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INDIRECT INTERACTIONS BETWEEN LOCALIZED MAGNETIC MOMENTS IN DOPED SEMICONDUCTORS

S. Lara* and R. Moreira Xavier
Centro Brasileiro de Pesquisas Físicas
Rio de Janeiro, Brasil

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ABSTRACT

The coupling between localized magnetic moments via conduction electrons is calculated taking into account the temperature and the mean free path of the electrons. For a fully degenerate electron gas and an infinite electronic mean free path the oscillatory RKKY interaction is obtained. On the limit of Boltzmann population and for infinite electronic mean free path the interaction can only be ferromagnetic. Taking into account the electronic mean free path the possibility of antiferromagnetism is restored. Furthermore the range of the interaction decreases.

Several intermediate cases and possible applications are discussed.

* Instituto de Física, Universidade Federal do Rio de Janeiro.

1. INTRODUCTION

It is well known that localized magnetic moments in metals are coupled via conduction electrons (1). This is known in the literature as the Ruderman-Kittel (1) or Ruderman-Kittel-Kasuya (3) - Yosida (4) interaction, s-f exchange, indirect enchange etc. The localized magnetic moments may be the moments of nuclei or incomplete 4f electron shells. The effects of collisions in this interaction have been studied by Kaplan (5) (electron--phonon scattering) and de Gennes (6) (electron-impurity scattering).

Baltensperger and de Graaf (7), Janak (8) and Darby (9) studied the indirect interaction through the medium of non-degenerate electron gas (a situation which may arise in doped semiconductors). We shall extend their work by taking into accout explicitly the electronic mean free path of the conduction electrons which arises in these systems essentially both by virtue of finite temperature and doping.

2. FORMULATION OF THE PROBLEM

We begin by considering a system of localized magnetic moments, at positions $\vec{R}_j$, with spins $\vec{S}_j$, interacting with the Bloch electrons (at position $\vec{x}$, spin $\vec{s}$)

$$< \vec{x} \mid \vec{k} > \; = \; \frac{1}{\sqrt{v}} \; u_{\vec{k}}(\vec{x}) \; e^{i\vec{k}.\vec{x}} \tag{1}$$

Where v is the crystal volume. The interaction between electrons and moments is described by

$$H = - \sum_{J} 2\Gamma \, (\vec{x} - \vec{R}_J) \, \vec{s}. \, \vec{S}_J \tag{2}$$

Here $(\vec{x} - \vec{R}_J)$ is the so-called exchange parameter between the localized moment $\vec{S}_J$ and the conduction electrons $\vec{s}$.

We define (here Ω is the atomic volume)

$$\Gamma(\vec{k}, \vec{k}') = \frac{V}{\Omega} < \vec{k}' | \Gamma(x) | \vec{k} > \tag{3}$$

which in general is a function of $\vec{k}$ and $\vec{k}'$. For the Fermi contact interactions between nuclei and electrons we have

$$\Gamma(\vec{x}) = \Gamma\delta(\vec{x})$$

so that

$$\Gamma(\vec{k}, \vec{k}') = \frac{V}{\Omega} \Gamma u^{*}_{\vec{k}'}(0)\, u_{\vec{k}}(0) = \Gamma_S \tag{4}$$

We shall assume that an incomplete 4f shell is also localized and that $\Gamma(\vec{k},\vec{k}')$ is a constant independent of $\vec{k}$ and $\vec{k}'$. Although there has been considerable effort in order to avoid these assumptions (10), (11), they are usually good approximations at least for rare-earth system (12).

We shall now apply perturbation theory. It is implicit in this approach that $\Gamma(\vec{k}, \vec{k}')$ is much smaller than the characteristic energies of the conduction states. This is, of course, true if one is considering the indirect interactions of nuclear moments but requires further analysis in the case of 4f-shells (13). However, in general, this is also a fair approximation (12) (13).

In a straightforward way one obtains

$$E = - \sum_{i \neq J} A_{iJ} \; \vec{S}_i . \vec{S}_J$$

$$A_{iJ} = - \frac{2\Gamma_s^2 \, \Omega^2}{V^2} \sum_{\vec{k} \neq \vec{k}'} \frac{e^{i(\vec{k}-\vec{k}') \, . \, \vec{R}_{iJ}}}{E(\vec{k}) - E(\vec{k}')} \; f(\vec{k}) [1 - f(\vec{k}')] \tag{5}$$

where $\vec{R}_{iJ} = \vec{R}_J - \vec{R}_i$ and $f(\vec{k})$ is the Fermi distribution function.

As usual eq. (5) is transformed into an integral over $\vec{k}$-space by means of a new set of variables

$$k_i = \frac{k_i}{\sqrt{m_i}} \qquad i = 1,2,3 \tag{6}$$

$$\rho_i = R_i \sqrt{m_i} \qquad i = 1,2,3 \tag{7}$$

$$\therefore \; \vec{k} . \vec{\rho} = \vec{k} . \vec{R} \tag{8}$$

and m_i is the effective mass in the i direction. If A_{ij} is then written in terms of the new variables, an integration over angular parts yields,

$$A_{iJ} = \frac{\Omega^2 \; \Gamma_s^2 \; m_x \; m_y \; m_z}{2\Pi^4 \; \rho^2} \, I \tag{9}$$

$$I = \int_0^\infty dk \int_0^\infty dk \; \frac{k \; k' \; \sin(k\rho) \; \sin(k'\rho)}{\varepsilon(k) - \varepsilon(k')} \; f(k) \tag{10}$$

The terms in $f(k)\ f(k')$ cancel out .

Here the perturbation theory has been applied on eigenstates (1).In a more realistic situation the total Hamiltonian should include also phonons (H_p) and electron-phonon (H_{ep}) interactions. Kaplan (5) has shown that the effect of $H_p + H_{ep}$ can be approximately evaluated by introducing relaxation processes which affect the perturbation as follows

$$I = \mathrm{Re} \int_0^\infty dk \int_0^\infty dk' \frac{k\ k' \sin(k\rho) \sin(k'\rho')}{\varepsilon(k) - \varepsilon(k') - i\delta^2} f(k) \tag{11}$$

Where $\delta^2 = \dfrac{2}{\hbar\tau}$

Complex corrections to the energies have been used by de Gennes (13) for a degenerate electron gas, to account for scattering processes due to impurities. So we shall assume that equation (11) is also valid in this case.

The integral in k' is analytic for $\varepsilon(k') = \dfrac{\hbar^2 k'^2}{2}$ and $\varepsilon(k) = \dfrac{\hbar^2 k^2}{2}$

$$I = - \frac{\Pi}{\hbar^2} \mathrm{Re} \int_0^\infty dk\ k \sin(k\rho)\ e^{-ik\rho\,[1 - (\delta/k)^2]^{1/2}}\ f(k) \tag{12}$$

which leads finally to

$$A_{iJ} = \frac{\Omega^2\ \Gamma_s^2\ m_x\ m_y\ m_z}{2\Pi^3\ \hbar^2\ \rho^2} \mathrm{Re} \int_0^\infty dk\ \ k\ \sin(k\rho) \exp\{\ ik\rho\ [1 - (\delta/k)^2]^{1/2}\}\ f(k) \tag{13}$$

For infinite electronic mean free path $\delta^2 \to 0$ it turns out:

$$\lim_{\delta^2 \to 0} A_{iJ} = \frac{\Omega^2 \, \Gamma_s^2 \, m_x \, m_y \, m_z}{4\Pi^3 \, \hbar^2 \, \rho^2} \int_0^\infty dk \;\; k \;\; \sin(2k\rho) \; f(k) \qquad (14)$$

This is the result obtained by W. Baltensperger and de Graaf (7) which includes only the effect of non-degenracy of the conduction band. For a null electronic mean free path $\delta^2 \to \infty \implies I \to 0$ and there are no interaction.

