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Distribution of switching fields in magnetic granular materials

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We present analytical calculations and kinetic Monte-Carlo modeling of rate-dependent behavior of switching field distributions (SFDs) in an ensemble of Stoner-Wohfarth particles, assuming distributions of anisotropies and volumes, and thermal activation included by the Néel-Brown theory. By applying probabilistic arguments, we show that the SFD can be self-consistently separated into the contribution from distributions of intrinsic properties of particles and the (irreducible) contribution resulting solely from thermal fluctuations, which is shown to become a significant effect at sweep rates relevant to the recording process. This provides a unifying framework for systematic analysis of different classes of systems. © *2012 American Institute of Physics*. [http://dx.doi.org/10.1063/1.4765085]

The intrinsic switching field distribution (SFD) is a fundamental characteristic of granular magnetic materials which, in addition to inter-granular interactions, governs the nature of the magnetization reversal.[1] As such it is an important technological design factor; for example, it determines the recording quality of perpendicular magnetic recording (PMR) materials.[2–4] It may also turn out to be an essential optimization criterion for the emerging heat assisted magnetic recording technology, where it is necessary to control the Curie temperature distribution as a result of the underlying distributions of intrinsic properties of grains.[5–7]

Formally, the SFD is defined as the probability distribution $D(H_{S,i})$ for grains i to switch at external field thresholds $H_{S,i}$ in the absence of inter-granular interactions. Consequently, an important task of materials characterization is the accurate identification of $D(H_{S,i})$ from hysteresis loops in the interacting case. Various methodologies have already been developed for this purpose with varying ranges of validity and sophistication.[8–12] Furthermore, the $D(H_{S,i})$—assuming it can be identified, depends on the distributions of grain volumes $D(V_i)$, anisotropy fields $D(H_{K,i})$, and angles $D(\psi_i)$. Also, due to the thermal relaxation, there is a further dependence on the measurement time scales such as the external field sweep rate $R = dH(t)/dt$.[13–18] Identifying general scaling laws relating all these distributions to the overall SFD at different R is essential, and this is the primary focus of the current letter.

Here, we study analytically, and by means of a numerical kinetic Monte-Carlo model, the sweep rate dependence of $D(H_{S,i})$ in an ensemble of non-interacting Stoner-Wohlfarth particles with thermal activation included by the Néel-Brown approach.[19,20] It is shown that the $D(H_{S,i})$ can be self-consistently separated into a contribution resulting from distributions of intrinsic properties $D(V_i)$, $D(H_{K,i})$, and $D(\psi_i)$, such as investigated, e.g., in Refs. 17 and 18, and a contribution resulting purely from thermal fluctuations, e.g., Refs. 13–16. The latter contribution is an irreducible thermodynamic phenomenon, becoming a very significant effect at sweep rates corresponding to the recording process.

To simplify notations it is convenient to define the "intrinsic property vector" of a grain $\vec{p}_i = (H_{K,i}, V_i, \psi_i)$ and joint probability distribution $D(\vec{p}_i) = D(H_{K,i}, V_i, \psi_i)$. Assuming Stoner-Wohlfarth particles, the energy barriers separating the "up" and "down" particle states depend on $\vec{p}_i$ and are given by $\Delta e_i = \beta_i(1 - H/(H_{K,i}\phi_i))^{\alpha_i}$. Here $\beta_i = K_i V_i / k_B T$ is the magnetic stability ratio with K_i, k_B, and T being the anisotropy and Boltzmann constants and temperature, and $H_{K,i} = 2K_i/\mu_0 M_s$ with M_s being the saturation magnetization of particles. The function $\phi_i = (\cos^{2/3}\psi_i + \sin^{2/3}\psi_i)^{-3/2}$ describes the angular contribution to the anisotropy field and is related to the exponent α_i by the Pfeiffer approximation $\alpha_i = 0.86 + 1.14\,\phi_i$.[21]

