Preisach Models and FORC Diagrams: A Critical Appraisal from a Physicist's Perspective

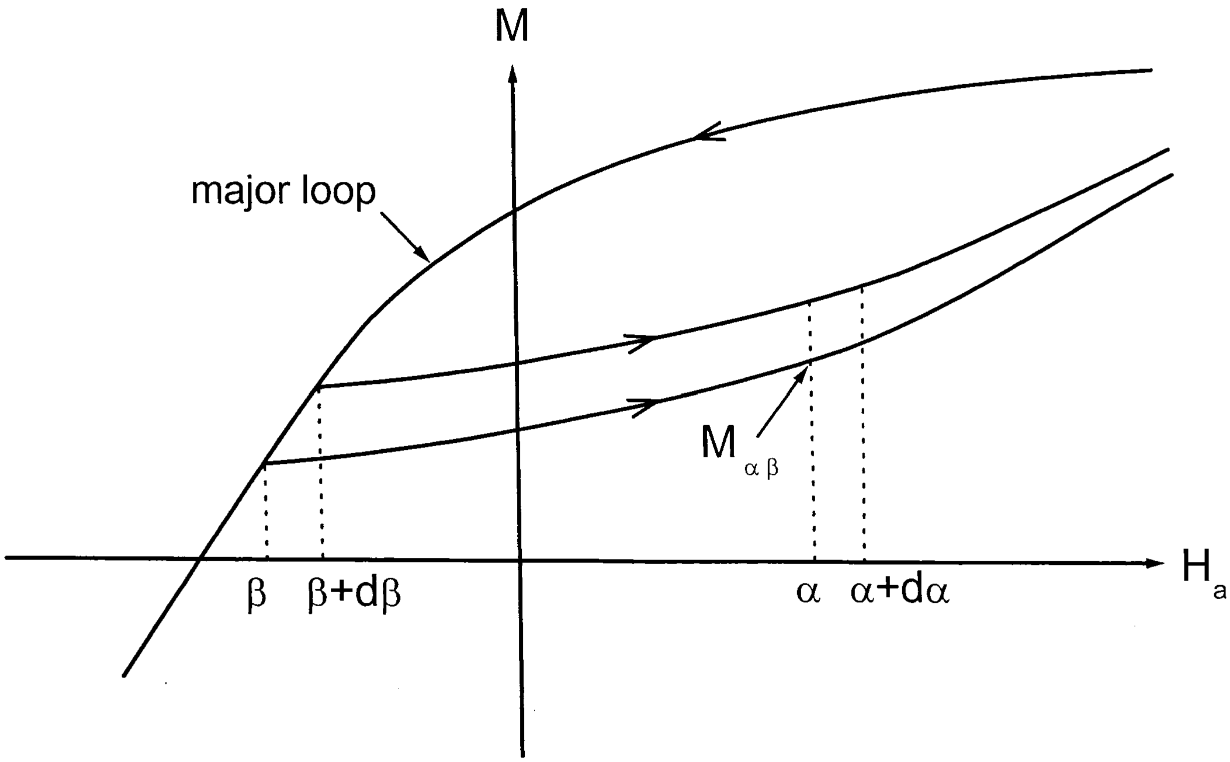
R M. Roshko

Department of Physics and Astronomy

University of Manitoba

Winnipeg, Manitoba, Canada

First Order Reversal Curve (FORC)



FORC Distribution

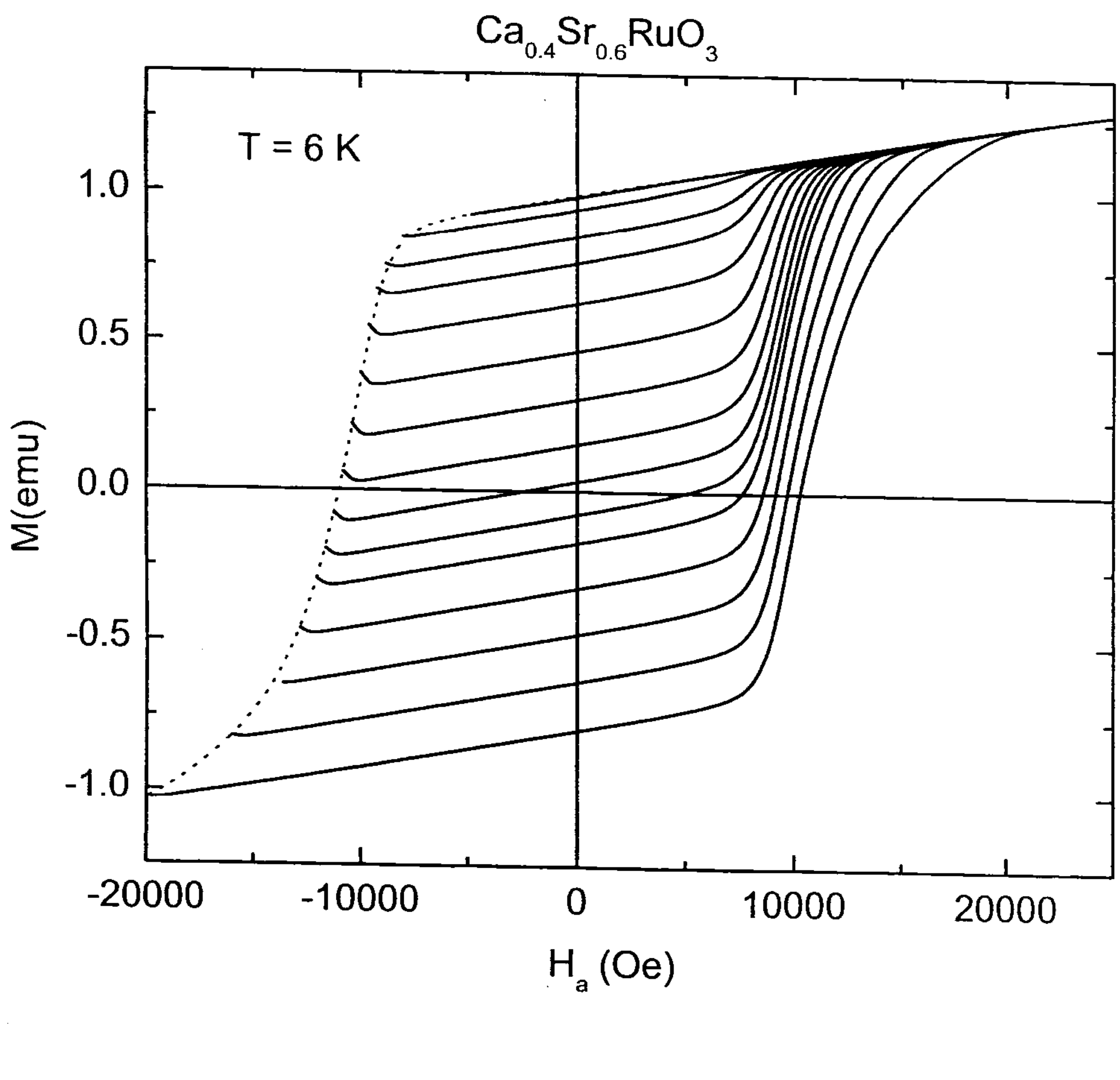
$$\rho(\beta,\alpha) = -\frac{\partial^2 M_{\alpha\beta}}{\partial\beta\,\partial\alpha}$$

transform variables: $H_d=(\alpha-\beta)/2$, $H_s=(\alpha+\beta)/2$

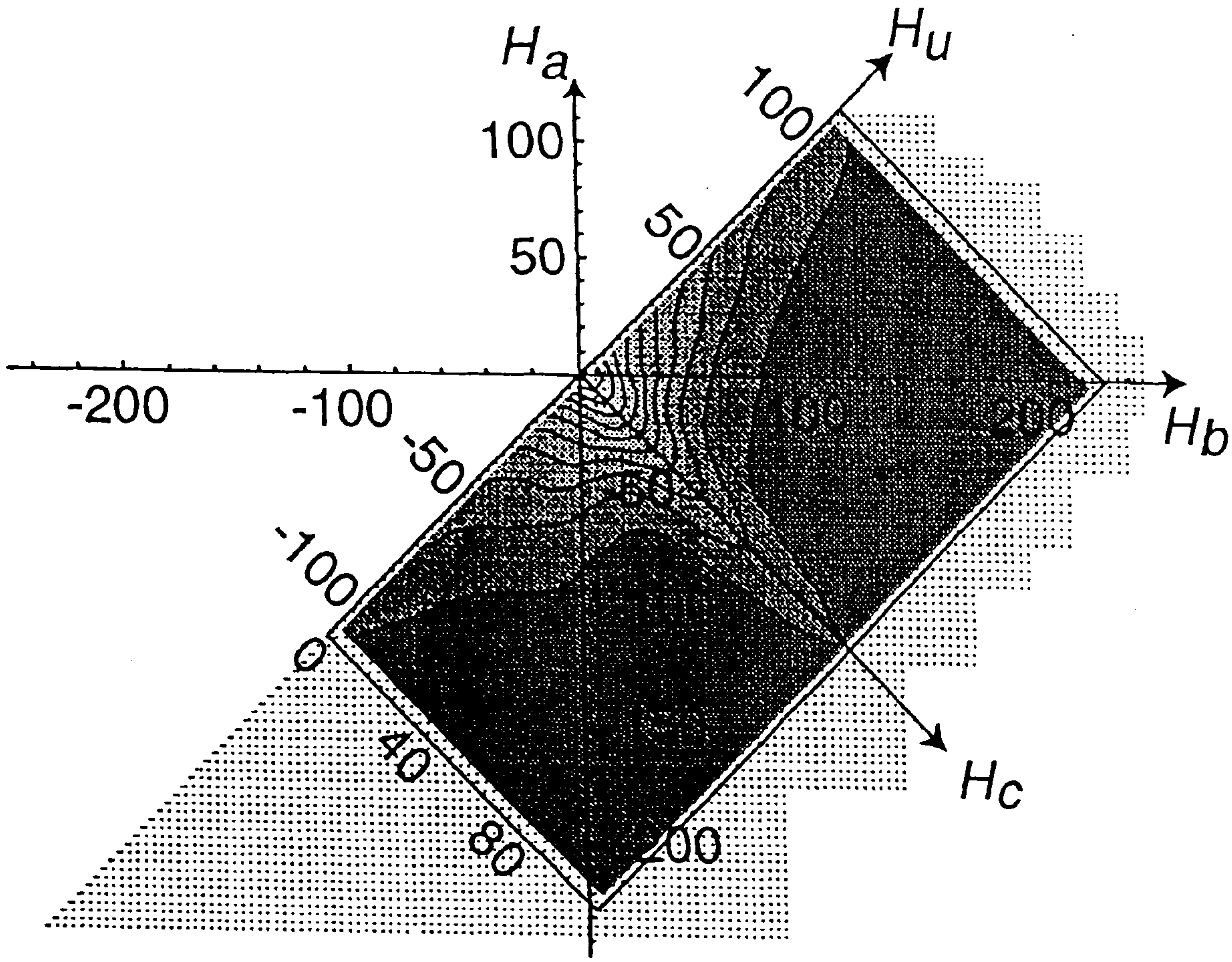
$$\rho(\beta,\alpha) \to \rho(H_d,H_S)$$

make contour plots of constant $\rho(H_d,H_S)$

A set of FORC's measured at T = 6K on an exchange-bond disordered ferromagnetic perovskite $Ca_{0.4}Sr_{0.6}RuO_3$ with a critical ordering temperature T_C = 70K:



A FORC diagram for a magnetite-bearing sediment



Roberts et al., J. Geophys. Res., Vol.105, p28461, 2000

But what physical insight is gained by measuring FORC's differentiating them, transforming the variables, and constructing contour plots? To answer these questions, it is necessary to appeal to the Preisach formalism, which is the source of inspiration for FORC diagrams.