In order to simplify the comparison with Baltensperger and de Graaf results let us introduce the variables

$$x = \frac{\hbar^2 \, k^2}{2k_B T}$$

$$\alpha = \frac{2\sqrt{2k_B T}}{\hbar} \, \rho$$

$$\beta = \frac{\hbar^2 \, \delta^2}{2k_B T}$$

$$\eta = \frac{E_F}{k_B T}$$

Note that we are assuming β to be independent of k. This hypothesis has been used by Kaplan (5).

With the new variables one obtains finally

$$A_{iJ} = \frac{\Pi\,\Omega^2\,\Gamma_s^2\,m_x\,m_y\,m_z}{2\,\hbar^2\,\rho^2}\;\frac{k_B T}{\hbar^2}\;G(\alpha,\beta,\eta)$$

where

$$G\,(\alpha,\beta,\eta) = 2\int_0^\infty dx\;\; f(x)\;\; \sin\frac{\alpha\,\sqrt{x}}{2}\;\; \mathrm{Re}\Phi$$

$$f(x) = \left[\,e^{x-\eta} + 1\,\right]^{-1}$$

$$\mathrm{Re}\Phi = \exp\left\{-\,\frac{\alpha}{2}\left[\,x^2 + \beta^2\,\right]^{1/4}\sin\frac{\Theta}{2}\right\}\cos\left\{\frac{\alpha}{2}\left[x^2 + \beta^2\,\right]^{1/4}\cos\frac{\Theta}{2}\right\}$$

$$\Theta = \mathrm{arc}\;\;\mathrm{tg}\;\frac{\beta}{x}$$

$$\eta = \frac{E_F}{k_B T}$$

$$\alpha = \frac{2\sqrt{2k_B T}}{\hbar}\,\rho$$

The variation of β gives us the effect of the electronic mean-free-path

Figures (1), (2), (3) and (4) show $G(\alpha)$ as a function of α for different η and β; with doping effects in mind, we illustrate the passage from Boltzmann-like behaviour to Fermi-like behaviour taking for the parameter η the following values:

η = -2 fig (1) (a and b)

η = 0 fig (2) (")

η = 2 fig (3) (")

η = 4 fig (4) (")

for β from 0 to 5

3. GENERAL DISCUSSION OF THE RESULTS

The interaction between two magnetic moments in a degenerate electron gas ($\eta \to \infty$) is oscillatory. For $\beta = 0$ this corresponds to the Ruderman-Kittel interaction (2).

$$A(\rho,\infty) = - \frac{\Gamma_s^2 \Omega^2 \; m_x \; m_y \; m_z}{16\Pi^3 \hbar^2 \; \rho^4} \left[\sin (2k_F\rho) - 2k_F\rho \; \cos (2k_F\rho) \right]$$

For the Boltzmann gas ($\eta \to -\infty$), and $\beta = 0$, the resulting interaction, namely (7),(8)

$$A (\rho, -\infty) = - \frac{\Gamma_s^2 \; \Omega^2 \; (m_x \; m_y \; m_z)^{1/2} \; n}{2 \; \Pi \; \hbar \; \rho} \exp \{- \frac{2k_B T\rho^2}{\hbar^2}\}$$

The density of particles n is related to η through

$$n = \frac{(m_x \; m_y \; m_z)^{1/2}}{(2\Pi)^2} \left(\frac{2k_B T}{\hbar^2} \right)^{3/2} F_{1/2} (\eta)$$

Where $F_{1/2}$ is a function tabulated by Mc Dougall and Stoner (14). In particular for $\eta \to -\infty$ (Boltzmann gas)

$$F_{1/2}(\eta \to -\infty) = \frac{\sqrt{\Pi}}{2} e^{\eta}$$

Figures 1 to 4 show intermediate cases and the effects of the electronic mean free path λ (a function of β, roughly $\lambda = \frac{1}{\Pi k_F \beta}$). It can be seen that for $\eta = 4$, $\beta = 0$, $G(\alpha)$ resembles the RKKY result. For $\eta = -2$, $G(\alpha)$ resembles ferromagnetic gaussian function.

In all cases the electronic mean-free-path has basically two effects: firstly it weakens the interaction and secondly it changes the phases of the oscillations. This is clearly seen in figures (1) to (4).

$G(\beta)$ is plotted (Fig. 5,6 and 7) for some values of α and η. It can be seen that $dG/d\beta$ is negative that is, G decreases with increasing β (at least for small values of α). This effect is more pronounced for $\eta = -2$ ("Boltzmann-like") than for $\eta = 4$ ("Fermi-like").

Furthemore it is seen that at the Boltzmann limit the presence of an electronic mean free path ($\beta \neq 0$) allows negative values for $G(\alpha)$, that is the oscillatory character and the possibility of antiferromagnetic coupling are restored. It should be remembered that for $\beta = 0$ the interaction is purely ferromagnetic for all distances (5), (6).

This is an interesting result. Indeed it shows that it is physically possible to vary the indirect interaction between two localized magnetic moments in a semiconductor by varying the electronic mean free path (for instance, by means of impurities chosen so that they do not interfere with the population of the conduction band). On the other hand a change in the population of the conduction band, keeping β constant, by optical means for example (9), may induce a variation in the strenght of the magnetic coupling. It should be noted that the indirect exchange results from the spin polarization of the conduction band (13). This can be observed in several ways, for example, through the changes in the magnetic moment and the indirect g-shifts(15) of the magnetic ions. The polarization also gives rise to a hyperfine field which can be measured by NMR or Mössbauer techniques.

4. NUMERICAL EXAMPLE: EPR STUDY OF MAGNETIC IMPURITIES IN A SEMICONDUCTOR

In an insulator or a semiconductor with an empty conduction band the interaction between localized magnetic moments may occur also by a virtual excitation of the conduction band due to interband matrix elements, the so-called Bloembergen-Rowland interaction (16). For a dilute system of magnetic impurities (magnetic moments $\sim 1\mu_B$) embedded in a semiconductor,the numerical values of the various interactions are estimated in table I for a semiconductor like germanium. Following Baltensperger and de Graaf (7) we take $m_x = m_y = m_z = \frac{1}{10} m$, $\Omega = 2.24 \times 10^{-23}$ cm³, $E_g = 0.8$ eV. The parameters are the distance R between two magnetic ions, the temperature T and the degeneracy. The exchange integral is of the order of a tenth of a electron Volt. Table I compares the various types of interactions for typical values of the four parameters involved: distance R, temperature T, degeneracy and the electronic mean free path effect in terms of the new variable β. It is seen that for R = 40Å all the interactions are of the same order of magnitude. It can be seen also, as mentioned before, that the electronic mean free path may change the magnitude and the sign of the interaction.

