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## INDIRECT EXCHANGE IN INSULATORS AND INTRINSIC SEMICONDUCTORS

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## ABSTRACT

The indirect interaction between localized magnetic moments in insulators and semiconductors is calculated at  $T = 0^{\circ}K$ . This interaction arises from virtual excitations of the valence electrons to the conduction band and is oscillatory as a function of distance. The passage from the case of small gap semiconductors to insulators is studied in detail, as a function of the energy gap and the effective masses. The Bloembergen-Rowland interaction is derived as a limiting case for insulators. Possible applications are also discussed.

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## 1. INTRODUCTION

It is well known that the localized magnetic moments of ions or of nuclei in metals are indirectly exchange-coupled through the conduction electrons <sup>1, 2</sup>. The proper calculation including both diagonal and off-diagonal effects was given by Kasuya <sup>3, 4</sup> and a little later by Yosida <sup>5</sup>. This interaction, because of its long-range and oscillatory nature, can give rise to complex ordered magnetic structures as shown, for instance, by rare-earth metals and alloys. However, in its original analytic form the theory necessarily involved several important approximations. This problem has been considered in detail by many authors - it is impossible to make justice to all of them - we shall mention only those really connected with this work. (See reference 6 and references therein).

In insulators there exists a similar interaction which arises from virtual excitations, through the energy gap, of the valence band electrons <sup>7</sup>. An evaluation of the integrals involved in a realistic case is not elementary, so Bloembergen and Rowland <sup>7</sup> calculated the interaction by invoking drastic approximations, the most important being that the energy gaps were assumed to be much greater than the valence band width. This necessarily excludes semiconductors.

The purpose of this paper is, essentially, to extend the Bloembergen-Rowland formula to intrinsic semiconductors, for  $T = 0^{\circ}\text{K}$ .

Besides the study of NMR linewidth in insulators and semiconductors (where the localized moments are nuclei) the indirect interaction studied here may play a role in the interpretation of the properties of magnetic semiconductors like Eu chalcogenides where the localized moments are the 4f

shells (EuO, EuS, EuSe and EuTe) 8, 9, 10.

## 2. FORMULATION OF THE PROBLEM

We begin by considering a system of localized spins  $\vec{S}_j$  in a semiconductor at the positions  $\vec{R}_j$ , which interacts with a valence band electron with spin  $\vec{s}$ , and wave function

$$\langle \vec{x} | \vec{k} \rangle = \frac{1}{\sqrt{V}} u_{\vec{k}}(\vec{x}) \quad (1)$$

through the potential

$$H = - \sum_j 2 \Gamma(\vec{x} - \vec{R}_j) (\vec{s} \cdot \vec{S}_j)$$

$\Gamma(\vec{x} - \vec{R}_j)$  is the exchange interaction between  $\vec{s}$  and  $\vec{S}_j$ . Now, in order to treat ellipsoidal energy surfaces, we can formally transform  $\vec{k}$  and  $\vec{R}$  spaces into  $\vec{k}'$  and  $\vec{\rho}$ -spaces such that  $\vec{k} \cdot \vec{R} = \vec{k}' \cdot \vec{\rho}$  by defining

$$k_i = \frac{k_i}{\sqrt{m_i}} \quad \text{and} \quad \rho_i = \sqrt{m_i} R_i \quad i = 1, 2, 3$$

Furthermore we shall assume that the dispersion relations  $\epsilon(k)$  in the valence band and  $\epsilon'(k')$  in the conduction band depend only on  $|\vec{k}|$  and  $|\vec{k}'|$  say  $\epsilon(\vec{k}) = k^2$  and  $\epsilon'(\vec{k}') = k'^2$ , i.e. are constants over ellipsoidal surfaces. This will enable us to perform the integrations over the angles in  $\vec{k}$  and  $\vec{k}'$ -spaces. We shall comment on this approximation later.

In a straightforward way, second order perturbation theory gives us

$$E_{ij} = - 2 A_{ij} \vec{S}_i \cdot \vec{S}_j$$

$$A_{ij} = \frac{8\Omega^2 (m_x m_y m_z m'_x m'_y m'_z)^{1/2}}{(2\pi)^4 \hbar^2 \rho \rho'} B \quad (2)$$

$\Omega$  is the volume of the elementary cell

$$B = - \int_0^{k_T} dk \int_0^{\infty} dk' \Gamma_S(\vec{k}, \vec{k}') \frac{kk' \sin k\rho \sin k'\rho'}{k^2 + k'^2 + \epsilon} \quad (3)$$

If  $\Gamma_S(\vec{k}, \vec{k}')$  is independent of  $k'$ , the integral in  $k'$  is analitic, that is

$$B = - \frac{\pi}{2} \Gamma \frac{\rho^2 k_T^4}{q^2} \int_0^1 dx x \sin(qx) \exp \left\{ -\rho' (k_T^2 x^2 + \epsilon)^{1/2} \right\}$$

$$\epsilon = \frac{2}{\hbar^2} E_g \quad (E_g \text{ is the energy gap})$$

$$q = k_T \rho$$

$\Gamma(\vec{k}, \vec{k}')$  is the so-called exchange interaction parameter. If  $\Gamma(\vec{x} - \vec{R}_j)$  is a delta-type interaction  $\Gamma\delta(\vec{x} - \vec{R}_j)$  (the Fermi contact interaction), this reduces to  $\Gamma_S(\vec{k}, \vec{k}') = \Gamma u_{\vec{k}}^*(0) v_{\vec{k}'}(0)$  which may be assumed to be independent of  $k$  and  $k'$  (this corresponds to the Wigner Seitz approximation).

