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THIRD ORDER CORRECTIONS AND FINITE CONDUCTION BAND EFFECTS ON THE INDIRECT EXCHANGE INTERACTION IN THE BLOEMBERGEN-ROWLAND APPROXIMATION

by

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ABSTRACT

We have investigated third order corrections and finite conduction band effects on the indirect exchange interaction assuming a large energy gap compared to the valence band width. We regain an oscillatory expression in which as opposed to the Bloembergen-Rowland formula the energy gap as well as the conduction band modifies both the magnitude and phase of the oscillations.

1 - INTRODUCTION

The polarization by local magnetic moments of conduction electrons in a metal results in the well known RKKY[1] indirect exchange interaction[1-16] which is the result of a perturbation calculation up to second order in the contact interaction . The RKKY mechanism has been noted however to be ineffective in semi-conductor insulators and alloys. But there is a corresponding effective interaction between local moments mediated by the polarization of valence band electrons through virtual transitions . An evaluation of the integrals in a realistic case is not elementary so Bloembergen and Rowland[2] calculated the interaction in second order by assuming that the energy gaps were much greater than the valence band width. The Bloembergen-Rowland (BR) formula was later extended to the semiconductors[5,8].

The third order perturbation series term of the RKKY interaction has been investigated and suggested[3] to be important. Estimates of the conduction band width in rare earth systems indicates that it is important to consider the fact that the band width is finite and should be considered in the indirect exchange interaction models[4]. We have investigated in this paper third order corrections and finite conduction band effects on the indirect exchange interaction in the BR approximation.

2 - FORMULATION OF THE PROBLEM AND DISCUSSION OF THE RESULTS

Using the formulation of second quantization the cotact term reads

$$H_{int} = \frac{1}{2\Omega} \sum_{n,k,k'} J \exp\left[i(k-k').R_n\right] \left[I_n^z (c^+_{k'+} c_{k+} - c^+_{k'-} c_{k-}) + \right.$$

$$\left. + I_n^+ c^+_{k'-} c_{k+} + I_n^- c_{k'+} c_{k-}\right] \qquad (1)$$

where J denotes the strength of the interaction, $\vec{R}_n$ the position, $\vec{I}_n$ the spin of nucleus n.

In second order the BR interaction yields[2]

$$H_2 = \frac{m' J^2 K_T^4}{(2\pi)^3} \sum_{n,m} F(K_T R_{mn}) \; \vec{I}_n . \vec{I}_m \; e^{-R_{mn}\sqrt{E}} \qquad (2)$$

$\hbar = 1$, k_T is the Brillouin zone radius $F(x)=(x\cos x-\sin x)/x^4$, m' and R_{nm} respectively denote effective mass and the distance between the spin n and m, $R_{nm} = |\vec{R}_n - \vec{R}_m|$, $E = \frac{2m'}{\hbar^2} E_g$, where E_g is the energy gap.

The third order correction reads

$$H_3 = \sum_{n,m}'' \frac{<0|H_{int}|n><n|H_{int}|m><m|H_{int}|0>}{(E_0 - E_n)(E_0 - E_m)} \qquad (3)$$

where the symbol $\sum''_{n,m}$ indicate a summation over all excited state unequal to the ground state $|0>$ where $|n> = c^+_{k_1\sigma_1} c_{k_2\sigma_2} |0>$ $|m>= = c^+_{k_3\sigma_3} c_{k_4\sigma_4} |0>$, $c_{k_i\sigma_i}$ (i = 1,2,3,4) are fermion operators, creation and annihilation operators for electron states of energy $\varepsilon_{\vec{k}_i\sigma_i}$. After some calculations one obtains[3]

$$H_3 = - \frac{m'^2 J^2}{2\Omega^3} \sum_{n,\ell,m} \left[I_n^z (I_\ell^+ I_m^- - I_\ell^- I_m^+) + I_n^+ (I_\ell^- I_m^z - I_\ell^z I_m^-) + \right.$$

