the present concept is greatly eased by the fact that the used nanomaterials, the device fabrication and characterization techniques and the detecting methods are well assessed, inexpensive, and easily available.

## ACKNOWLEDGMENTS

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There are no conflicts to declare.

## DATA AVAILABILITY

The data that support the findings of this article are not publicly available. The data are available from the authors upon reasonable request.

## APPENDIX A: DWS AND RATE EQUATIONS

It is recalled that each tag is ideally comprised of  $Z = 16 \times 16 = 256$  identical square MUs of side  $B = 2 \times 10^3$  nm and local nanoparticle surface density  $f_s$  (see Fig. 1). The total side of the device is  $\sqrt{Z}B = 3.2 \times 10^4$  nm = 32  $\mu$ m. The nanoparticles dispersed in any MU are distributed in size according to Eq. (1), with average magnetic diameter is  $\langle D_m \rangle = 22.5$  nm and magnetic volume  $\langle V_m \rangle \approx \frac{\pi}{6} \langle D_m \rangle^3$ . The saturation magnetization is  $M_s$  ( $=350$  emu/cm<sup>3</sup> for magnetite).

Magnetic nanoparticles characterized by uniaxial anisotropy are described as double-well systems (DWSs, [86,87]). The reversal of the magnetic moment  $\mu$  of magnitude  $\mu = M_s V_m$  takes place through a thermally activated jump over the total anisotropy barrier between the two energy minima [see panels (b.1)–(b.3) of Fig. 2]:

$$E_B = K_{\text{tot}} V_m - \mu \cdot \mathbf{H}, \quad (\text{A1})$$

where  $K_{\text{tot}}$  is the total anisotropy energy density,  $V_m$  the particle's magnetic volume,  $\mathbf{H}$  the applied magnetic field. In the rate-equation approach, the total anisotropy constant  $K_{\text{tot}} = [K_{\text{eff}} + e_{\text{dip}}]$  is comprised of the single-particle effective anisotropy  $K_{\text{eff}}$  and of the term  $e_{\text{dip}}$ , which accounts for the effect of magnetic dipolar interactions on the nanoparticle [70]. We take  $K_{\text{eff}} = 3 \times 10^5$  erg/cm<sup>3</sup>, a value appropriate to magnetite nanoparticles at room temperature.

### 1. Magnetic dipolar contribution to the energy barrier

The dipolar term  $e_{\text{dip}}$  is evaluated starting from the dipole-dipole interaction [48] between magnetic moments  $\mu_i, \mu_j$  in the MU:

$$E_{ij} = \frac{\mu_i \mu_j}{d_{ij}^3} [\cos \theta_i \cos \theta_j + \sin \theta_i \sin \theta_j \cos(\phi_i - \phi_j) - 3 \cos \theta_i \cos \theta_j], \quad (\text{A2})$$

where the  $\theta_i, \phi_i$  angles are defined in Fig. 2, panel (a), and  $d_{ij}$  is the center-to-center distance between the nanoparticles. Assuming that the random quantities  $\mu_i, \theta_i, \phi_i, d_{ij}$ , are uncorrelated, the average value of Eq. (A2) is

$$\begin{aligned} \langle E_{ij} \rangle &= \langle \mu_i \mu_j \rangle \left\langle \frac{1}{d_{ij}^3} \right\rangle [\langle \cos \theta_i \rangle \langle \cos \theta_j \rangle + \langle \sin \theta_i \rangle \langle \sin \theta_j \rangle \\ &\quad \times \langle \cos(\phi_i - \phi_j) \rangle - 3 \langle \cos \theta_i \rangle \langle \cos \theta_j \rangle] \\ &= -2 \langle \mu \rangle^2 \left\langle \frac{1}{d_{ij}^3} \right\rangle \langle \cos \theta \rangle^2, \end{aligned} \quad (\text{A3})$$

where  $\langle \mu \rangle = M_s \langle V_m \rangle$  and the average of the cosine term is made over all angles between the easy axis of particles and the  $z$  axis.