It should be remembered that for a moment of $1\ \mu_B$ the energy of 6.7×10^{-8} eV coresponds to a width of one Oersted.

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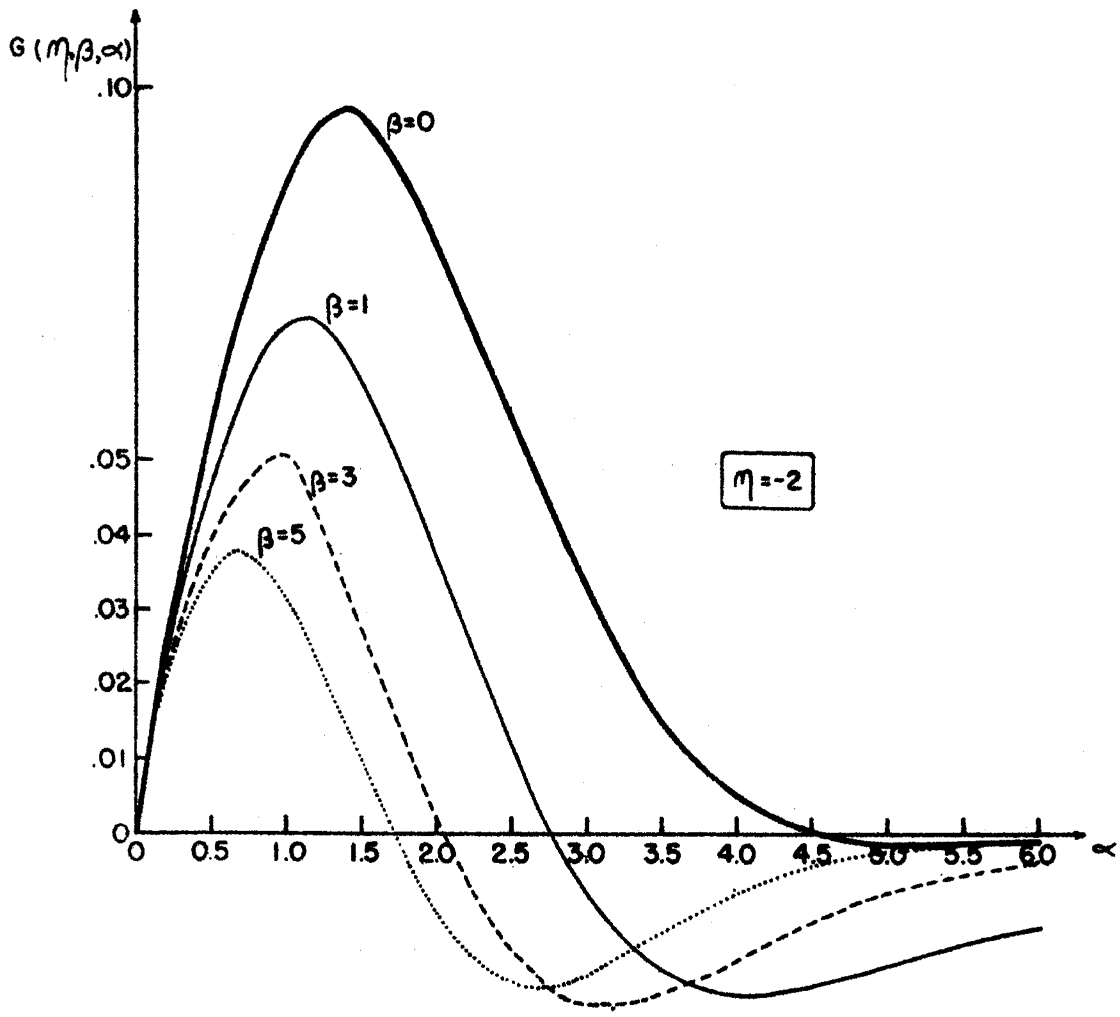

G (η,β,α)
.10
.05
.04
.03
.02
.01
0
0
0.5
1.0
1.5
2.0
2.5
3.0
3.5
4.0
4.5
5.0
5.5
6.0
α
β=0
β=1
β=3
β=5
η=-2