$$\left. + I_n^- (I_\ell^z I_m - I_\ell^+ I_m^z)\right]$$

$$\times\left\{\sum_{k_1,k_2,k_3,k_4} \delta_{k_1,k_3}\left[\frac{\exp\left[i(\vec{k}_1-\vec{k}_2)\cdot\vec{R}_n + i(\vec{k}_2-\vec{k}_4)\cdot R_\ell + i(\vec{k}_4-\vec{k}_3)\cdot R_m\right]}{(k_2^2-k_1^2)(k_4^2-k_3^2)}\right.\right.$$

$$\left.\left. + \delta_{k_2,k_4}\frac{\exp\left[i(\vec{k}_1-\vec{k}_2)\cdot\vec{R}_n + i(\vec{k}_3-\vec{k}_1)\cdot\vec{R}_\ell + i(\vec{k}_4-\vec{k}_3)\cdot R_m\right]}{(k_2^2-k_1^2)(k_4^2-k_3^2)}\right]\right\} \tag{4}$$

We note that in the BR approximation lower filled bands are assumed to have such a large difference with the conduction band that their contribution is neglected. Converting summations into integration by using $\sum_k = \left[\Omega/(2\pi)^3 \int dk\right]$ and with the aid of the relations

$$\left[I_J^z, I_k^+\right] = \delta_{jk}\, I_j^+, \quad \left[I_j^z, I_k^-\right] = -\delta_{jk}\, I_j^-, \quad \left[I_j^+, I_k^-\right] = 2\delta_{kj} I_j^z\ , \tag{5}$$

$$\sum_{n,m,\ell} = \sum_{n,m,\ell} \delta_{nm}\delta_{m\ell} + \sum_{n\neq\ell,m}{}' \delta_{nm} + \sum_{m\neq\ell,n}{}' \delta_{n\ell} + \sum_{n\neq m,\ell}{}' \delta_{m\ell} + \sum_{n\neq m\neq\ell}{}''$$

eq. (4) can be written in the form[3]

$$H_3 = -\sum_n A_{nn}\, \vec{I}_n\cdot\vec{I}_n + \sum_{n\neq\ell}{}' B_{n\ell}\, \vec{I}_n\cdot\vec{I}_\ell - 2\sum_{\ell\neq m} C_{mn}\, \vec{I}_m\cdot\vec{I}_n \tag{6}$$

$$A_{nn} = \frac{m'^2 J^3}{(2\pi^2)^3}\left[\int_0^\infty \frac{dk_1 k_1^2}{(k_1^2+E)^2} \int_0^{k_T} k_2^2\, dk_2 \int_0^{k_T} dk_4 k_4^2\right.$$

$$\left. + \int_0^{k_T} dk_2 k_2^2 \int_0^\infty \frac{dk_1 k_1^2}{k_1^2+E} \int_0^\infty \frac{dk_3 k_3^2}{k_3^2+E}\right] \tag{7}$$

$$B_{n\ell} = \frac{m'^2 J^3}{R^2_{n\ell}(2\pi^2)^3} \left[\int_0^\infty \frac{dk_1 k_1^2}{(k_1^2+E)^2} \int_0^{k_T} dk_2 k_2 \sin(k_2 R_{n\ell}) \int_0^{k_T} dk_4 k_4 \sin(k_4 R_{n\ell}) \right.$$

$$\left. + \int_0^{k_T} dk_2 k_2^2 \cdot \int_0^\infty \frac{dk_1 k_1 \sin(k_1 R_{n\ell})}{(k_1^2 + E)} \int_0^\infty \frac{dk_3 \sin(k_3 R_{n\ell})}{(k_3^2 + E)} \right]$$

$$C_{mn} = \frac{m'^2 J^3}{R^2_{mn}(2\pi^2)^3} \left[\int_0^\infty \frac{dk_1 k_1 \sin(k_1 R_{mn})}{(k_1^2+E)^2} \int_0^{k_T} dk_2 k_2^2 \int_0^{k_T} dk_4 k_4 \sin(k_4 R_{mn}) \right.$$

$$\left. + \int_0^{k_T} dk_2 k_2 \sin(k_2 R_{mn}) \int_0^\infty \frac{dk_1 k_1^2}{(k_1^2+E)} \int_0^\infty \frac{dk_3 k_3 \sin(k_3 R_{mn})}{(k_3^2 + E)} \right]$$

$E = \frac{2m'}{\hbar^2} E_g$, E_g is the energy gap, k_T is the Brillouin zone radius.

