

MINISTÉRIO DA CIÊNCIA E TECNOLOGIA



**CBPF**

**CENTRO BRASILEIRO DE PESQUISAS FÍSICAS**

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**Notas de Física**

CBPF-NF-009/88

PHENOMENOLOGICAL THEORY OF FERROMAGNETS  
WITHOUT ANISOTROPY

by

W. Baltensperger and J.S. Helman

RIO DE JANEIRO  
1988

## ABSTRACT

For a ferromagnet without anisotropy, which may be a spin  $1/2$  lattice or an amorphous substance, the distribution of magnetization depends on the shape of the sample. The phenomenological equations of micromagnetics which contain the exchange energy and the magnetic energy are generalized to allow for a change in the magnitude of the magnetization. This permits the treatment of line singularities in the field free configurations with circular concentric magnetization lines. For an ellipsoid with a rotation axis the free energy of such an arrangement is compared with that of the homogeneously magnetized sample, which is the stable form for a sufficiently small sample (roughly with a radius  $\leq 20$  lattice constants).

Key-words: Magnetic order; Anisotropy; Domains.

PACS: 75.30.Kz, 75.30.Gw, 75.60.Ch.

## 1 INTRODUCTION

In the usual ferromagnets the anisotropy energy determines the domain structure, in particular the width of the Bloch walls. In the absence of an anisotropy energy this width becomes infinitely large. The magnetic structure then depends on the shape of the specimen. In this work aspects of this question are investigated.

A spin 1/2 ferromagnet with isotropic exchange cannot have any anisotropy. Similarly a spin 1 ferromagnet, where the spins are in sites of cubic symmetry, will not show anisotropy. Magnetic dipole-dipole couplings introduce anisotropies in the 1 K energy range. Known spin 1/2 ferromagnets are mostly Cu chlorides and bromides, e.g.  $(\text{CH}_3\text{NH}_3)_2\text{CuCl}_4$  with  $T_c = 8.9$  K or  $(\text{C}_2\text{H}_5\text{NH}_3)_2\text{CuBr}_4$  with  $T_c = 10.7$  K. These are layer structures. The substance  $\text{CuSeO}_4$  has a  $T_c = 26$  K and the ferromagnet  $\text{Cu}_2\text{SeO}_4$  has a  $T_c = 59$  K. In an amorphous ferromagnet the anisotropy of the various sites tends to cancel each other. Ferromagnetic metal glasses exist<sup>1</sup>, such as  $\text{Fe}_{80}\text{B}_{20}$  with  $T_c = 600$  K.

This work uses phenomenological equations which are often referred to as micromagnetics<sup>2-4</sup>. The treatment, however, is generalized to the case where the modulus of the local magnetization can differ from the saturation value.

The absence of anisotropy is also a property of ferrofluids<sup>5</sup>, which, however, are more complex since the density of magnetic particles can vary in space.

The paper will consider two simple magnetic structures in samples with rotational symmetry and compare the relative stability of these structures. Only equilibrium states will be studied.

## 2 THE MODEL

We use a phenomenological description of the system with a continuously varying magnetization  $\vec{M}(\vec{x})$ . The free energy density, which is a function of  $\vec{M}(\vec{x})$ , has the contributions

$$F = F_1 + F_2 + F_3 \quad (1)$$

$F_1$  is the leading term of the exchange energy due to inhomogeneities of wavelengths that are long compared to the lattice constant<sup>6</sup>,

$$F_1 = C \sum_{\alpha, \nu} \frac{\partial M_\alpha}{\partial x_\nu} \cdot \frac{\partial M_\alpha}{\partial x_\nu} \quad (2)$$

It is derived from a Heisenberg exchange energy  $J \sum \vec{S}_i \cdot \vec{S}_j$ , where the sum extends over the nearest neighbor bonds. The spin density is related to the magnetization by  $\vec{M}/(g \mu_B)$ , where  $g$  is the magneto-mechanical ratio and  $\mu_B$  the Bohr magneton. Then for a simple cubic lattice  $C = J Z a^5 / (g \mu_B)^2$ , where  $Z=6$  is the coordination number and  $a$  the lattice constant.