We consider a swept field experiment with time-dependent external field $H(t) = R(t - t_0) - H_{\rm sat}$, where t_0 is the reference time at the start corresponding to the negative saturation field $-H_{\rm sat}$ and we take $R > 0$. We first evaluate the probability $P_i(H)$ of finding an external field value H at which the grain reverses to its down state.[22] The solution follows from the master equation valid for PMR media,[2] namely, $dP_i(H(t))/dt = R \cdot dP_i(H)/dH = -w_i(H)P_i(H)$. The $w_i = f_0 \exp(-\Delta e_i)$ is the Arrhenius relaxation rate for transitions out of the negative state and we assume constant attempt frequency $f_0 = 10^9\ {\rm s}^{-1}$. Solving for $P_i(H)$ allows to find the magnetization as $M_i(H) = 1 - 2P_i(H)$

$$M_i(H) = 1 - 2\exp\Big(-f_0 R^{-1} I_i(-H_{\rm sat}, H)\Big), \qquad (1a)$$

$$I_i = \int_{-H_{\rm sat}}^{H} \exp\left(-\beta_i\left(1 - \frac{H'}{H_{K,i}\phi_i}\right)^{\alpha_i}\right) dH', \qquad (1b)$$

which can be, for arbitrary α_i, integrated by standard numerical techniques. Equations (1) define the relationship between the grain magnetization, the external field applied at the sweep rate R, and the intrinsic properties $\vec{p}_i$. Typical shapes of single-particle $M_i(H)$ for different β_i and fixed R are plotted in Fig. 1(a). Similarly, Fig. 1(b) shows the normalized field-derivative of $M_i(H)$, which directly corresponds to the

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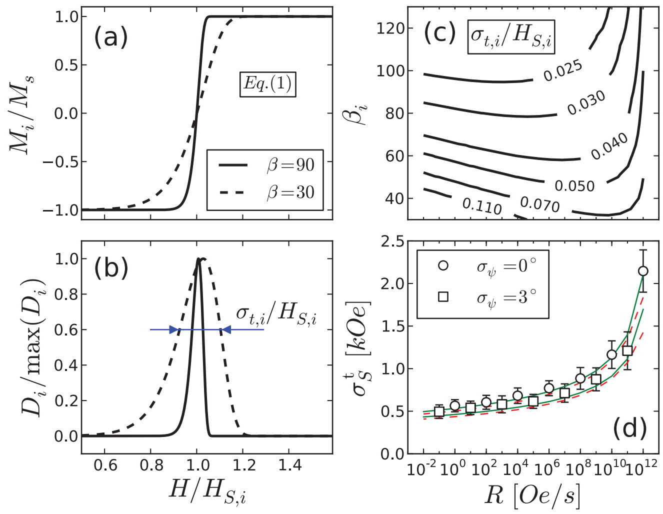

FIG. 1. (a) M_i vs H dependence calculated according to Eqs. (1) for $\beta_i = 30$ and 90. (b) Normalized probability density $D_i(H)$ defining the thermal broadening width $\sigma_{t,i}$. (c) Contour plot of the relative thermal broadening $\sigma_{t,i}/H_{S,i}$ for variable β_i and R. (a)–(c) assume $\sigma_\psi = 3°$. (d) Thermal broadening contribution in Eq. (2) computed as an ensemble average $\langle\sigma_{t,i}^2\rangle$ (symbols), using an equivalent-particle approximation $\sigma_i^2(\langle\vec{p}_i\rangle)$ assuming 100 grains in the averaging (solid lines), and Eq. (7) (dashed lines), with $\beta = 80$, $\sigma_K/H_K = 0.05$, $\sigma_V/V = 0.3$ and $\sigma_\psi = 0°, 3°$. Other parameters used in (a)–(d): $H_K = 40$ kOe and $V \sim 400\,\mathrm{nm}^3$.