Straigthforward integration yields

$$A_{nn} = \frac{m'^2 J^3 k_T^6}{9(2\pi)^5 \sqrt{E}} + \text{divergent term}$$

$$B_{n\ell} = \frac{m'^2 J^3 k_T^6}{(2\pi)^5 \sqrt{E}} \left\{ \left[xF(x) \right]^2 + \left[\frac{\pi \sqrt{E}}{3k_T x^2} e^{-2R_{n\ell}\sqrt{E}} \right] \right\} \tag{8}$$

$$C_{mn} = \frac{-m'^2 J^3 k_T^6}{3(2\pi)^5 \sqrt{E}} \left[xF(x) \right] e^{-R_{mn}\sqrt{E}} + \text{divergent term}$$

$$x = k_T R_{n\ell}$$

Divergencies[16] in third order of perturbation theory of the indirect exchange interaction has been attributed at least in part to arise from a Kondo effect and incorrent account of the motion of the atoms due to phonons.

The fact that A_{nn} is infinite is not interesting since A_{nn} is a self energy, range independent, and does not give interaction between two different spins[3].

In order to make (C'_{mn}) the second divergent term of C_{mn} convergent we replace the upper limit ∞ for k_1 and k_3 by a $k_{cutt\text{-}off} = k_{c.o.}$ and obtain

$$C'_{mn} = \frac{m'^2 J^3}{R_{mn}^2 (2\pi^2)^3} \left[\frac{\sin k_T R_{mn} - k_T R_{mn} \cos k_T R_{mn}}{R_{mn}^2} \right]$$

$$\times \left[k_{c.o.} - \sqrt{E} \arg tg \left(\frac{k_{c.o.}}{\sqrt{E}}\right) \right] \int_0^{k_{c.o.}} \frac{dk_3 k_3 \sin(k_3 R_{mn})}{(k_3^2 + E)} \qquad (9)$$

For large $k_{c.o.}$, replacing the upper limit $k_{c.o.}$ for k_3 by ∞ the leading term of C_{mn} becomes

$$C'_{mn} = \frac{-m'^2 J^3 k_T^4 \, k_{c.o.}}{16\pi^5} F(k_T R_{mn}) \, e^{-R_{mn}\sqrt{E}} \qquad (10)$$

Thus in third order the indirect exchange interaction has for large $k_{c.o.}$ the asymptotic form

$$H_{int} = \frac{m'^2 J^3 k_T^4 \, k_{c.o.}}{8\pi^5} \sum_{n,m}{}' \; F(k_T R_{mn}) \, e^{-R_{mn}\sqrt{E}} \; \vec{I}_m \cdot \vec{I}_n \qquad (11)$$

Comparing with the usual BR result eq. (2) we find then to be of the same order if

$$k_{c.o.} = \frac{\pi^2}{m'J}$$

In EuO and EuS for example, we have[7] (EuO) = 0.32 eV and Γ(EuS) = 0.2 eV; $J = \Omega\Gamma$, where Ω is the atomic volume, α'(EuO) = 0.4 and α'(EuS) = 0.3, $\alpha' = m_e/m'$, m_e is a free electron mass $k_T = 2\pi/a_0\ (3/\pi)^{1/3}$, a_0 is the lattice parameter k_T(EuO) = 1.20 Å^{-1} , k_T(EuS) = 1.04 Å^{-1}. Using these values we obtain $k_{c.o.}$(EuO) = 2.29 k_T(EuO) $k_{c.o.}$(EuS) = 2.05 k_T(EuS).

Thus for these systems to which the BR model has been applied[7] the third order contribution will be of the same order of magnitude as the second order contribution if $k_{c.o.}$ is of the same order of magnitude of k_T.

At least a rough estimate of the conduction band width in these systems indicates that it is important to consider the fact that the band width is finite[4].

We thus considered it of interest to derive a general formula for the third order contribution introducing the finite conduction band width ($E_g + \hbar^2 W^2/2m'$) as a variable parameter.

