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Band ferromagnetism in systems of variable dimensionality

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The Stoner instability of the paramagnetic state, yielding to the occurence of ferromagnetism, is reviewed for electron density of states reflecting changes in the dimensionality of the system. The situations treated are one-dimensional (1D), two-dimensional (2D) and three-dimensional (3D) cases and also a special case where the density of states has a parabolic shape near the Fermi level, and is half-filled at equilibrium. We recover the basic results obtained in the original work of E.C. Stoner [Proc. Roy. Soc. London A **165**, 372 (1938)]; also we demonstrate that in 1D and 2D case, whenever the Stoner criterion is satisfied, the system evolves spontaneously towards maximum polarization ξ allowed by Hund's rules. For the 3D case, the situation is that: (i) when the Stoner criterion is satisfied, but the ratio between the Hubbard repulsion energy U and the Fermi energy ε_F is between 4/3 and 3/2, the system evolves towards a ferromagnetic state with incomplete polarization (the polarization parameter ξ is between 0 and 1); (ii) when $U/\varepsilon_F > 3/2$, the system evolves towards maximum polarization. This situation was also recognized in the original paper of Stoner, but with no further analysis of the obtained polarizations nor comparison with experimental results. We apply the result of calculation in order to predict the Hubbard interaction energy. Finally, for the case of half-filled parabolic density of states a result quite similar with the case of the free electron 3D DOS is obtained, but here the Hubbard energy has to be compared with the band half-widh δ, whereas the absolute value of the Fermi energy has no relevance in this model. The Stoner criterion is $U/\delta > 4/3$ and for $U/\delta < 2$ the total energy is minimized for incomplete polarization ξ. We compare the results obtained from the present theoretical study with the newest available experimental data.

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1. Introduction. The Stoner model

The basics of the Stoner model regarding the band ferromagnetism consists in rigid displacement of energy bands corresponding to the ↑ and ↓ spins [1]. In this paper we review the general physics of the zero temperature Stoner model and apply it to several forms of the electron density of states (DOS), modeling systems of variable dimensionality.

We further call the sub-band ↑ as of majority charge carriers, and the sub-band ↓ as of minority charge carriers. The densities of states are equal:

$$g_\uparrow(\varepsilon) = g_\downarrow(\varepsilon) = g(\varepsilon) \tag{1}$$

but the occupancies of energy bands are not equal (Fig. 1):

$$\varepsilon_F^\uparrow \neq \varepsilon_F^\downarrow \neq \varepsilon_F \neq \varepsilon_F^\uparrow \tag{2}$$

This will be subsequently understood as the *rigid band displacement model*. An alternate model for band ferromagnetism is the Hirsch model [2-4], where the shape of the DOS changes for ↑ and ↓ electrons, whereas the bands are not mutually displaced. However, detailed calculations of spin-dependent DOS [5] tend to confirm rather the validity of the Stoner model than that of the Hirsch model of ferromagnetism, at least for 3*d* metals.

Let ξ be the fraction of electrons which pass from the sub-band ↓ into sub-band ↑ (Fig. 1a).
Following Wohlfarth [3,6]:

$$n_\uparrow = \frac{n}{2}(1+\xi) = \int_0^{\varepsilon_F^\uparrow} g(\varepsilon)\, d\varepsilon \tag{3a}$$

$$n_\downarrow = \frac{n}{2}(1-\xi) = \int_0^{\varepsilon_F^\downarrow} g(\varepsilon)\, d\varepsilon \tag{3b}$$

It is straightforward to see that $\varepsilon_F^\uparrow$ and $\varepsilon_F^\downarrow$ are functions of ξ, then the bands are splitted by the value:

$$\Delta\varepsilon_S(\xi) = \varepsilon_F^\uparrow(\xi) - \varepsilon_F^\downarrow(\xi) \tag{4}$$

The conservation of electron number is written as:

$$\frac{n}{2}\xi = \int_{\varepsilon_F}^{\varepsilon_F^\uparrow} g(\varepsilon)\, d\varepsilon = \int_{\varepsilon_F^\downarrow}^{\varepsilon_F} g(\varepsilon)\, d\varepsilon \tag{5}$$