Micromagnetics

The goal is a point-by-point reconstruction of the magnetization vector field $\vec{M}(\vec{r})$ with a spatial resolution which is intermediate between interatomic distances and some characteristic length scale over which the magnetization changes significantly. The material is subdivided into volume elements dV, each with a local magnetization vector $\vec{M}(\vec{r}) = M_S \hat{\vec{m}}(\vec{r})$ and a local anisotropy energy $f_{AN}(\vec{M}(\vec{r}))$.

The Gibbs free energy $G_L(\vec{M}(\vec{r}), \vec{H}_a, T)$ is a functional of the magnetization vector field $\vec{M}(\vec{r})$:

$$G_L(\vec{M}(\vec{r}), \vec{H}_a, T) = \int d^3r [A(\vec{\nabla}\vec{M}(\vec{r}))^2 + f_{AN}(\vec{M}(\vec{r}))$$

exchange energy   anisotropy energy

$$-\tfrac{1}{2}\vec{M}(\vec{r}) \cdot \vec{H}_{mag} - \vec{M}(\vec{r}) \cdot \vec{H}_a$$

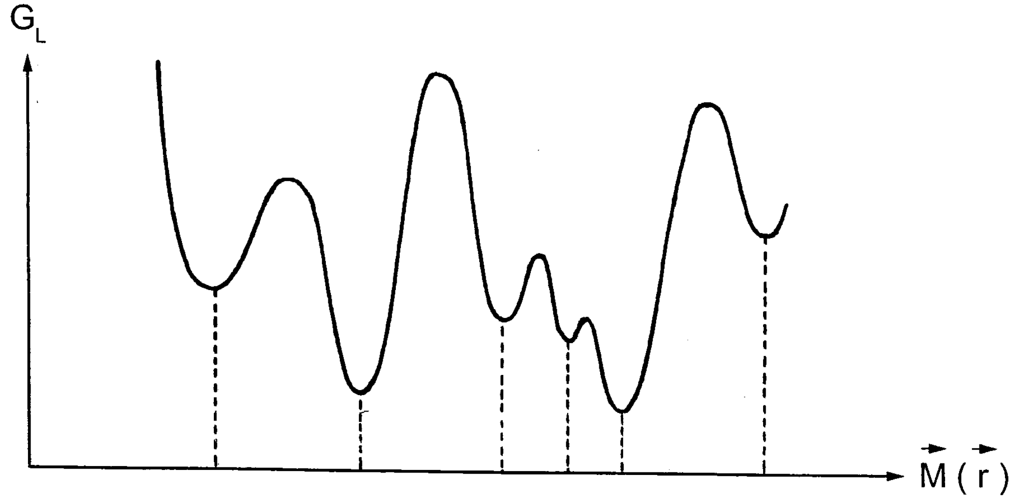
magnetostatic energy   applied field energy

where the magnetostatic field $\vec{H}_{mag}$ is given by

$$\vec{H}_{mag} = \int \frac{\rho_m(\vec{r} - \vec{r}\,')}{|\vec{r} - \vec{r}\,'|^3} dV' + \int \frac{\sigma_m(\vec{r} - \vec{r}\,')}{|\vec{r} - \vec{r}\,'|^3} dS'$$

where $\rho_m = -\vec{\nabla}' \cdot \vec{M}(\vec{r}\,')$ and $\sigma_m = \vec{M}(\vec{r}\,') \cdot \hat{n}$

As a consequence of the natural structural disorder which is present in all real materials, the micromagnetic energy functional $G_L(\vec{M}(\vec{r}), \vec{H}_a, T)$ yields an extremely complicated hypersurface when plotted in the infinite dimensional functional space of all magnetization configurations $\vec{M}(\vec{r})$, with an enormous number of local minima, maxima, and saddle points. Minimization of G_L yields the metastable states of the material under given external conditions $(\vec{H}_a, T)$



When the local metastable minimum occupied by the system is transformed into a saddle point by the action of the external field $\vec{H}_a$ the system experiences a Barkhausen instability and evolves rapidly toward a new metastable minimum.

The Preisach Formalism

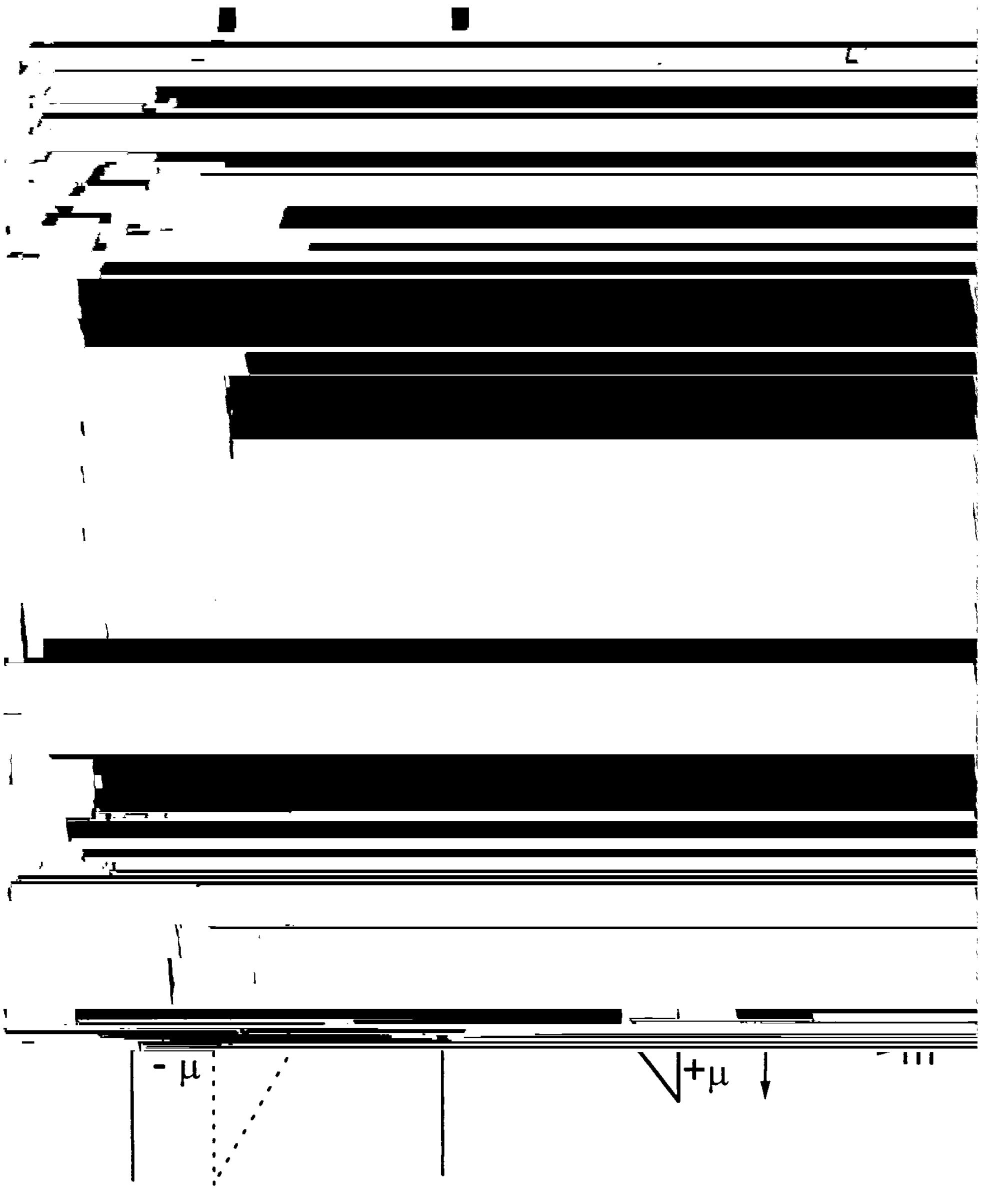
The Preisach approach postulates that the micromagnetic free energy functional can be decomposed into an ensemble of elementary two level (double well) subsystems (TLS). Each TLS incorporates the two characteristic energies fundamental to all metastable systems which exhibit Barkhausen instabilities and hysteresis:

(a) the free energy stored reversibly in a transition from one metastable state to another, and
(b) the energy dissipated irreversibly as heat during the transition

The Preisach formalism is an energy-based description of hysteresis, and does not require that the material under investigation be decomposable into discrete physical entities, like magnetic particles.