The modulus of the magnetization  $M = |\vec{M}(\vec{x})|$  can differ from  $M_0$ , the equilibrium value in an infinitely long cylinder in the absence of a magnetic field. The corresponding increase in the free energy density is written in the form

$$F_2 = \frac{1}{2\chi_0} (M - M_0)^2 \quad (3)$$

Here  $\chi_0^{-1} = (\partial^2 F / \partial^2 M)_T = (\partial H / \partial M)_T$  is the reciprocal high field susceptibility at temperature  $T$ .

In an ellipsoid the equilibrium magnetization is directed a-

long the longest axis. The magnetic field inside the sample is  $\vec{H} = -N\vec{M}$ , where  $N$  is the demagnetization factor. This leads to an additional contribution to the free energy density<sup>4</sup>

$$F_3 = \frac{1}{2} N M^2. \quad (4)$$

Actually there is a problem with this expression in connection with the phenomenological property (3). Electrodynamics defines the magnetic energy as  $\int_{B_0} \vec{H} \cdot d\vec{B}$ . The question is which value of  $B_0$  to use, when the phenomenological description of the substance presupposes a magnetization.

For the purpose of the problem at hand, we shall simply consider the magnetic energy to vanish in the field free magnetic configuration. When the ellipsoidal sample is homogeneously magnetized, on the other hand, the magnetization will very closely be  $M_0$ , for which case the micromagnetic expression (4) holds.

When the sample has a general shape, there is no local relation between  $\vec{H}$  and  $\vec{M}$ , so that no free energy density exists which depends on the local magnetization only.

### 3 FIELD FREE MAGNETIC CONFIGURATION

For a rotationally symmetric sample there is an obvious way to avoid a magnetic field altogether ( $\vec{H} \equiv 0$ ), namely with circular concentric magnetization lines (Fig.1). In order to compare the relative stability of this arrangement with that of a homogeneous magnetization, the corresponding total free energies will be calculated.

With cylindrical coordinates  $r, \phi, z$  the free energy density

(1) takes the form

$$F(r, M(r, z), T) = C \left[ \left( \frac{\partial M}{\partial r} \right)^2 + \frac{M^2}{r^2} + \left( \frac{\partial M}{\partial z} \right)^2 \right] + \frac{1}{2\chi_0} (M - M_0)^2 \quad (5)$$

and the free energy of the sample is

$$F = 2\pi \int F(r, M(r, z), T) r \, dr \, dz \quad (6)$$

where the integral extends over the volume of the sample.  $F$  is a functional to be minimized with respect to  $M(r, z)$ . This variational problem leads to the Euler equation<sup>7</sup>

$$\frac{\partial}{\partial r} \left[ \frac{\partial (rF)}{\partial (\partial M / \partial r)} \right] + \frac{\partial}{\partial z} \left[ \frac{\partial (rF)}{\partial (\partial M / \partial z)} \right] - \frac{\partial (rF)}{\partial M} = 0 \quad (7)$$

with prescribed boundary values  $M(r, z)$  at the surface and the symmetry axis of the sample. The resulting partial differential equation for  $M(r, z)$  is not separable and we use an approximation in which the  $z$ -dependence of  $M$  is at first neglected. This is equivalent to have the sample sliced in disks of width  $\Delta z$  and radius  $R(z)$  and the integral

$$F_z = 2\pi \int_0^{R(z)} F(r, M(r), T) r \, dr \quad (8)$$

minimized for each  $z$ . Then, the Euler equation (7) reduces to the ordinary inhomogeneous differential equation

$$\frac{d^2 M}{dr^2} + \frac{1}{r} \frac{dM}{dr} - M \left( \frac{1}{r^2} + \frac{1}{2C\chi_0} \right) = - \frac{M_0}{2C\chi_0} \quad (9)$$

with fixed boundary values  $M(0)$  and  $M(R(z))$ . Equation (9) becomes,

with the dimensionless variables

$$\rho = \kappa r \quad (10)$$

$$\mu = M/M_0$$

where  $\kappa = (2C\chi_0)^{-(1/2)}$ , (11)

$$\ddot{\mu} + \frac{\dot{\mu}}{\rho} - \mu(1/\rho^2 + 1) = -1 \quad (12)$$

where  $\dot{\mu} = d\mu/d\rho$ .

