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by

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Centro Brasileiro de Pesquisas Físicas

Rio de Janeiro, Brazil

INTRODUCTION

The study of the electronic structure of actinide metals has been the subject of increasing interest in recent years. The peculiar magnetic properties of these metals have been discussed by Jullien et al.[1] in terms of the virtual bound state model of Anderson, extended to include the existence of two local levels (d and f) and phenomenological hybridization among them. Latter on, Continentino et al.[2] introduced a band picture to describe the electron-correlations in these metals using the Roth[3] approxi mation. The advantage of the band picture is that it takes into account the fact that due to the larger spatial extension (as compared to the rare-earths) the f-states overlap with each other an important amount. Recently Jullien et al.[4] introduced also the band picture (within the tight binding frame

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work) and described f-d hybridization in terms of transfer integrals between f states and neighbouring d like states. They succeed also in describing the anomalous magnetic behaviour of actinide metals essentially in terms of band-parameters and Coulomb correlations. It is the purpose of this work to give a description (in terms of the Kishore and Joshi [5] model) of a three band problem, namely a broad s-band which overlaps high-density d and f bands. We hope that such a picture approaches a more realistic description of actinide metals, particularly in order to discuss the role of the broad s-band in the electronic structure. Since the broad s-band played a role in producing virtual bound states in the first Jullien et al. [1] approach, it is interesting to discuss its effect within the band model, at least in the hybridized density of states. The plan of this paper is as follows: firstly we discuss the one-electron propagators G^{ss}, G^{dd} and G^{ff} calculated within the Hartree-Fock approximation. Secondly the method of Kishore and Joshi [5] is extended to include the three band model. The third part discuss the numerical results obtained for homothetic bands and model density of states and finally the results are discussed (in particular the implications of the mixings in the Stoner critetion are qualitatively discussed).

1. FORMULATION OF THE PROBLEM

We start discussing the model hamiltonian we adopt to describe hybridization effects for the three band metal. Written the Wannier representation one has:

$$\mathcal{H} = \mathcal{H}_0 + \mathcal{H}_c + \mathcal{H}_h \tag{1}$$

where

$$\mathcal{H}_0 = \sum_{i,j,\sigma} T_{ij}^{(s)} c_{i\sigma}^+ c_{j\sigma} + \sum_{i,j,\sigma} T_{ij}^{(d)} d_{i\sigma}^+ d_{j\sigma} + \sum_{i,j,\sigma} T_{ij}^{(f)} f_{i\sigma}^+ f_{j\sigma} \qquad (2\text{-a})$$

$$\mathcal{H}_c = U_d \sum_i n_{i\uparrow}^{(d)} n_{i\downarrow}^{(d)} + U_f \sum_i n_{i\uparrow}^{(f)} n_{i\downarrow}^{(f)} + I_{df} \left\{ \sum_i n_{i\uparrow}^{(d)} n_{i\downarrow}^{(f)} + \sum_i n_{i\downarrow}^{(d)} n_{i\uparrow}^{(f)} \right\} \qquad (2\text{-b})$$

$$\mathcal{H}_h = \sum_{i,\sigma} \left\{ V_{sd} c_{i\sigma}^+ d_{i\sigma} + V_{ds} d_{i\sigma}^+ c_{i\sigma} \right\} + \sum_{i,\sigma} \left\{ V_{sf} c_{i\sigma}^+ f_{i\sigma} + V_{fs} f_{i\sigma}^+ c_{i\sigma} \right\}$$

$$\qquad (2\text{-c})$$

$$+ \sum_{i,j,\sigma} \left\{ V_{df}(R_i - R_j) d_{i\sigma}^+ f_{j\sigma} + V_{fd}(R_i - R_j) f_{i\sigma}^+ d_{j\sigma} \right\}$$

The Hamiltonian $\mathcal{H}_0$ describe three non-hybridized s,d and f bands, shown schematically in fig. 1. We assume that the f band is a narrow band (as suggested by the relatively small overlap of f atomic states). The d and s bands are transition metal like bands.

The Hamiltonian $\mathcal{H}_c$ accounts for Coulomb repulsions among d and f electrons; one recognizes intra-orbital d-d, f-f, and inter-orbital interactions d-f. Finally $\mathcal{H}_h$ describes the usual hybridization effects among s-d and s-f electrons; we note that these hybridization matrix elements are considered for simplicity to be k independent. We also include a k dependent $V_{df}(k)$ hybridization, whose derivation within the tight binding approximation has been given recently [4]. Later on we will use a constant V_{df}, which is the mean value calculated in the region of overlap of the d and f bands. This simplifying assumption is used because it provides analitically simple

results for the density of state.

We emphasize that the purpose of this work is to set up the formalism and the numerical problems that arise in the three band problem, and to study the effect of the hybridization in the behaviour of the density of states.

In order to solve the problem described in (1), the natural way is to use the Green's function method, from which the density of states is easily derived.

We use the general equation of motion for the green's function $<<A;B>>_\omega$

$$\omega <<A;B>>_\omega = \frac{1}{2\pi} <[A,B]_+> + <<[A,\mathcal{H}] ; B>>_\omega \tag{3}$$

and the interesting propagators for this problem are $<<c_{i\sigma}; c_{j\sigma}^+>>_\omega$, $<<d_{i\sigma}; d_{j\sigma}^+>>_\omega$ and $<<f_{i\sigma}; f_{j\sigma}^+>>_\omega$, since from these one derives the density of states $n_s(\omega)$, $n_d(\omega)$ and $n_f(\omega)$.

a) DETERMINATION OF THE s-ELECTRON PROPAGATOR

Using equation (3) and the Hamiltonian (1) ones gets:

$$\omega <<c_{i\sigma}; c_{j\sigma}^+>>_\omega = \frac{1}{2\pi} \delta_{ij} + \sum_\ell T_{i\ell}^{(s)} <<c_{\ell\sigma}; c_{j\sigma}^+>>_\omega + V_{sd} <<d_{i\sigma}; c_{j\sigma}^+>>_\omega + V_{sf} <<f_{i\sigma}; c_{j\sigma}^+>>_\omega$$

$$\tag{4-a}$$

For the new Green's function introduced by the hybridization terms one gets:

$$\omega <<d_{i\sigma}; c_{j\sigma}^+>>_\omega = \sum_\ell T_{i\ell}^{(d)} <<d_{\ell\sigma}; c_{j\sigma}^+>>_\omega + U_d <<n_{i-\sigma}^{(d)} d_{i\sigma}; c_{j\sigma}^+>>_\omega + I_{df} <<n_{i-\sigma}^{(f)} d_{i\sigma}; c_{j\sigma}^+>>_\omega$$

$$+ V_{ds} <<c_{i\sigma}; c_{j\sigma}^+>>_\omega + \sum_\ell V_{df}(R_i - R_\ell) <<f_{\ell\sigma}; c_{j\sigma}^+>>_\omega \tag{4-b}$$

and

$$\omega<<f_{i\sigma};c^+_{j\sigma}>>_\omega = \sum_\ell T^{(f)}_{i\ell} <<f_{\ell\sigma};c^+_{j\sigma}>>_\omega + U_f<<n^{(f)}_{i-\sigma}f_{i\sigma};c^+_{j\sigma}>>_\omega + I_{df}<<n^{(d)}_{i-\sigma}f_{i\sigma};c^+_{j\sigma}>>_\omega$$

$$+ V_{fs}<<c_{i\sigma};c^+_{j\sigma}>>_\omega + \sum_\ell V_{fd}(R_i-R_\ell)<<d_{\ell\sigma};c^+_{j\sigma}>>_\omega \qquad (4\text{-c})$$

In equations (4-b) and (4-c) the Coulomb interaction terms introduce higher order propagators; these are treated in the simplest approximation namely the Hartree-Fock approximation. One gets the following results:

$$<<n^{(d)}_{i-\sigma}d_{i\sigma};c^+_{j\sigma}>>_\omega = <n^{(d)}_{-\sigma}><<d_{i\sigma};c^+_{j\sigma}>>_\omega$$

$$<<n^{(f)}_{i-\sigma}d_{i\sigma};c^+_{j\sigma}>>_\omega = <n^{(f)}_{-\sigma}><<d_{i\sigma};c^+_{j\sigma}>>_\omega \qquad (5)$$

$$<<n^{(f)}_{i-\sigma}f_{i\sigma};c^+_{j\sigma}>>_\omega = <n^{(f)}_{-\sigma}><<f_{i\sigma};c^+_{j\sigma}>>_\omega$$

$$<<n^{(d)}_{i-\sigma}f_{i\sigma};c^+_{j\sigma}>>_\omega = <n^{(d)}_{-\sigma}><<f_{i\sigma};c^+_{j\sigma}>>_\omega$$

Translational invariance being used to write

$$<n^{(\alpha)}_{i-\sigma}> = <n^{(\alpha)}_{-\sigma}> \quad \text{where} \quad \alpha = d, f$$

With the approximations (5) one gets using equations (4):

$$\omega<<d_{i\sigma};c^+_{j\sigma}>>_\omega = \sum_\ell T^{(d)}_{i\ell}<<d_{\ell\sigma};c^+_{j\sigma}>>_\omega + \left\{U_d<n^{(d)}_{-\sigma}> + I_{df}<n^{(f)}_{-\sigma}>\right\}<<d_{i\sigma};c^+_{j\sigma}>>_\omega$$

$$+ V_{ds}<<c_{i\sigma};c^+_{j\sigma}>>_\omega + \sum_\ell V_{df}(R_i-R_\ell)<<f_{\ell\sigma};c^+_{j\sigma}>>_\omega \qquad (6\text{-a})$$

$$\omega<<f_{i\sigma};c^+_{j\sigma}>>_\omega = \sum_\ell T^{(f)}_{i\ell}<<f_{\ell\sigma};c^+_{j\sigma}>>_\omega + \left\{U_f<n^{(f)}_{-\sigma}> + I_{df}<n^{(d)}_{-\sigma}>\right\}<<f_{i\sigma};c^+_{j\sigma}>>_\omega$$

$$+ V_{fs}<<c_{i\sigma};c^+_{j\sigma}>>_\omega + \sum_\ell V_{fd}(R_i-R_\ell)<<d_{\ell\sigma};c^+_{j\sigma}>>_\omega \qquad (6\text{-b})$$