FIG. 1a

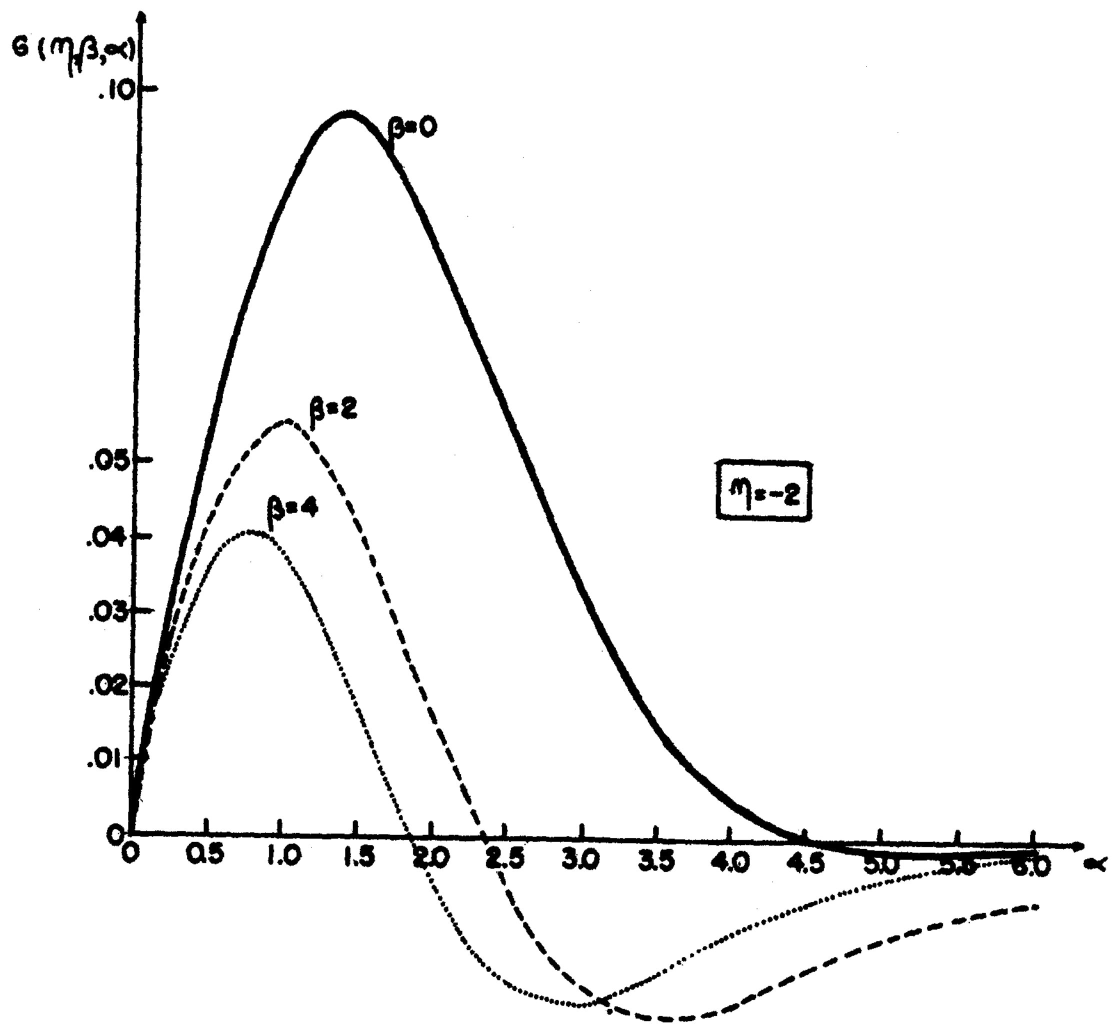

G (η,β,α)
.10
.05
.04
.03
.02
.01
0
β=0
β=2
β=4
η=-2
0
0.5
1.0
1.5
2.0
2.5
3.0
3.5
4.0
4.5
5.0
5.5
6.0
α