$\mu(0)$  must be set equal to zero to avoid a divergent  $F_z$ . The value of  $\mu_p = \mu(P)$ , where  $P = \kappa R$  is the largest  $\rho$ , will be fixed arbitrarily at first.  $F_z$  becomes a function of  $\mu_p$ , the minimum of which will be determined in a second step.

The solution of (12) with the prescribed boundary conditions is<sup>6</sup>

$$\begin{aligned} \mu(\rho) = & \mu_p \frac{I_1(\rho)}{I_1(P)} + \left[ K_1(\rho) - \frac{K_1(P)}{I_1(P)} I_1(\rho) \right] \int_0^\rho I_1(\xi) \xi d\xi \\ & + I_1(\rho) \int_\rho^P \left[ K_1(\xi) - \frac{K_1(P)}{I_1(P)} I_1(\xi) \right] \xi d\xi \end{aligned} \quad (13)$$

where  $I_1$  and  $K_1$  are the modified Bessel functions<sup>9</sup>. The first term, which contains  $\mu_p$ , is the solution of the homogeneous equation which satisfies the boundary conditions. The second and third terms together are the solution of the inhomogeneous equation which vanishes at the boundaries.

Expressed in dimensionless variables  $F = C M_0^2 \kappa^2 f(\rho)$  with

$$f(\rho) = \dot{\mu}^2 + \mu^2/\rho^2 + (1-\mu)^2. \quad (14)$$

Therefore, using (12),

$$\rho f(\rho) = \frac{d}{d\rho} (\rho \mu \dot{\mu}) + (1-\mu)\rho. \quad (15)$$

For a rotationally symmetric ellipsoid of radius  $\alpha$  and semiaxis  $\beta$ , both in units of  $\kappa^{-1}$ , the total free energy of the state with rotational magnetization becomes in this zeroth approximation

$$F^{(0)} = 2C_1 \int_0^\beta d\zeta \int_0^{\alpha(1-\zeta^2/\beta^2)^{1/2}} f(\rho) \rho d\rho \quad (16)$$

where  $C_1 = 2\pi C M_0^2 / \kappa$ .

With (13) and the relation for the Wronskian  $\dot{I}_1(\rho) K_1(\rho) - \dot{K}_1(\rho) I_1(\rho) = 1/\rho$ ,  $\dot{\mu}(P)$  becomes

$$\dot{\mu}(P) = \mu_P \frac{\dot{I}_1(P)}{I_1(P)} - \frac{1}{P I_1(P)} \int_0^P I_1(\zeta) \zeta d\zeta. \quad (17)$$

The condition

$$\partial F^{(0)} / \partial \mu_P = 0 \quad (18)$$

yields

$$\mu_P = \frac{1}{P \dot{I}_1(P)} \int_0^P I_1(\zeta) \zeta d\zeta. \quad (19)$$

Thus from (17)  $\dot{\mu}(P) = 0$ , which means that (18) is equivalent to this boundary condition. In view of (15)  $F^{(0)}$  given by (16) reduces to (using the change of variables  $P = \alpha(1-\zeta^2/\beta^2)^{1/2}$ )

$$F^{(0)} = \frac{2C_1 v}{\alpha^3} \int_0^\alpha dP \frac{P}{\sqrt{\alpha^2 - P^2}} g(P) \quad (20)$$



$$\text{where } v = \beta \alpha^2 \text{ and } g(P) = \int_0^P (1-\mu) \rho \, d\rho. \quad (21)$$