Equations (6) and (4-a) form a closed set which determine the propagator $<<c_{i\sigma};c^+_{j\sigma}>>$ Now we solve by Fourier transformation, introducing the

Hartree-Fock renormalized d and f energies.

$$E_{k\sigma}^{(d)} = \epsilon_k^{(d)} + U_d <n_{-\sigma}^{(d)}> + I_{df} <n_{-\sigma}^{(f)}>$$

$$E_{k\sigma}^{(f)} = \epsilon_k^{(f)} + U_f <n_{-\sigma}^{(f)}> + I_{df} <n_{-\sigma}^{(d)}>$$

(7)

One gets

$$(\omega-\epsilon_k^{(s)})G_{kk'}^{ss}(\omega) = \frac{1}{2\pi}\delta_{kk'} + V_{sd}\, G_{kk'}^{ds}(\omega) + V_{sf}G_{kk'}^{fs}(\omega) \quad (8\text{-}a)$$

$$(\omega-E_{k\sigma}^{(d)})G_{kk'}^{ds}(\omega) = V_{ds}\, G_{kk'}^{ss}(\omega) + V_{df}(k)G_{kk'}^{fs}(\omega) \quad (8\text{-}b)$$

$$(\omega-E_{k\sigma}^{(f)})G_{kk'}^{fs}(\omega) = V_{fs}G_{kk'}^{ss}(\omega) + V_{fd}(k)G_{kk'}^{ds}(\omega) \quad (8\text{-}c)$$

The $G_{kk'}^{ss}(\omega)$ propagator is then:

$$\left\{ \omega-\epsilon_k^{(s)} - \frac{|V_{sd}|^2}{\omega-E_{k\sigma}^{(d)} - \dfrac{|V_{df}(k)|^2}{\omega- E_k^{(f)}}} - \frac{|V_{sf}|^2}{\omega-E_{k\sigma}^{(f)} - \dfrac{|V_{df}(k)|^2}{\omega- E_{k\sigma}^{(d)}}} \right\} G_{kk'}^{ss}(\omega) = \frac{1}{2\pi}\delta_{kk'} \quad (9)$$

or alternatively

$$G_{kk'}^{ss}(\omega) = \frac{1}{2\pi}\delta_{kk'} \cdot \frac{1}{\omega-\epsilon_k^{(s)} - \dfrac{|V_{sd}|^2}{\omega-E_{k\sigma}^{(d)} - \dfrac{|V_{df}(k)|^2}{\omega- E_{k\sigma}^{(f)}}} - \dfrac{|V_{sf}|^2}{\omega-E_{k\sigma}^{(f)} - \dfrac{|V_{df}(k)|^2}{\omega-E_{k\sigma}^{(d)}}}} \quad (10)$$

We have dropped out the terms $V_{sd}\, V_{df}(k)V_{fs}$ and $V_{sf}\, V_{fd}(k)\, V_{ds}$ from the

propagator, because we can choose V_{sd}, $V_{df}(k)$, V_{fs}, V_{sf}, $V_{fd}(k)$, V_{ds} which are in general complex numbers, in such a way that V_{sd} $V_{df}(k)$ V_{fs} and V_{sf} $V_{fd}(k)$ V_{ds} can be taken zero; and the values of $|V_{sf}|^2$ $|V_{sd}|^2$ and $|V_{df}(k)|^2$ finite. Later we will refer again to these terms. So we can rewrite the s-propagator:

$$G_{kk'}^{ss}(\omega) =$$

$$= \frac{\delta_{kk'}}{2\pi} \frac{(\omega-\varepsilon_{k\sigma}^{(d)})(\omega-E_{k\sigma}^{(f)})-|V_{df}(k)|^2}{(\omega-\varepsilon_k^{(s)})(\omega-E_{k\sigma}^{(d)})(\omega-E_{k\sigma}^{(f)})-|V_{df}(k)|^2(\omega-\varepsilon_k^{(s)})-|V_{sd}|^2(\omega-E_{k\sigma}^{(f)})-|V_{sd}|^2(\omega-E_{k\sigma}^{(d)})}$$

$$(11)$$

One should note that equation (11) is identical to that obtained for three hybridized bands in the absence of Coulomb repulsions; the only effect associated to it being the renormalization of f and d energies as defined in (7).

b) DETERMINATION OF THE f PROPAGATOR

Using the equation of motion (3) and the Hartree-Fock approximation (5), together with translation invariance one gets:

$$\omega \langle\langle f_{i\sigma}; f_{j\sigma}^+ \rangle\rangle_\omega = \frac{1}{2\pi} \delta_{ij} + \sum_\ell T_{i\ell}^{(f)} \langle\langle f_{\ell\sigma}; f_{j\sigma}^+ \rangle\rangle_\omega +$$

$$+ \left\{ U_f \langle n_{-\sigma}^{(f)}\rangle + I_{df} \langle n_{-\sigma}^{(d)}\rangle \right\} \langle\langle f_{i\sigma}; f_{j\sigma}^+ \rangle\rangle_\omega$$

$$+ V_{fs} \langle\langle c_{i\sigma}; f_{j\sigma}^+ \rangle\rangle_\omega + \sum_\ell V_{fd}(R_i - R_\ell) \langle\langle d_{\ell\sigma}; f_{j\sigma}^+ \rangle\rangle_\omega \qquad (12\text{-}a)$$

$$\omega<<c_{i\sigma};f_{j\sigma}^{+}>>_{\omega} = \sum_{\ell} T_{i\ell}^{(s)}<<c_{\ell\sigma};f_{j\sigma}^{+}>>_{\omega} + V_{sf} <<f_{i\sigma};f_{j\sigma}^{+}>>_{\omega} + V_{sd}<<d_{i\sigma};f_{j\sigma}^{+}>>_{\omega}$$

$$(12\text{-}b)$$

$$\omega<<d_{i\sigma};f_{j\sigma}^{+}>>_{\omega} = \sum_{\ell} T_{i\ell}^{(d)}<<d_{\ell\sigma};f_{j\sigma}^{+}>>_{\omega} + \left\{ U_d<n_{-\sigma}^{(d)}> + I_{df} <n_{-\sigma}^{(f)}> \right\}<<d_{i\sigma};f_{j\sigma}^{+}>>_{\omega}$$

$$+ V_{ds} <<c_{i\sigma};f_{j\sigma}^{+}>>_{\omega} + \sum_{\ell} V_{df}(R_i-R_\ell)<<f_{\ell\sigma}; f_{j\sigma}^{+}>>_{\omega}$$

Again the coupled system (12) provides the solution for the propagator $<<f_{i\sigma}; f_{j\sigma}^{+}>>$. Using Fourier transformation and the definition of the renormalized energies (7) one gets:

$$(\omega-E_k^{(f)})G_{kk'}^{ff}(\omega) = \frac{1}{2\pi} \delta_{kk'} + V_{fs} G_{kk'}^{sf}(\omega) + V_{fd}(k)G_{kk'}^{df}(\omega) \qquad (13\text{-}a)$$

$$(\omega-\varepsilon_k^{(s)})G_{kk'}^{sf}(\omega) = V_{sf} G_{kk'}^{ff}(\omega) + V_{sd} G_{kk'}^{df}(\omega) \qquad (13\text{-}b)$$

$$(\omega-E_{k\sigma}^{(d)})G_{kk'}^{df}(\omega) = V_{ds} G_{kk'}^{sf}(\omega) + V_{df}(k) G_{kk'}^{ff}(\omega) \qquad (13\text{-}c)$$

the solution of the coupled system (13) gives as solution

$$G_{kk'}^{ff}(\omega) =$$

$$= \frac{\delta_{kk'}}{2\pi} \frac{(\omega-E_{k\sigma}^{(d)})(\omega-\varepsilon_k^{(s)})-|V_{sd}|^2}{(\omega-\varepsilon_k^{(s)})(\omega-E_{k\sigma}^{(d)})(\omega-E_{k\sigma}^{(f)})-|V_{sd}|^2(\omega-E_{k\sigma}^{(f)})-|V_{df}(k)|^2(\omega-\varepsilon_k^{(s)})-|V_{sf}|^2(\omega-E_{k\sigma}^{(d)})}$$

$$(14)$$

where we have dropped out the terms of third order in the mixings.

c) DETERMINATION OF THE d-PROPAGATOR

Quite similarly to the previous cases one gets the following coupled system

$$(\omega-E_{k\sigma}^{(d)})G_{kk'}^{dd}(\omega) = \frac{1}{2\pi}\delta_{kk'} + V_{ds}\,G_{kk'}^{sd}(\omega) + V_{df}(k)G_{kk'}^{fd}(\omega) \qquad (15\text{-}a)$$

$$(\omega-\epsilon_{k}^{(s)})G_{kk'}^{sd}(\omega) = V_{sd}\,G_{kk'}^{dd}(\omega) + V_{sf}\,G_{kk'}^{fd}(\omega) \qquad (15\text{-}b)$$

$$(\omega-E_{k\sigma}^{(f)})G_{kk'}^{fd}(\omega) = V_{fs}\,G_{kk'}^{sd}(\omega) + V_{fd}(k)G_{kk'}^{dd}(\omega) \qquad (15\text{-}c)$$

by solving equation (15) one gets:

$$G_{kk'}^{dd}(\omega) =$$

$$= \frac{\delta_{kk'}}{2\pi}\frac{(\omega-\epsilon_{k}^{(s)})(\omega-E_{k\sigma}^{(f)})-|V_{sf}|^{2}}{(\omega-\epsilon_{k}^{(s)})(\omega-E_{k\sigma}^{(d)})(\omega-E_{k\sigma}^{(f)})-|V_{sd}|^{2}(\omega-E_{k\sigma}^{(f)})-|V_{df}(k)|^{2}(\omega-\epsilon_{k}^{(s)})-|V_{sf}|^{2}(\omega-E_{k\sigma}^{(d)})}$$

$$(16)$$

where like in the others propagators we have also dropped out the terms $V_{sd}\,V_{df}(k)\,V_{fs}$ and $V_{sf}\,V_{fd}(k)\,V_{ds}$.