### 3. COMMENTS ON THE APPROXIMATIONS

The simple derivation used here involves perturbation theory, so it is only valid provided that  $\Gamma_S(\vec{k}, \vec{k}')$  is much smaller in magnitude than the characteristic energies of a conduction particle. This is of course true if

one is considering the problem of indirect interactions of nuclear moments (which, by the way, was the problem originally studied by Ruderman-Kittel and Bloembergen-Rowland). We shall assume, as usual <sup>2, 3, 4</sup> that this is also true for localized electron shells.

In principle, of course, one may use a double integral to evaluate the interaction for arbitrary  $\epsilon(\vec{k})$  and  $\epsilon'(\vec{k}')$  instead of  $k^2$  and  $k'^2$ . In a real situation where  $\epsilon(\vec{k})$  and  $\epsilon'(\vec{k}')$  depend on the direction in  $\vec{k}$ -space, a better estimate of the total energy may be obtained by a weighted sum over the Brillouin zone

$$\epsilon = \sum_i P_i \epsilon_i(\vec{k})$$

$P_i$  being proportional to the degeneracy and the relative volume in the Brillouin zone of a particular direction for which the energy is assumed to depend only on  $|\vec{k}|$ .

However we shall use  $\epsilon(\vec{k}) = k^2$  and  $\epsilon'(\vec{k}') = k'^2$ , which are not very strong assumptions. It is reasonable to assume that the interaction is not very sensitive to the details of the conduction band structure. This comes from the fact that in perturbation theory the most important matrix elements are those connecting the top of the valence band to the bottom of the conduction band. In these regions the dispersion relations may be assumed to be parabolic. In metals a computation of the indirect interactions using non-spherical Fermi surface <sup>11</sup> has shown that the details of the dispersion relations are not very important in the final result and that the parabolic approximation is qualitatively good. Furthermore the success of the Ruderman-Kittel interaction in the qualitative inter-

pretation of several properties of rare-earth metals and alloys<sup>12, 13</sup> induces us to believe that the Bloembergen-Rowland scheme can be equally useful in the study of the magnetic properties of rare-earth non-conducting compounds<sup>13</sup>, specially magnetic semiconductors<sup>8</sup>.

#### 4. GENERAL DISCUSSION OF THE RESULTS

The indirect interaction between localized magnetic moments in a semiconductor is given by equations (2) and (3). Of course we need to consider only the interaction in the direction of principal axes,  $m_x$  and  $m'_x$  which shall be assumed to be parallel. This results from the fact that the principal axes are determined by the symmetries of the crystal. So,

$$A_{ij} = - \frac{2 \Omega^2 \Gamma_s^2 k_T^4}{(2\pi)^3 \hbar^2} M' \Phi \quad (4)$$

$$\Phi = \frac{1}{q^2} \int_0^1 dx x \sin(qx) \exp\{-\rho'(k_T^2 x^2 + \epsilon)^{1/2}\}$$

$$M' = \left( \frac{m_x}{m'_x} \right)^{1/2} \frac{(m_x m_y m_z m'_x m'_y m'_z)^{1/2}}{m_x^2 + m_y^2 + m_z^2}$$

and  $M' = m'$  in the case of spherical Fermi surfaces, where  $m_x = m_y = m_z = m$ ,  $m'_x = m'_y = m'_z = m'$ .

We shall study numerically this integral in the form

$$\Phi = \frac{1}{q^2} \int_0^1 dx x \sin(qx) e^{-A} \quad (5)$$

$$A = q [y' + \gamma^2 x^2]^{1/2}$$

where

$$\gamma^2 = \frac{\alpha_X}{\alpha'_X} = \frac{m'_X}{m_X} \quad \alpha_X = \frac{m_e}{m_X} \quad \text{and} \quad \alpha'_X = \frac{m_e}{m'_X}$$

For spherical Fermi surfaces  $\gamma^2$  indicates roughly the ratio of the valence to conduction band widths.

$$y' = \frac{E_g}{\alpha'_X E_T} = \gamma^2 \frac{E_g}{\alpha_X E_T} = \gamma^2 y$$

$E_g$  is the energy gap, and  $\alpha_X E_T$  is roughly the valence band width. We shall treat  $y'$  and  $\gamma^2$  as independent parameters.

For a typical intrinsic semiconductor  $\gamma^2$  may be

$$.1 \leq \gamma^2 \leq 1$$

and in an insulator one has  $\gamma^2 \sim 0$  and  $y'$  is finite. In this case equation 5 gives us the Bloembergen-Rowland<sup>7</sup> result

$$\phi = -e^{-q\sqrt{y'}} F(kR) \quad F(x) = \frac{x \cos x - \sin x}{x^4}$$

$F(x)$  is the Ruderman-Kittel function<sup>1, 2</sup>.