Using Eq. (A3), the total dipolar energy of the MU can be written as

$$E_{\text{dip}} = \sum_{i \neq j} E_{ij} \approx \frac{N(N-1)}{2} \langle E_{ij} \rangle \approx \frac{N^2}{2} \langle E_{ij} \rangle, \quad (\text{A4})$$

where  $N$  is the number of nanoparticles in the MU and the sum is over all the  $[N \times (N-1)/2]$  particle pairs. Such a quantity is related to the local surface density by the relation:

$$N = \frac{4B^2 f_s}{\pi \langle D_m \rangle^2}. \quad (\text{A5})$$

Therefore, the dipolar energy density (energy per magnetic nanoparticle and per unit volume of the nanoparticle) can be written as

$$\begin{aligned} e_{\text{dip}} &= \frac{E_{\text{dip}}}{N \langle V_m \rangle} \\ &= N M_s^2 \langle V_m \rangle \left\langle \frac{1}{d_{ij}^3} \right\rangle \langle \cos \theta \rangle^2 \\ &= \frac{2}{3} B^2 \langle D_m \rangle M_s^2 \left\langle \frac{1}{d_{ij}^3} \right\rangle f_s \langle \cos \theta \rangle^2. \end{aligned} \quad (\text{A6})$$

Therefore, the expression of the total energy barrier is

$$\begin{aligned} E_B(\theta) &= (K_{\text{eff}} + e_{\text{dip}}) \langle V_m \rangle - \mu \cdot \mathbf{H} \\ &= \left[ K_{\text{eff}} + \frac{2}{3} B^2 \langle D_m \rangle M_s^2 \left\langle \frac{1}{d_{ij}^3} \right\rangle f_s \langle \cos \theta \rangle^2 \right] \\ &\quad \times \langle V_m \rangle - M_s \langle V_m \rangle H \cos \theta. \end{aligned} \quad (\text{A7})$$

The barrier's height is determined by different geometric and physical parameters and is particularly sensitive to the distance between particles through the term  $\langle 1/d_{ij}^3 \rangle$  and to the  $\langle \cos \theta \rangle^2$  average.

### 2. Solution of the rate equations

Magnetic rate equations are exploited to predict the time evolution of the population of nanoparticles in potential wells 1,2 [i.e., to know which magnetic moments are instant by instant directed along the direction 1 or 2 in particles whose easy axis makes the angle  $\theta$  with the  $z$  axis; see Fig. 2, panels (a),(b)]. The nanoparticle population in each well changes through activated jumps over the energy barrier  $E_B$  by effect of both absolute temperature  $T$  and magnetic field  $H$ ; the kinetics also depends on the angle  $\theta$  between the easy axis and the magnetic field. In this paper, the temperature is always assumed to be constant ( $T = 300$  K).

Let us consider the particular subset of all the  $N_\theta$  particles in the MU whose easy axis makes the angle  $\theta$  with the field ( $0 \leq \theta \leq \pi$ ). The fractional populations  $n_i(\theta, t) = N_i(\theta, t)/N_\theta$  ( $i = 1, 2$ ) evolve according to the rate equations:

$$\begin{cases} \frac{dn_1(\theta, t)}{dt} = -\frac{1}{\tau_1(\theta)}n_1(\theta, t) + \frac{1}{\tau_2(\theta)}n_2(\theta, t) \\ \frac{dn_2(\theta, t)}{dt} = \frac{1}{\tau_1(\theta)}n_1(\theta, t) - \frac{1}{\tau_2(\theta)}n_2(\theta, t) \end{cases}, \quad (\text{A8})$$

where the relaxation times for the activated  $1 \rightarrow 2$  and  $2 \rightarrow 1$  processes are

$$\tau_i(\theta) = \tau_0 \exp\left(\frac{E_{Bi}(\theta)}{k_B T}\right) \quad (i = 1, 2) \quad (\text{A9})$$

with  $\tau_0 \approx 1 \times 10^{-9}$ .  $E_{Bi}(\theta)$  is the energy barrier [see Eq. (A7)] as seen from well  $i$ . By definition,  $n_2(\theta, t) = 1 - n_1(\theta, t)$ .

When  $H = 0$  the energies of the two wells are the same and  $E_{B1} = E_{B2} = E_B$  [as in panels (b.1) and (b.3) of Fig. 2]. When a constant, uniform field directed along the positive  $z$  axis (the *conditioning field*) is applied to the device for a finite time  $t_c$  (the *conditioning time*) the exact solution of the rate equations is:

$$n_1(\theta, t_c) = \frac{1}{2} \exp\left(-\frac{t_c}{\tau_{\text{eff}}(\theta)}\right) + n_{1\text{eq}}(\theta) \left[1 - \exp\left(-\frac{t_c}{\tau_{\text{eff}}(\theta)}\right)\right] \quad (\text{A10})$$

where the effective time constant is

$$\tau_{\text{eff}}(\theta)^{-1} = \tau_1^{-1}(\theta) + \tau_2^{-1}(\theta). \quad (\text{A11})$$

and  $n_{1\text{eq}}(\theta)$  is the equilibrium value of the fractional population of well 1 at  $T = 300$  K in the presence of the conditioning field.