d) DISCUSSION OF THE OBTAINED PROPAGATORS

Firstly let us compare the results embodied in equations (11), (14) and (16): one notes that the ss, dd and ff propagators have the same

denominators and consequently the same poles. This just means that the energies of the system are contained in a single equation describing three hybridized bands. However different propagators have differents residues, just meaning that the s,d and f characters of the hybridized bands are not necessarily identical. A very clear picture of the mixing effects separated from intraband correlations is provided by the following; consider for instance the free ss propagator without hybridization:

$$G_0^s(\omega) = \frac{1}{\omega - \epsilon_k^s}.$$ The G_k^{ss} propagator (10) reads:

$$G_{kk'}^{ss}(\omega) = \frac{\delta_{kk'}}{2\pi} \, G_0^s(\omega) \, \frac{1}{1 - x \, G_0^s(\omega)} \tag{17-a}$$

where x is defined as

$$x = |V_{sd}|^2 \tilde{G}_k^{(d)}(\omega) + |V_{sf}|^2 \, \tilde{G}_k^{(f)}(\omega) \tag{17-b}$$

In the Hartree-Fock approximation adopted here the propagators $\tilde{G}^d$ and $\tilde{G}^f$ are defined:

$$\tilde{G}_k^{(d)}(\omega) = \frac{1}{\omega - E_{k\sigma}^{(d)} - \dfrac{|V_{df}(k)|^2}{\omega - E_{k\sigma}^{(f)}}} \quad \text{and} \quad \tilde{G}_k^{(f)}(\omega) = \frac{1}{\omega - E_{k\sigma}^{(f)} - \dfrac{|V_{df}(k)|^2}{\omega - E_{k\sigma}^{(d)}}} \tag{17-c}$$

which in the absence of d-f mixing reduce to G_0^d and G_0^f, which are similar to $G_0^{(s)}$ expect that $\epsilon_k^{(s)}$ is replaced by $E_{k\sigma}^{(d)}$ and $E_{k\sigma}^{(f)}$.
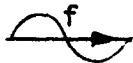
Expression (17-a) can be rewritten as:

$$G_{kk'}^{ss}(\omega) = \frac{1}{2\pi}\,\delta_{kk'}\;G_0^{(s)}(\omega)\sum_{n=0}^{\infty} x^n(G_0^{(s)}(\omega))^n$$

$$= \frac{1}{2\pi}\,\delta_{kk'}\;G_0^{(s)}(\omega)\sum_{n=0}^{\infty}(|V_{sd}|^2\,\tilde{G}^{(d)}(\omega) + |V_{sf}|^2\,\tilde{G}^{(f)}(\omega))^n(G_0^{(s)}(\omega))^n \quad (17\text{-}d)$$

Equation (17-d) can be put in pictorial terms defining free propagator as lines with arrows and a letter specifying its nature (s, d, f); the mixing matrix elements are represented by dotted lines. One gets:
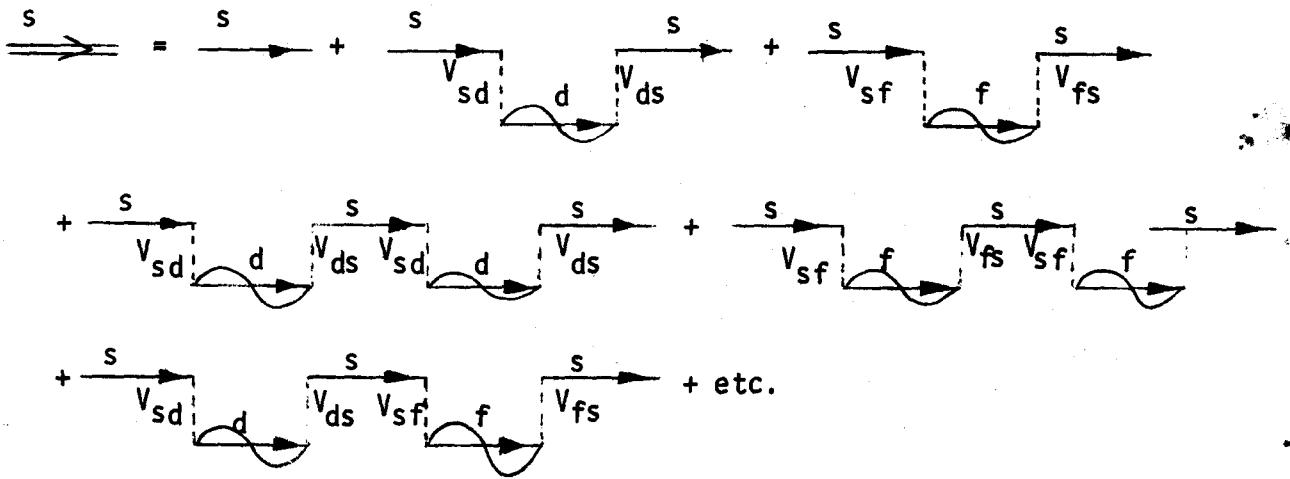
The propagator $\tilde{G}^d$ and $\tilde{G}^f$ are represented by and respectively. Because of d-f hybridization each of these propagator can be written in a way similar to that of the $G_{kk}^{ss}(\omega)$ as in expression (17-d). In a pictorial form:

+ etc.
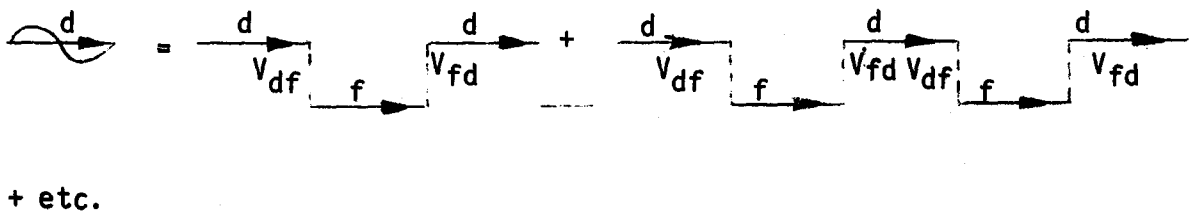
It should be emphasized that the processes depicted above are perfectly general in the following sense; using appropriate free propagators d and f and effective mixing matrix elements this series of diagram can always be drawn [6]. Quite similar diagrams can be written for d and f propagators. From this picture one sees the possibles improvements of the Hartree-Fock approach for the three band problem; one replaces the $\tilde{G}^d$, $\tilde{G}^f$ propagators by new ones describing more in detail the effect of electron correlations (for instance the Roth method [3] for narrow bands). Also one replaces the mixing matrix elements by effective ones (including correlations effects)[6] in these conditions the general effect of mixing is still described by these diagrams.

2. EQUATION FOR THE POLES; HYBRIDIZED BAND DISPERSION RELATIONS

One gets the energies for this three-band problem from the roots of the third order equation:

$$(\omega - \varepsilon_k^{(s)})(\omega - E_{k\sigma}^{(d)})(\omega - E_{k\sigma}^{(f)}) - |V_{sd}|^2(\omega - E_{k\sigma}^{(f)}) - |V_{sf}|^2(\omega - E_{k\sigma}^{(d)}) - |V_{df}(k)|^2(\omega - E_k^{(s)}) = 0$$

$$(18)$$

For each value of k one gets from (18) three solutions corresponding to the three branches of the hybridized bands. Equation (18) is general in the sense that the non-hybridized s,d and f bands have general dispersion relations. An schematic picture of these dispersion relations is shown in fig. (1), and the general result one obtains is that within the constant mixing elements scheme (from now on we take $V_{df}(k)$ constant equal to $\overline{V}_{df}$, which is a mean value calculated in the region of overlap of the d and f bands),

crossing of bands are supressed (fig. 1). Also one obtains a shift in energy of the lower and higher bands.

At this point it is useful to introduce a trick firstly used by Kishore and Joshi [5] to obtain simple results for the density of states curve; this trick consists in adopting a model for the non-hybridized bands consisting of homothetic bands.

One chooses a certain dispersion relation ε_k and defines:

$$\varepsilon_k^{(s)} = \varepsilon_k$$

$$\varepsilon_k^{(d)} = A\varepsilon_k + A'$$

$$\varepsilon_k^{(f)} = B\varepsilon_k + B'$$

(19)

Then only one dispersion relation ε_k specifies all the three bands, the bottom of the d and f bands being specified by A' and B'. The width of the d and f bands are specified through the coefficients A and B, which are taken less than 1. For this choice, the bands d and f defined in (19) are narrower than the s band, and if A > B one has a f band narrower than the d band. Then equation (19) qualitatively describe the situation expected for an actinide metal where a narrow f band is superposed to a transition metal like s and d bands. Figure 1 shows for the case of parabolic bands the dipersion rela tions as derived from (18). As it will be shown below the band model (19) provides a simple and analytical results for the density of states.