The relationship between the Preisach formalism and micromagnetics is analagous to that between thermodynamics and statistical mechanics.

irreversible subsystems



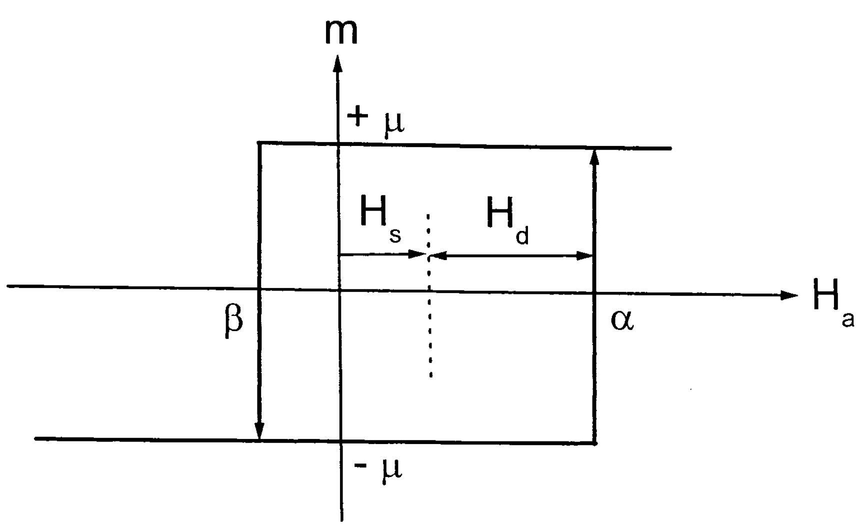
$W_{-}=\mu\alpha$

$W_{+}=-\mu\beta$

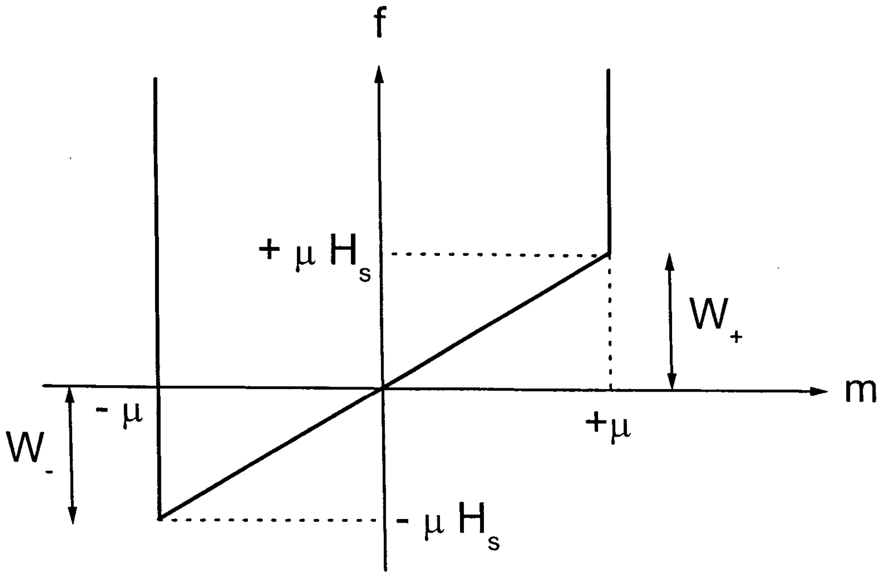
$W_d=\mu H_d$

$W_s=2\mu H_s$

$H_a>0$



reversible subsystems



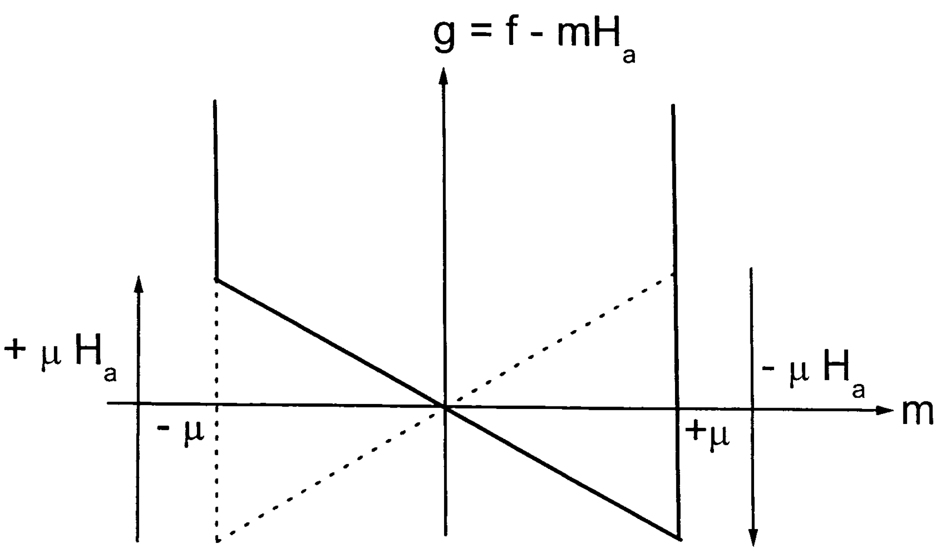
$W_- = \mu\alpha$

$W_+ = -\mu\beta$

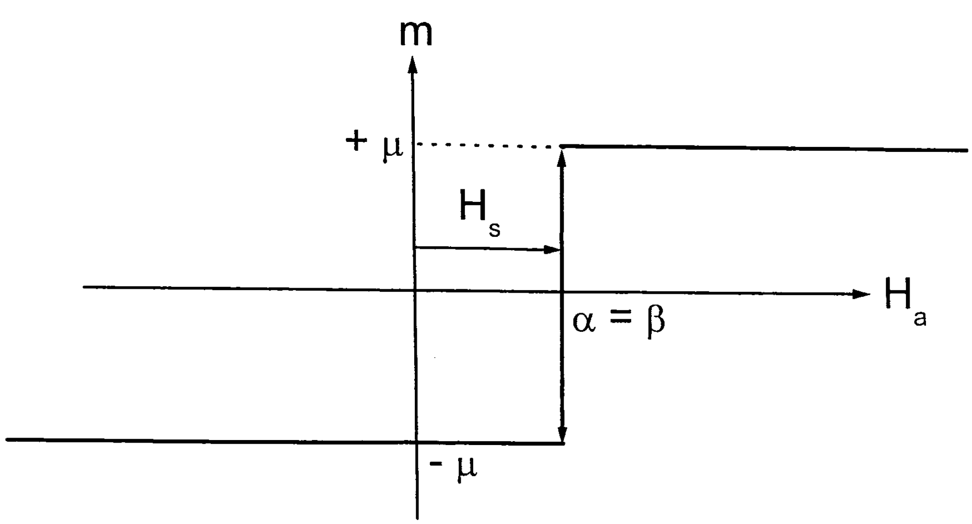
$\alpha = \beta$

$W_d = 0$

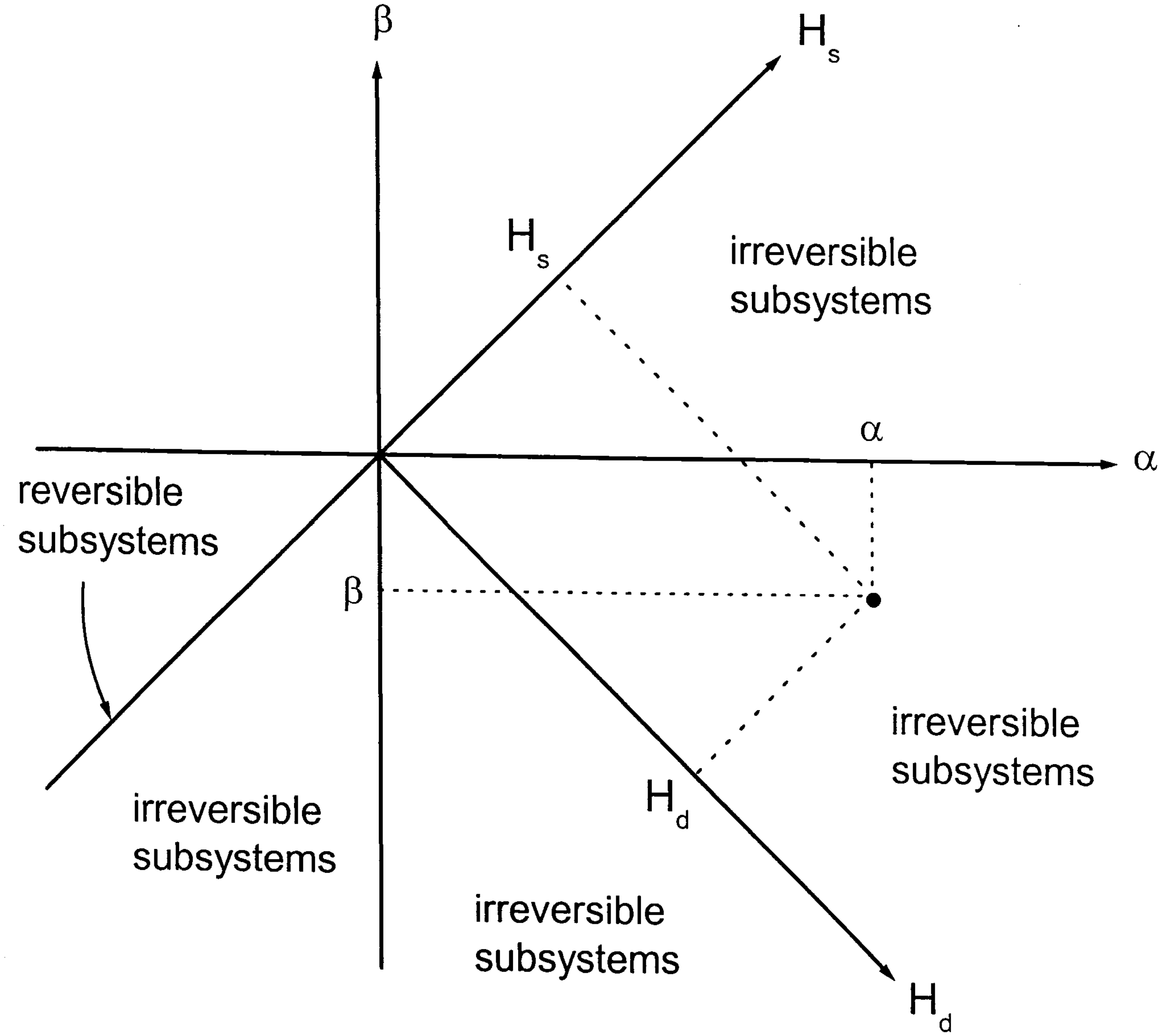
$W_s = 2\mu H_s$



$H_a > 0$



Preisach plane



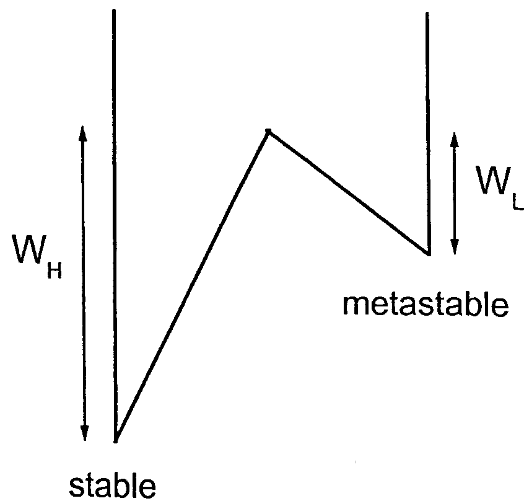
$$\alpha = H_s + H_d$$

$$\beta = H_s - H_d$$

$$H_d = \frac{\alpha - \beta}{2} = \frac{1}{\sqrt{2}}[\alpha \cos(\pi/4) - \beta \cos(\pi/4)]$$

$$H_s = \frac{\alpha + \beta}{2} = \frac{1}{\sqrt{2}}[\alpha \cos(\pi/4) + \beta \cos(\pi/4)]$$

energy barriers of a TLS in a field H_a:

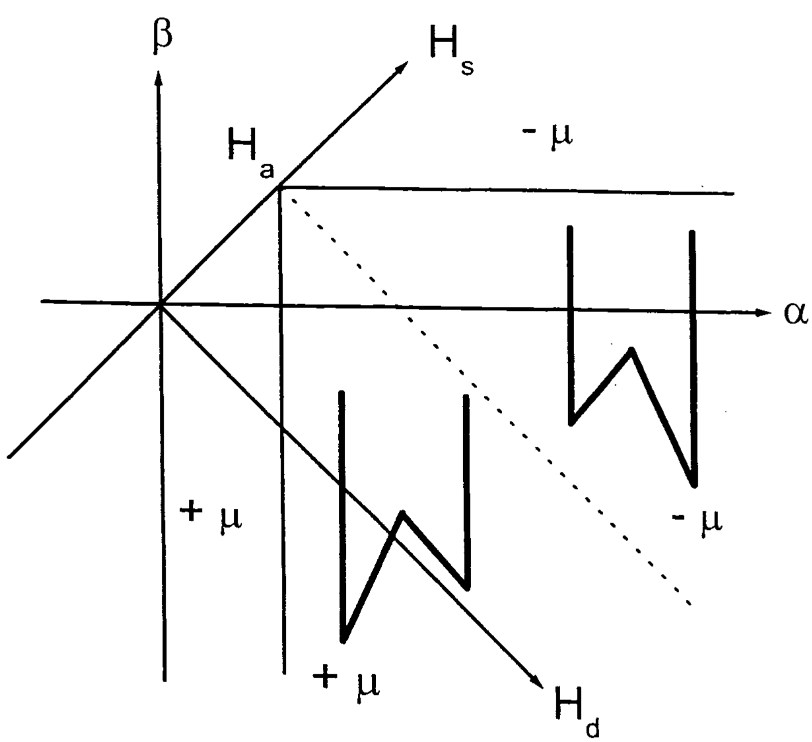


$W_H = \mu\ (H_d + |H_a - H_s|)$

$W_L = \mu\ (H_d - |H_a - H_s|)$

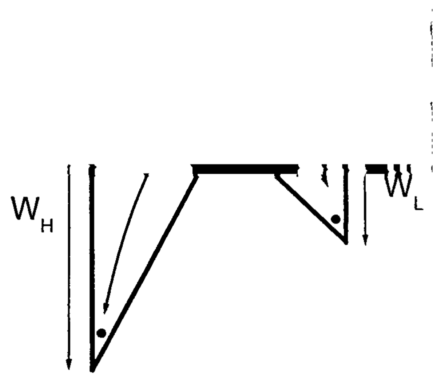
$W_T = kT\ ln\ (t_{exp} / \tau_0) = \mu\ H_T$

In the Preisach plane:



The Preisach model exhibits nonlocal memory, in the sense that there are many different metastable configurations of Preisach elements for each macroscopic system state, as specified by (M,Ha).

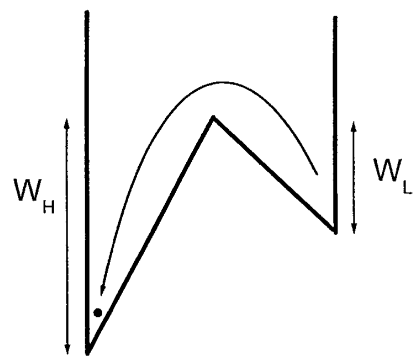
thermal equilibrium



$W_T > W_H$ and $W_T > W_L$

$m = \mu \tanh [\mu (H_a - H_s) / kT]$

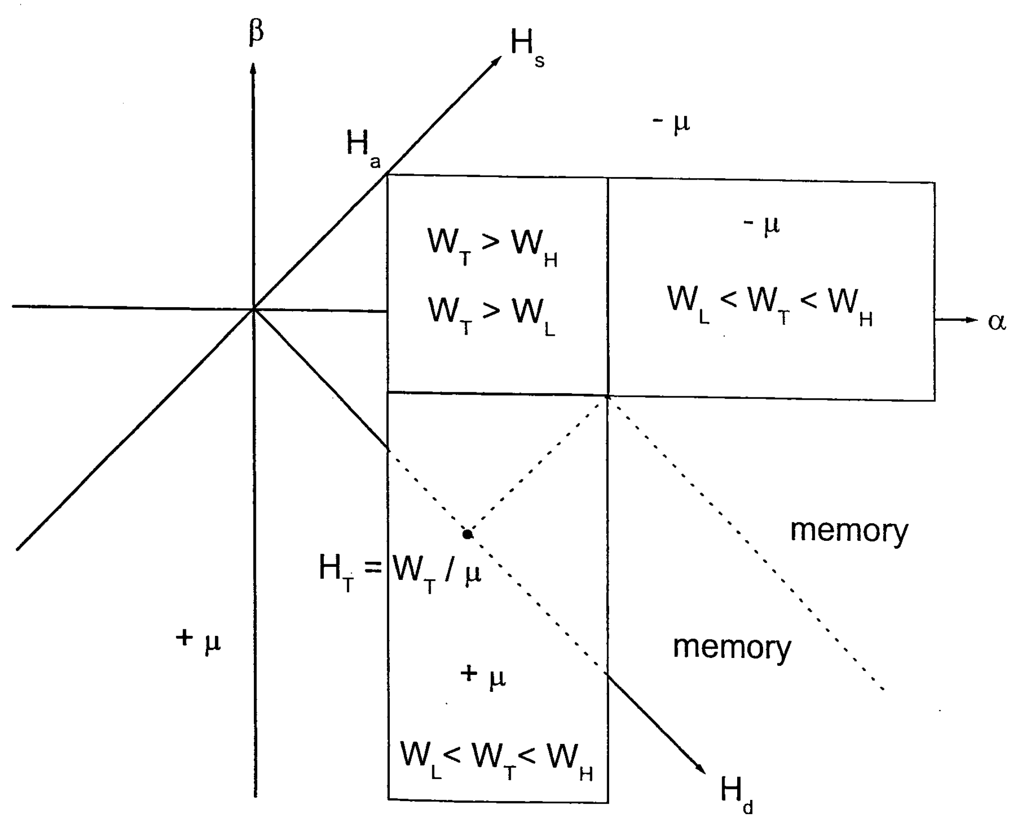
unidirectional thermal activation into the ground state



$W_L < W_T < W_H$

$m=+\mu$ or $m=-\mu$

In the Preisach plane:



If the irreversible TLS's have a density $n_{irrev}(H_d,H_s)$ such that

$$\int_0^{\infty} dH_d \int_{-\infty}^{\infty} dH_s \, n_{irrev}(H_d,H_s) = N_{irrev}$$

and if each TLS has a moment $\mu(H_d,H_s)$,then the contribution to the total moment from the irreversible TLS's is:

$$M_{irrev}(H_a,T) = \int_0^{\infty} dH_d \int_{-\infty}^{\infty} dH_s \, \underset{\substack{\uparrow \\ \text{history dependent}}}{(\pm 1)} \mu(H_d,H_s) \, n_{irrev}(H_d,H_s)$$

Since $\mu(H_d,H_s)$ $n_{irrev}(H_d,H_s)$ always appear as a product, we can define an average TLS moment $\bar{\mu}$ and a Preisach probability density $\rho_{irrev}(H_d,H_s)$ for irreversible TLS's as follows:

$$\mu(H_d,H_s) \, n_{irrev}(H_d,H_s) \Rightarrow [\bar{\mu}] \, [\, N_{irrev} \, \rho(H_d,H_s) \,]$$

Then

$$M_{irrev}(H_a,T) = N_{irrev} \bar{\mu} \int_0^{\infty} dH_d \int_{-\infty}^{\infty} dH_s (\pm 1) \underset{\substack{\uparrow \\ \text{Preisach density}}}{\rho_{irrev}(H_d,H_s)}$$

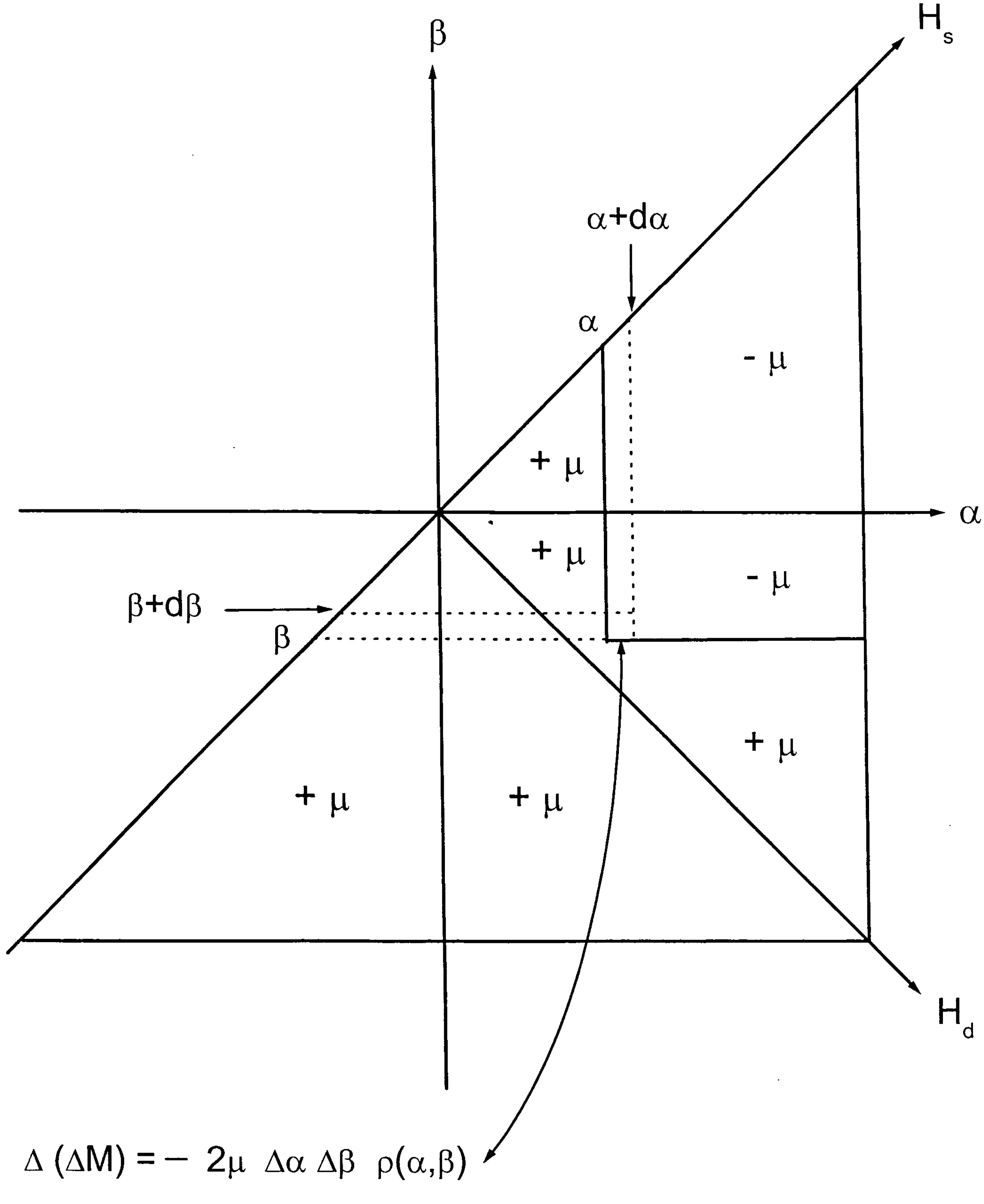
Similarly, for reversible TLS's

$$M_{rev}(H_a,T) = N_{rev}\bar{\mu}\int_{-\infty}^{\infty} dH_s(\pm 1)\rho_{rev}(H_s)$$