We take now into consideration the contribution to the free energy,  $F^{(1)}$ , coming from the  $z$ -dependence of the magnetization. From Eqs. (6) and (7) and using the dimensionless variables (11) we have

$$F^{(1)} = 2C_1 \int_0^\beta d\zeta \int_0^P \left[ \frac{\partial \mu_P(\rho)}{\partial z} \right]^2 \rho \, d\rho. \quad (22)$$

Here we write  $\mu_P(\rho) \equiv \mu(\rho)$  to emphasize that the  $z$ -dependence of  $\mu$  comes entirely from  $P(\zeta)$ . From (13) and (17), and using the relation for the Wronskian of the Bessel functions, we have

$$\frac{\partial \mu_P(\rho)}{\partial \zeta} = \frac{I_1(\rho)}{I_1(P)} \frac{d\mu_P}{dP} \frac{dP}{d\zeta}, \quad (23)$$

and using properties of the Bessel functions we can write (22) in the form

$$F^{(1)} = \frac{C_1 v \eta^2}{\alpha^4} \int_0^\alpha dP P (\alpha^2 - P^2)^{1/2} \cdot \left( 1 - \mu_P(1 + 1/P^2) \right)^2 \left[ \frac{I_1^2(P)}{I_1^2(P)} (1 + 1/P^2) - 1 \right] \quad (24)$$

with  $\eta = \alpha/\beta$ .

Fig. 2 shows the dimensionless magnetization  $\mu$  as a function of the dimensionless distance  $\rho$  for various radii  $P$ .

Fig. 3 shows the dimensionless free energies  $F^{(0)}$  and  $F^{(1)}$  in units of  $(2/3)C_1 v$  as a function of  $\alpha$ . Asymptotically (for  $\alpha \gg 1$ )  $F^{(0)} \propto \log \alpha / \alpha^2$  and  $F^{(1)} \propto 1/\alpha^2$  at constant  $\eta$ . The exact solution  $F$  certainly satisfies the inequalities:  $F^{(0)} < F < F^{(0)} + F^{(1)}$ .

## 4 HOMOGENEOUSLY MAGNETIZED SAMPLE

For an ellipsoid without external magnetic field, the formula (4) gives a free energy density (1)

$$F_m = \frac{N M^2}{2} . \quad (25)$$

The total free energy of the magnetized ellipsoid with radius  $\alpha$  and semiaxis  $\beta$  (in units of  $\kappa^{-1}$ ) becomes

$$F_m = C_2 \alpha \beta^2 N \quad (26)$$

where  $C_2 = (2\pi/3)M_0^2/\kappa^3 = (2/3)\chi_0 C_1$ . Therefore to compare the free energies of the parallel and the rotational magnetized states, it is convenient to write

$$F_m = (2/3)C_1 \alpha^2 \beta N \chi_0 . \quad (27)$$

It has been shown<sup>3</sup> that for particles of any shape, which are sufficiently small so that the magnetization is homogeneous, the magnetic energy of the particle is equal to that of a suitable defined ellipsoid. Since the free energy of the state with rotational magnetization can be evaluated for any rotationally symmetric body (with  $\alpha = \alpha(\zeta)$  in (16)), this theorem can be used to compare the free energies.

## 5 A NUMERICAL EXAMPLE

A numerical comparison between  $F_r = F^{(0)} + F^{(1)}$  and  $F_m$  involves the dimensionless numbers  $\eta = \alpha/\beta$ ,  $\chi_0$  and  $\kappa a$ . From (11)

$$(\kappa a)^2 = \frac{(g\mu_B)^2}{2\chi_0 J Z a^3} = \frac{1^\circ \text{K}}{2\chi_0 T_c} \quad (28)$$

$\chi_0$  is obtained from molecular field or experiment.  $N(\eta)$  is taken from Ref. 10. In Fig. 4,  $F_m$  and  $F_r$  are plotted using  $\chi_0 = 0.1$  and  $\eta = 4$  which corresponds to a rather flat disk, with<sup>10</sup>  $N(\eta) = 1.88$ . The transition at  $\alpha \approx 6$  means that in a sample of radius  $\kappa^{-1}\alpha$  larger than  $6 \kappa^{-1}$ , the state with rotational magnetization is stable.