It is the effective time constant  $\tau_{\text{eff}}$  that determines the relaxation time for magnetic-moment reversal. Although for simplicity the sole dependence of  $\tau_{\text{eff}}$  on the angle  $\theta$  is explicitly reported in Eqs. (A10) and (A11), this parameter is a function of both temperature and nanoparticle diameter  $D_m$  (through the energy barrier  $E_{Bi}$ ). When no magnetic field is applied, the energy barrier is no longer dependent of  $\theta$ , so that  $\tau_1 = \tau_2$  and the effective relaxation time becomes

$$\tau_{\text{eff}}^0 = \frac{\tau_1}{2} = \frac{1}{2} \tau_0 \exp\left[\frac{(K_{\text{eff}} + e_{\text{dip}})\langle V_m \rangle}{k_B T}\right] \quad (\text{A12})$$

and is a function of temperature and nanoparticle diameter only.

According to Eq. (A10), the fractional population  $n_1(\theta, t)$  of well 1 is assumed to be equal to  $\frac{1}{2}$  when  $t_c = 0$ ;

this means that before the conditioning treatment the two directions of the magnetic moment have the same probability of being observed in the MU. According to Eq. (A10),  $n_1(\theta) \rightarrow n_{1\text{eq}}(\theta)$  for a very long conditioning time. The equilibrium value  $n_{1\text{eq}}(\theta)$  is obtained from the rate equations putting the derivatives with respect to time to zero:

$$n_{1\text{eq}}(\theta) = \frac{\tau_1(\theta)}{\tau_1(\theta) + \tau_2(\theta)}. \quad (\text{A13})$$

The equilibrium fractional population is  $n_{1\text{eq}} \approx 0$  when  $\tau_1 \ll \tau_2$  and  $n_{1\text{eq}} \approx 1$  when  $\tau_2 \ll \tau_1$ .

The behavior of a DWS assembly with randomly distributed easy axes is obtained by averaging the solutions of the rate equations over all  $\theta$  angles. In three dimensions, the average of a  $\theta$ -dependent quantity  $g(\theta)$  is the sum:  $\sum_1^N g(\theta_i) \sin(\theta_i) / \sum_1^N \sin(\theta_i)$  over  $N$  discrete angles in the interval  $-\pi/2 \leq \theta_i \leq \pi/2$ . In this work,  $N$  has been fixed to 181; the relative difference between the average done with  $N = 181$  and with  $N = 1801$  is negligible ( $< 2 \times 10^{-3}$ ).

### 3. Validity of rate equations

The rate-equation approach is a simplifying approximation to the Fokker-Planck equation for the double-well problem. In magnetic nanoparticle systems, rate equations are shown to naturally emerge from the Fokker-Planck equation when the ratio  $E_B/k_B T \equiv K_{\text{eff}}V/k_B T$  is significantly larger than unity; therefore the validity of the approach at a given temperature depends on both magnetic anisotropy and nanoparticle size.

In rate equations containing harmonic magnetic fields, detailed balancing is achieved only when  $\omega\tau_c \ll 1$  where  $\omega$  is the driving-field frequency and  $\tau_c$  is the characteristic relaxation time of the system. In magnetic nanoparticles, the free diffusion time of magnetization is  $\tau_c = M_s V(1 + \alpha^2)/2\gamma\alpha k_B T$  where  $\gamma$  is the electron gyromagnetic ratio and  $\alpha$  is the Gilbert's damping constant appropriate to describe systems with intermediate-to-high damping ( $\alpha \approx 1$ ) such as magnetic nanoparticles. Using  $\alpha = 0.5$  one gets  $\tau_c = 1.06 \times 10^{-9}$  s for  $D = 15$  nm for magnetite NPs. In this case, the driving-field frequency should be much lower than  $1.50 \times 10^8$  Hz to guarantee detailed balancing. All of the above requirements are satisfied in the operating conditions of the present paper.