The Bloembergen-Rowland approximation has been used in the study of the magnetic properties of Eu-chalcogenides<sup>14, 15, 16</sup> and  $\text{EuB}_6$ <sup>17</sup>. Now if one intends to apply the indirect interaction to the study of the magnetic semiconductors, it is important to know which are the relevant parameters, independently of the approximations involved in the band structure calculation.

The relative sensitivity of  $\phi$  on the several parameters involved can be seen more vividly in figs. 1 to 7.



$\phi$  is plotted in figures 1, 2 and 3 for different values of  $y'$  and  $\gamma^2$ , illustrating the passage from a small gap semiconductor ( $y' \approx .1$ ) to an insulator ( $y' \approx 2$ ).

In fig. 1 one plots  $\phi$  versus  $q$ , taking  $\gamma^2 = 0.1$ . In general the interaction  $\phi$  in the small gap semiconductor ( $y' \approx .1$ ) is stronger although for special values of  $q$  and in very limited regions, corresponding approximately to the zeroes of  $\phi(q)$ , (for instance  $q \approx 4.9$ ) this may not be true as can also be seen in figure 1. In figs. 2 and 3 one plots  $\phi$  versus  $q$  for  $\gamma^2 = .3$  and  $\gamma^2 = .6$  respectively. The general trend is the same as in fig. 1 but now the relative effect is more pronounced.

In figs. 4 and 5, and in figs. 6 and 7 the interaction  $\phi$  is plotted against  $\gamma^2$  and  $y'$  respectively taking  $q$  as a parameter.

For large values of  $q$  the interaction depends both on  $\gamma^2$  and  $y'$  but, for a given  $q$ , the dependence with  $y'$  is always greater than the dependence with  $\gamma^2$ , as can be seen by comparing (4a) with (6a) ( $q = .4$ ) and (4b) with (6b) ( $q = 1.9$ ) or (5a) with (7a) ( $q = 4.9$ ) and (5b) with (7b) ( $q = 9.9$ ).

Furthermore the dependence of  $\phi$  with  $\gamma^2$  becomes weaker as  $y'$  increases. That is, the indirect interaction becomes less dependent on  $\gamma^2$  in the passage from a semiconductor to an insulator. Finally in an insulator  $y'$  is the only meaningful parameter, as shown before.

## 5. CONCLUDING REMARKS

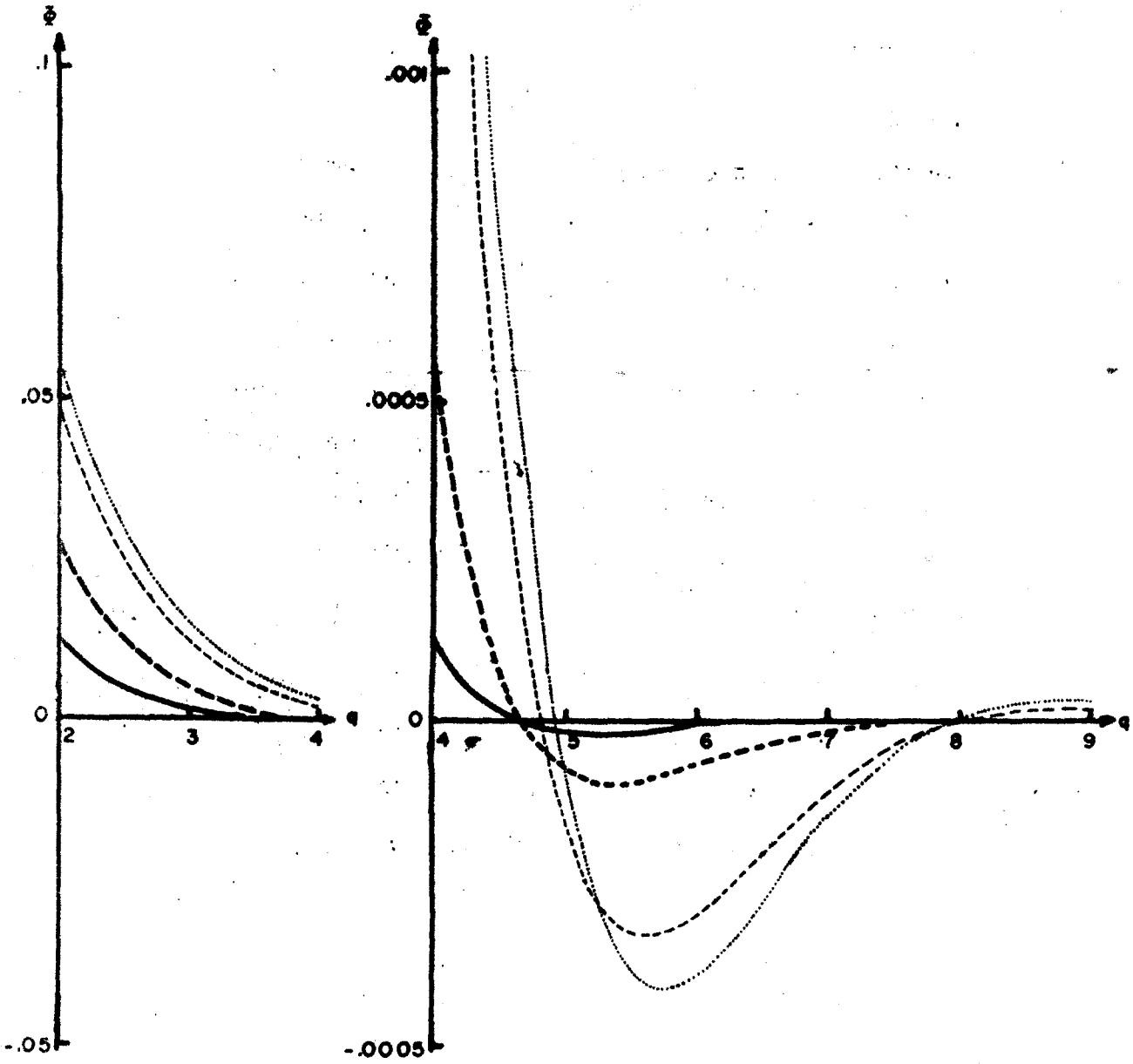
We have shown that in a semiconductor, the indirect interaction between localized magnetic moments depends on  $\gamma^2 = \alpha/\alpha'$  ( $\alpha$  is the effective mass in the valence band as defined by  $E = \alpha \frac{\hbar^2 k^2}{2m}$  and  $\alpha'$  corresponds to the conduction band) and  $y' = E_g/\alpha'$  ( $E_g$  is the energy gap), but the variation with  $y'$  is always dominant. This is a very interesting result, for it shows that the details of the valence band, which determine  $\alpha$ , are less important than the energy gap and the structure of the conduction band or, more exactly than  $E_g/\alpha'$ , in the variation of the magnetic properties of rare earth semiconducting compounds.