In our third order model with a finite conduction band we replace all upper limits ∞ in A_{nn}, $B_{n\ell}$ and C_{mn} by a finite W.

Straightforward integration then yields

$$A_{nn} = \frac{m'^3 J^3}{(2\pi^2)^3} \left\{ \frac{k_T^6}{9} \left[\frac{1}{2\sqrt{E}} \text{ arctg } \frac{W}{\sqrt{E}} - \frac{W}{2(W^2+E)} \right] + \frac{k_T^3}{3} \left[W - \sqrt{E} \text{ arctg } \frac{W}{\sqrt{E}} \right]^2 \right\}$$

$$B_{n\ell} = \frac{m'^2 J^3}{R_{n\ell}^2 (2\pi^2)^3} \left\{ \frac{1}{R_{n\ell}^4} \left[\frac{1}{2\sqrt{E}} \text{ arctg } \frac{W}{\sqrt{E}} - \frac{W}{2(W^2+E)} \right] \left[\sin(k_T R_{n\ell}) \right.\right.$$

$$\left.\left. - k_T R_{n\ell} \cos(R_{n\ell} k_T) \right]^2 + \frac{k_T^3}{3} \left[(B - \frac{\pi}{2}) \cosh(R_{n\ell}\sqrt{E}) + D \sinh(R_{n\ell}\sqrt{E}) \right]^2 \right\} \tag{13}$$

$$C_{mn} = \frac{m'^2 J^3}{R_{mn}^4 (2\pi^2)^3} \left[\sin(R_{mn} k_T) - R_{mn} k_T \cos(k_T R_{mn}) \right] \left\{ \frac{k_T^3}{3} \right.$$

$$\times \left[\left(\frac{R_{mn}}{4\sqrt{E}}\right) \left[\sinh(R_{mn}\sqrt{E})(\pi - 2B) - 2D\cosh(R_{mn}\sqrt{E}) \right] \right.$$

$$\left. - \frac{1}{2} \frac{\sin(R_{mn} W)}{W^2 + E} \right] + \left[W - \sqrt{E} \text{ arctg}\left(\frac{W}{\sqrt{E}}\right) \right] \left[(B - \frac{\pi}{2}) \cosh(R_{mn}\sqrt{E}) + \right.$$

$$\left.\left. + D \text{ senh}(R_{mn}\sqrt{E}) \right] \right\}$$

$$B = \mathrm{Re}\,\mathrm{Si}(u)\Big|_{k=0}^{k=W} \quad \text{where Si is the sine integral function}$$

$$D = \mathrm{Im}\,c_i(u)\Big|_{k=0}^{k=W} \quad \text{where } c_i \text{ is the cosine integral function}$$

$$u = R(\sqrt{E}\, i + k) \qquad R = R_{mn}, R_{n\ell}$$

We shall study numerically C_{mn} and $B_{n\ell}$ in the form

$$C_{mn} = A\phi_C$$

$$\phi_C = \left[-F(q)\right] \left\{ \frac{q}{12\sqrt{y'}} \left[(\pi-2B)\sinh(q\sqrt{y'}) - 2D\cosh(q\sqrt{y'})\right] - \frac{\sin(qz)}{2(z^2+y')} + \left[z-\sqrt{y'}\ \mathrm{arctg}\left(\frac{z}{\sqrt{y'}}\right)\right]\left[(B-\frac{\pi}{2})\cosh(q\sqrt{y'}) + D\,\mathrm{senh}(R_{mn}\sqrt{E})\right] \right\}$$

$$B_{n\ell} = A\phi_B$$

$$\phi_B = \left\{ \left[qF(q)\right]^2 \left[-\frac{z}{2(z^2+y')} + \frac{1}{2\sqrt{y'}}\ \mathrm{arctg}\ \left(\frac{z}{\sqrt{y'}}\right)\right] + \frac{1}{3q^2}\left[(B-\frac{\pi}{2})\ \cosh(q\sqrt{y'}) + D\sinh(q\sqrt{y'})\right]^2 \right\}$$