Note that the energy per unit volume of the homogeneously magnetized sample,  $F_m/v$ , is constant, while the energy per unit volume of the rotationally magnetized sample vanishes like  $\log \alpha/\alpha^2$ . Thus, the latter state will always be more stable for a large enough radius  $\alpha$ . This critical radius is not very sensitive to the value of  $\chi_0$  (depends only logarithmically on  $\chi_0$ ) and is proportional to  $T_c^{1/2}$ . For the above example, using  $T_c = 100$  K, we have  $\kappa^{-1}\alpha \approx 27$  lattice constants  $a$ .

## ACKNOWLEDGEMENTS

We are grateful to Prof. H.J. Güntherodt and to Dr. F. Hulliger for indicating possible substances. One of the authors (W.B.) thanks the hospitality extended to him during his visit at the Centro Brasileiro de Pesquisas Físicas (CBPF), Rio de Janeiro, Brazil.

## FIGURE CAPTIONS

- Fig. 1 - Cross section of a rotationally symmetric ellipsoid with closed magnetic lines.
- Fig. 2 - Dimensionless magnetization  $\mu$  as a function of the dimensionless radial coordinate  $\rho$  for several values of the radius  $P$ .
- Fig. 3 - Free energy per unit volume (in units of  $(2/3)C_1 v$ , see text) of a symmetric ellipsoid of small axis  $\beta$  and large radius  $\alpha$  with rotational magnetization, as a function of  $\alpha$ , for  $\eta = \alpha/\beta$  constant. ———: neglecting the z-dependence of the magnetization  $M$ , -.-.-.-: correction due to the z-dependence of  $M$ .
- Fig. 4 - Free energy per unit volume (in units of  $(2/3)C_1 v$ , see text) of a symmetric ellipsoid of axes  $\beta$  and  $\alpha$ , as a function of the radius  $\alpha$ .  $\eta = \alpha/\beta = 4$ . ———: rotational magnetization, -.-.-.-: homogeneous magnetization.

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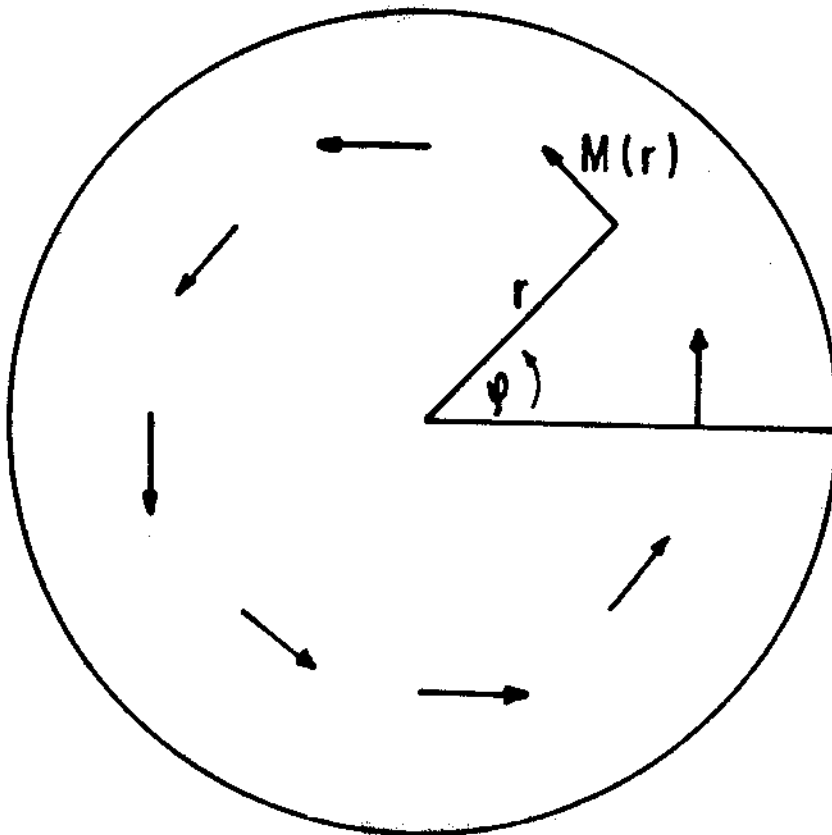