3. CALCULATION OF THE DENSITY OF STATES

In this part we generalize Kishore and Joshi [5] procedure to calculate the state density for hybridized bands. Let $\omega_k^{(1)}$, $\omega_k^{(2)}$ and $\omega_k^{(3)}$ be the roots of equation (18); the ff propagator for instance, may be rewritten in terms of simple fractions as:

$$G_k^{ff}(\omega) = \frac{1}{2\pi} \frac{(\omega-\varepsilon_k^{(s)})(\omega-E_{k\sigma}^{(d)})-|V_{sd}|^2}{(\omega-\omega_k^{(1)})(\omega-\omega_k^{(2)})(\omega-\omega_k^{(3)})}$$

$$= \frac{1}{2\pi}\left\{ \frac{A_k^{(1)}}{\omega-\omega_k^{(1)}} + \frac{A_k^{(2)}}{\omega-\omega_k^{(2)}} + \frac{A_k^{(3)}}{\omega-\omega_k^{(3)}} \right\} \qquad (20)$$

where the $\omega_k^{(i)}$'s are the roots of the third degree equation (18) with $V_{df}(k)$ constant. The coefficients A_k's may be determined through equating the right and left hand sides of equation (20). If one introduces α as defined by

$$\alpha = (\omega_k^{(1)})^2(\omega_k^{(2)}-\omega_k^{(3)})+(\omega_k^{(2)})^2(\omega_k^{(3)}-\omega_k^{(1)})+(\omega_k^{(3)})^2(\omega_k^{(1)}-\omega_k^{(2)}) \qquad (21-a)$$

one gets for these coefficients

$$A_k^{(1)} = \frac{1}{\alpha}(\omega_k^{(2)} - \omega_k^{(3)})f_k(\omega_k^{(1)})$$

$$A_k^{(2)} = \frac{1}{\alpha}(\omega_k^{(3)} - \omega_k^{(1)})f_k(\omega_k^{(2)}) \qquad (21-b)$$

$$A_k^{(3)} = \frac{1}{\alpha}(\omega_k^{(1)} - \omega_k^{(2)})f_k(\omega_k^{(3)})$$

where $f_k(\omega)$ is the following auxiliary function

$$f_k(\omega) = (\omega-\varepsilon_k^{(s)})(\omega-E_k^{(d)})-|V_{sd}|^2 \qquad (21-c)$$

The coefficients A_k's defined in (21-b) can be put in a simple form if one notes that

$$\alpha = - (\omega_k^{(1)} - \omega_k^{(2)})(\omega_k^{(2)} - \omega_k^{(3)})(\omega_k^{(3)} - \omega_k^{(1)}) \qquad (21\text{-}d)$$

so that some simplifications occur; one gets finally the symmetric form:

$$A_k^{(1)} = \frac{f_k(\omega_k^{(1)})}{(\omega_k^{(1)} - \omega_k^{(2)})(\omega_k^{(1)} - \omega_k^{(3)})}$$

$$A_k^{(2)} = \frac{f_k(\omega_k^{(2)})}{(\omega_k^{(2)} - \omega_k^{(1)})(\omega_k^{(2)} - \omega_k^{(3)})} \qquad (22)$$

$$A_k^{(3)} = \frac{f_k(\omega_k^{(3)})}{(\omega_k^{(3)} - \omega_k^{(1)})(\omega_k^{(3)} - \omega_k^{(2)})}$$

Next step is then to calculate $F_\omega\{G_k^{ff}(\omega)\} = i \lim_{\epsilon \to 0} \{G_k^{ff}(\omega+i\epsilon) - G_k^{ff}(\omega-i\epsilon)\}$ in order to obtain the density of states, one gets:

$$F_\omega\left\{G_k^{ff}(\omega)\right\} = \left| \frac{f_k(\omega)}{(\omega-\omega_k^{(2)})(\omega-\omega_k^{(3)})} \right| \delta(\omega-\omega_k^{(1)}) + \left| \frac{f_k(\omega)}{(\omega-\omega_k^{(1)})(\omega-\omega_k^{(3)})} \right| \delta(\omega-\omega_k^{(2)}) +$$

$$+ \left| \frac{f_k(\omega)}{(\omega-\omega_k^{(1)})(\omega-\omega_k^{(2)})} \right| \delta(\omega-\omega_k^{(3)}) \qquad (23)$$

The absolute values ensuring that each term corresponds to the density of

Finally substituting in expression (21-c) the energies ε_k^s and $E_{k\sigma}^{(d)}$ by the model band structure energies one gets a new function $\bar{f}(\omega, \varepsilon_k)$; taking this into account and using the definition $N(\omega) = \sum \delta(\omega-\varepsilon_k)$ one gets finally:

$$n_f^{(1)}(\omega) = \frac{|\bar{f}(\omega, g^{(1)}(\omega))|}{AB|(g^{(3)}(\omega) - g^{(1)}(\omega))(g^{(2)}(\omega) - g^{(1)}(\omega)|} N(g^{(1)}(\omega))$$

$$n_f^{(2)}(\omega) = \frac{|\bar{f}(\omega, g^{(2)}(\omega))|}{AB|(g^{(3)}(\omega) - g^{(2)}(\omega))(g^{(1)}(\omega) - g^{(2)}(\omega))|} N(g^{(2)}(\omega)) \qquad (28)$$

$$n_f^{(3)}(\omega) = \frac{|\bar{f}(\omega, g^{(3)}(\omega))|}{AB|(g^{(2)}(\omega) - g^{(3)}(\omega))(g^{(1)}(\omega) - g^{(3)}(\omega))|} N(g^{(3)}(\omega))$$

Equations (28) are the analytical expressions for the contributions of the f electrons to the density of states of each of the three branches of the hybrid bands *. It remains only to determine the equation which the functions $g_i(\omega)$ must satisfy, in order to completely determine the state density. From equations (25), (18) and (19) by equating equal powers of ε_k on the right and left hand sides one obtains relations among the roots $g_i(\omega)$, namely

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* Similar expressions hold for the s and d density of states, just changing the function $\bar{f}(\omega, g^{(i)}(\omega))$.

$$g^{(1)}(\omega)+g^{(2)}(\omega)+g^{(3)}(\omega) = \alpha(\omega) = \frac{1}{AB}\left[\omega(A+B+AB) - (A'B + B'A)\right] \qquad (29\text{-}a)$$

$$g^{(1)}(\omega)g^{(2)}(\omega)+g^{(1)}(\omega)g^{(3)}(\omega)+g^{(2)}(\omega)g^{(3)}(\omega) = \frac{1}{AB}\left[\omega^2(A+B+1)- (A'+B'+AB'+A'B)\right.$$

$$\left.- B|V_{sd}|^2 - A|V_{sf}|^2 - |\bar{V}_{df}|^2 + A'B'\right] = \beta(\omega) \quad (29\text{-}b)$$

$$g^{(1)}(\omega)g^{(2)}(\omega)g^{(3)}(\omega) = \frac{1}{AB}\left[\omega^3-\omega^2(A'+B')-\omega(|V_{sd}|^2+|V_{sf}|^2+ |\bar{V}_{df}|^2- A'B')\right.$$

$$\left.+ B'|V_{sd}|^2 + A'|V_{sf}|^2\right]= \gamma(\omega) \qquad (29\text{-}c)$$

Using the properties of algebraic equations, from (29) one derives the follow ing third degree equation for $g(\omega)$:

$$g^3(\omega) - \alpha(\omega) \, g^2(\omega) + \beta(\omega) \, g\,(\omega) - \gamma(\omega) = 0 \qquad (30)$$

From (30), for a given value of ω, which defines the functions α, β, γ one gets the corresponding g's, and equation (30) together with equations (28) solve completely the problem of determining the density of states.

4. NUMERICAL RESULTS

Now we present some numerical results obtained from the model developped above. Since no attempt of self-consistency is made in this work, we restrict ourselves to discuss the dispersion relations, band widths and the shape of the density of states.

a) *Dispersion Relations*

Firstly using equation (18) we calculate the energy values in presence

of hybridization; the new dispersion relations are directly obtained from:

$$\omega^3 - (\varepsilon_k^{(s)} + E_k^{(d)} + E_k^{(f)})\omega^2 + (\varepsilon_k^{(s)} E_{k\sigma}^{(d)} + \varepsilon_k^{(s)} E_{k\sigma}^{(f)} + E_{k\sigma}^{(d)} E_{k\sigma}^{(f)} - |V_{sd}|^2 - |V_{st}|^2 - |\overline{V}_{df}|^2)\omega$$

$$+ (|V_{sd}|^2 E_{k\sigma}^{(f)} + |V_{sf}|^2 E_{k\sigma}^{(d)} + |\overline{V}_{df}|^2 \varepsilon_k^{(s)} -$$

$$- \varepsilon_k^{(s)} E_{k\sigma}^{(d)} E_{k\sigma}^{(f)}) = 0 \tag{31}$$

where the $\varepsilon_k^{(s)}$, $E_{k\sigma}^{(d)}$ and $E_{k\sigma}^{(f)}$ are the unhybridized dispersion relations. From (31) one obtains numerically, as a function of k, the energies associated to the three branches of the hybridized bands. We adopt here the approximation of homothetic bands (19), the spin dependence of the d and f bands being incorporated in the constants A' and B' of (19). Fig. 1 shows the dispersion relations for non-overlapping d and f bands. Such a situation was chosen in order to emphasize the role of $|\overline{V}_{df}|$ mixing in a pictorially simpler situation, but in fact the physically interesting situation corresponds to overlapping d and f bands. Finally fig. 1 was constructed adopting parabolic ε_k curves.

In that figure one sees clearly the main effects of hybridization, namely:

i) The crossing regions of the unperturbed bands (as expected from general arguments) disappear when $|V_{sd}|^2$ and $|V_{sf}|^2$ are finite. In the case they are zero the crossing regions persist.

ii) The top and the bottom of the unperturbed band structure are shifted by the hybridization this effect is enhanced due to the assumption of k independent matrix elements $|V_{sd}|^2$, $|V_{sf}|^2$ and $|\overline{V}_{df}|^2$.

iii) In figure 1 one takes $|V_{ds}|^2 = 0.8$ and $|V_{sf}|^2 = 0.1$, consequently one observes that the splitting of the bands at the crossing is larger for s-d than for s-f crossing. Then we take $|V_{ds}|^2 = 0.1$ and $|V_{sf}|^2 = 0.8$ and observe the opposite result, for the same value of $|\overline{V_{df}}|^2$.