This shows that in band structure calculations one should emphasize the detailed structure of the bottom of the conduction band and the values of the energy gaps. Furthermore this analysis shows that the Bloembergen-Rowland approximation already contains the relevant parameters for insulators.

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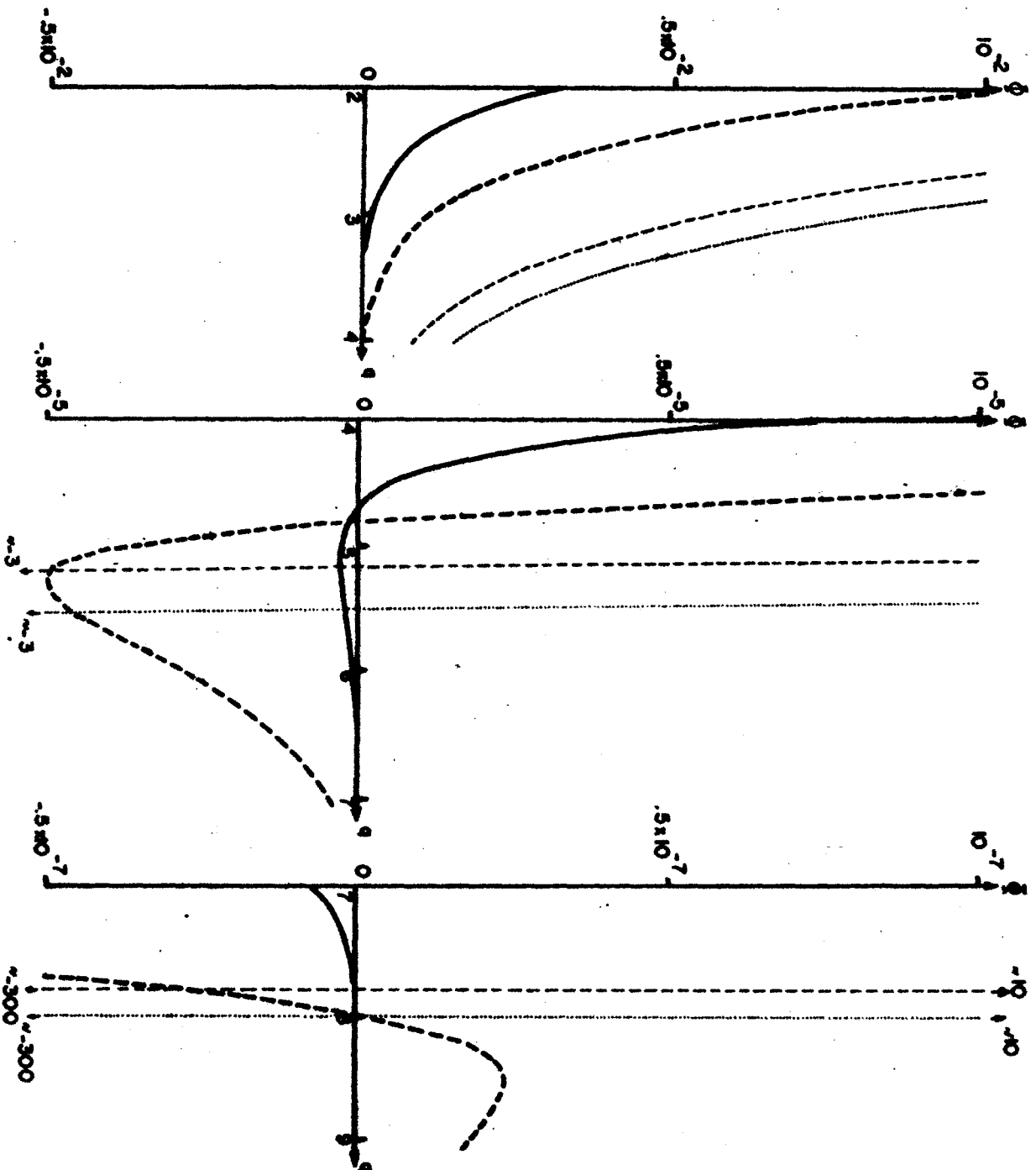
*The authors are grateful to A. A. Gomes, X. A. da Silva and A. P. Guimarães for most enlightening discussions and comments on the manuscript.*