Fig. 1

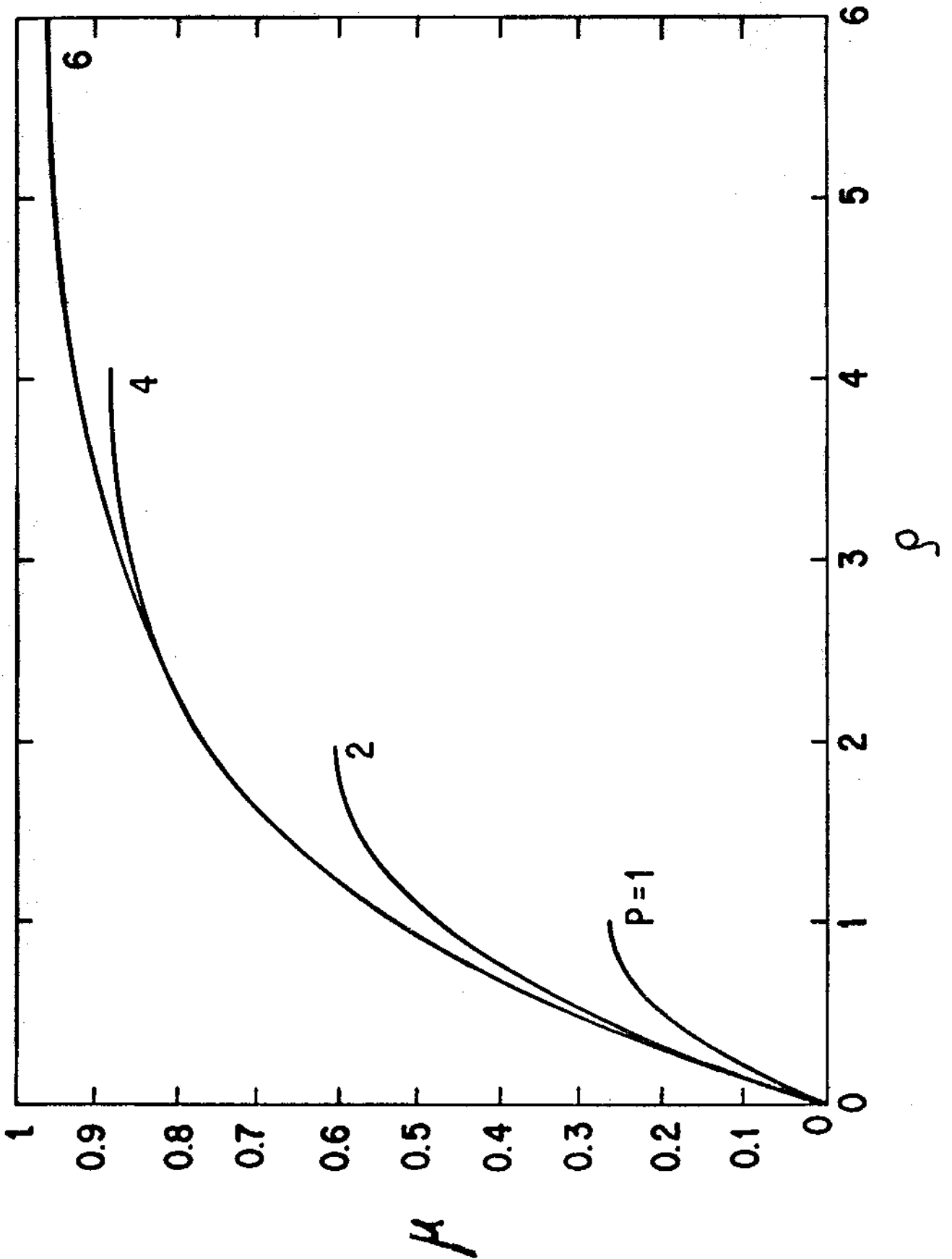


Fig. 2