In the curves of figure 1, the only effect of $|\overline{V_{df}}|^2$ is to repel the unperturbed d and f bands, since these bands do not overlap.

b) *Solutions for the g(ω) Functions*

In order to calculate the density of states, one needs as a function of energy ω the values of the function g(ω) defined through equations (29) and (30). The coefficients of the third order equation (30) are defined in (29) as function of ω in terms of the mixing coefficients, the "effective masses" A, B and the band positions A', B'. In general one has to assign values to these parameters and numerically solve equation (30) for several values of ω. It is however interesting to see the nature of the solutions of (30) in the limit of zero hybridization.

One can easily check that for $|V_{sd}|^2 = |V_{sf}|^2 = |V_{df}|^2 = 0$ one finds three solutions for g(ω):

$$g^{(1)}(\omega) = \omega$$

$$g^{(2)}(\omega) = \frac{\omega - A'}{A} \tag{32-a}$$

$$g^{(3)}(\omega) = \frac{\omega - B'}{B}$$

These solutions correspond to the three unperturbed bands namely:

These solutions correspond to the three unperturbed bands namely:

$$\omega = \epsilon_k$$

$$\frac{\omega - A'}{A} = \epsilon_k \qquad \text{or} \qquad \omega = A\,\epsilon_k + A' \qquad (32\text{-}b)$$

$$\frac{\omega - B'}{B} = \epsilon_k \qquad \text{or} \qquad \omega = B\,\epsilon_k + B'$$

If one plots these solutions as a function of ω one gets the straight lines of fig. 2. These solutions will now be compared to those obtained in presence of mixing. In this case one expects that the crossing of these curves will disappear, and this is confirmed by numerical calculation (Fig. 2).

The interpretation of this fact goes as follows:

From Fig. 2 one sees that the first branch of the $g(\omega)$ curve starts following $g(\omega) = \omega$, which corresponds to s-like states. When the crossing point with the f-band is approached, f states begin to be admixed in the band which becomes gradually f-like. This corresponds in the $g(\omega)$ curve to become parallel to the f-band straight line. Then in the first band one passes continuously from s-like states to pure f-like states. Consequently one expects that the s density of states will have important values at the beginning of the hybrid band and tends to zero at the end where f states are predominant. The reverse behaviour is expected for f states.

Now in the second branch one starts with a strong f-character, then

s-like character and finally d-like behaviour. This corresponds to the intermediate branch of fig. 2.

Finally in the third branch one passes from d-like character to s-like character.

Using these results one can roughly describe the behaviour of the density of states. The detailed calculations of the density of states made in part d) will confirm these predictions. We can observe also in this figure the shift in the unperturbed d and f bands caused by the strong d-f hybridization. We have analysed here the non-physical case of non-overlapping d and f bands, which however clarifies the effects described above.

c) *Determination of the Band Edges*

Firstly we recall that the density of states $N(\omega)$ involved in equation (28) is defined in a certain energy range and zero outside it. Let us take for this range the energies between 0 and Δ. Since the density of states for the perturbed bands involve $N|g(\omega)|$ the important range of ω is such that $g(\omega)$ in this range varies between 0 and Δ. This requirement defines the band edges; in order to obtain explicitly the energies E_b^i and E_t^i one proceeds as follows: The energies E_b^i are the solutions of the equation:

$$g(E_b^i) = 0 \tag{33-a}$$

If this condition is imposed in equation (30) it follows directly

$$\gamma(E_b^i) = 0 \tag{33-b}$$

which from (29) is a third degree equation whose roots determine the energy of the bottom of the three sub-bands. Quite similarly, to obtain the

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If this condition is imposed in equation (30) it follows directly

$$\gamma(E_b^i) = 0 \qquad\qquad (33\text{-}b)$$

which from (29) is a third degree equation whose roots determine the energy of the bottom of the three sub-bands. Quite similarly, to obtain the

energy of the tops of the bands one imposes.

$$g(E_t^i) = \Delta \qquad\qquad (33\text{-}c)$$

Imposing this condition in equation (30) one gets:

$$\Delta^3 - \alpha(E_t^i)\,\Delta^2 + \beta(E_t^i)\Delta - \gamma(E_t^i) = 0 \qquad\qquad (33\text{-}d)$$

From (29) one sees that (33-d) is a third order equation, determining the three energies of the top of the three hybrid bands.

d) *Density of States*

In this part of the work we intend to discuss the effect of d-f hybridization in the structure of the density of states and the consequent implications on the condition for magnetism in actinides. We consider two kinds of band structure namely a square density of states (Figs. 3 and 4) and a parabolic one (Figs. 5 to 8). The square density of states is known to be a not very good one in analysing magnetic effects, however in a first approach we can learn something about the effect of V_{df}. These figures are constructed numerically evaluating expressions similar to (28) as a function of ω.

In fig. 3 we show the unperturbed s, d and f bands together with the new hybrid bands. In each branch of hybrid bands it is represented the s, d and f contributions to the density of states of this band. We treat the physical case of overlopping bands and here $|V_{sd}| = |V_{sf}| = 0.1$ and $|\overline{V}_{df}| = = 0$. In figure 4 we have the same case but now there is a strong hybridization ($|\overline{V}_{df}| = 0.8$) between d and f bands. The effect of this hybridization is essentially to create a "repulsion" between the d and f density of states, distributing more uniformly the number of electron

states over the range of values of ω. When $|\overline{V_{df}}|$ vanishes these states are concentrated in the energy region of the unperturbed band. This effect becomes more clearly when we treat the more realistic case of parabolic bands. (Figs. 5 to 8).

In Fig. 5 and 6 we have non-overlapping d and f bands. When $|\overline{V_{df}}|$ is turned off the density of states of the hybrid bands remains finite only over the unperturbed bands. Turning on d-f hybridization the d and f bands "repel" each other, and its mechanism lowers the density of states by spreading out these states ina a wider range of energy.

Figs. 7 and 8 describe the situation of overlapping d-f bands. Comparison between figs. 6 and 8 show that the effects are much more spectacular in the case of overlapping bands.

Now we can see that the role V_{df} is fundamental in alaysing the conditions for magnetism. In another paper [6] we have obtained a Stoner like criterion with an effective "exchange interaction" for the occurrence of magnetism in actinides. In this approach we can see that density of states in the Fermi level is, together with the effective Coulomb repulsion, the essential parameter in determining the conditions for magnetic instabilities.

Now in turns out visible the role of V_{df} in studying magnetism, since it lowers the density of states by distributing the electronic states over a range of energy. In fact if in fig. 8 the Stoner criterion is satisfied for the unperturbed f density of states, even keeping constant the value of the Coulomb repulsion, $|\overline{V_{df}}|$ mixing drastically reduces the density of states. This reduction tends to inhibit magnetism, and the Stoner criterion is only satisfied if the Coulomb repulsion is increased.
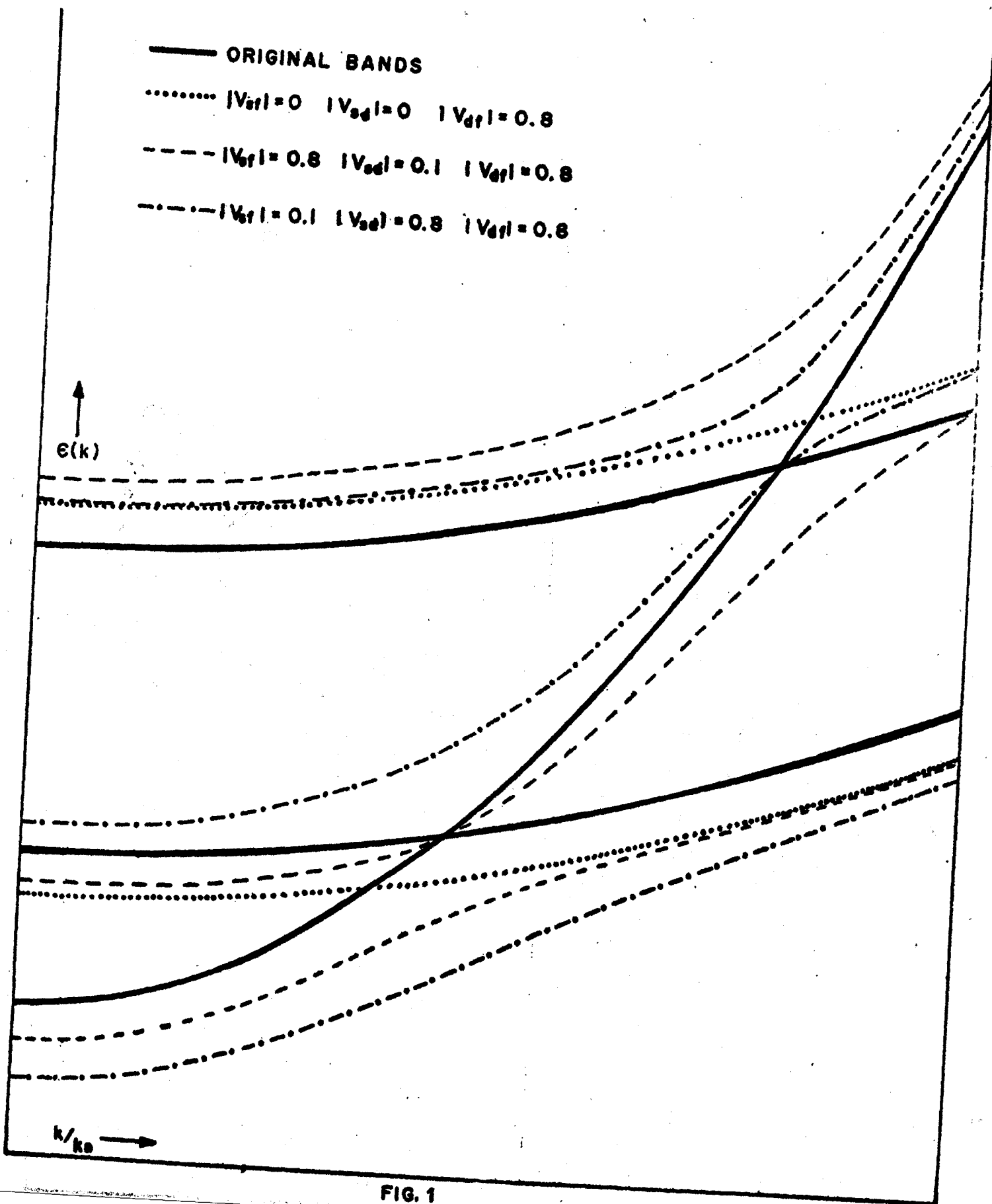
CONCLUSIONS