$$A = \frac{m'^2 J^3 k_T^5}{(2\pi^2)^3}$$

$$B = \sum_{k=1}^{\infty} \frac{(-1)^{k+1}\ (q\sqrt{z^2+y'})^{2k-1}}{(2k-1)!\,(2k-1)} \cos(2k-1)\theta \qquad \theta = \mathrm{arctg}\left(\frac{\sqrt{y'}}{z}\right)$$

$$D = \theta + \sum_{k=1}^{\infty} \frac{(-1)^k (\sqrt{y' + z^2}\, q)^{2k}}{(2k)!(2k)!} \sin(2k\theta)$$

$$\phi_{BR} = -F(q)\exp(-q\sqrt{y'})$$

$$q = k_T R$$

$$y' = \frac{E_g}{\alpha' E_T} \qquad E_T = \frac{\hbar^2 k_T^2}{2m_e} \qquad y = \frac{E_g}{\alpha E_T}$$

$$\alpha' = m_e/m' \qquad y' = \gamma^2 y \qquad \gamma^2 = \alpha'/\alpha$$

$$z = W/k_T$$

We note that Si(u), (B = ReSi(u)) is an entire function of u; on the other hand ci(u), (D = Im ci(u)) is an infinitely many-valued function of u whose circuit relation is $ci(ue^{im\pi}) = ci(u) + m\pi i$. We have used the principal part (m = 0) in our calculations of ϕ_B, ϕ_C vs q (Figs. 1-3) since for each curve, $\theta = \text{arctg}\,(\sqrt{y'}/z)$ is mantained fixed for a given y' and z.

In Figs. 1 and 2 we plot ϕ_C vs q for different values of z (conduction band width parameter) and y' (energy gap parameter). The Bloembergen-Rowland formula (ϕ_{BR}) is given for comparison. It may be noted that the third order perturbation series term correction with finite conduction band effects included (ϕ_C) is also oscillatory with q (distance between localized magnetic moments) as in the BR model. The phase, frequency and magnitude of the oscillations are however strongly modified. The term ϕ_B (Fig. 3) mainly damps without changing the sign of the exchange interaction.

As expected both $d\phi_B/dy'$ and $d\phi_C/dy'$ are negative, that is the interaction decreases with increasing values of y' (energy gap). On the other hand both $d\phi_B/dz$ and $d\phi_C/dz$ are positive, i.e., the interaction increases with increasing values of z (band width parameter). We also note that ϕ_B, ϕ_C and ϕ_{BR} are of the same order of magnitude when z is of the same order as k_T.

Of interest is the fact that opposed to the second order BR term, the energy gap in the third order term (ϕ_C) not only damps but modifies as well the phase of the oscillations (sign of the exchange interaction).

Our calculations indicate that ϕ_C also oscillates with z (band width parameter), i.e., the exchange interaction is alternatively ferromagnetic and antiferromagnetic.

Band theoretical parameters such as the energy gap and conduction band width thus appears to play a more important role in our third order term and may thus be useful for correlating the band structure of rare earth systems with their magnetic properties.

The influence of third order matrix elements of the form $\langle n|H_{int}|m\rangle$ where $|n\rangle$ and $|m\rangle$ are excited states are not taken into account in the BR formula which is obtained by a perturbation calculation up to second order. The explicit calculation of these matrix elements in the present paper suggest that they may considerable contribute to the interaction between two nuclear or ironic spins.

We would like to note however that other factors may also significantly affect the exchange interaction, such as finite temperatures, mean free path effects, non-spherical Fermi surfaces, a more realistic expression for the exchange integral and higher order contributions. Some of these effects are presently under investigation by our group.