### 4. Calculation of the $\langle \cos \theta \rangle$ average

The  $\langle \cos \theta \rangle$  term appearing in the energy barrier [Eqs. (A6) and (A7)] is calculated by integrating over all values of the Euler angles  $\theta, \phi$ . The integration over the angle  $\phi$  is trivial because  $\phi$  never appears in the expression of the energy. The average over  $\theta$  makes use of a weighing function  $w(\theta, t_c)$ , which gives the fractional population of nanoparticles whose easy-axis direction is characterized by

the angle  $\theta$ :

$$\begin{aligned}\langle \cos \theta \rangle &= \frac{1}{2\pi} \int_0^\pi \int_0^{2\pi} \cos \theta w(\theta, t_c) \sin \theta d\theta d\phi \\ &= \int_0^\pi \cos \theta w(\theta, t_c) \sin \theta d\theta.\end{aligned}\quad (\text{A14})$$

By definition, the weighing function  $w(\theta, t_c)$  is strictly related to the quantity  $n_1(\theta, t_c)$ :

$$\begin{cases} w(\theta, t_c) = n_1(\theta, t_c) & (0 \leq \theta < \pi/2) \\ w(\theta, t_c) = n_2(\theta, t_c) \equiv [1 - n_1(\pi - \theta, t_c)] & (\pi/2 < \theta \leq \pi) \end{cases} \quad (\text{A15})$$

Before the start of the conditioning treatment  $w(\theta) = \frac{1}{2}$ . It is easy to show that the weighing function is always normalized:

$$\begin{aligned}\int_0^\pi w(\theta, t_c) \sin \theta d\theta &= \int_0^{\pi/2} n_1(\theta, t_c) \sin \theta d\theta + \int_{\pi/2}^\pi [1 - n_1(\pi - \theta, t_c)] \\ &\times \sin \theta d\theta = 1.\end{aligned}\quad (\text{A16})$$

Using Eq. (A15), the last integral in Eq. (A14) is transformed into

$$\langle \cos \theta \rangle = 2 \int_0^{\pi/2} \cos \theta n_1(\theta, t_c) \sin \theta d\theta - \frac{1}{2}. \quad (\text{A17})$$

Therefore, the averaged cosine takes values between 0 and  $\frac{1}{2}$ . The lower limit corresponds to the result expected

before applying the conditioning field,  $w(\theta) \equiv n_1(\theta, 0) = \frac{1}{2}$  for all angles  $\theta$ ; the upper limit corresponds to the case where the moments of all particles in the MU point along the direction making the smallest angle with  $\mathbf{H}$ , i.e.,  $w(\theta) = 1$  for  $\theta < \pi/2$  and  $w(\theta) = 0$  for  $\theta > \pi/2$ . This happens for  $t_c \rightarrow \infty$  under a field sufficiently strong to completely empty well 2. A pictorial representation of the effect is given in panel (c) of Fig. 2.

## 5. Magnetic remanence

After the conditioning time  $t_c$ , the magnetic field is removed; the populations in wells 1 and 2 do not change further because activated jumps between energy wells are again forbidden by the height of the energy barrier [see panel (b.3) of Fig. 2]. The magnetic moment of each particle lies on the easy axis and the magnetic remanence (i.e., the value of the magnetization along the  $z$  axis when  $H = 0$ ) is

$$M_r(\theta) = \begin{cases} M_s[n_1(\theta, t_c) \cos \theta + n_2(\theta, t_c) \cos(\pi - \theta)] & (0 \leq \theta < \pi/2) \\ M_s[n_2(\theta, t_c) \cos \theta + n_1(\theta, t_c) \cos(\pi - \theta)] & (\pi/2 < \theta < \pi) \end{cases}.$$