Thus

$$n\xi = \int_{\varepsilon_F^{\downarrow}}^{\varepsilon_F^{\uparrow}} g(\varepsilon)\, d\varepsilon \tag{6}$$

The intuitive reason of occurence of the band splitting and, furthermore, of ferromagnetic ordering is the *intra-site* Coulomb repulsion [7,8]. The opposite spin electrons, ↑ and ↓, may occupy the same orbital onto the same site, but with the cost of an energy increase due to the Coulomb repulsion. Hirsch [2] also pointed out that the exchange inter-site integral acts eventually only on the *direct* energy term, in case of hybridation, and not on the *exchange* term, where just the Coulomb interaction is the leading one. This situation is trated in the Hubbard model [9,10].

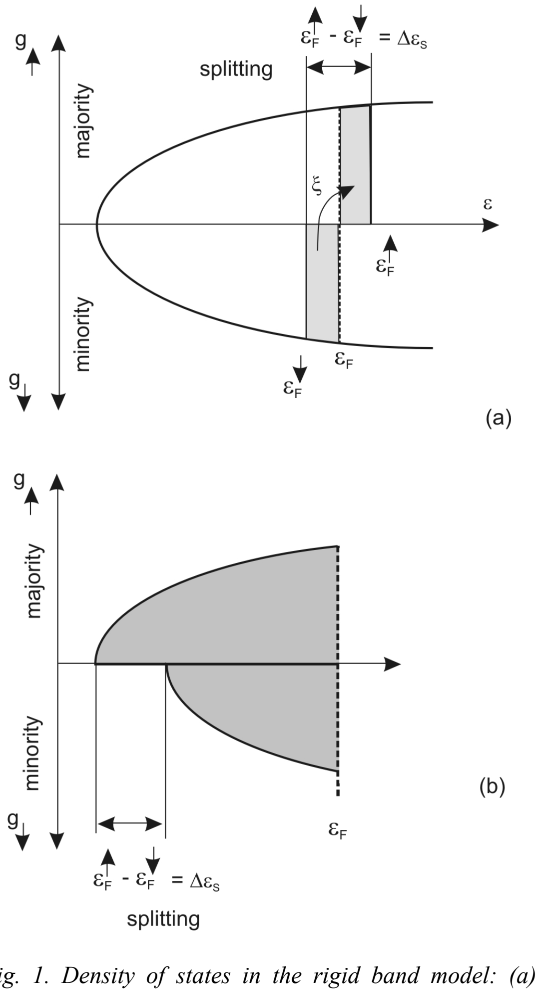

Fig. 1. Density of states in the rigid band model: (a) assuming two different Fermi levels for the two sub-bands; (b) assuming the same Fermi energy, which is actually the real physical case. The two situations are mathematically equivalent, but the formalism developed based on model (a) is easiest for computations.

2. The Hubbard model

In the original work of Stoner [1] the interaction term, proportional to the square of magnetization of the system, is introduced phenomenologically. Wohlfarth [6] took a step further by considering the many-electron Heitler-London model and connecting the interaction to the one- and two-electron hopping integrals. The microscopic origin of the quadratic term was demonstrated two decades after Stoner's work in the Hubbard model we discuss briefly in the following.

The Hubbard model describes the *hopping* process between sites in the presence of a Coulomb repulsion [9-11].

$$\begin{aligned}\widehat{\mathcal{H}} &= T\sum_{\langle ij\rangle} \hat{c}^{\dagger}_{i\sigma}\hat{c}_{j\sigma} + U\sum_{i}\hat{n}_{i\uparrow}\hat{n}_{i\downarrow} \\ &= T\sum_{\langle ij\rangle}\left(\hat{c}^{\dagger}_{i\uparrow}\hat{c}_{j\uparrow} + \hat{c}^{\dagger}_{i\downarrow}\hat{c}_{j\downarrow}\right) + U\sum_{i}\hat{c}^{\dagger}_{i\uparrow}\hat{c}_{i\uparrow}\hat{c}^{\dagger}_{i\downarrow}\hat{c}_{i\downarrow}\end{aligned} \tag{7}$$