probability density $D_i(H) = dP_i(H)/dH$ for the reversal field to the downward state given the intrinsic properties $\vec{p}_i$ and sweep rate R. It can be seen in these two figures that as β decreases the broadening of the magnetization transition region becomes more pronounced, which is a direct consequence of the energy barrier reduction enhancing the thermal relaxation effect. The magnetization transition can be quantified by an average field $\{H\}_i$ equal to the mean of $D_i(H)$ and defining the switching threshold of a particle, i.e., $H_{S,i} = \{H\}_i$, and the thermal fluctuation-induced broadening of the transition around $H_{S,i}$, equal to the variance $\sigma_{t,i}^2 = \{H^2\}_i - \{H\}_i^2$ of $D_i(H)$. Both $H_{S,i}$ and $\sigma_{t,i}^2$ depend on the sweep rate R. Note that in the definitions above we denoted by $\{\}_i$ the field-averages over the single-particle $D_i(H)$, to be distinguished below from the averages over the intrinsic properties distribution $D(\vec{p}_i)$, which will be denoted by triangular brackets $\langle\rangle$.

Assuming an ensemble of non-interacting particles with $D(\vec{p}_i)$, the overall probability density $D(\vec{p}_i, H)$ to observe the switching of a particle with properties $\vec{p}_i$ to occur at the field H is the product $D(\vec{p}_i)D_i(H)$.[23] Then the average switching field in the ensemble is obtained as $H_S = \int_{-\infty}^{\infty}\int_{-\infty}^{\infty} H D_i(H) D(\vec{p}_i)\, dH\, d\vec{p} = \langle\{H\}_i\rangle = \langle H_{S,i}\rangle$, where we used the field-independence of $D(\vec{p}_i)$ to sequence integrations during the averaging procedure. Similarly, the overall switching field variance can be calculated as $\sigma_S^2 = \langle\{H^2\}_i\rangle - \langle\{H\}_i\rangle^2$. Thus, these relations indicate that $D(\vec{p}_i)D_i(H)$ is the probability distribution governing the statistical behavior of switching fields of particles and as such it directly defines the intrinsic SFD: $D(H_{S,i}) \equiv D(\vec{p}_i)D_i(H)$, essentially expressing the implicit dependence of $H_{S,i}$ on $\vec{p}_i$ and on the thermal contribution entering through $D_i(H)$.

The starting point to study the variance σ_S^2 of the $D(H_{S,i})$, which is the practically relevant quantity,[1–3] is its definition in the preceding paragraph, which we rewrite as $\sigma_S^2 = \langle\{H^2\}_i\rangle - \langle\{H\}_i^2\rangle + \langle\{H\}_i^2\rangle - \langle\{H\}_i\rangle^2$ or equivalently as

$$\sigma_S^2 = (\sigma_S^{\mathrm{t}})^2 + (\sigma_S^{\mathrm{in}})^2. \tag{2}$$

In Eq. (2), $(\sigma_S^{\mathrm{t}})^2 = \langle\sigma_{t,i}^2\rangle = \langle\{H^2\}_i - \{H\}_i^2\rangle$ is the overall broadening due to thermal fluctuations and it is an irreducible thermodynamic effect which exists independently of $D(\vec{p}_i)$, i.e., also in the uniform case. On the other hand, the $(\sigma_S^{\mathrm{in}})^2 = \langle H_{S,i}^2\rangle - \langle H_{S,i}\rangle^2$, although also thermally dependent, appears only in the presence of $D(\vec{p}_i)$ and vanishes in the uniform case. The fact that these two contributions are separable is a direct consequence of the statistically uncorrelated nature of the distribution $D(\vec{p}_i)$ and thermal fluctuations that aid the reversal process over the activation barriers of grains.

To evaluate these contributions to the SFD, we first seek an approximate solution of Eqs. (1) for that field $H_{M,i}$ inside the transition region, which corresponds to a particular magnetization value M. Setting $M_i(H_{M,i}) = M$ in Eqs. (1) and substituting in the integral $h' = H'/(H_{K,i}\phi_i)$ gives after arranging $R\ln(2/(1-M)) \cdot (f_0 H_{K,i}\phi_i)^{-1} = I_i(-\infty, h_{M,i})$, where $h_{M,i} = H_{M,i}/(H_{K,i}\phi_i)$. In the integral I_i, for practical reasons, we also replaced the lower boundary $-H_{\mathrm{sat}} \to -\infty$ which, given the location of the transition region around $H_{M,i} \gg 0$ and its finite width, has no effect on the solution. Next, the requirement of a sufficiently narrow transition width implies the approximation that only a small neighborhood $h_{M,i} - h^* < h_{M,i} < h_{M,i} + h^*$ contributes to the magnetization switching. Given the smooth and monotonic variation of the integrand in the integral $I_i(-\infty, h_{M,i})$ in the entire integration range, this in turn means that only a small interval $(h_{M,i} - h^*, h_{M,i})$ within the integration range contributes to the solution for $M_i(H_{M,i}) = M$. Thus, substituting $h' = h_{M,i} - h^*$ and expanding to the first order in the small variable h^* gives