Therefore,

$$M_r(\theta) = \begin{cases} M_s[2n_1(\theta, t_c) - 1] \cos \theta \\ M_s[2n_2(\theta, t_c) - 1] \cos \theta \equiv M_s[2n_1(\pi - \theta, t_c) - 1] |\cos \theta| \end{cases},$$

where the first line applies when  $(0 \leq \theta < \pi/2)$ , and the second one when  $(\pi/2 < \theta < \pi)$ . The overall remanence of the MU is obtained by integration:

$$M_r = \frac{1}{2} \int_0^\pi M_r(\theta) \sin \theta d\theta = \int_0^{\pi/2} M_r(\theta) \sin \theta d\theta \quad (\text{A18})$$

the second equality being allowed because the integrand function  $M_r(\theta) \sin \theta$  is symmetric around the vertical axis

through  $\theta = \pi/2$ . As a result,

$$\begin{aligned}M_r &= M_s \int_0^{\pi/2} [2n_1(\theta, t_c) - 1] \cos \theta \sin \theta d\theta \\ &= M_s \left[ 2 \int_0^{\pi/2} n_1(\theta, t_c) \cos \theta \sin \theta d\theta - \frac{1}{2} \right] = M_s \langle \cos \theta \rangle.\end{aligned}\quad (\text{A19})$$

The behavior of  $M_r$  as a function of  $t_c$  for magnetite nanoparticles of average magnetic diameter  $\langle D_m \rangle = 22.5$

nm submitted to a conditioning field  $H = 300$  Oe at room temperature is shown in Fig. 8.

The asymptotic value attained for  $t_c \rightarrow \infty$  is close but not exactly equal to  $M_s/2 = 175$  Oe because in this case the conditioning field is not sufficiently strong to completely empty well 2. Figure 8 shows that a conditioning time of  $t_c = 300$  s is enough to generate a sufficiently high remanence signal at room temperature;  $t_c$  values orders of magnitude longer would only give rise to a limited increment of  $M_r$ , resulting in an exceedingly long conditioning process.

$$f_L(d_{ij}) = \begin{cases} \frac{2\pi d_{ij}}{B^2} - \frac{8d_{ij}^2}{B^3} + \frac{2d_{ij}^3}{B^4} \\ -\frac{2d_{ij}}{B^2} \left[ 2 + \frac{d_{ij}^2}{B^2} - 4 \sqrt{\left( \frac{d_{ij}^2}{B^2} - 1 \right)} - 2 \arcsin \left( \frac{d_{ij}^2}{B^2} - 1 \right) \right] \end{cases},$$

where the first expression applies to the case ( $d_{ij} \leq B$ ) and the second one to the case ( $d_{ij} \geq B$ ). The  $f_L(d_{ij})$  function is plotted in panel (b) of Fig. 3 (full line in color). The excellent agreement between the frequency distribution function  $\tilde{f}_L(d_{ij})$  of the center-to-center distances  $d_{ij}$  between the computer-generated particles and the analytic  $f_L(d_{ij})$  function shows that complete spatial randomness is indeed achieved when the nanoparticle positions in the MU

## APPENDIX B: PROBABILITY DENSITY FUNCTION FOR A UNIFORM POISSON PROCESS

When  $N$  pointlike magnetic nanoparticles are randomly distributed on the surface of a MU (a square of side  $B = 2 \times 10^3$  nm) in such a way that the condition of complete spatial randomness (aka homogeneous spatial Poisson process) is satisfied, the PDF  $f_L(d_{ij})$  for the generic interparticle distance  $d_{ij}$ , whose upper limit is  $\sqrt{2}B$ , is predicted to be [97]

are generated by the computer without introducing in the simulation any algorithm to mimic local inhomogeneities of the nanoparticle distribution.

## APPENDIX C: PEARSON'S LINEAR CORRELATION BETWEEN LOCAL SURFACE DENSITIES

For each tag a column vector with  $Z = 256$  rows is created, whose elements are the values of the local surface density of nanoparticles  $f_s$  in all the MUs of the device. The linear correlation coefficient  $r_{mn}$  between any two column vectors ( $m, n$ ) of the set is calculated through the standard expression [102]:

$$r_{mn} = \frac{\sum_{k=1}^Z \left[ (f_{sk}^{(m)} - \langle f_s \rangle) \times (f_{sk}^{(n)} - \langle f_s \rangle) \right]}{\sqrt{\sum_{k=1}^Z (f_{sk}^{(m)} - \langle f_s \rangle)^2} \times \sqrt{\sum_{k=1}^Z (f_{sk}^{(n)} - \langle f_s \rangle)^2}}, \quad (\text{C1})$$

where  $f_{sk}^{(m)}$  and  $f_{sk}^{(n)}$  are the local surface densities of the  $k$ th MU of devices  $m$  and  $n$ . The total number of  $r_{mn}$  values is  $[N_D \times (N_D - 1)/2]$ .