The above discussed numerical results confirm, even in presence of the broad s-band, the role of V_{df} in the study of the magnetic properties of actinide metals. The presence of hybridization with the s-band (though $|V_{sf}|$ and $|V_{sd}|$ do not modify the general behaviour of the density of states, this supporting the approach of Jullien et al.[4], in considering only d and f bands. On the other hand, the formalism set up in this work can be used to introduce spin-orbit effects. In fact, as shown by Jullien et al.[8] the behaviour of actinide impurities in transition metals can be explained if spin-orbit effects are introduced, and these effects seem to be important also in pure metals. In this context, a model is suggested where two splitted f bands interact (through mixing and Coulomb correlation) with a d-band, and again a three band model is obtained. The only difference respect to the present case is that now the three bands are Hartree-Fock renormalized, and consequently the self-consistency problem is a numerically very difficult one.

* * *

ORIGINAL BANDS

$|V_{sf}| = 0$ $|V_{sd}| = 0$ $|V_{df}| = 0.8$

$|V_{sf}| = 0.8$ $|V_{sd}| = 0.1$ $|V_{df}| = 0.8$

$|V_{sf}| = 0.1$ $|V_{sd}| = 0.8$ $|V_{df}| = 0.8$

$\epsilon(k)$

k/k_0

FIG. 1
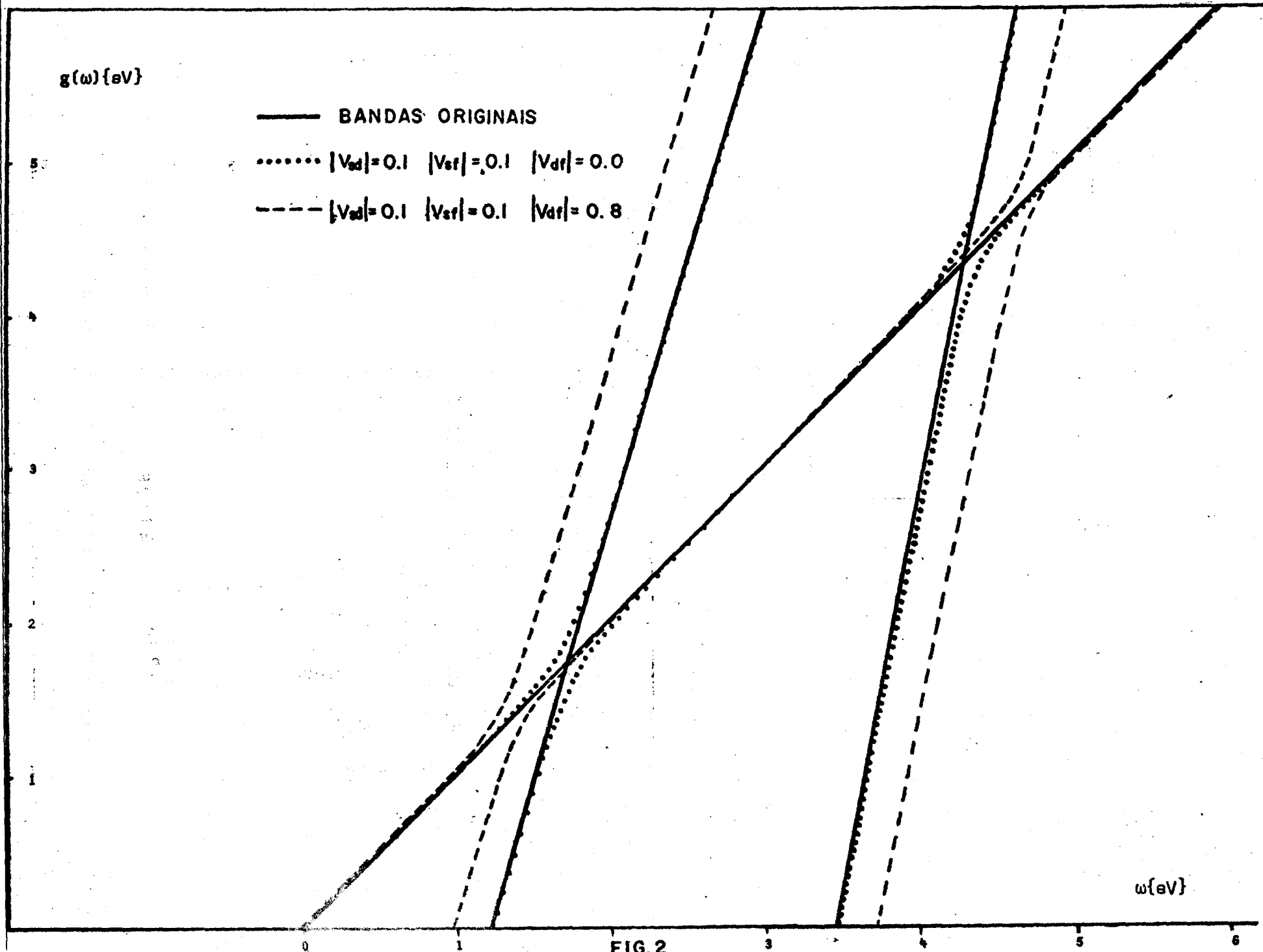
FIG. 2

FIG. 3

Legend:
- ——— NON HYBRIDIZED BANDS
- – – – FIRST HYBRIDIZED BAND
- ······· SECOND HYBRIDIZED BAND
- –·–·– THIRD HYBRIDIZED BAND
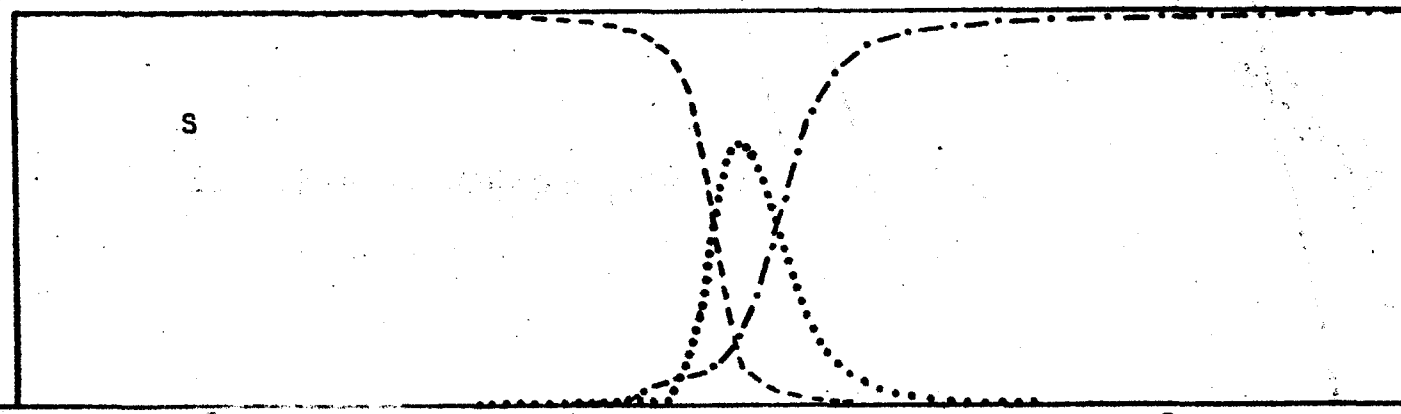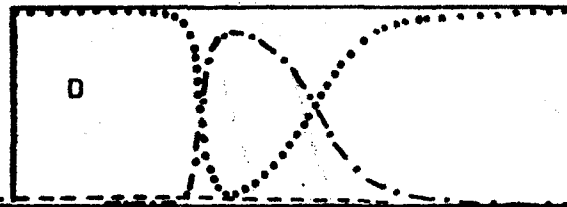
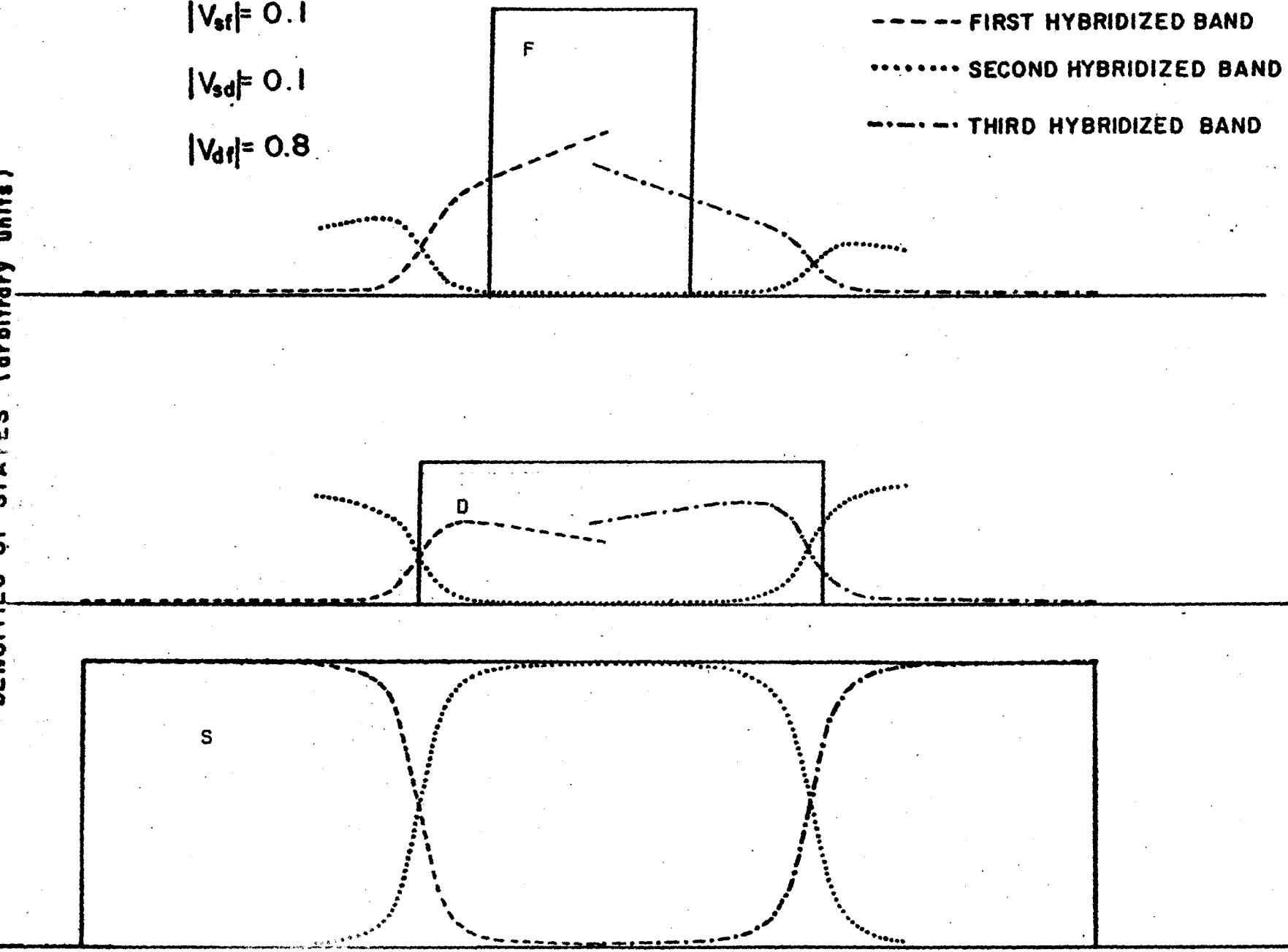$|V_{df}| = 0.0$