The total moment is:

$$M(H_a,T) = M_{irrev}(H_a,T) + M_{rev}(H_a,T)$$

Deriving the Preisach density from FORCs



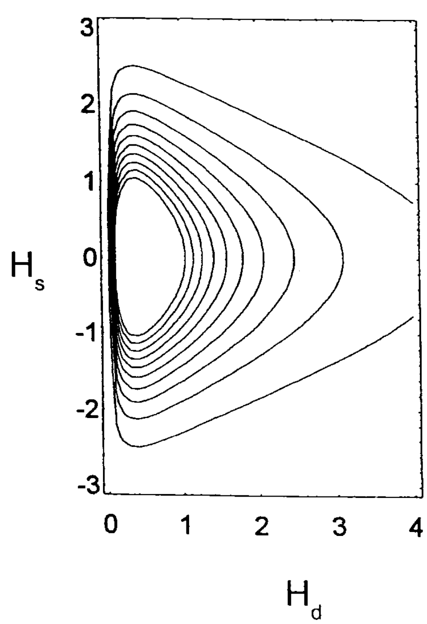
$$\Delta\,(\Delta M) = -\,2\mu\;\Delta\alpha\;\Delta\beta\;\;\rho(\alpha,\beta)$$

$$\rho(\alpha,\beta) = -\,\frac{1}{2\mu}\,\frac{\partial^2 M}{\partial\beta\,\partial\alpha}$$

For the classical Preisach model with no mean field effects, the FORC distribution is proportional to the Preisach density, and contour plots of these two distributions will have precisely the same shape. If the irreversible Preisach density is the product of a lognormal distribution of dissipation fields and a Gaussian distribution of bias fields:

$$\rho_{irrev}(H_d,H_s) = (2\pi\sigma_d^2 H_d^2)^{-1/2} \exp[-(\ln(H_d/H_{d0}))^2/2\sigma_d^2](2\pi\sigma_s^2)^{-1/2} \exp(-H_s^2/2\sigma_s^2)$$

then



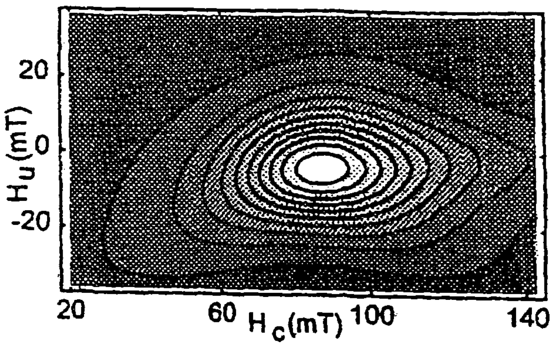
$H_{d0} = 1.0$

$\sigma_d = 1.0$

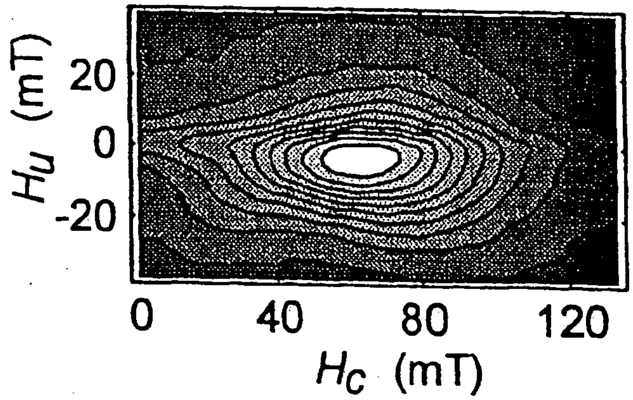
$\sigma_s = 1.0$

The contours are concentric, and symmetric about the H_s -axis.

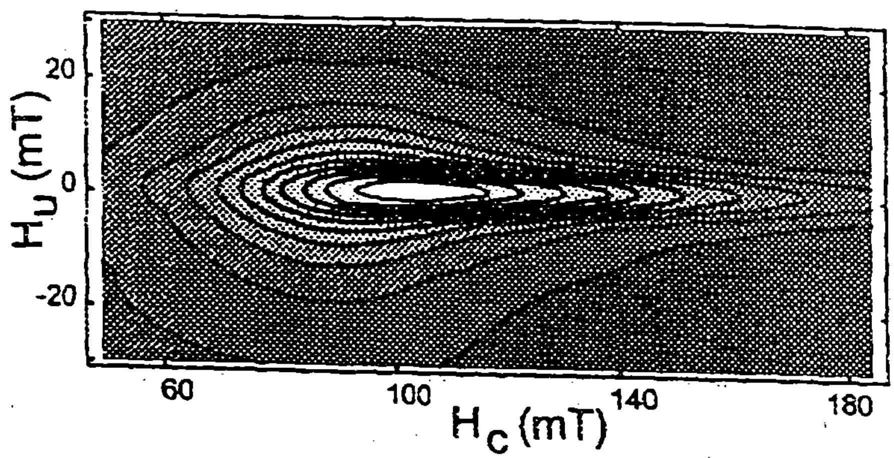
Measured FORC Diagrams



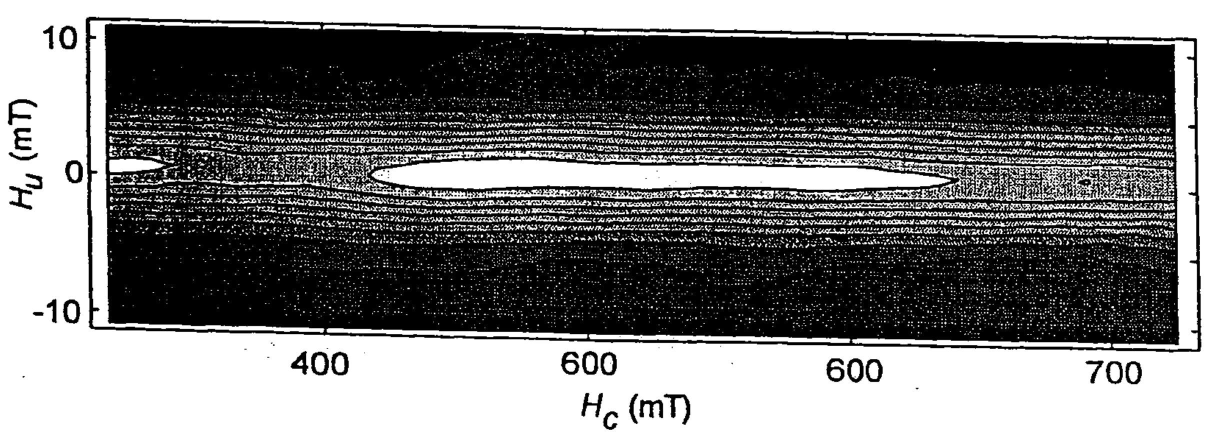
floppy disk



lake sediment



Kodak SD particles



Bulgarian brick

Return Point Memory and Congruency

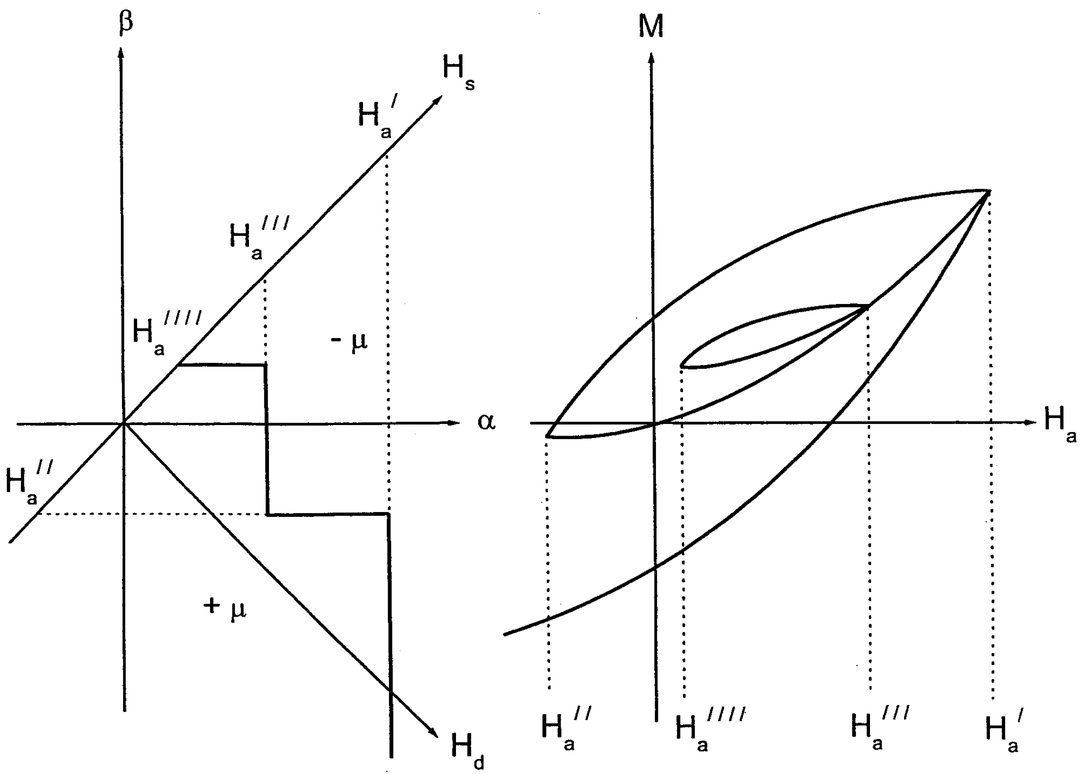
As a consequence of the internal mathematical structure of the Preisach formalism, all magnetization curves predicted by the model possess two properties, independent of the details of their shape:

(a) return point memory, which means that a field sweep from saturation to $H_a^{/}$, followed by field cycling from $H_a^{/}$ to $H_a^{//}$ and back to $H_a^{/}$ wipes out memory of the intermediate reversal to $H_a^{//}$.