$\sigma^2 = .1$

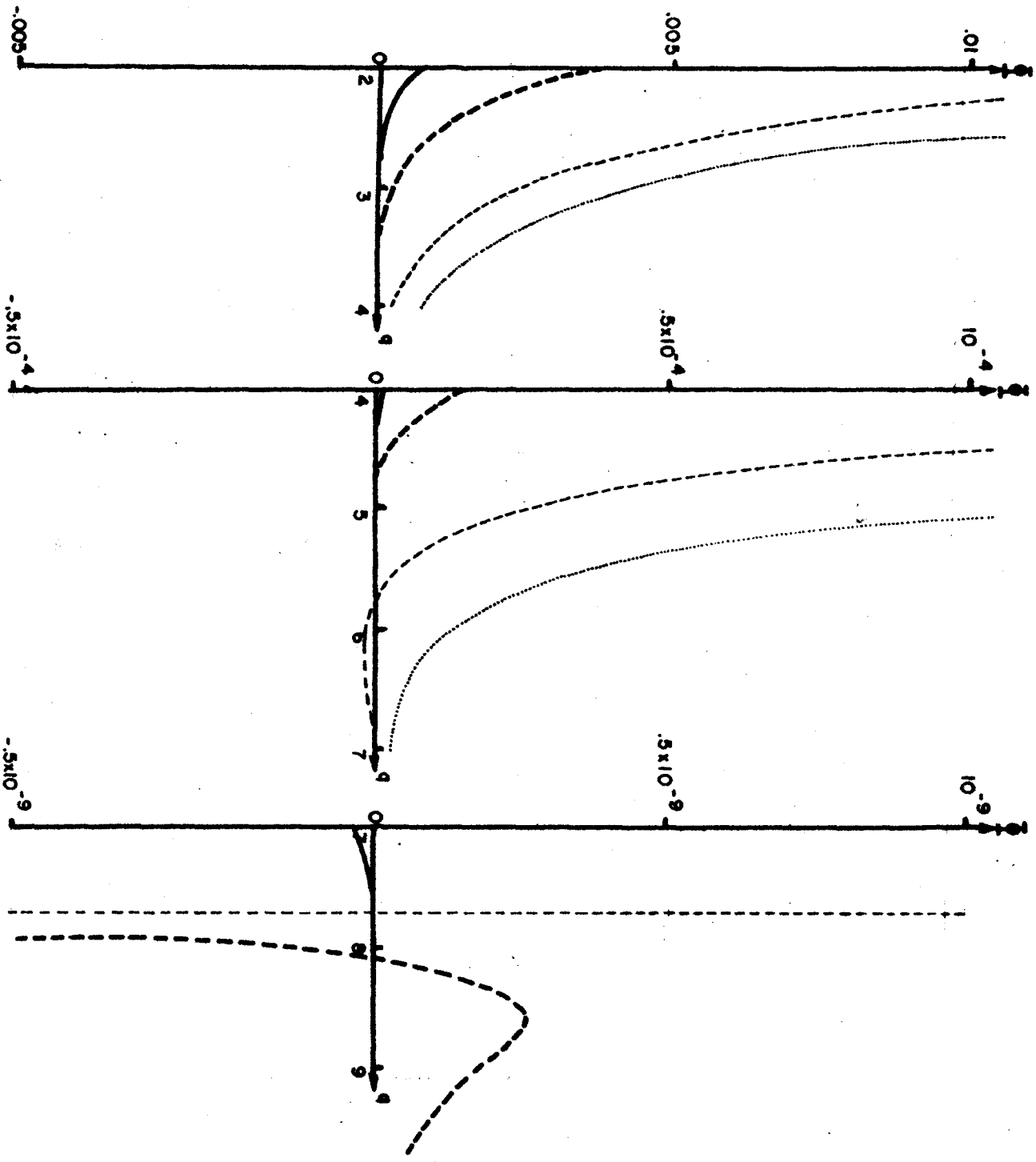
|                          |           |
|--------------------------|-----------|
| $y=0 \Rightarrow y'=.1$  | —————     |
| $y=4 \Rightarrow y'=.4$  | - - - - - |
| $y=1 \Rightarrow y'=.1$  | - · - · - |
| $y=5 \Rightarrow y'=.05$ | · · · · · |

FIG. 1



$\delta = 10^{-2}$   
 $y = 10 \Rightarrow y' = 3$   
 $y = 4 \Rightarrow y' = 1.2$   
 $y = 1 \Rightarrow y' = .3$   
 $y = .5 \Rightarrow y' = .15$

FIG. 2



$y = 10 \Rightarrow y' = 0$   
 $y = 4 \Rightarrow y' = 2.4$   
 $y = 1 \Rightarrow y' = .6$   
 $y = .5 \Rightarrow y' = .3$