$\langle ij\rangle$ indicates that the sum is performed over the nearest neighbours (in 1D, it means that

$$\sum_{\langle ij\rangle}\hat{c}^{\dagger}_{i\sigma}\hat{c}_{j\sigma} = \sum_{i}\sum_{j=i\pm1}\hat{c}^{\dagger}_{i\sigma}\hat{c}_{j\sigma}).$$

As usual $\hat{c}^{\dagger}_{i\sigma}$ and $\hat{c}_{i\sigma}$ are creation, respectively anihilation operators of electrons on site i with spin σ, and $\hat{n}_{i\sigma} = \hat{c}^{\dagger}_{i\sigma}\hat{c}_{i\sigma}$ are electron density operators.
If the states are localized on centers, such as

$$\psi_{i\sigma}(x) = \langle x|\psi_{i\sigma}\rangle = \langle x|\hat{c}^{\dagger}_{i\sigma}|\phi_0\rangle \longrightarrow \delta(x-x_i), \tag{8}$$

then the average value of the repulsion term on the vacuum state $\langle\phi_0|$ becomes [12]:

$$\begin{aligned}\langle\phi_0|U\sum_{i}\hat{n}_{i\uparrow}\hat{n}_{i\downarrow}|\phi_0\rangle &= U\sum_{i}N_{i\uparrow}N_{i\downarrow} \\ &= UN_{\uparrow}N_{\downarrow} \\ &= U\Omega^2 n_{\uparrow}n_{\downarrow}\end{aligned} \tag{9}$$

with N_σ being the total electron numbers, n_σ the corresponding electron densities, and Ω the crystal volume.
Further,

$$\begin{aligned}n_{\uparrow}n_{\downarrow} &= \frac{1}{4}\left[\left(n_{\uparrow}+n_{\downarrow}\right)^2 - \left(n_{\uparrow}-n_{\downarrow}\right)^2\right] \\ &= \frac{1}{4}\left[n^2 - \left(n_{\uparrow}-n_{\downarrow}\right)^2\right]\end{aligned} \tag{10}$$

The term $\frac{U\Omega^2}{4}n^2$ does not depend of spin polarization, and it will be involved in the kinetic term (it may correspond to introduction of a chemical potential). The term $-\frac{U\Omega^2}{4}\left(n_{\uparrow}-n_{\downarrow}\right)^2$ corresponds to the decrease of total energy due to spin polarization, thus to a ferromagnetic ordering.

3. The total interacting energy of the system

For the whole crystal, the interaction implies an energy decrease:

$$\begin{aligned}\Delta V &= -\frac{U\Omega^2}{4}\left(n_\uparrow - n_\downarrow\right)^2 \\ &= -\frac{U\Omega^2}{4}\left(\int\limits_0^{\varepsilon_F^\uparrow(\xi)} g(\varepsilon)\ d\varepsilon - \int\limits_0^{\varepsilon_F^\downarrow(\xi)} g(\varepsilon)\ d\varepsilon\right)^2 \\ &= -\frac{U\Omega^2}{4}\left(\int\limits_{\varepsilon_F^\downarrow(\xi)}^{\varepsilon_F^\uparrow(\xi)} g(\varepsilon)\ d\varepsilon\right)^2 < 0\end{aligned} \tag{11}$$

Also for the whole crystal, the kinetic energy will be given by

$$E_C = \Omega\left(\int\limits_0^{\varepsilon_F^\uparrow(\xi)} \varepsilon g(\varepsilon)\ d\varepsilon + \int\limits_0^{\varepsilon_F^\downarrow(\xi)} \varepsilon g(\varepsilon)\ d\varepsilon\right) \times N \tag{12}$$

We have multiplied by N, because otherwise the energy is related to its amount for a single electron.