The random quantity  $r \equiv r_{mn}$  defined in Eq. (C1) is distributed between  $-1$  and  $+1$ . The values of  $r$  can be reported on a histogram whose height  $a(r)$  is equal to the number of pairs of devices whose correlation coefficients fall in the interval between  $r - \Delta r/2$  and  $r + \Delta r/2$ . The sum of all  $a(r)$  values is therefore equal to  $[N_D \times (N_D - 1)/2]$ .

In the case of uncorrelated random variables, the linear correlation coefficients are expected to be distributed around zero according to Student's  $t$ -distribution law [81]

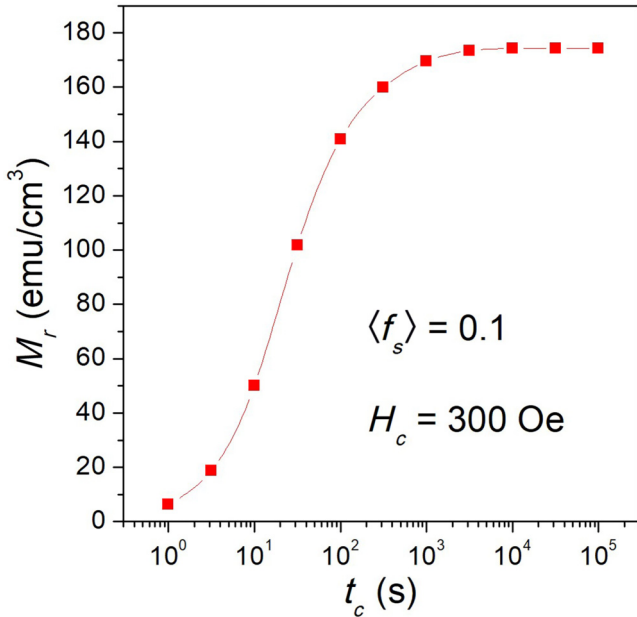


FIG. 8. Effect of conditioning time  $t_c$  on the development of the magnetic remanence of a distribution of  $\text{Fe}_3\text{O}_4$  nanoparticles of magnetic diameter  $D_m = \langle D_m \rangle = 22.5$  nm submitted to the conditioning field  $H_c$ .