FIG. 3

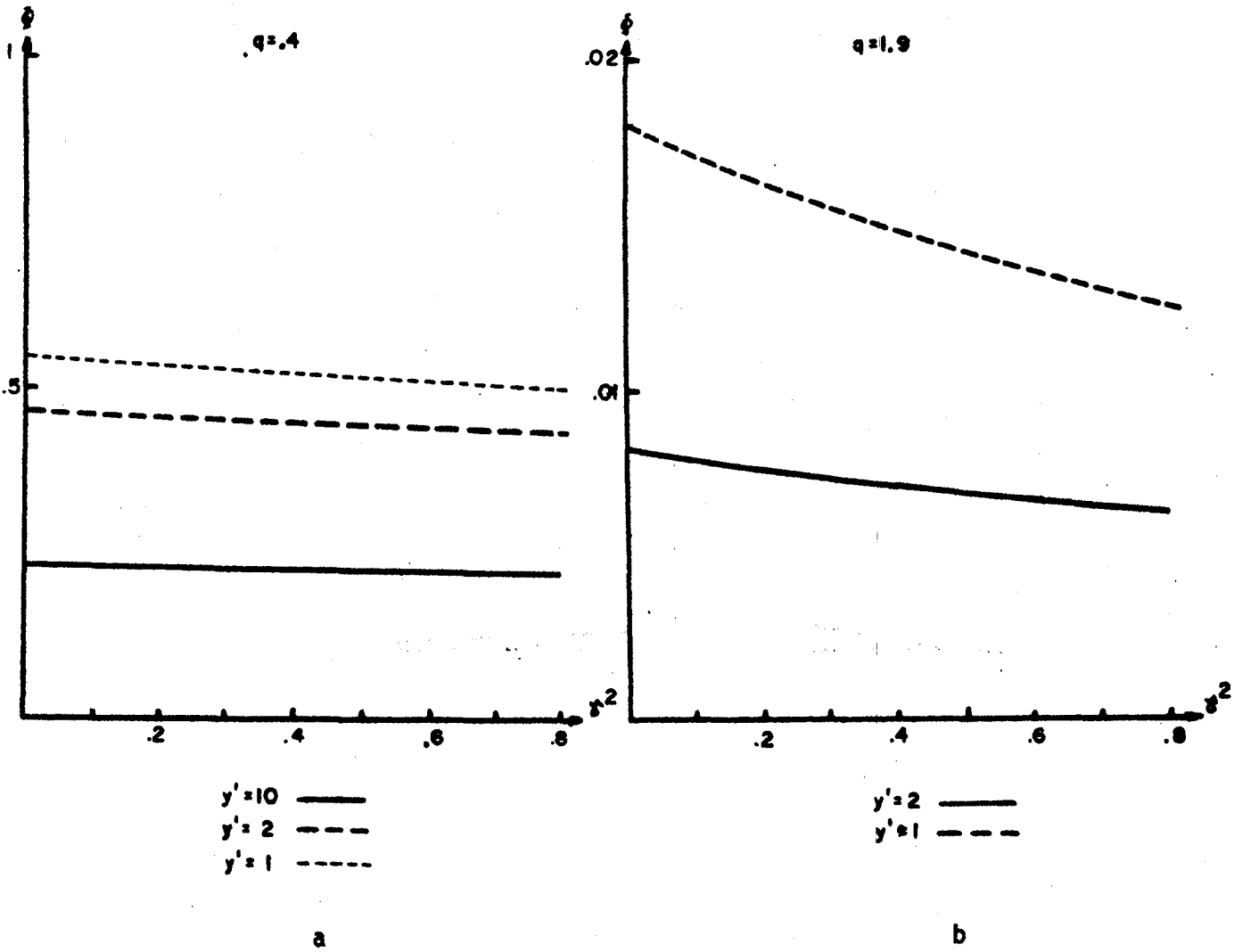


FIG. 4

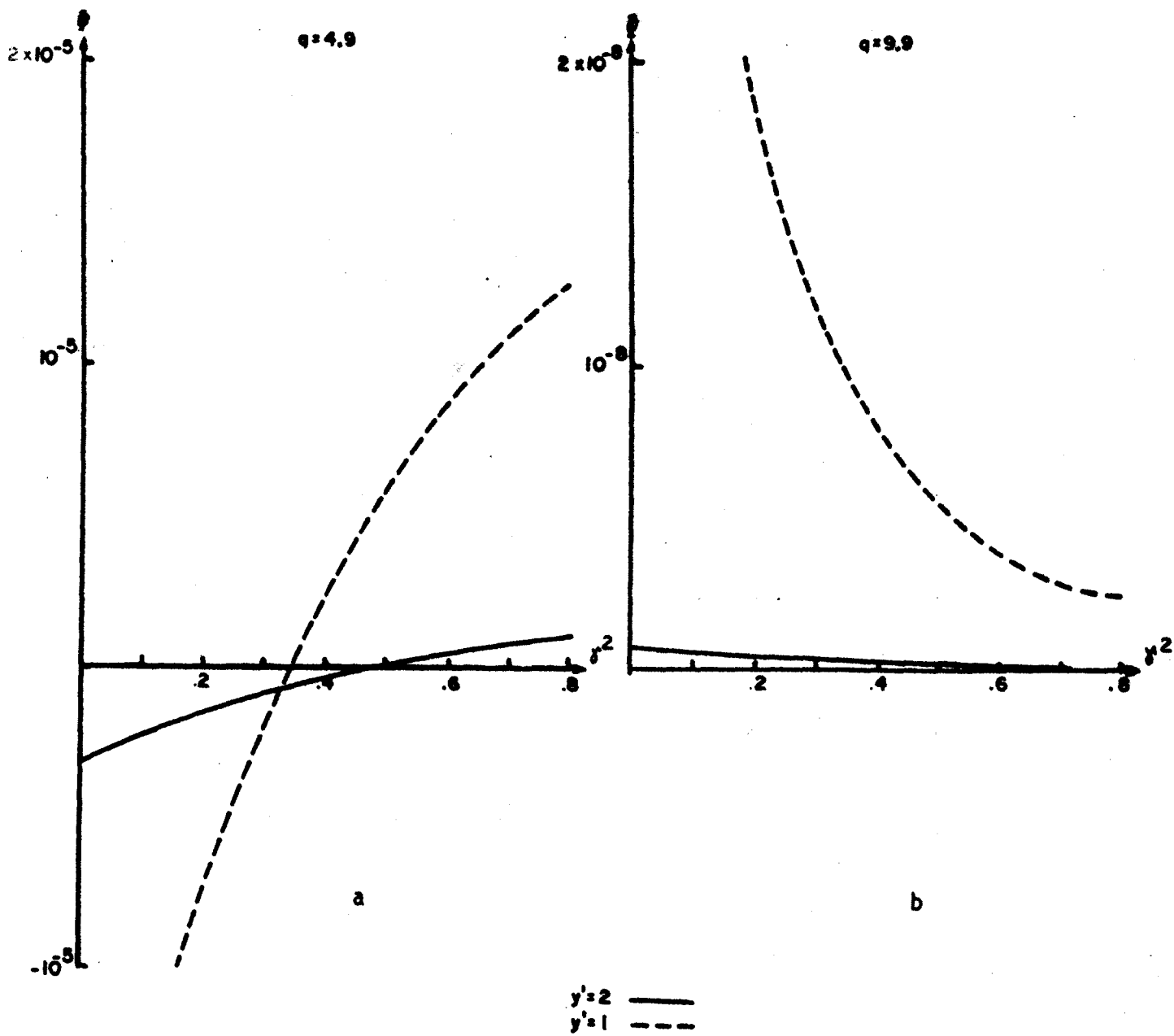


FIG. 5