$|V_{sd}| = 0.1$

$|V_{sf}| = 0.1$

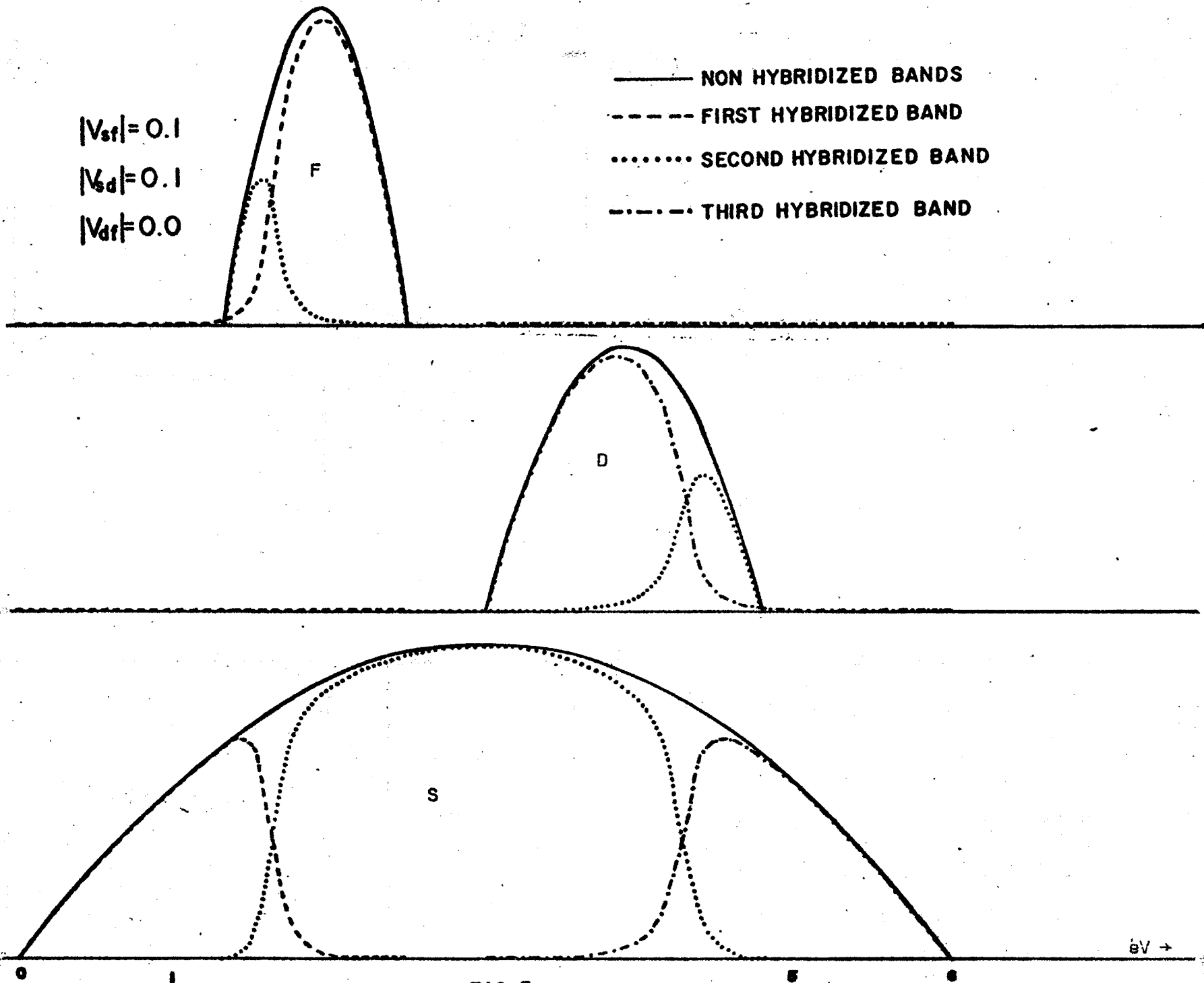DENSITIES OF STATES (arbitrary units)

eV →
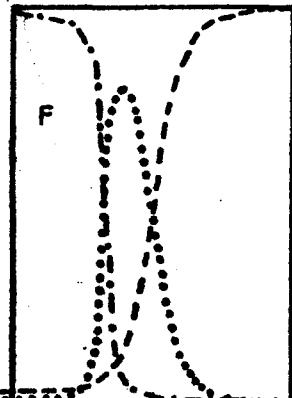
FIG. 4

FIG. 5

DENSITIES OF STATES (arbitrary units)

$|V_{sf}| = 0.1$

$|V_{sd}| = 0.1$

$|V_{df}| = 0.0$

———————— NON HYBRIDIZED BANDS

– – – – – FIRST HYBRIDIZED BAND

············ SECOND HYBRIDIZED BAND

–·–·–·– THIRD HYBRIDIZED BAND

F

D

S

eV →

130

FIG. 6

$|V_{sf}| = 0.1$

$|V_{sd}| = 0.1$

$|V_{df}| = 0.8$

NON HYBRIDIZED BANDS

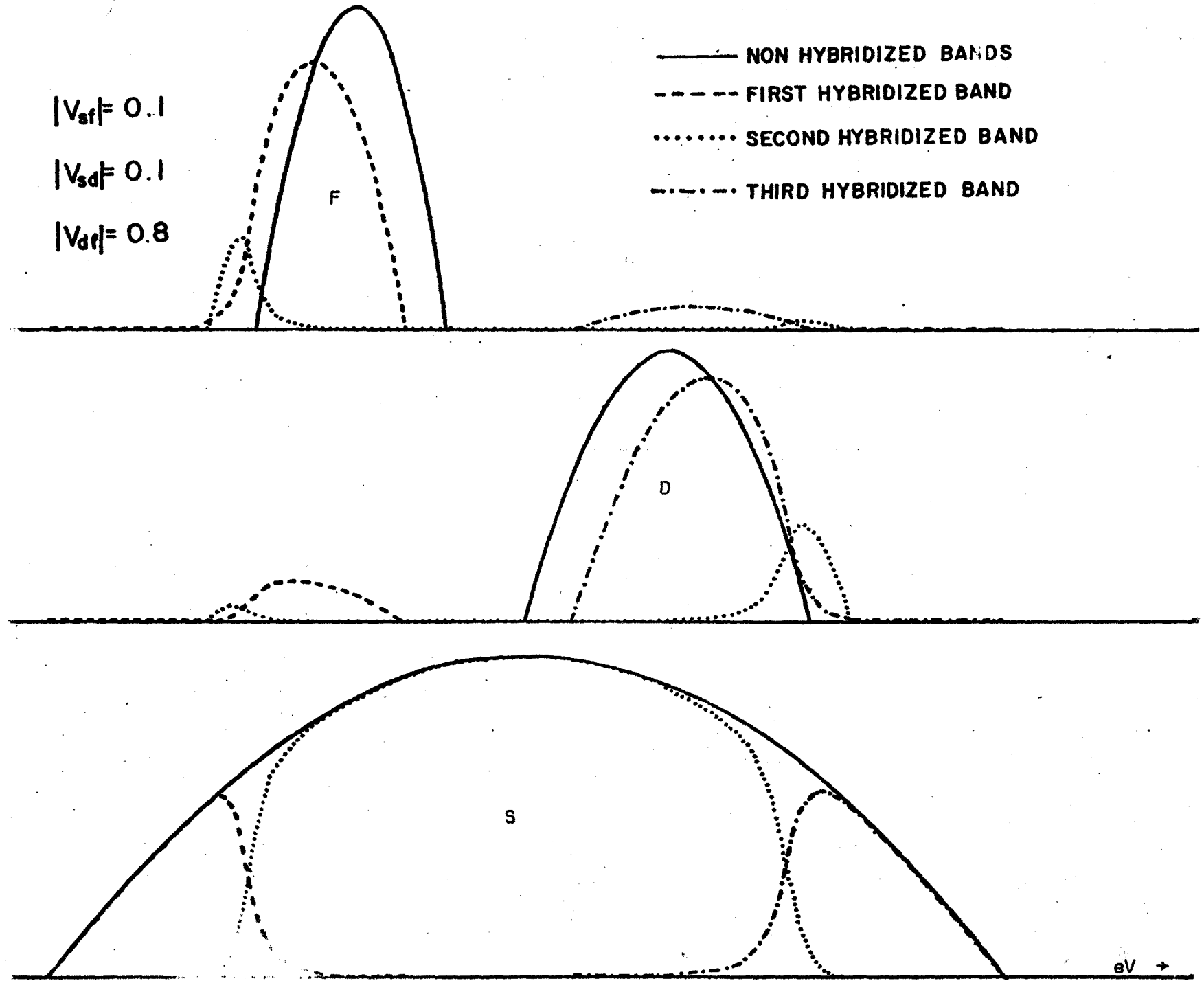FIRST HYBRIDIZED BAND

SECOND HYBRIDIZED BAND

THIRD HYBRIDIZED BAND

DENSITIES OF STATES (arbitrary units)