$$\begin{aligned}\frac{R\ln(2/(1-M))}{f_0 H_{K,i}\phi_i} &= \int_0^\infty \exp\Big(-\beta_i (x_i + h^*)^{\alpha_i}\Big)dh^*,\\ &\approx \int_0^\infty \exp(-\beta_i x_i^{\alpha_i} - \beta\alpha_i h^* x_i^{\alpha_i - 1})dh^*,\\ &= \exp(-\beta_i x_i^{\alpha_i})/(\beta_i \alpha_i x_i^{\alpha_i - 1}),\end{aligned}$$

where we put $x_i = 1 - h_{M,i} = 1 - H_{M,i}/(H_{K,i}\phi_i)$. Returning the substitutions for x_i and $h_{M,i}$ and rearranging gives

$$H_{M,i} = H_{K,i}\phi_i - H_{K,i}\phi_i(\ln\tau_i/\beta_i)^{1/\alpha_i}, \tag{3a}$$

$$\tau_i = \frac{f_0 R^{-1}}{\ln(2/(1-M))}\frac{H_{K,i}}{\beta_i}\frac{\phi_i}{\alpha_i}\left(1 - \frac{H_{M,i}}{H_{K,i}\phi_i}\right)^{1-\alpha_i}, \tag{3b}$$

which is the sought solution describing the rate-dependence of $H_{M,i}$ corresponding to M inside the transition region. Equations (3) will now be used to derive expressions for σ_S^{in} and σ_S^{t} entering in Eq. (2).

(a) *Broadening due to distributions of intrinsic properties*: σ_S^{in}. The variance $(\sigma_S^{\mathrm{in}})^2$ can be calculated by employing the standard error-propagation approach.[24] In this approach, the rate-dependent relation for the switching threshold $H_{S,i}(R)$, obtained after setting $M = 0$ in Eqs. (3)[25] is viewed as a transformation law between the random variable $H_{S,i}$ and the $H_{K,i}$, V_i, and α_i, and the goal is to evaluate the variance $(\sigma_S^{\mathrm{in}})^2$ knowing the σ_K^2, σ_V^2, and σ_α^2. Assuming that the random variables $H_{K,i}$, V_i, and α_i are uncorrelated, the error-propagation formula reads

$$(\sigma_S^{\mathrm{in}})^2 = \left|\frac{\partial H_S}{\partial H_K}\right|^2 \sigma_K^2 + \left|\frac{\partial H_S}{\partial V}\right|^2 \sigma_V^2 + \left|\frac{\partial H_S}{\partial \alpha}\right|^2 \sigma_\alpha^2, \tag{4}$$

where the derivatives are calculated from the mean value equation for H_S consistent with Eqs. (3) with $M = 0$

$$H_S = H_K\phi - H_K\phi(\ln\tau/\beta)^{1/\alpha}, \tag{5a}$$

$$\tau = \frac{f_0}{R\ln 2}\frac{H_K}{\beta}\frac{\phi}{\alpha}\left(1 - \frac{H_S}{H_K\phi}\right)^{1-\alpha}. \tag{5b}$$