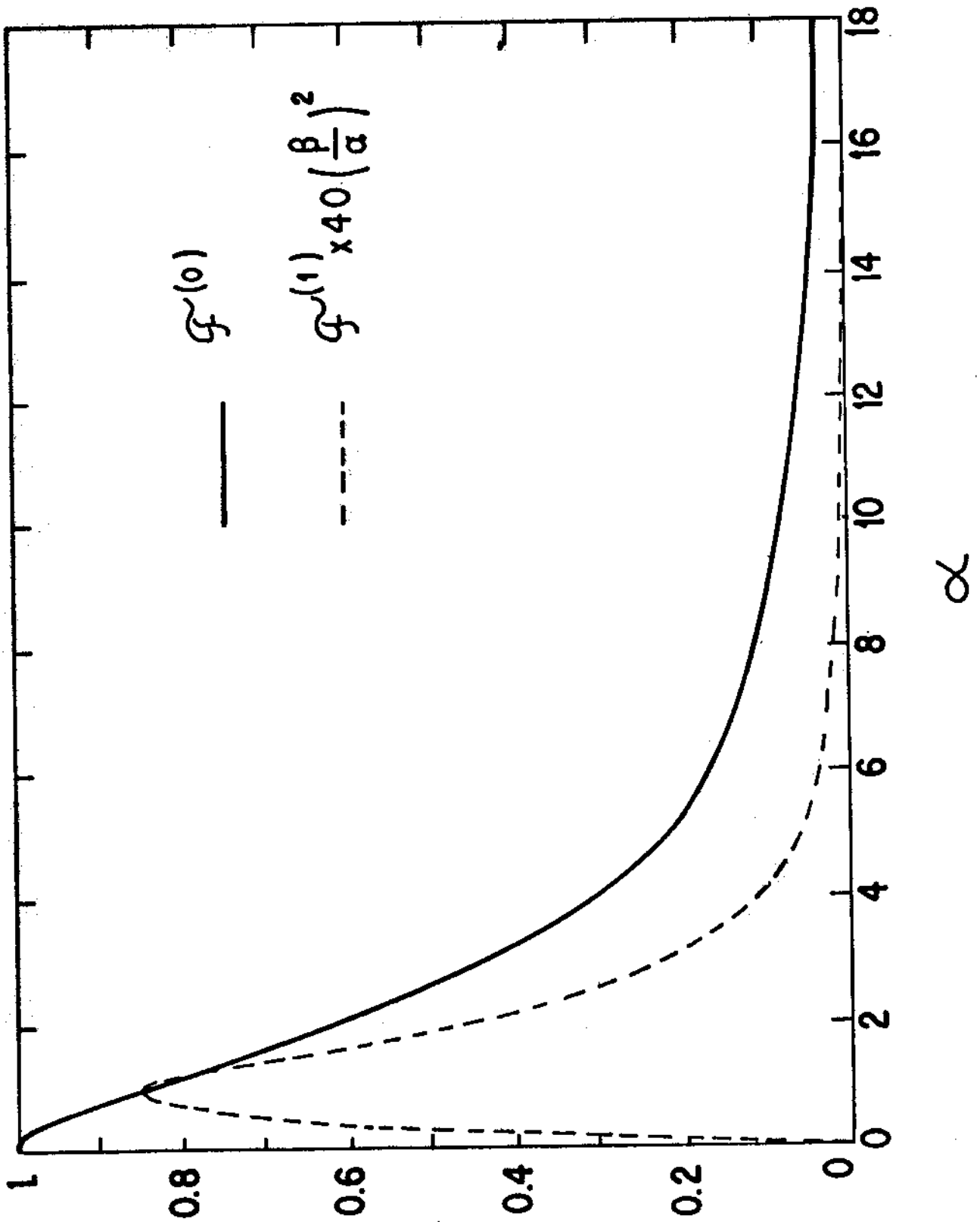


Fig. 3

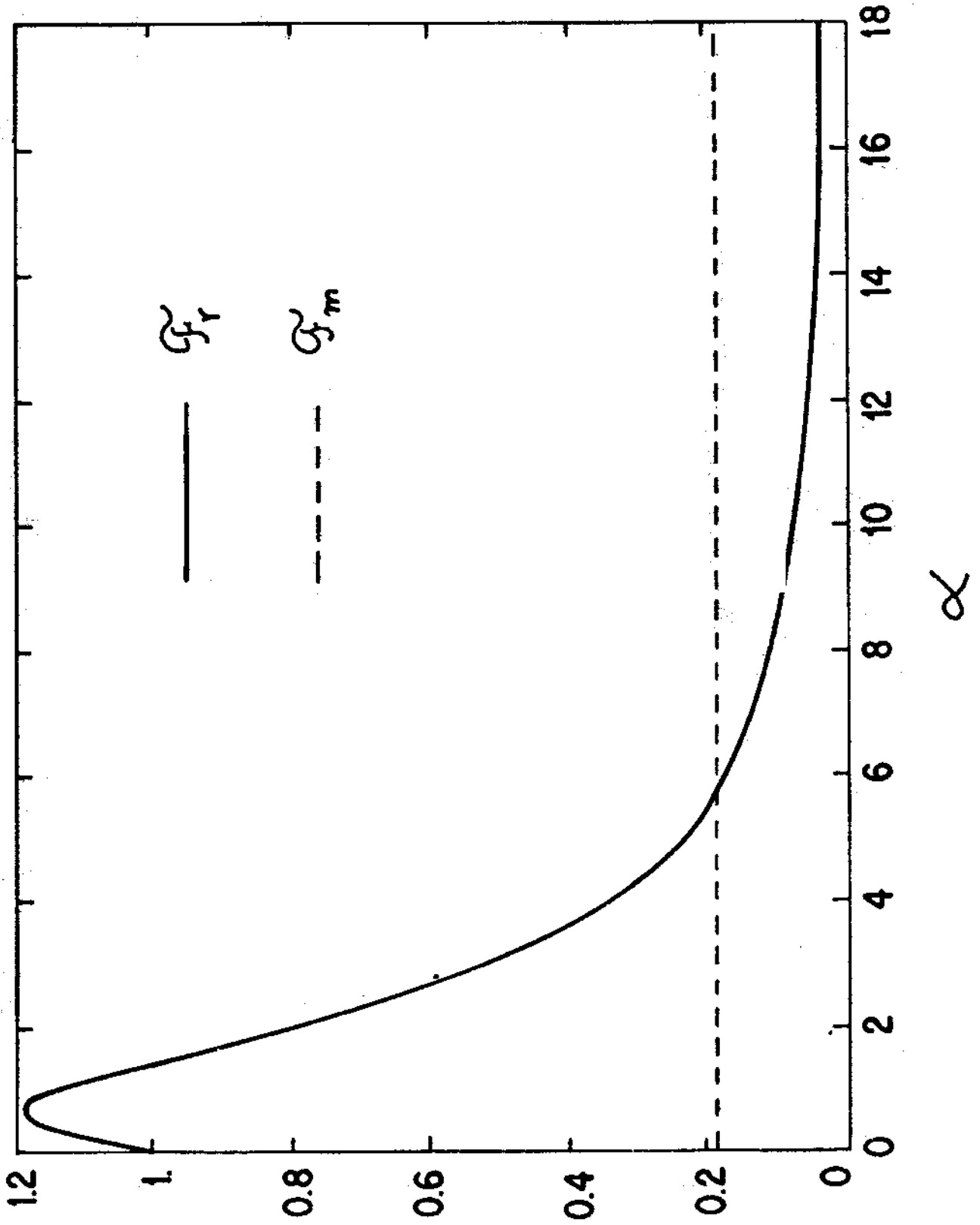


Fig. 4



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