eV →

131

FIG 7

$|V_{sf}| = 0.1$

$|V_{sd}| = 0.1$

$|V_{df}| = 0.0$

NON HYBRIDIZED BANDS
FIRST HYBRIDIZED BAND
SECOND HYBRIDIZED BAND
THIRD HYBRIDIZED BAND

DENSITIES OF STATES (arbitrary units)

eV →

0   1   5   6

132

$|V_{sf}| = 0.1$

$|V_{sd}| = 0.1$

$|V_{df}| = 0.8$

——— NON HYBRIDIZED BANDS

– – – FIRST HYBRIDIZED BAND

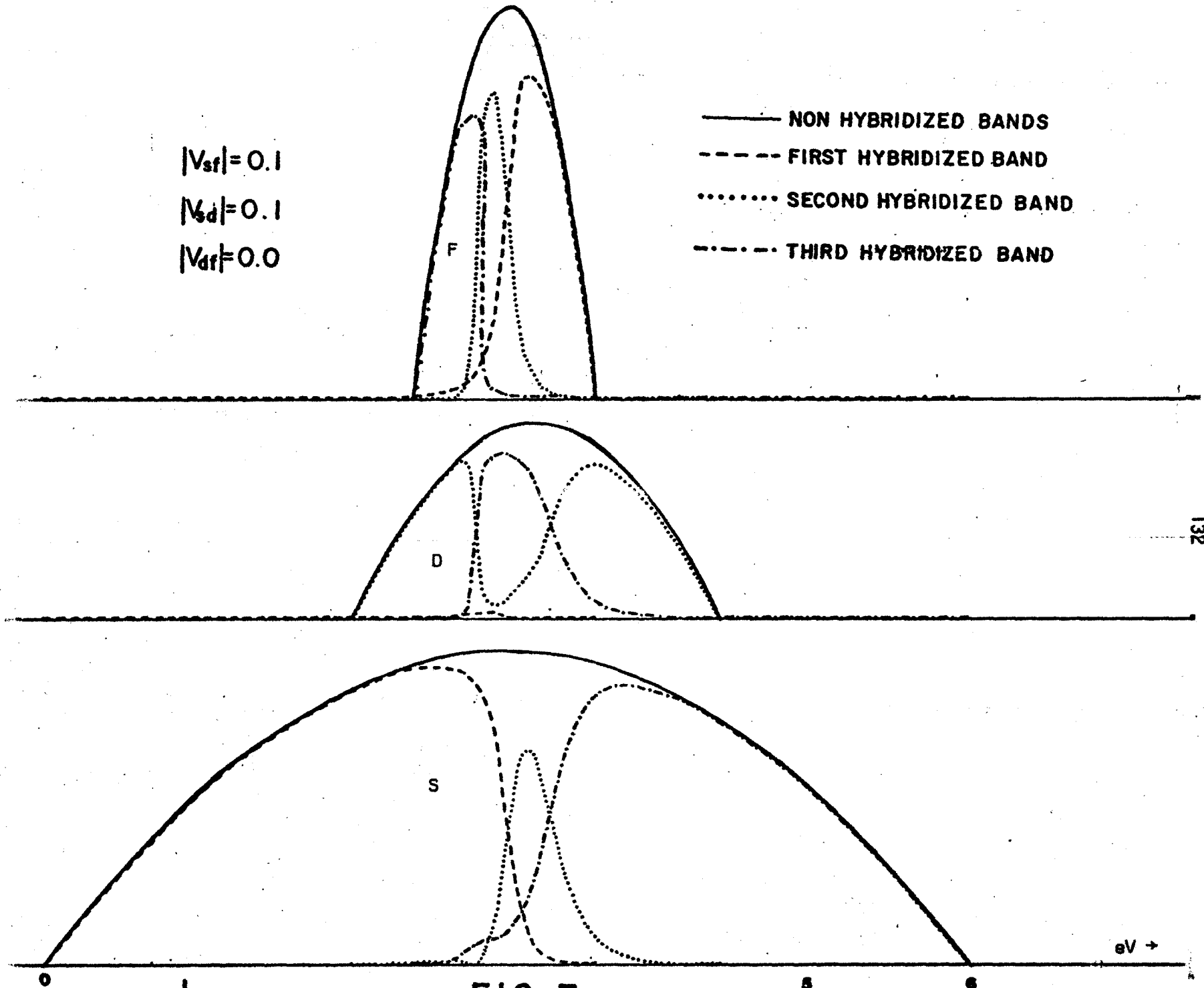········· SECOND HYBRIDIZED BAND

–·–·– THIRD HYBRIDIZED BAND

DENSITIES OF STATES (arbitrary units)

eV →

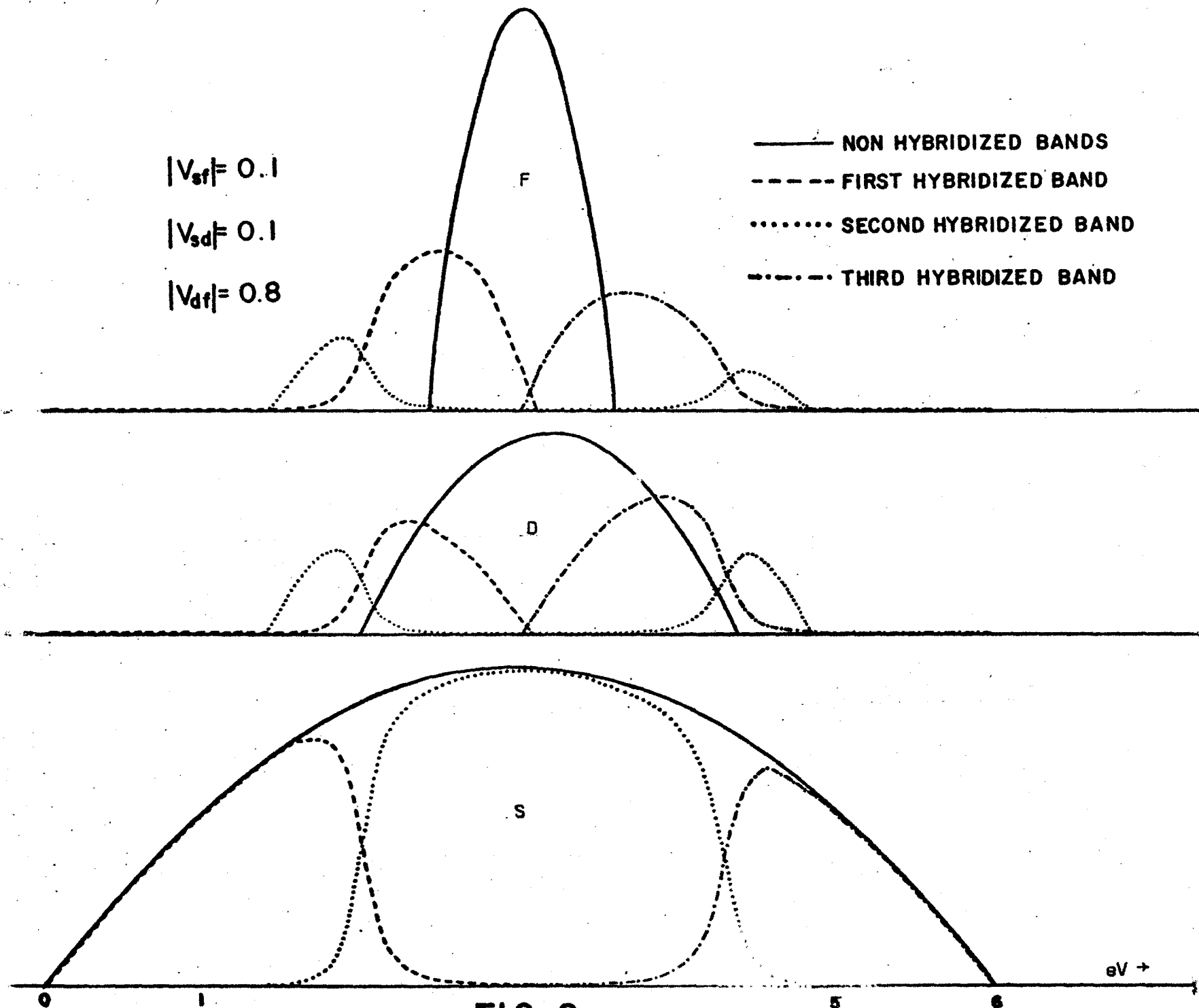0   1   5   6

FIG. 8

133

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