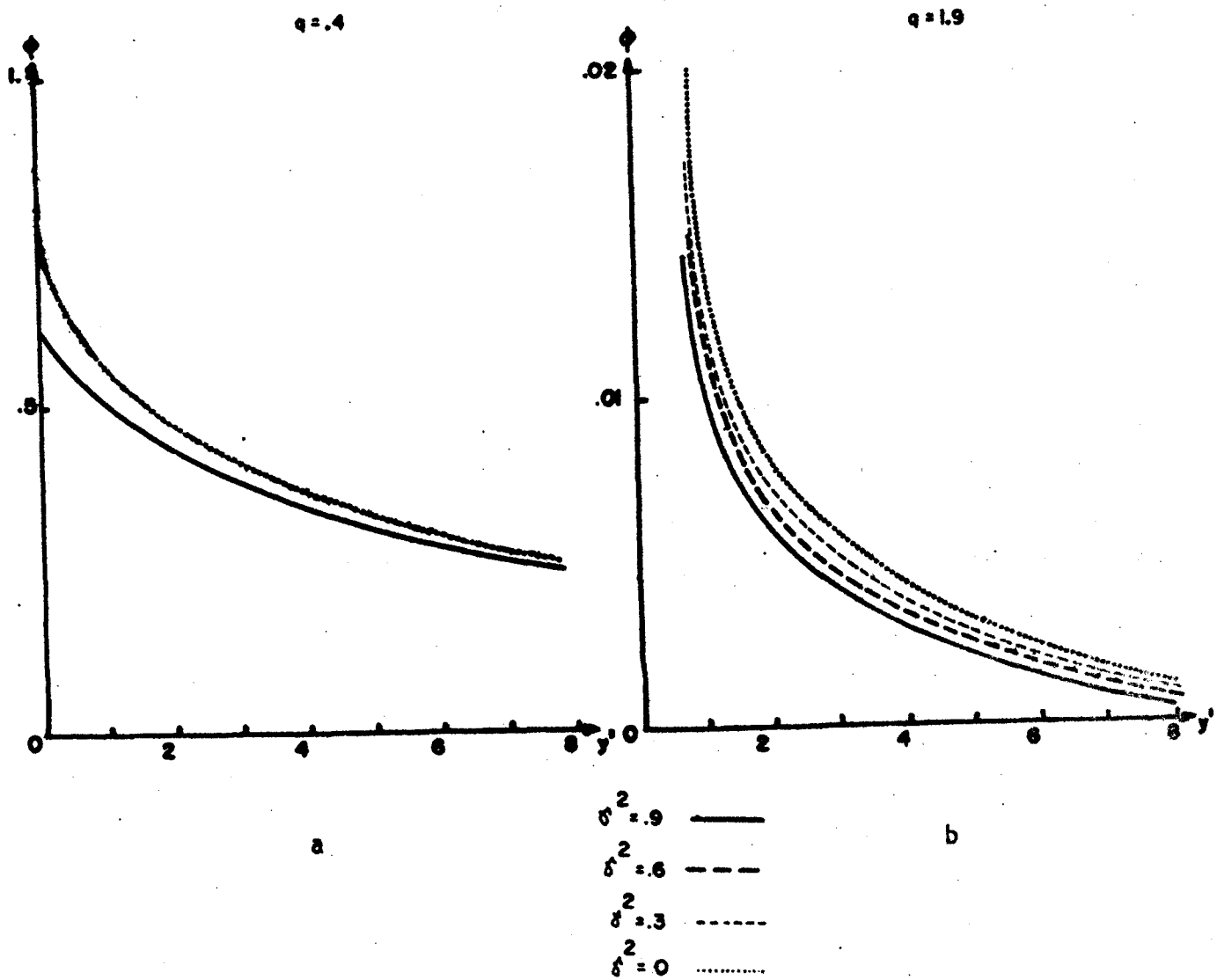
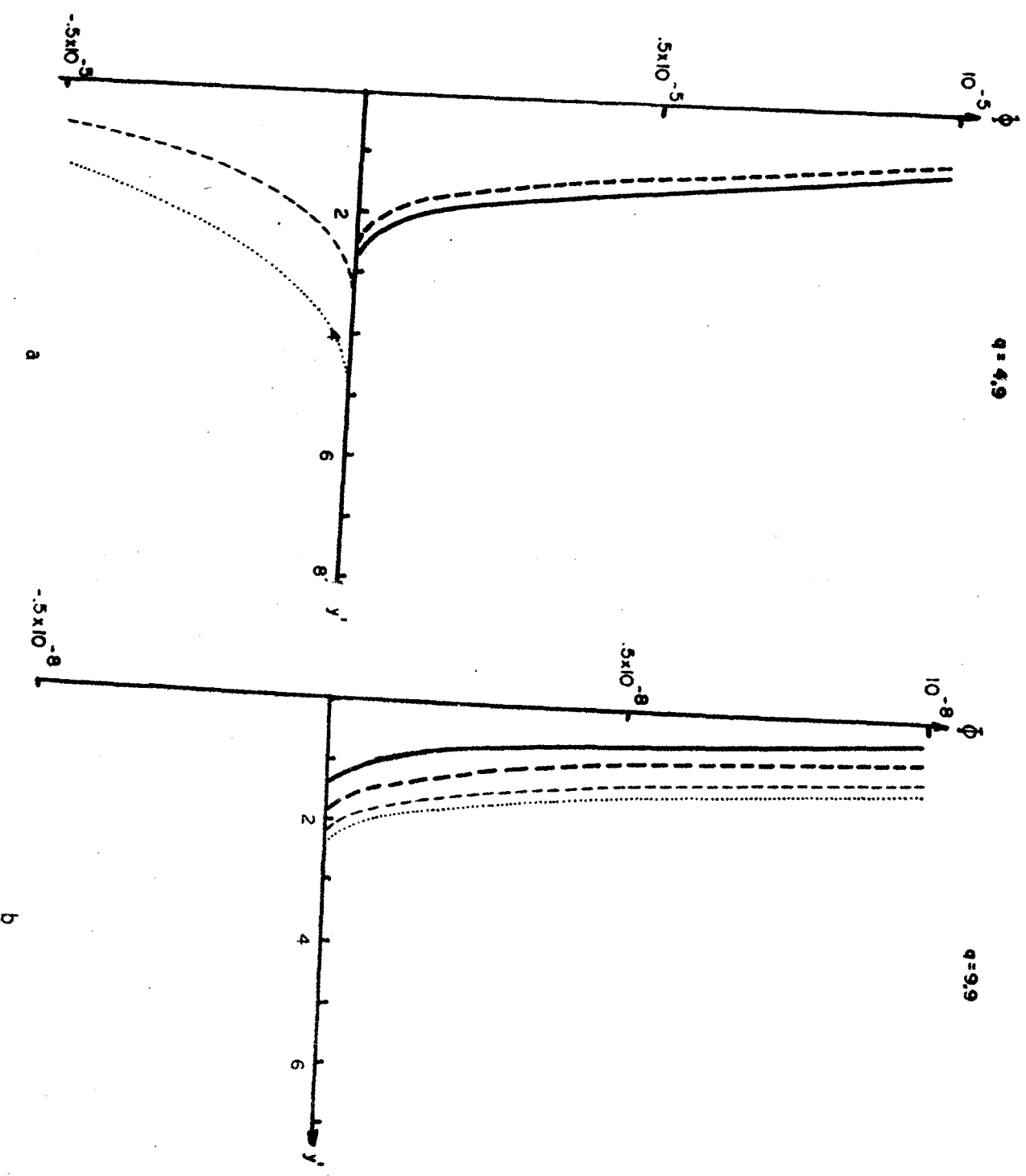


FIG. 6





- $r^2 = .9$  ————
- $r^2 = .6$  - - - -
- $r^2 = .3$  - - - -
- $r^2 = 0$  .....

FIG. 7

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