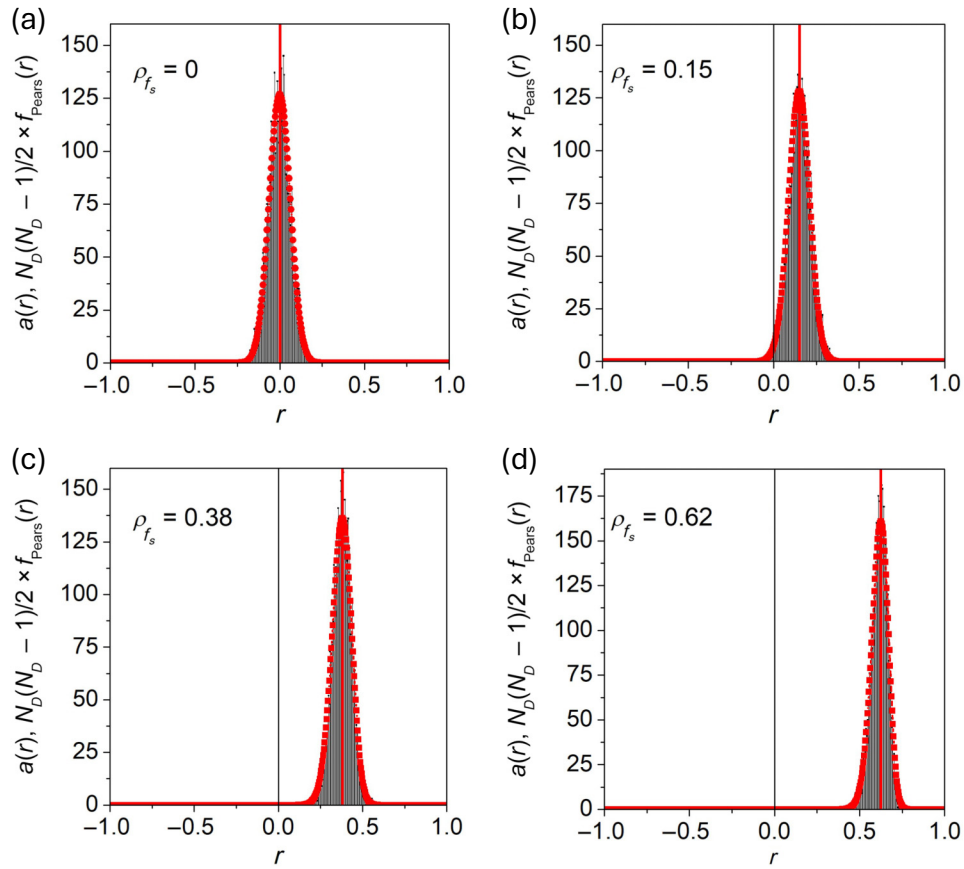


FIG. 9. Distribution of Pearson's correlation coefficient for the fraction of tag surface occupied by the nanoparticles in the magnetic units, as obtained from the pairwise comparison of the computer-generated tag set; black lines, normalized histograms; red lines, theoretical predictions from the  $t$ -Student's distribution law; the parameter  $\rho_{f_s}$  is a measure of the degree of correlation between tags,  $\rho_{f_s} = 0$  meaning uncorrelated tags.

for the random variable  $t = r\sqrt{(Z-2)/(1-r^2)}$ , resulting in

$$f_P^{(\text{unc})}(r) = \frac{1}{\sqrt{2\pi}} \left(1 + \frac{r^2}{1-r^2}\right)^{-\frac{(Z-1)}{2}} \approx \frac{1}{\sqrt{2\pi}} \exp\left[-\frac{(Z-2)}{2} \frac{r^2}{1-r^2}\right]. \quad (\text{C2})$$

The standard error of the distribution is predicted to be

$$\langle r^2 \rangle^{\frac{1}{2}} = \sqrt{\frac{2 \ln 2}{2 \ln 2 + (Z-2)}}. \quad (\text{C3})$$

The histogram obtained by analyzing a set of  $N_D$  computer-generated tags is reported in panel (a) of Fig. 9 [the histogram is normalized, i.e., the sum of all heights  $a(r)$  is set equal to 1]. In this particular case, the computer was instructed to generate the nanoparticle patterns of all the MUs of the  $N_D$  tags without any particular constraint or prescription. The  $r$  coefficients turn out to be symmetrically distributed around  $r = 0$  with low dispersion.

The  $f_P^{(\text{unc})}(r)$  function is superimposed to the computer-generated histogram (red line). The histogram perfectly matches in position and width the expected behavior of the distribution of the linear correlation coefficients, indicating that the  $(m, n)$  column vectors formed with the  $f_s$  values of the MUs are indeed uncorrelated random variables.

On the other hand, the computer can produce a set of  $N_D$  tags containing a partial correlation of the  $f_s$  random variables according to a procedure outlined in the Supplemental Material [98] and making use of a triangular  $2 \times 2$  Cholesky matrix. The preset degree of correlation between  $f_s$  values is expressed by the parameter  $\rho_{f_s}$  ( $0 \leq \rho_{f_s} \leq 1$ ),  $\rho_{f_s} = 0$  corresponding to completely uncorrelated variables. Three examples of histograms corresponding to partial (positive) correlation are reported in panels (b)–(d) of Fig. 9. The histograms are now centered at some positive value of  $r$  and are no longer symmetric around their peak values.

In the case of correlation, the variable  $t$  of Student's distribution becomes  $t = (r - \rho_{f_s})\sqrt{\frac{Z-2}{1-r^2}}$ , and Eq. (C2)

transforms into

$$f_P^{(\text{cor})}(r) = \frac{1}{\sqrt{2\pi}} \left( 1 + \frac{(r - \rho_{fs})^2}{1 - r^2} \right)^{-\frac{(Z-1)}{2}} \\ \approx \frac{1}{\sqrt{2\pi}} \exp \left[ -\frac{(Z-2)}{2} \frac{(r - \rho_{fs})^2}{1 - r^2} \right]. \quad (\text{C4})$$

The  $f_P^{(\text{cor})}$  functions corresponding to three selected values of  $\rho_{fs}$  are superimposed to the corresponding computer-generated histograms (red lines). Also in the case of partial correlation between  $(m, n)$  column vectors, the agreement between computer-generated histograms and the theoretical prediction of Eq. (C4) is excellent in position and shape for all values of  $\rho_{fs}$ .

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