FIG. 1b

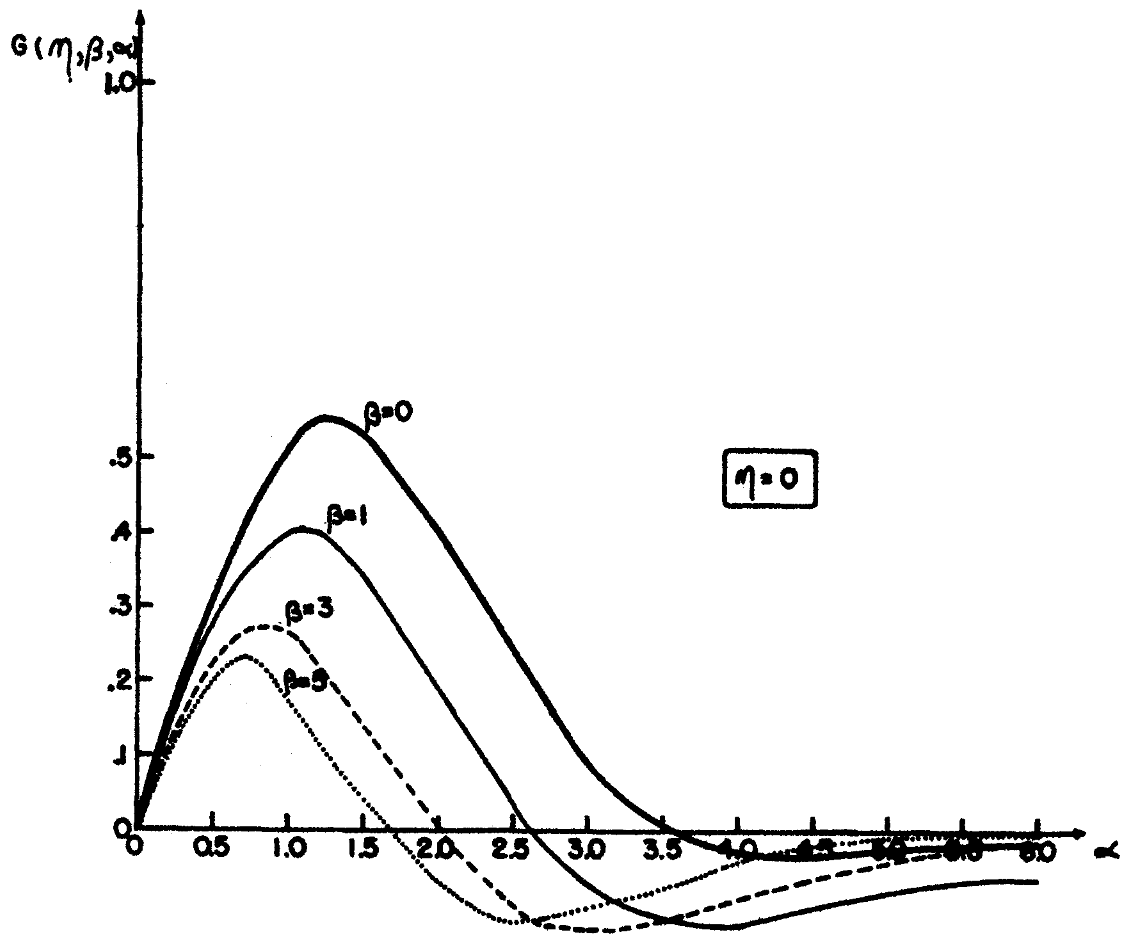


FIG. 2a

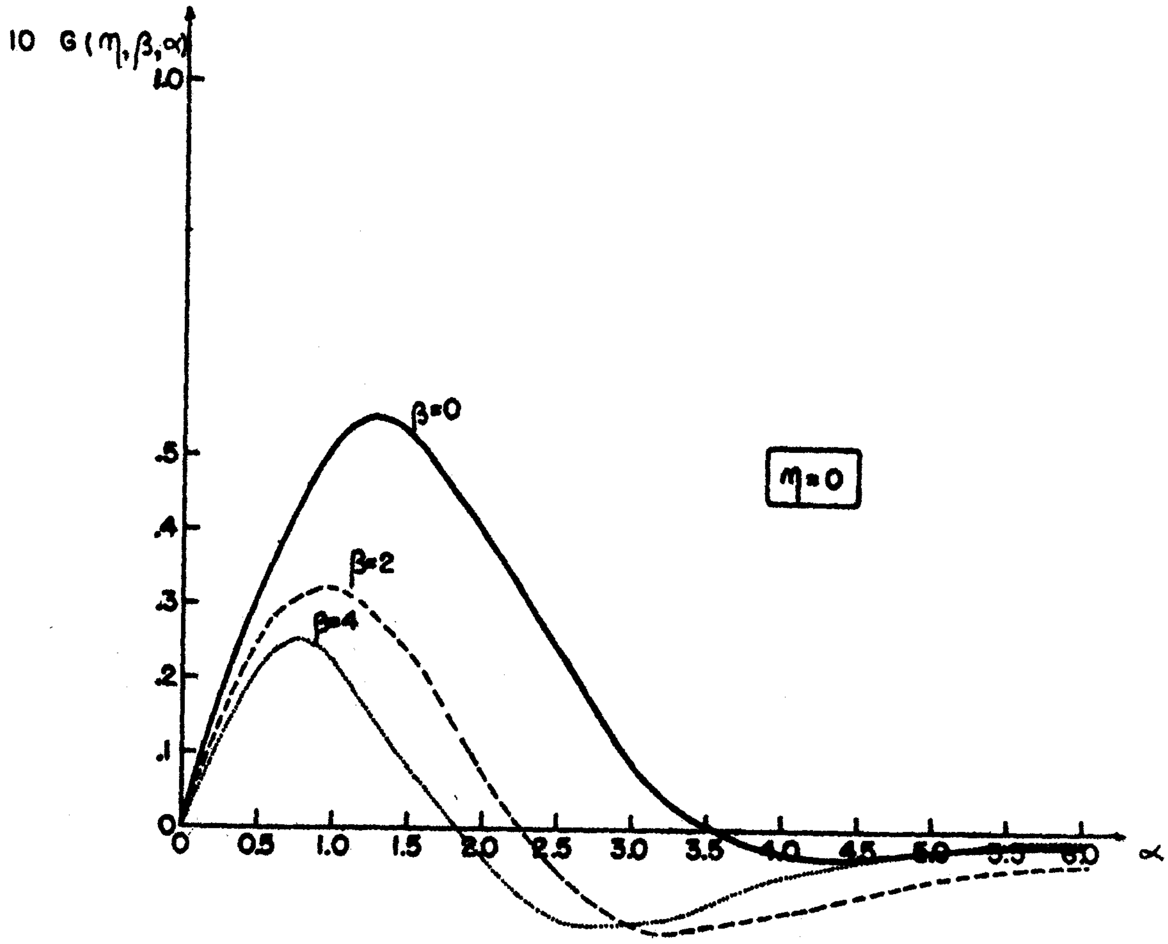


FIG. 2b

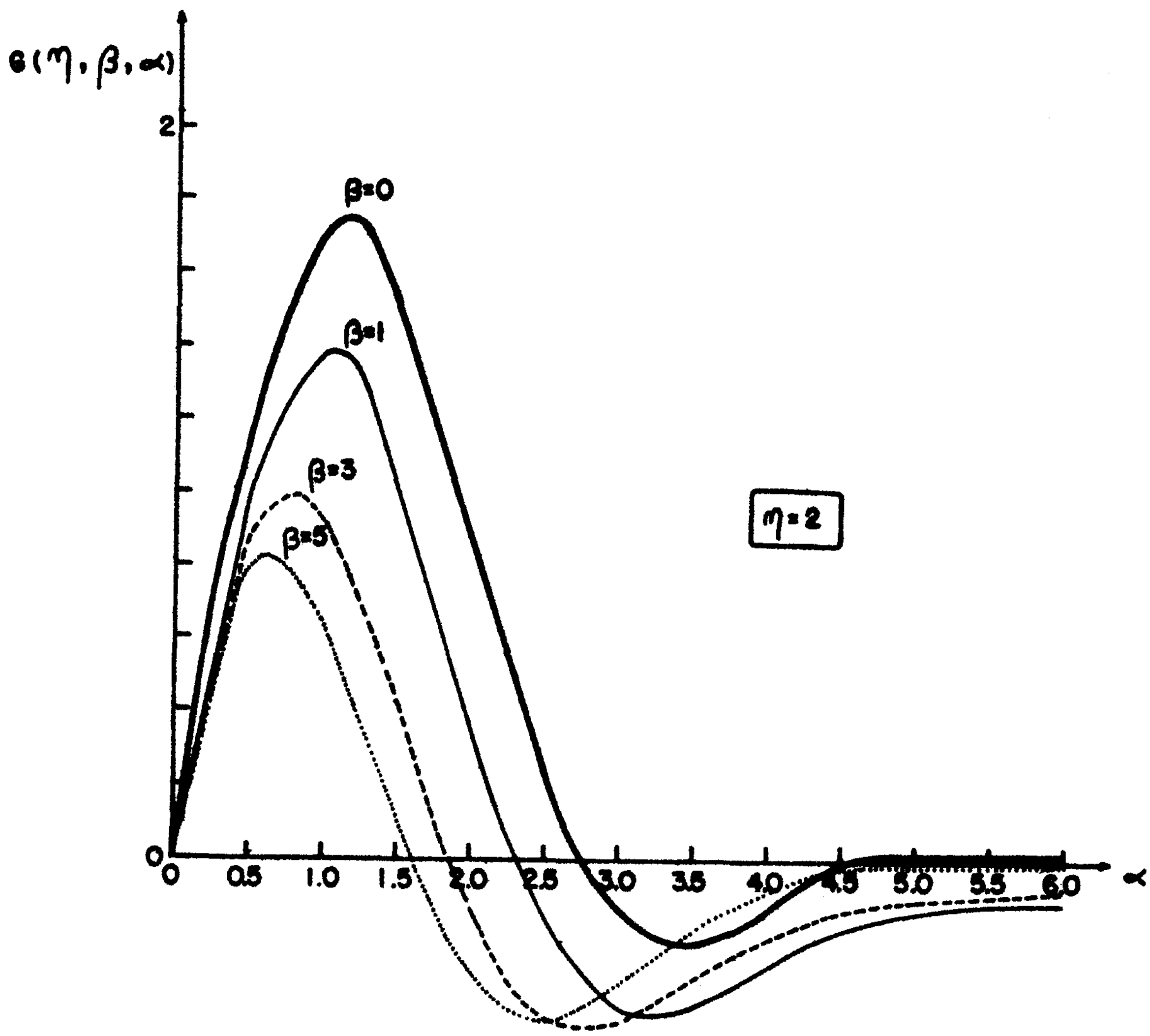


FIG. 3a

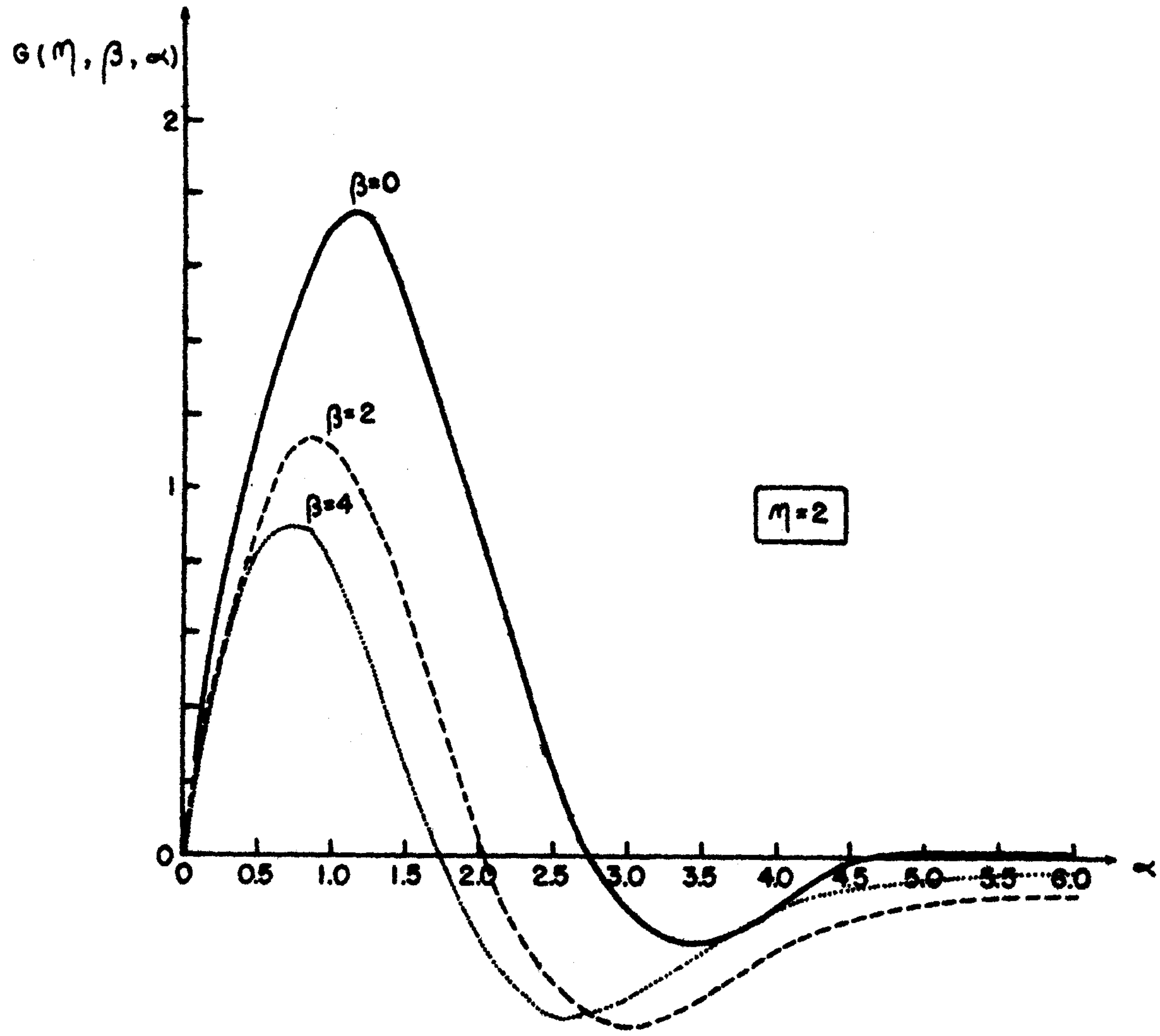


FIG. 3b

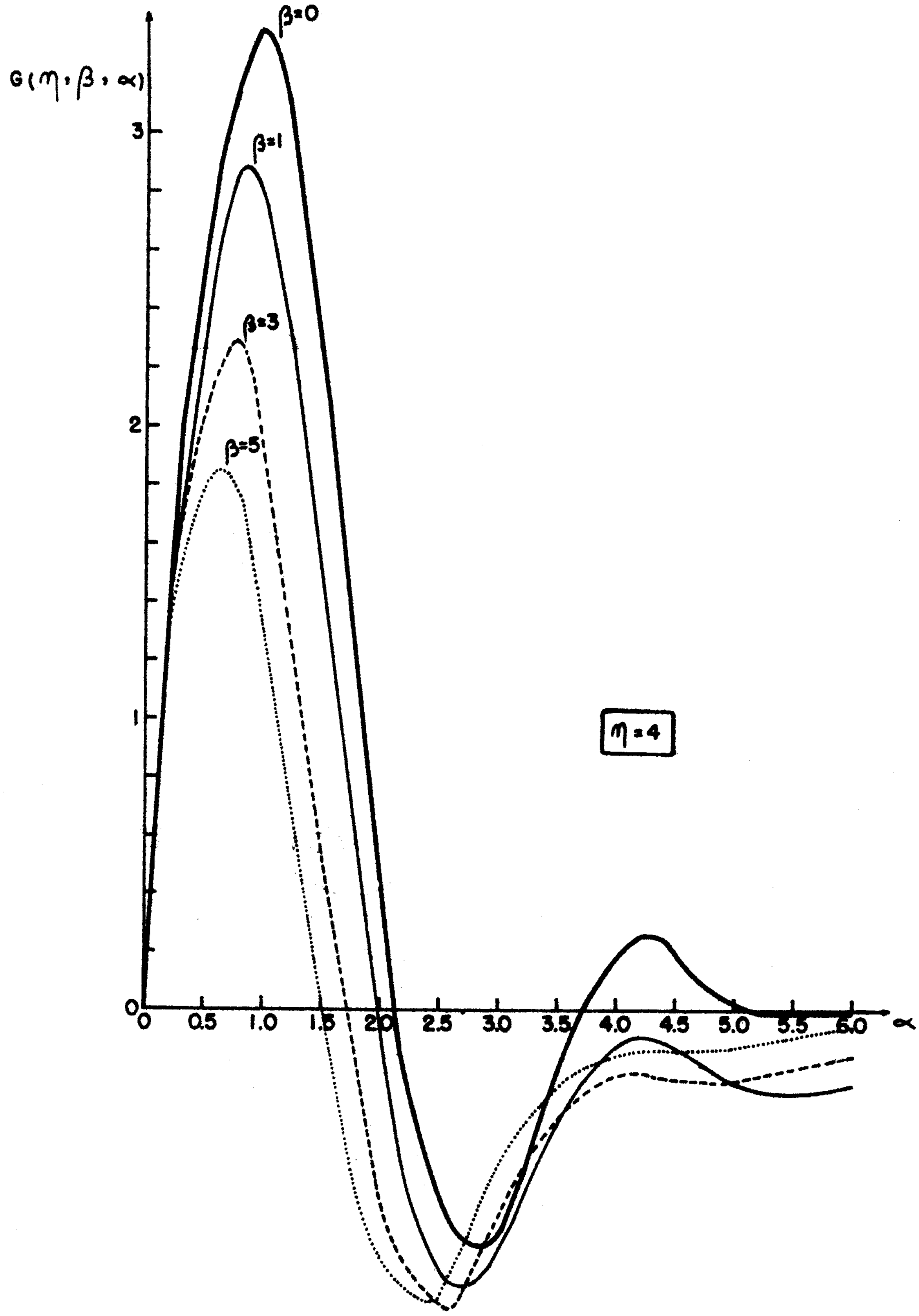


FIG. 4a

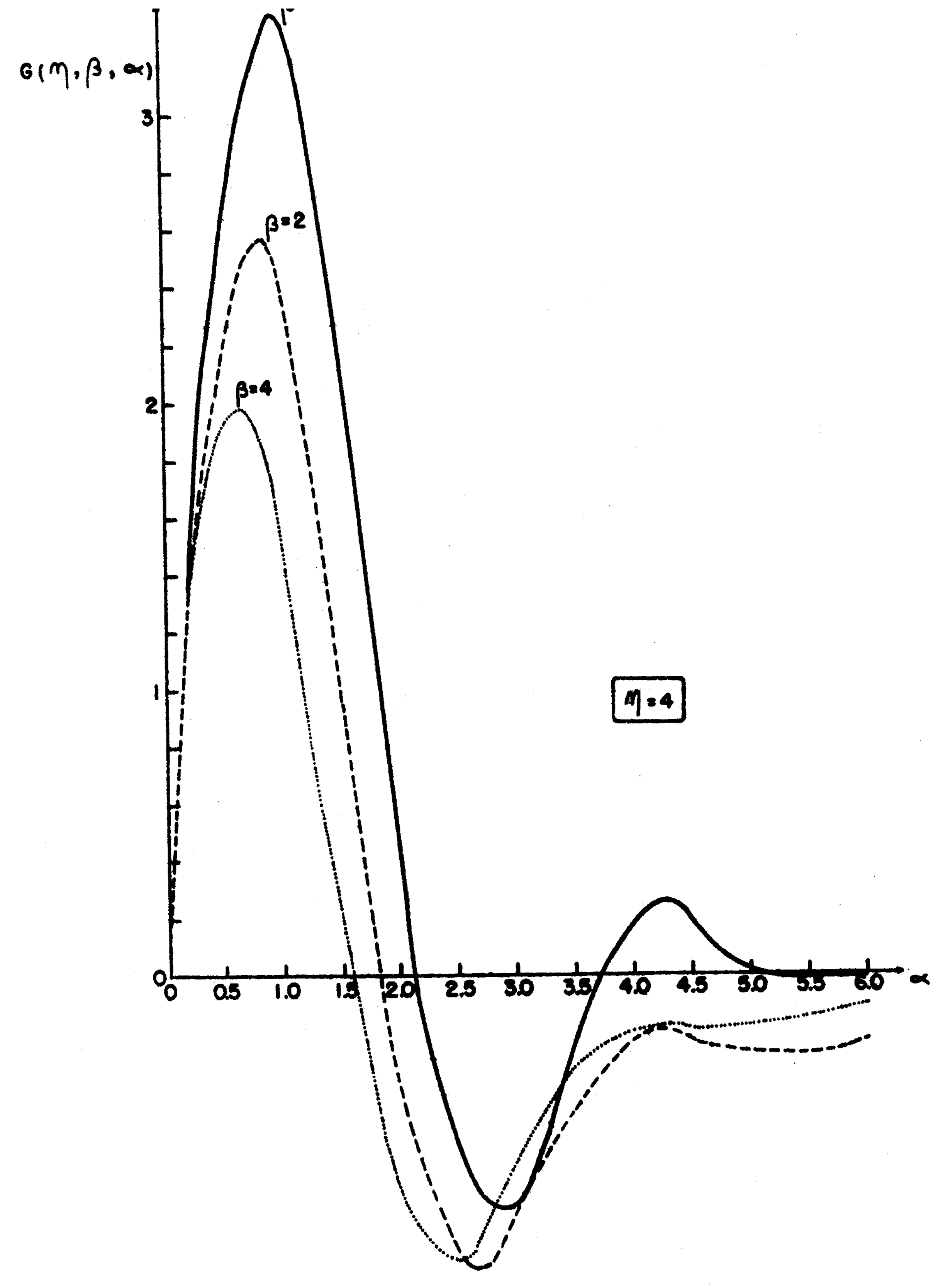


FIG. 4b

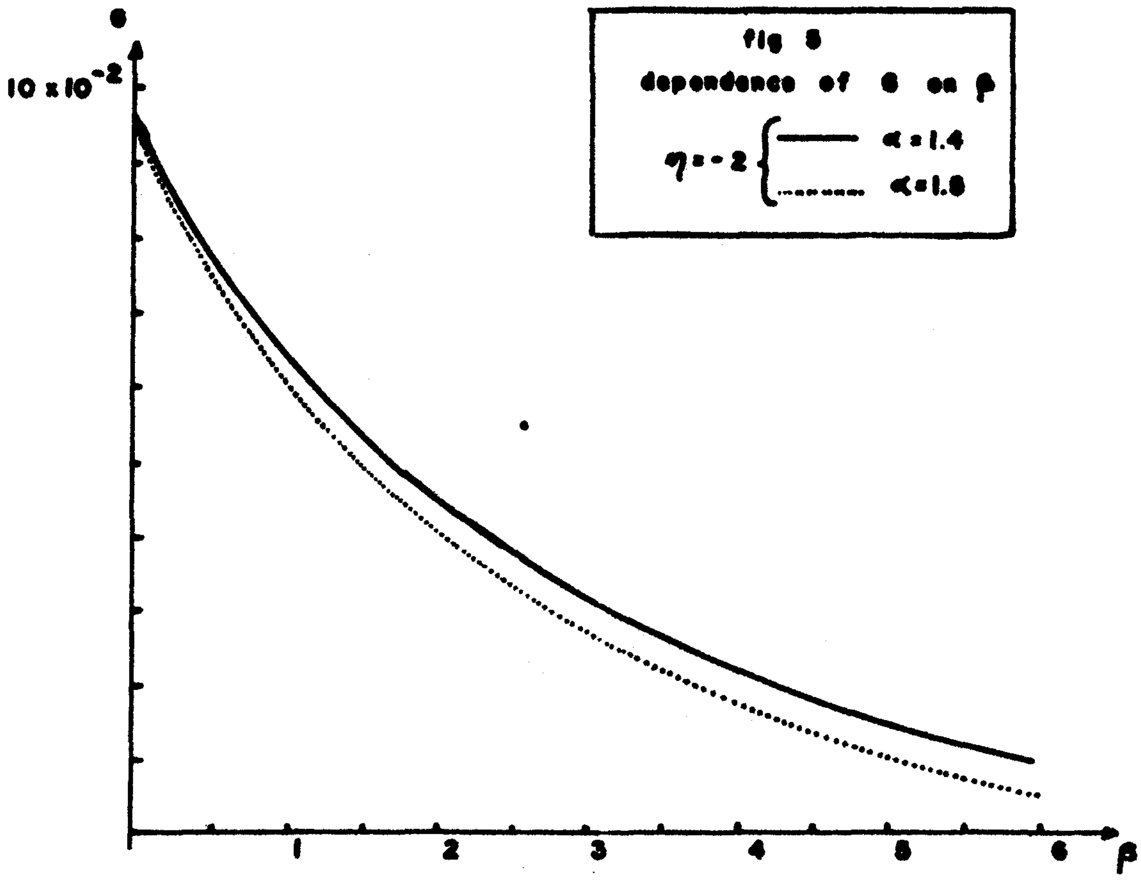


fig 5
dependence of σ on β

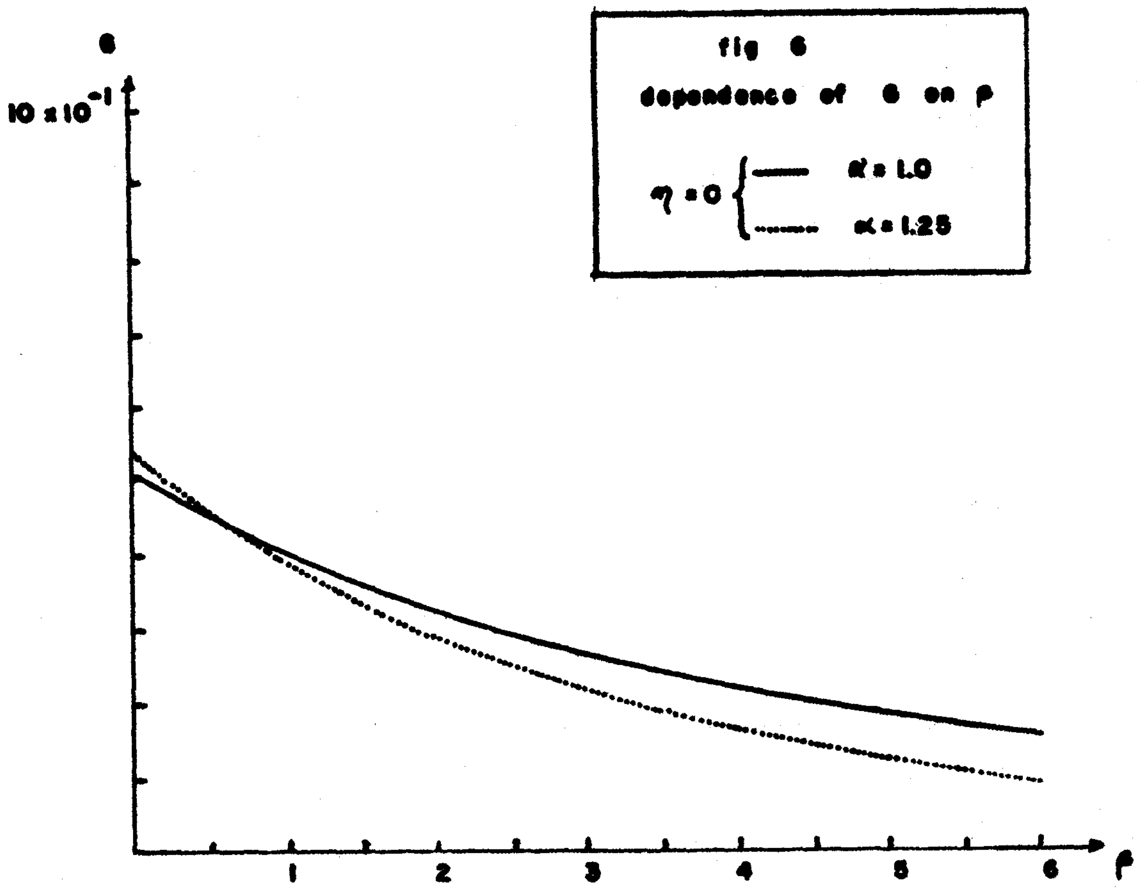