The variables H_S, H_K, $\beta = KV/k_BT$, ϕ, and α are mean values of their respective probability distributions, with ϕ, α tabulated in Table I. Equations (5) is the solution describing the rate-dependence of the mean switching threshold H_S for the swept field experiment,[26] and resembles the well-known Sharrock equation for the magnetization relaxation experiment.[27] It simplifies for slower rates when in Eq. (5b) the term $H_S/H_K\phi < 1$ and can be neglected. Thus, an implicit differentiation of general Eqs. (5) with respect to H_K, V, and α gives straightforwardly

$$\frac{H_K}{H_S}\frac{\partial H_S}{\partial H_K} = 1 + \frac{\beta x^{1+\alpha}}{(1-x)(\alpha\Omega - 1)}, \tag{6a}$$

$$\frac{V}{H_S}\frac{\partial H_S}{\partial V} = \frac{x\Omega}{(1-x)(\alpha\Omega - 1)}, \tag{6b}$$

$$\frac{\alpha}{H_S}\frac{\partial H_S}{\partial \alpha} = \alpha^* + x\frac{1 - \alpha^* + \alpha\Omega\ln x}{(1-x)(\alpha\Omega - 1)}, \tag{6c}$$

where $x = 1 - H_S/(H_K\phi) = (\ln\tau/\beta)^{1/\alpha}$ following Eq. (5a), and we introduced $\Omega = 1 + \beta x^\alpha$ and also $\alpha^* = \alpha\phi^{-1}d\phi/d\alpha$. The values of all α-related quantities are given in Table I. Equations (6) define the derivatives in Eq. (4) and rely on the solution of Eqs. (5). We note that the present standard assumption of uncorrelated $H_{K,i}$, V_i, and α_i is in fact not necessary. The error-propagation equation (4) can be extended to include the relevant correlation coefficients once they have been quantified experimentally or by independent theories.[24,28]

b) *Broadening due to thermal fluctuations*: σ_S^{t}. It turns out that $\sigma_S^{\mathrm{t}} > 0$ even in the uniform case when $\vec{p}_i = \vec{p}$ for all i. The importance of this contribution for a single grain is shown in Fig. 1(c) as contour plot of the ratio $\sigma_{t,i}/H_{S,i}$ for different β_i and sweep rates R. It suggests that the broadening effect due to thermal fluctuations contributes significantly at fast rates R, which is consistent with the recent work.[16] According to Eq. (2), $(\sigma_S^{\mathrm{t}})^2$ needs to be evaluated as an ensemble average $\langle\sigma_{t,i}^2\rangle$. However, due to the uncorrelated nature of thermal contributions from individual particles and for sufficiently narrow $D(\vec{p}_i)$, it is possible to relate $\langle\sigma_{t,i}^2\rangle \approx \sigma_t^2(\langle\vec{p}_i\rangle)$, where $\sigma_t^2(\langle\vec{p}_i\rangle)$ is the thermal broadening for an equivalent particle with properties set to an ensemble average $\langle\vec{p}_i\rangle$. This suggests that to compute the $(\sigma_S^{\mathrm{t}})^2$, it is sufficient to evaluate σ_t^2 from $D_i(H)$ based on Eqs. (1) only once assuming $\langle\vec{p}_i\rangle$, and explicit averaging requiring computations of $\sigma_{t,i}^2$ for individual particles is not necessary. Fig. 1(d) confirms the equivalence between $\langle\sigma_{t,i}^2\rangle$ and $\sigma_t^2(\langle\vec{p}_i\rangle)$ for typical PMR media.

Given the validity of the equivalent particle picture, we can obtain the rate-dependence of σ_S^{t} by first dividing two mean value Eqs. (3), similar to Eqs. (5) but one for $M(H_S + \delta_1 H_K\phi) = M_1$ and the other for $M(H_S - \delta_2 H_K\phi) = M_2$, which after arranging reads

$$\left(1 + (\alpha - 1)x^{-1}\delta\right)\exp(\beta\alpha x^{\alpha-1}\delta) = C, \tag{7}$$

where $\delta = \delta_1 + \delta_2$ and the constant $C = \ln(2/(1-M_1))/\ln(2/(1-M_2))$. We neglected all higher but first order terms in δ and as before we put $x = 1 - H_S/(H_K\phi) = (\ln\tau/\beta)^{1/\alpha}$. Then the σ_S^t is obtained as $\sigma_S^t \approx \delta H_K\phi$, and arguments using Gaussian approximation allow finding optimum behavior for $M_{1,2} = \pm(1 - \mathrm{e}^{-1/2})$, which gives $C \approx 3.3$. For slower rates, the pre-exponential term can be neglected, simplifying Eq. (7) to $\delta \approx x^{1-\alpha}\ln C/(\alpha\beta)$.