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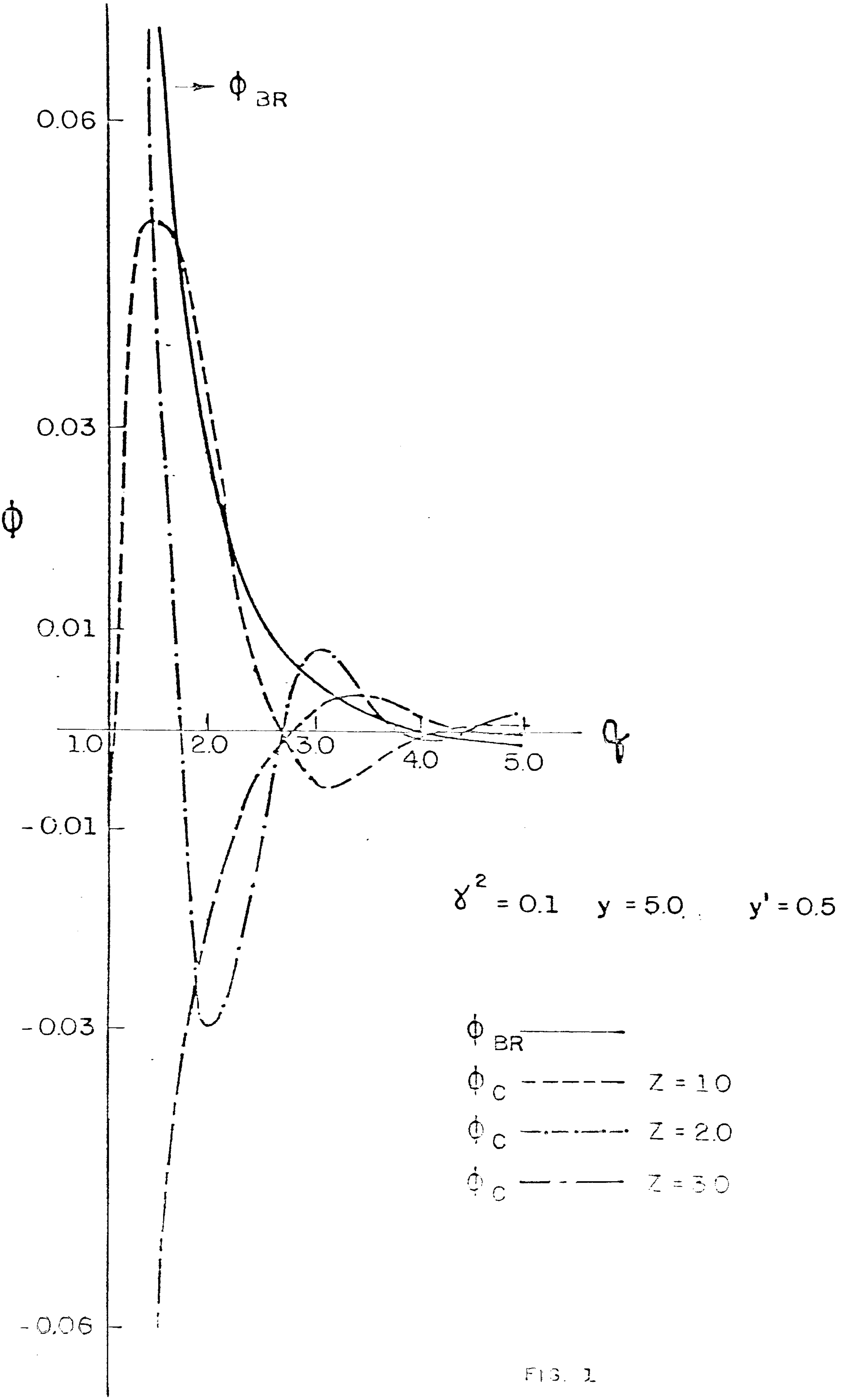
FIGURE CAPTIONS

Fig. 1 - ϕ_C and ϕ_{BR} vs q (distance between magnetic moments) , y = 5.0 (energy gap) and several values of z (conductor band width parameter).