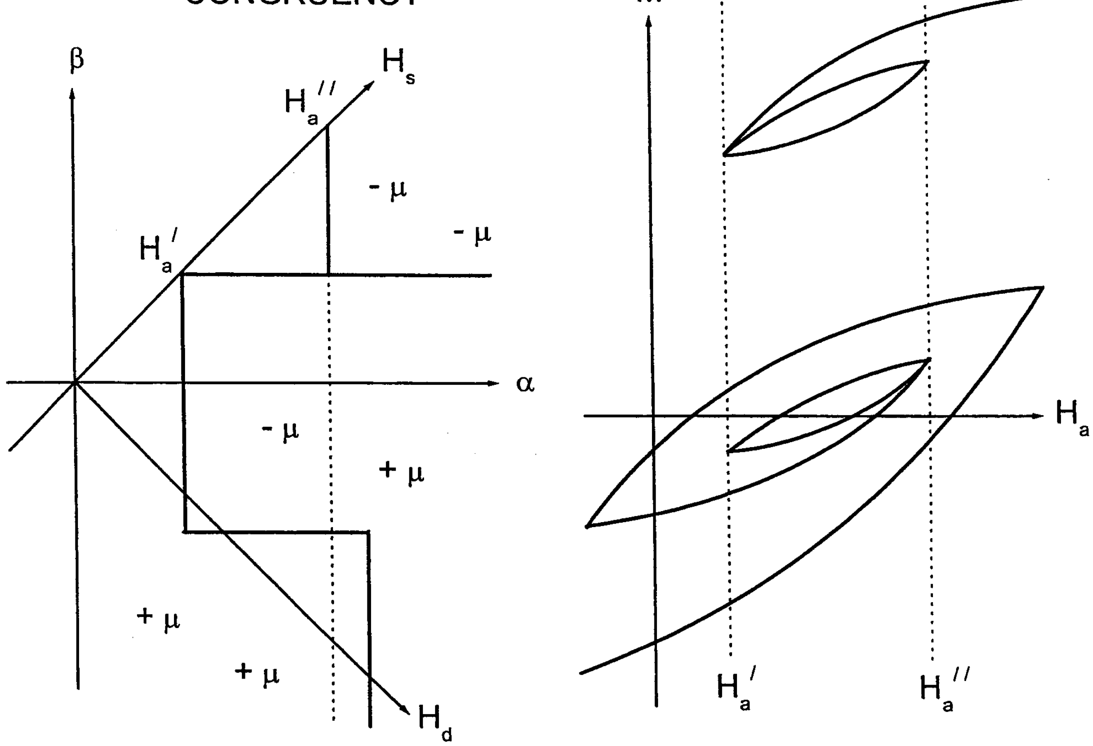
(b) congruency, which means that minor hysteresis loops bounded by the same upper and lower reversal fields are geometrically congruent, provided that the field sequence originated from saturation

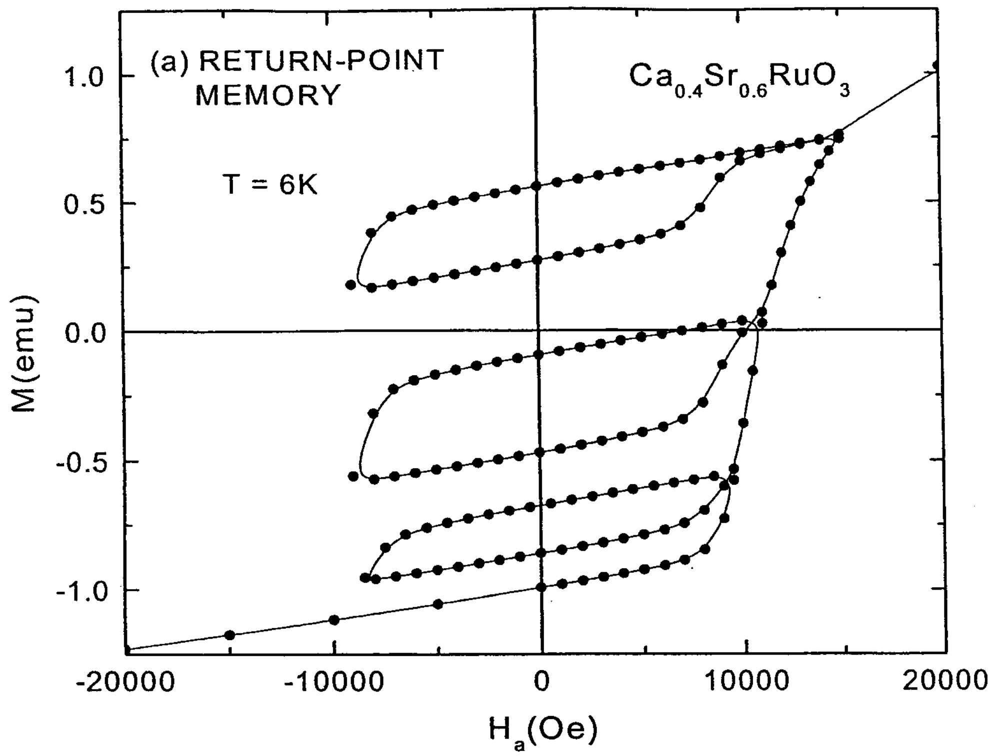
Return point memory and congruency are the macroscopic manifestation of the existence of the Preisach distribution. They are necessary and sufficient conditions for the decomposition of the micromagnetic free energy of a real hysteresis transducer into elementary TLS's.

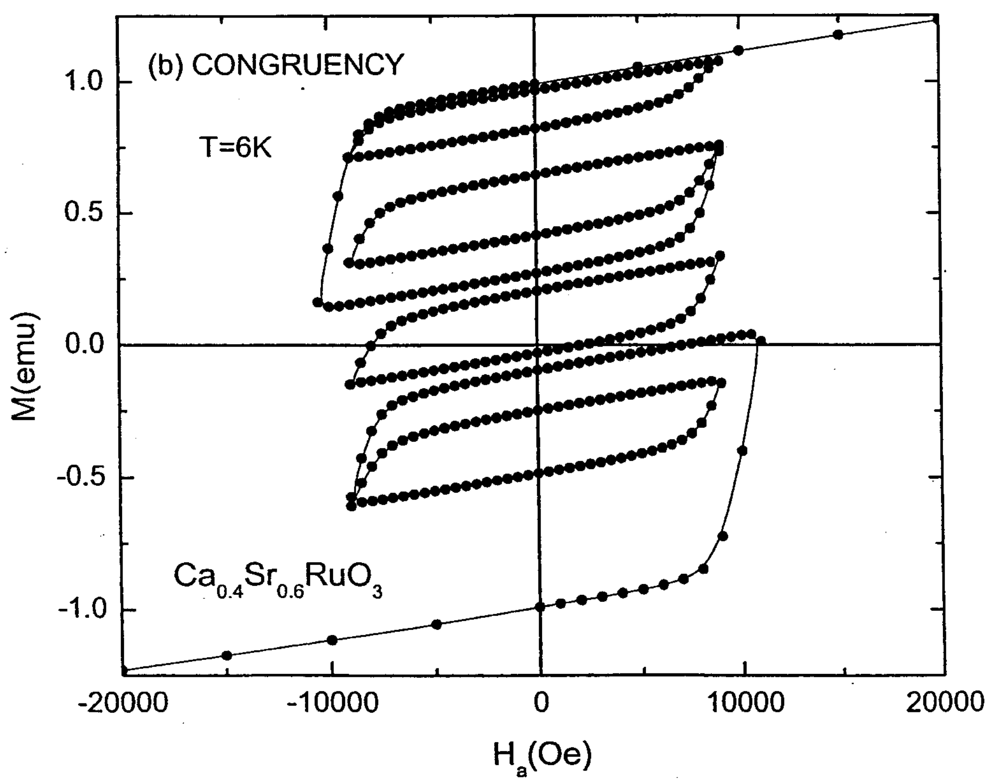
RETURN POINT MEMORY



CONGRUENCY




(a) RETURN-POINT MEMORY
Ca$_{0.4}$Sr$_{0.6}$RuO$_3$
T = 6K
M(emu)
H$_a$(Oe)
1.0
0.5
0.0
-0.5
-1.0
-20000
-10000
0
10000
20000


(b) CONGRUENCY
T=6K
Ca$_{0.4}$Sr$_{0.6}$RuO$_3$
M(emu)
H$_a$(Oe)
1.0
0.5
0.0
-0.5
-1.0
-20000
-10000
0
10000
20000

Mean Field Effects

Various generalizations of the Preisach formalism have been proposed. The most important of these are mean field models, according to which the excitation fields α and β of each TLS vary in proportion to the total induced moment M of the system as follows:

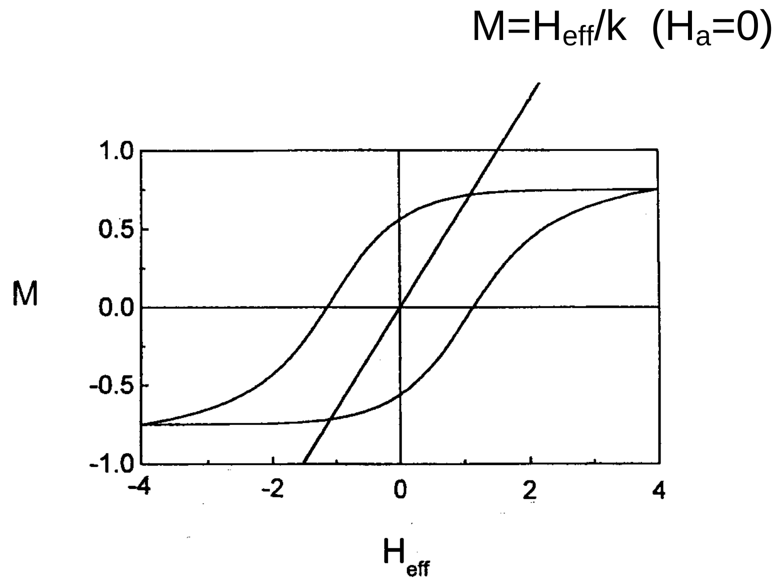
$$(\alpha, \beta) \rightarrow (\alpha - kM, \beta - kM)$$