To validate formula (2) with Eqs. (4)–(7), we computed rate-dependent room-temperature hysteresis loops by employing kinetic Monte-Carlo Stoner-Wohlfarth modeling developed earlier.[29] We considered sweep rates R in the interval $10^{-2} - 10^{12}$ Oe/s and all combinations of values $\sigma_K/H_K = 0.0, 0.05, 0.1, 0.2$, $\sigma_V/V = 0.0, 0.1, 0.2, 0.3$ (Lognormal distribution), and $\sigma_\psi = 0°, 3°$ (Gaussian distribution), which includes the range relevant for PMR. The distribution of switching thresholds $D(H_{S,i})$ was obtained by histogramming the fractions of particles switched at every field step along the hysteresis loop, which then allowed a direct evaluation of the mean switching field H_S and variance σ_S^2. Fig. 2 shows the dependences of $H_S(R)$ and $\sigma_S(R)$, respectively, demonstrating excellent agreement between the analytical calculations and the modeling. Generally, we found that for all input parameter choices the relative error remains below 8%.

Fig. 2(a) shows that increasing σ_ψ reduces $H_S(R)$, which is a result of the reduction of energy barriers. The behavior of $\sigma_S(R)$ shown in Fig. 2(b) is more subtle indicating a crossing point at a certain R. It is furthermore demonstrated that including σ_S^{t} in analytical calculations is essential. Fig. 3 shows typical behaviors of $\sigma_S(R)$ and $(\sigma_S/H_S)(R)$, and shows a change from an increasing trend for $\sigma_K \gtrsim \sigma_V$ towards a decreasing trend for $\sigma_K \ll \sigma_V$, proceeding through a plateau observed for certain optimal $\sigma_K < \sigma_V$. This behavior can be confirmed by exploring the limit $\sigma_V, \sigma_\psi \to 0$, when for sufficiently slow sweep rates, such that $\beta x^2 \gg 1$, Eqs. (4) and

TABLE I. Statistical properties of α-related quantities evaluated by generating histograms following Gaussian distribution of ψ_i with standard deviation σ_ψ. The ϕ, α, σ_α, and α^* are defined in the text.

σ_ψ	ϕ	α	σ_α	ϕ/α	α^*
0°	1	2	0	1/2	1.754
1°	0.9257	1.9153	0.0430	0.4831	1.8159
2°	0.8874	1.8716	0.0629	0.4737	1.8531
3°	0.8579	1.8380	0.0771	0.4660	1.8845
5°	0.8128	1.7866	0.0968	0.4537	1.9380
10°	0.7381	1.7014	0.1231	0.4315	2.0437