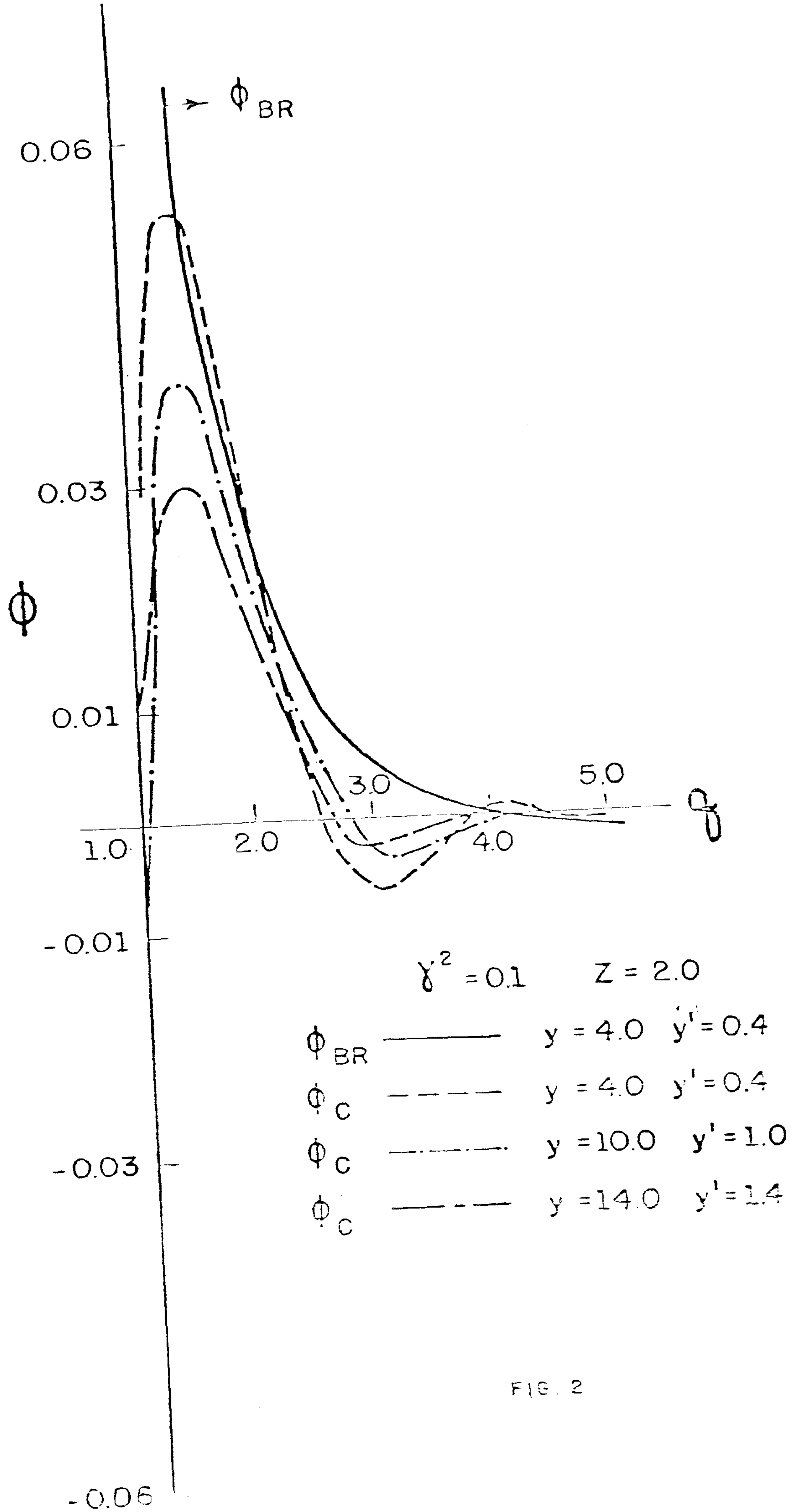
Fig. 2 - ϕ_C and ϕ_{BR} vs q (distance between magnetic moments) , z = 2.0 (conduction band width parameter) and several values of y (energy gap).

Fig. 3 - ϕ_B vs q (distance between magnetic moments), z = 2.0 (conduction band width parameter) and several values of y (energy gap).