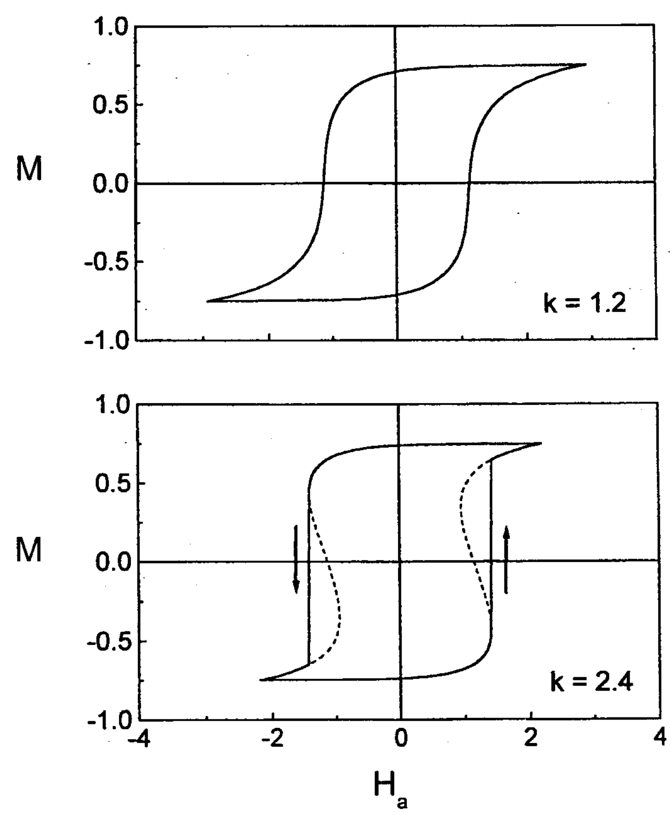
where k (>0 or <0) is the mean field parameter. This generates a global shift of the entire Preisach density parallel to the H_s -axis:

$$\rho(H_d, H_s) \rightarrow \rho(H_d, H_s + kM)$$

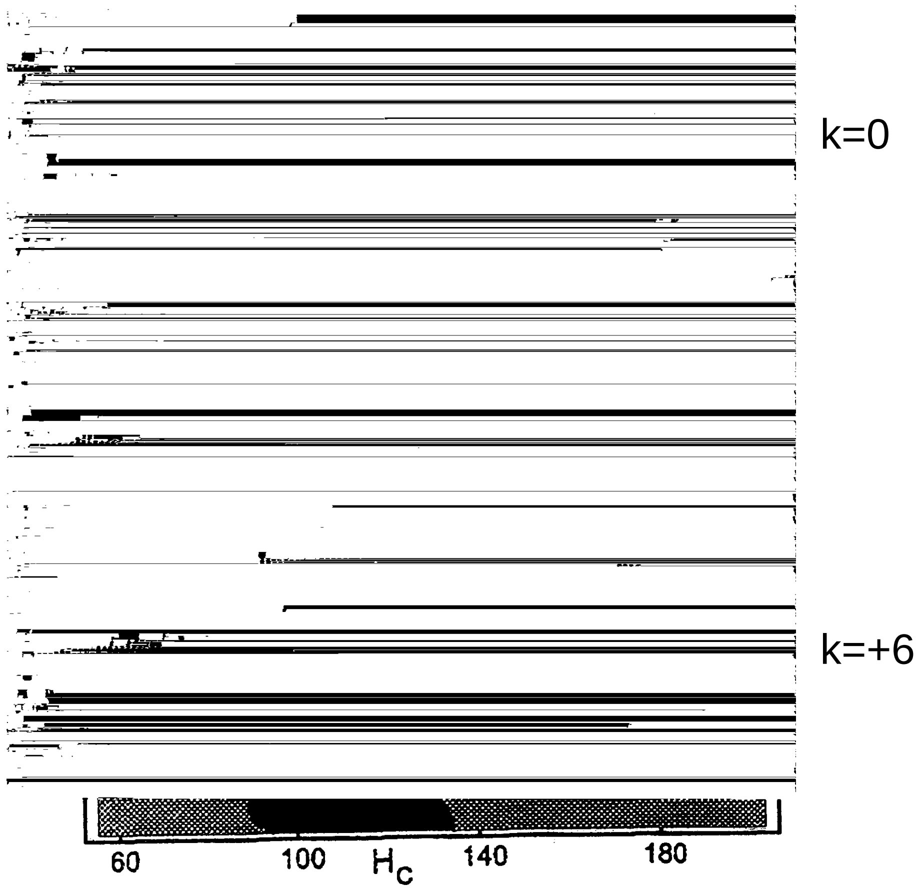
This is equivalent to assuming that the Preisach density $\rho(H_d, H_s)$ is stationary, and that each TLS experiences an effective magnetizing field $H_{eff} = H_a + kM$. The model will exhibit return point memory and congruency with respect to the effective field H_{eff}, but not with respect to the applied field H_a.

The hysteresis loop $M(H_a)$ is obtained from the loop $M(H_{eff})$ by a skew transformation, and minor loops exhibit skew congruency when plotted as a function of H_a.





When mean field effects are present, the FORC distribution is no longer proportional to the Preisach density, and FORC contours are distorted with respect to Preisach contours:

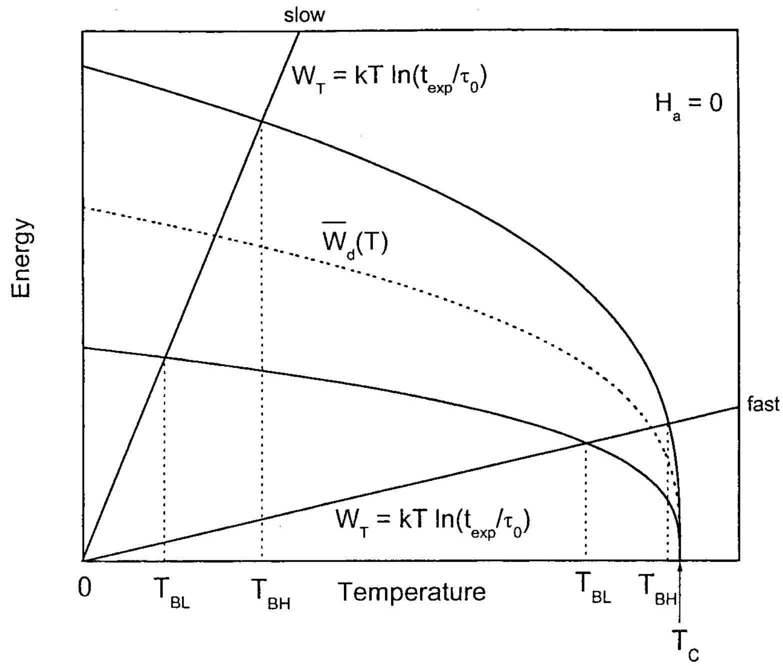


(Pike et al., J. Appl. Phys., Vol.85, p6660, 1999 with lognormal dissipation distribution and Gaussian bias distribution)

Temperature, Time, and Superparamagnetism

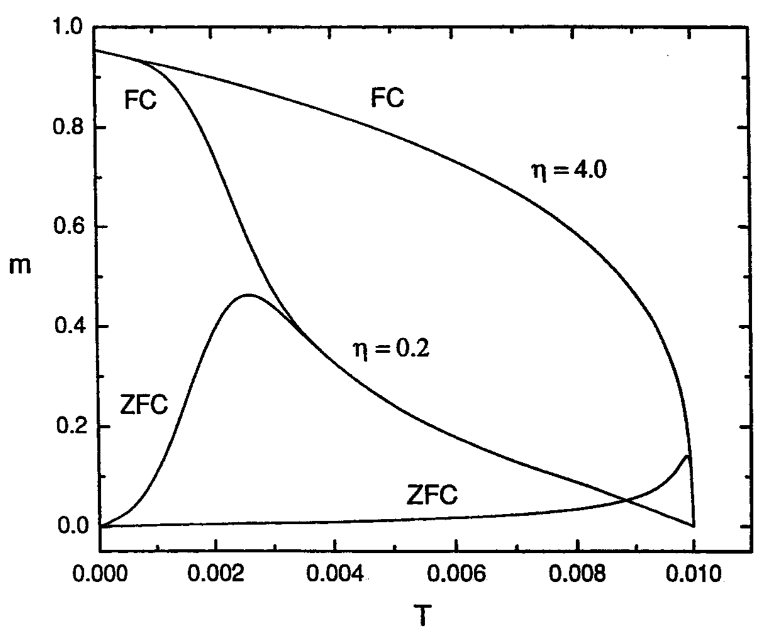
The physical origin of the temperature dependence of the magnetic response is potentially two-fold : (a) explicit modifications of the free energy landscape, and hence of the spectrum of metastable state excitation barriers, with temperature, and (b) thermal fluctuations, which reduce all energy barriers by the time dependent thermal fluctuation energy $W_T = kT \ln(t_{exp}/\tau_0)$, and which drive the system towards equilibrium. We thus assume that the system can be characterized at each temperature T by a set of intrinsic metastable state excitation barriers, which define the characteristics of the irreversible response in the hypothetical limit where all changes in temperature or field are accomplished instantaneously. Thermal fluctuations then distort the magnetic response with respect to this ideal limit by introducing a dependence on measuring time, and hence on sweep rate.

The relationship between these two temperature dependences, for a system with a critical ordering temperature T_c, can be illustrated as follows:

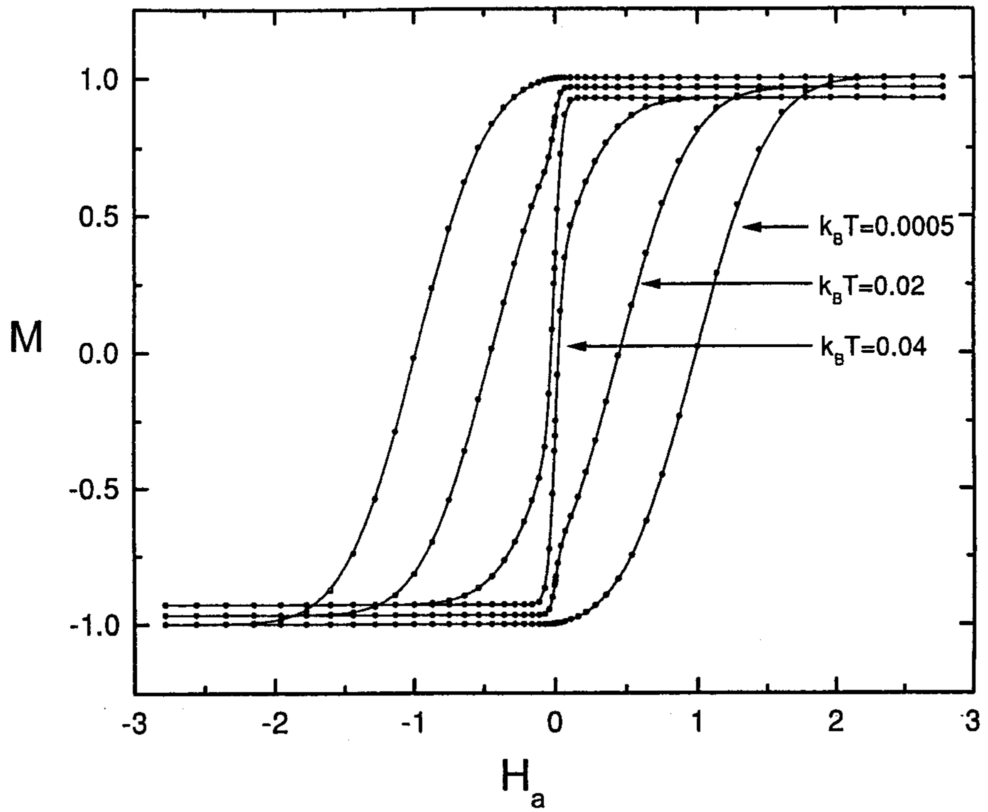


In the "fast' limit, the magnetic response is dissipation-dominated, while in the "slow" limit, it is fluctuation-dominated.