Without spin polarization, $\varepsilon_F^\uparrow(0) = \varepsilon_F^\downarrow(0) = \varepsilon_F$, resulting that:

$$E_C^{(0)} = 2\Omega N \int\limits_0^{\varepsilon_F} \varepsilon g(\varepsilon)\ d\varepsilon \tag{13}$$

The variation of the kinetic energy will thus be written as

$$\Delta E_C = E_C - E_C^{(0)} = \Omega N\left(\int\limits_{\varepsilon_F}^{\varepsilon_F^\uparrow(\xi)} \varepsilon g(\varepsilon)\ d\varepsilon - \int\limits_{\varepsilon_F^\downarrow(\xi)}^{\varepsilon_F} \varepsilon g(\varepsilon)\ d\varepsilon\right) \tag{14}$$

Then, we compute the variation of total energy in presence of ferromagnetic polarization:

$$\begin{aligned}\Delta E(\xi) &= \Delta E_C(\xi) + \Delta V(\xi) \\ &= \Omega N\left(\int\limits_{\varepsilon_F}^{\varepsilon_F^\uparrow(\xi)} \varepsilon g(\varepsilon)\ d\varepsilon - \int\limits_{\varepsilon_F^\downarrow(\xi)}^{\varepsilon_F} \varepsilon g(\varepsilon)\ d\varepsilon\right) \\ &\quad - \frac{U\Omega^2}{4}\left(\int\limits_{\varepsilon_F^\downarrow(\xi)}^{\varepsilon_F^\uparrow(\xi)} g(\varepsilon)\ d\varepsilon\right)^2\end{aligned} \tag{15}$$

We will further use the *density of energy per electron and per unit volume*, which is definited as

$$\Delta w = \frac{1}{\Omega N}\Delta E \tag{16}$$

Extracting ξ from eq. (6) and including it in eq. (16), also keeping in mind that $N = n\Omega$, results in:

$$\Delta w(\xi) = \left(\int\limits_{\varepsilon_F}^{\varepsilon_F^\uparrow(\xi)} \varepsilon g(\varepsilon)\ d\varepsilon - \int\limits_{\varepsilon_F^\downarrow(\xi)}^{\varepsilon_F} \varepsilon g(\varepsilon)\ d\varepsilon\right) - \frac{nU}{4}\xi^2 \tag{17}$$

where

$$n = \int\limits_0^{\varepsilon_F^\uparrow(\xi)} g(\varepsilon)\ d\varepsilon + \int\limits_0^{\varepsilon_F^\downarrow(\xi)} g(\varepsilon)\ d\varepsilon \tag{18}$$

4. The Stoner instability

Being given a polarization ξ and once known the density of states $g(\varepsilon)$, the Fermi levels $\varepsilon_F^\uparrow$ şi $\varepsilon_F^\downarrow$, and the value of the splitting, $\Delta\varepsilon_S(\xi)$ may be univoquely defined via eqs. (4), (5), and (6).
Thus one may compute the dependence of the energy on the polarization ξ, via eq. (17). The extremum condition for the energy yields the equilibrium value of the polarization $\xi = \xi_0$

$$\begin{aligned}0 &= \left(\frac{\partial\Delta w}{\partial\xi}\right)_{\xi_0} \\ &= \varepsilon_F^\uparrow(\xi_0)g(\varepsilon_F^\uparrow(\xi_0))\left(\frac{\partial\varepsilon_F^\uparrow(\xi)}{\partial\xi}\right)_{\xi_0} + \varepsilon_F^\downarrow(\xi_0)g(\varepsilon_F^\downarrow(\xi_0))\left(\frac{\partial\varepsilon_F^\downarrow(\xi)}{\partial\xi}\right)_{\xi_0} \\ &\quad - \frac{nU}{2}\xi_0\end{aligned} \tag{19}$$