Φ_{BR}
0.06
0.03
Φ
0.01
1.0
2.0
3.0
4.0
5.0
q
-0.01
-0.03
-0.06
$\gamma^2 = 0.1$ $y = 5.0$ $y' = 0.5$
Φ_{BR}
Φ_C Z = 1.0
Φ_C Z = 2.0
Φ_C Z = 3.0

FIG. 1



Φ BR
0.06
0.03
0.01
-0.01
-0.03
-0.06
Φ
1.0
2.0
3.0
4.0
5.0
ϑ
γ² = 0.1 Z = 2.0
Φ BR y = 4.0 y' = 0.4
Φ C y = 4.0 y' = 0.4
Φ C y = 10.0 y' = 1.0
Φ C y = 14.0 y' = 1.4

FIG. 2

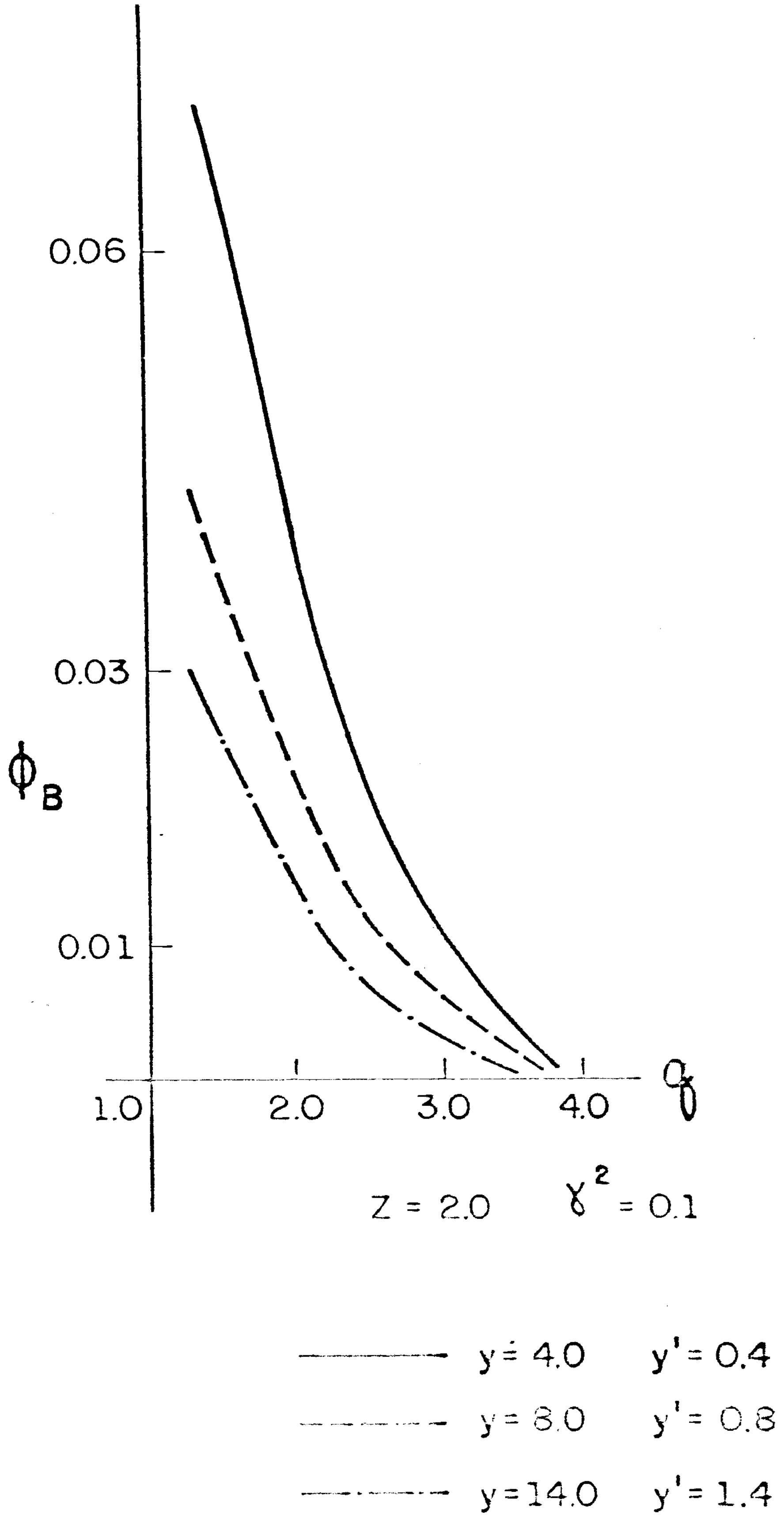


FIG. 3