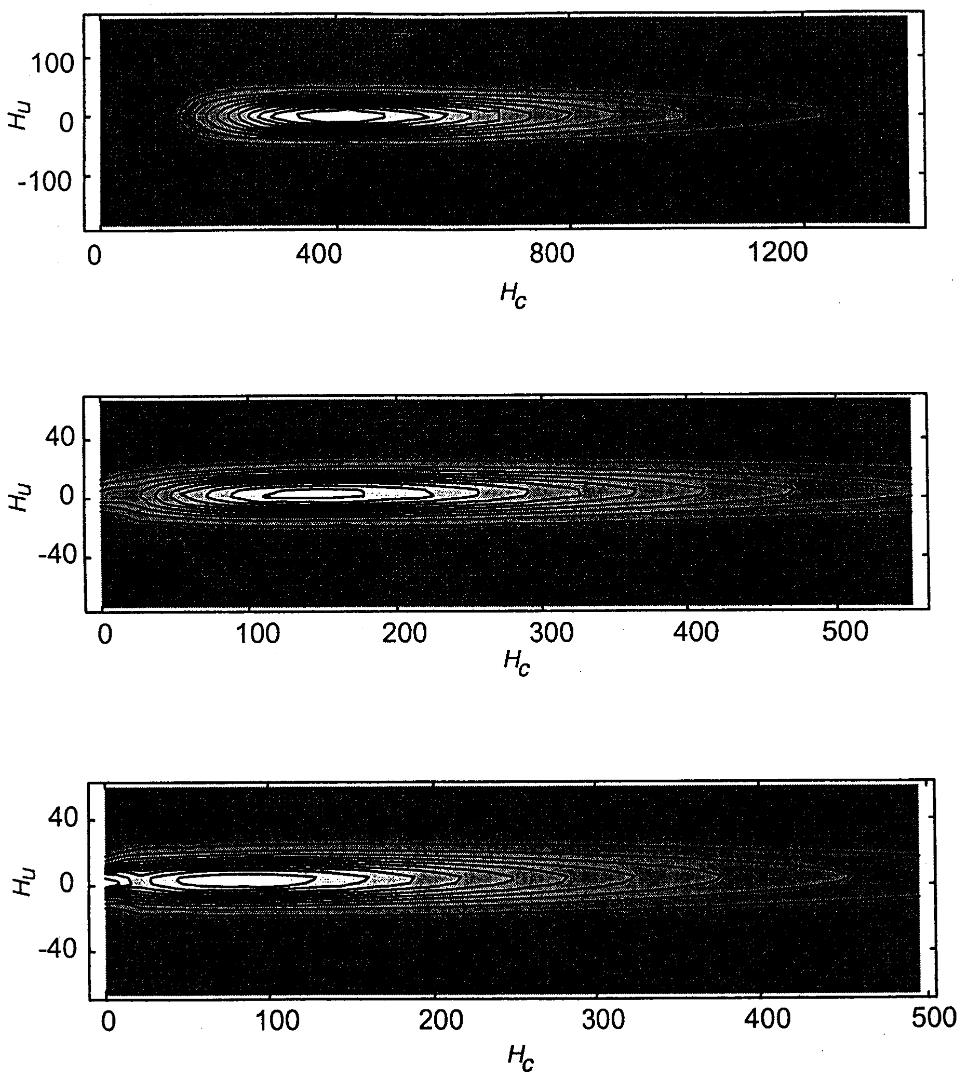
The Preisach model predicts the following temperature dependences for the field cooled (FC) moment and the zero field cooled (ZFC) moment in these two limits:



In the fluctuation-dominated regime, the hysteresis loop exhibits “wasp-waisting" at higher temperatures:



When thermal fluctuations are incorporated into the Preisach formalism, the model continues to exhibit return point memory and congruency. Thus, it is still possible to extract a Preisach density from FORCs. However, it must be emphasized that this procedure yields, not the original "intrinsic" Preisach density $\rho(H_d,H_s)$, but rather an effective Preisach density $\rho_{eff}(H_d,H_s)$ which defines an effective set of excitation barriers which reproduce all of the characteristics of the original model hysteresis loop. In this case, FORC diagrams will not yield an image of the intrinsic Preisach elements (individual particles, clusters of particles, domain wall pinning sites).

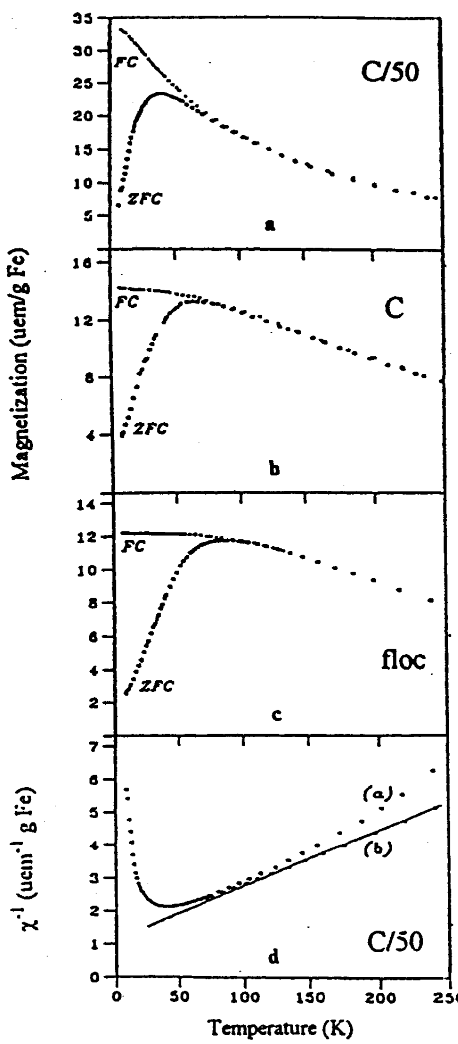


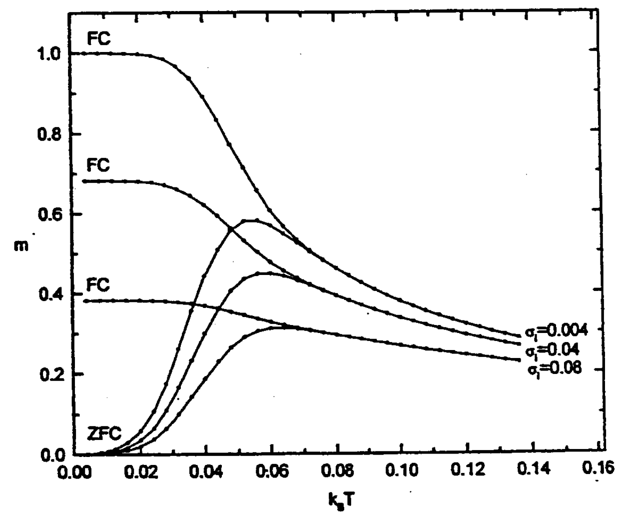
(Pike et al., Geophys. J. Int., Vol.145, p721, 2001)

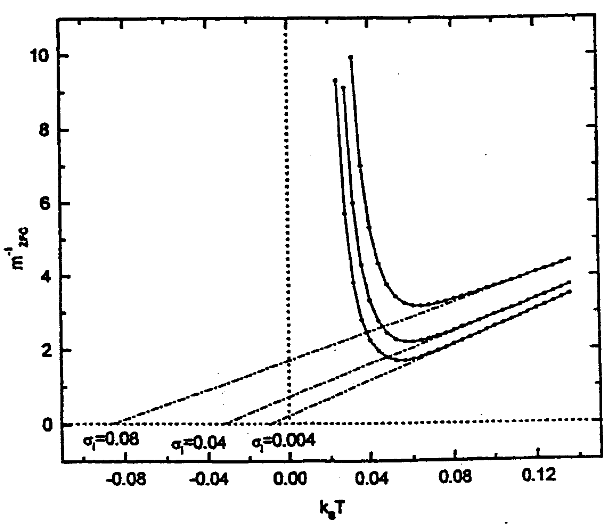
Are bias fields H_S a measure of interactions?

measurements of the FC and ZFC moment of polymer suspensions of γ-Fe_2O_3 particles (Prene at al., IEEE Trans. Magn., Vol.29, p.2658, 1993)

Preisach simulations of the FC and ZFC moment of a fluctuation-dominated system with different dispersions σ_s of bias fields







Concluding Thoughts

- FORC diagrams have been proposed as a sensitive diagnostic tool for characterizing and interpreting the hysteresis properties of magnetic materials, which are not constrained by the "artificial" physical postulates and mathematical structure of the Preisach formalism, and which are capable of identifying interparticle interaction effects, of discriminating between single domain, multidomain, and superparamagnetic grains in particle assemblages, and of recognizing and quantifying specific moment reversal mechanisms (DW pinning, vortex annihilation and nucleation,...)

- the program involves constructing model hysteresis nonlinearities based on "microscopic" physical pictures, cataloguing their structural signatures in a FORC diagram, and then searching for these features in measured FORC diagrams

- FORC diagrams also play a pivotal role in the Preisach formalism where, under certain conditions, they provide an undistorted image of the Preisach spectrum of characteristic energies $\rho(H_d,H_s)$

- however, the formalism is not limited to this simple correspondence, and generalizations have been proposed which retain the conceptual simplicity and symmetry and mathematical integrity of the original model, while predicting FORC contours which are distorted with respect to the Preisach contours, and which simulate intricate structural features observed in measured FORC diagrams

- furthermore, the TLS substructure of the Preisach formalism has the flexibility to transcend simple field histories, and to unite hysteresis and thermal effects into a common framework which can model the field and temperature dependence of a variety of standard experimental protocols and response functions, like the field cooled (FC) moment, the zero field cooled (ZFC) moment, the TRM, and the IRM.

- all FORC based identifications of interaction effects are Preisach based, in the sense that these interpretations rely exclusively on the Preisach TLS structure

- similarly, all FORC based identifications of SD and SP behaviour are Preisach based

- the language of FORC diagrams is implicitly TLS language, since the representation is always in terms of two characteristic fields, and in this sense, the entire construction implies some sort of decomposition into bistable entities

- many physically motivated generalizations of the Preisach formalism have been proposed (TLS's with a more refined free energy profile and with energy barriers which are nonlinear functions of H_a, applied field dependent and magnetization dependent Preisach densities, vector models), and their implications for FORC diagrams have yet to be explored in detail.