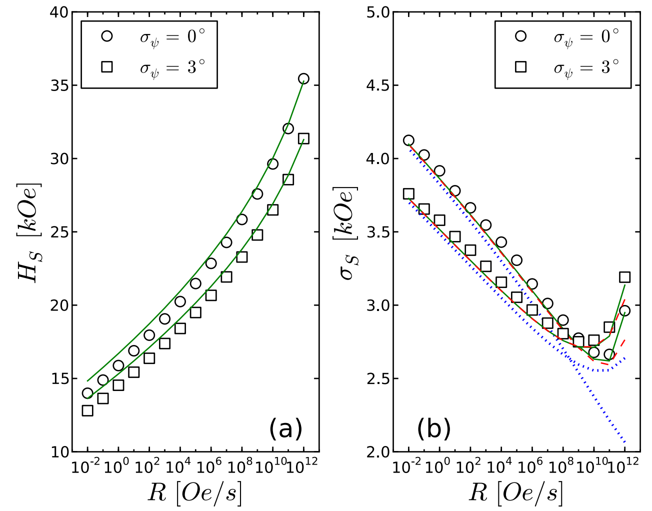

FIG. 2. (a) H_S vs R from Eqs. (5a) and (5b) σ_S vs R from Eq. (2) with $\sigma_S^{\rm in}$ from Eqs. (4)–(6) and σ_S^t obtained numerically using an equivalent particle approximation (solid lines), from Eq. (7) (dashed line), and with σ_S^t ignored (dotted lines); assuming $\sigma_K = 0.05$, $\sigma_V = 0.3$, and $\sigma_\psi = 0°, 3°$. The symbols correspond to H_S and σ_S from kinetic Monte-Carlo simulations. Other parameters: $H_K = 40\,$kOe, $\beta = 80$, and $V \sim 400\,\text{nm}^3$.

(6a) reduce to an approximate form $\sigma_S^{\rm in}/\sigma_K \approx 1 - x/2$, recovering the previous result.[30] Similarly, for $\sigma_K, \sigma_\psi \to 0$, Eqs. (4) and (6b) give $\sigma_S^{\rm in}/\sigma_V \approx (H_K/V)x/2$, indeed showing an opposite trend as a function of R and confirming the competing tendencies of the distributions of $D(H_{K,i})$ and $D(V_i)$ during the thermally activated reversal.

In summary, the present work develops a unifying framework, successfully validated by the kinetic Monte-Carlo modeling, which links the major contributions to the SFD of a PMR medium. The expansion technique leading to Eqs. (3) is essential and it can be extended beyond the assumption of constant f_0, which becomes relevant at high R.[16] Such analysis is more extensive and is not presented here due to the lack of space. The analytical model allows a self-consistent separation of the overall variance σ_S^2 of the SFD into two thermally dependent contributions. The first, $(\sigma_S^{\rm t})^2$, is solely due to thermal fluctuations and the second, $(\sigma_S^{\rm in})^2$, results from intrinsic distributions of $H_{K,i}$, V_i, and ψ_i. The $(\sigma_S^{\rm in})^2$ vanishes in the uniform case and, consequently, $(\sigma_S^{\rm t})^2$ represents the minimum SFD achievable for a given recording medium, becoming increasingly important for high sweep rates such as of the recording process. Thus, the present analytical approach demonstrates how to rescale the SFD determined from quasi-static measurements to obtain realistic values relevant during the recording process.

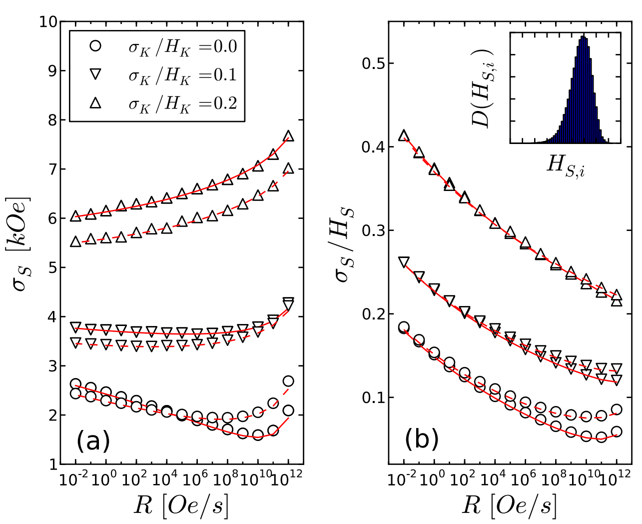

FIG. 3. (a) Dependence σ_S vs R and (b) σ_S/H_S vs R for $\sigma_V/V = 0.2$, $\sigma_\psi = 0°$ (solid lines) and 3° (dashed lines), and σ_K/H_K as shown, calculated using Eqs. (4)–(7). Data points are from the kinetic Monte-Carlo computations. Other parameters used in (a) and (b): $H_K = 40\,$kOe, $\beta = 80$, and $V \sim 400\,\text{nm}^3$. Inset: An illustration of a typical distribution $D(H_{S,i})$.

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definition was in terms of the mean of $D_i(H)$. However, we confirmed that mean and median of are very close for the parameter set used here.
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