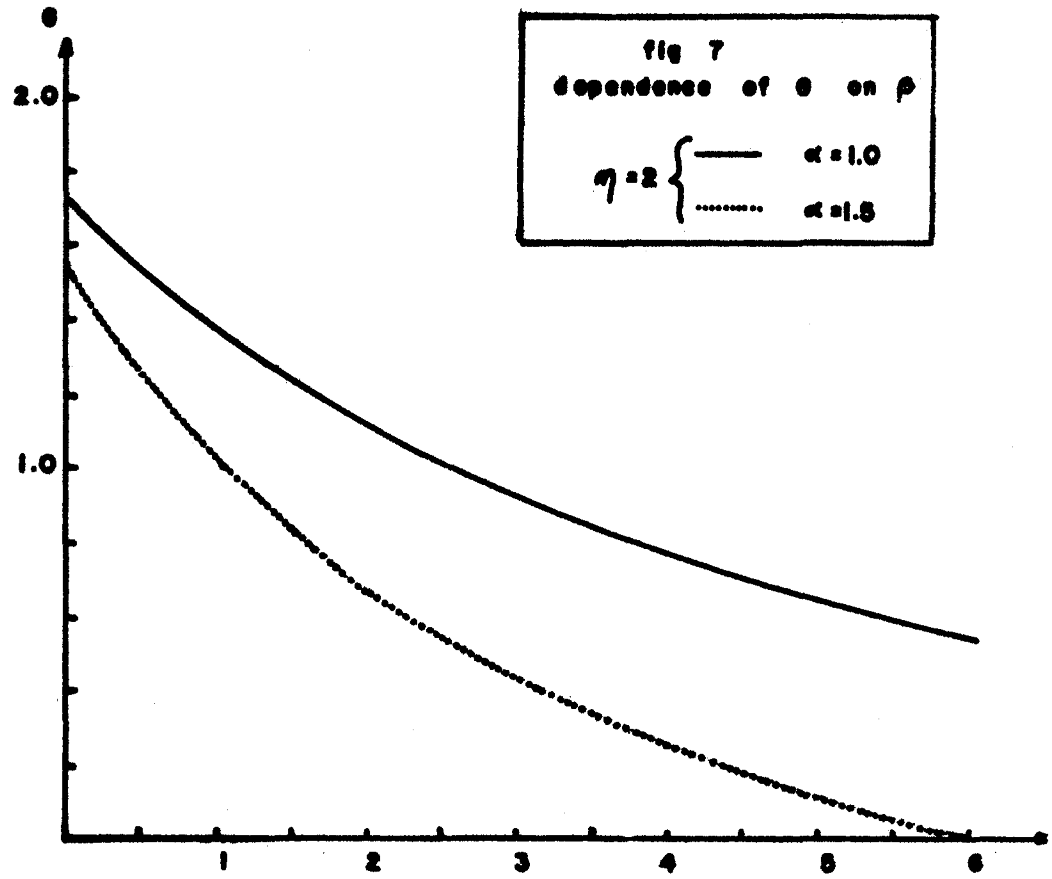


fig 6
dependence of G on ρ
η = 0
α = 1.0
α = 1.25
10 x 10⁻¹
1
2
3
4
5
6
ρ


θ
2.0
1.0
1
2
3
4
5
6
fig 7
dependence of θ on β
η = 2
α = 1.0
α = 1.5

$R(\mathring{A})$	10				40			
$T(^{o}k)$	100		500		100		500	
α	0.30		0.67		1.2		2.7	
η	2	-1	2	-1	2	-1	2	-1
n	$4.3x10^{17}$	$5.0x10^{16}$	$4.8x10^{18}$	$5.6x10^{17}$	$4.3x10^{17}$	$5.0x10^{16}$	$4.8x10^{18}$	$5.6x10^{17}$
A/Γ_s^2 $\beta = 0$	$4.3x10^{-8}$	$5.1x10^{-9}$	$4.2x10^{-7}$	$5.2x10^{-8}$	$6.4x10^{-9}$	$8.8x10^{-10}$	$1.2x10^{-9}$	$2.0x10^{-9}$
A/Γ_s^2 $\beta = 1$	$4.0x10^{-8}$	$4.8x10^{-9}$	$3.7x10^{-7}$	$4.4x10^{-8}$	$4.8x10^{-9}$	$6.4x10^{-10}$	$-7.0x10^{-9}$	$4.2x10^{-11}$
A/Γ_s^2 $\beta = 2$	$3.9x10^{-8}$	$4.5x10^{-9}$	$3.3x10^{-7}$	$3.9x10^{-8}$	$3.7x10^{-9}$	$4.8x10^{-10}$	$-9.8x10^{-9}$	$-6.8x10^{-10}$
A/Γ_s^2 $\beta = 4$	$3.6x10^{-8}$	$4.2x10^{-9}$	$2.7x10^{-7}$	$3.2x10^{-8}$	$2.2x10^{-9}$	$2.9x10^{-10}$	$-1.05x10^{-8}$	$-1.0x10^{-9}$
$\lvert A'\rvert/\Gamma_s^2$	3.5×10^{-5}				7.1×10^{-9}			
A"	5.4×10^{-8}				8.4×10^{-10}			

TABLE 1

$A = A_{iJ}$ is the indirect interaction.

A' is the Bloembergen - Rowland interaction.

$A" = \mu_B^2 / R^3$ is the magnetic dipole interaction.

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