Deriving also the relations (5), one obtains:

$$\frac{n}{2} = g(\varepsilon_F^\uparrow(\xi))\frac{\partial\varepsilon_F^\uparrow(\xi)}{\partial\xi} = -g(\varepsilon_F^\downarrow(\xi))\frac{\partial\varepsilon_F^\downarrow(\xi)}{\partial\xi} \tag{20}$$

valid for any value of ξ, i.e. also for $\xi = \xi_0$.
Replacing (20) for $\xi = \xi_0$ in eq. (19), we obtain:

$$0 = \left(\frac{\partial\Delta w}{\partial\xi}\right)_{\xi_0} = \frac{n}{2}\left[\varepsilon_F^\uparrow(\xi_0) - \varepsilon_F^\downarrow(\xi_0)\right] - \frac{nU}{2}\xi_0 \tag{21}$$

Thus, one obtains a general equation:

$$\Delta\varepsilon_S(\xi_0) = \varepsilon_F^\uparrow(\xi_0) - \varepsilon_F^\downarrow(\xi_0) = U\xi_0 \tag{22}$$

This is an interesting result: the value of the band splitting (experimentaly accessible) is gived by the product between the Coulomb repulsion parameter and the fraction of band polarization (experimentaly accessible, as well). We shall use this formula especially for $\xi_0 \neq 0$, the case of $\xi_0 = 0$ being trivial (see below).

It is obvious that using eq. (22), an energy extremum is realized also for $\xi = 0$. The next step is to see in what conditions the state with $\xi = 0$ (which corresponds to paramagnetism, the absence of polarization) is unstable. This is the *Stoner instability* of the paramagnetic state, which requires that the second derivative of energy with respect to ξ to be negative:

$$0 > \frac{1}{n}\frac{\partial^2 \Delta w}{\partial \xi^2} = \frac{1}{2}\left(\frac{\partial \varepsilon_F^{\uparrow}}{\partial \xi} - \frac{\partial \varepsilon_F^{\downarrow}}{\partial \xi}\right) - \frac{U}{2} \quad (23)$$

By using eqs. (20),

$$\frac{\partial \varepsilon_F^{\uparrow}}{\partial \xi} = \frac{n}{2g(\varepsilon_F^{\uparrow}(\xi))}; \quad -\frac{\partial \varepsilon_F^{\downarrow}}{\partial \xi} = \frac{n}{2g(\varepsilon_F^{\downarrow}(\xi))} \quad (24)$$

Finally, we evaluate this second derivative in $\xi = 0$:

$$\frac{1}{n}\left(\frac{\partial^2 \Delta w}{\partial \xi^2}\right)_{\xi=0} = \frac{n}{2g(\varepsilon_F)} - \frac{U}{2} < 0 \quad (25)$$

where we replaced $\varepsilon_F^{\uparrow}(0) = \varepsilon_F^{\downarrow}(0) = \varepsilon_F$.

It results that paramagnetic state cannot be realized (the energy is not stable in $\xi = 0$) unless $g(\varepsilon_F)U < n$, corresponding to

$$\left(\frac{\partial^2 \Delta w}{\partial \xi^2}\right)_{\xi=0} > 0 \quad (26)$$

Contrary (the interesting case for ferromagnetism), if

$$g(\varepsilon_F)U > n, \quad (27)$$

the paramagnetic state is unstable. A ferromagnetic state with $\xi \neq 0$ will become the stable state of the system. Eq. (27) is called *the Stoner criterion* (1938).

If the DOS is normalized per electron, i.e. $\tilde{g}(\varepsilon_F) = \frac{1}{n}g(\varepsilon_F)$, the Stoner criterion may be written as [4,11]:

$$\tilde{g}(\varepsilon_F)U > 1 \quad (28)$$

5. Intuitive way of deduction of the Stoner criterion

This way is the usual one presented in textbooks [11,13] and we will briefly mention it for historical reasons. (Note also that we identified a few errors in the derivation presented in [13].) From Fig. 1 it results that a number of $\frac{n}{2}\xi$ eletrons inside unit volume pass from sub-band ↓ into sub-band ↑. Their average kinetic energy varies as follows:

$$\frac{\varepsilon_F^{\downarrow} + \varepsilon_F}{2} \longrightarrow \frac{\varepsilon_F + \varepsilon_F^{\uparrow}}{2} \quad (29)$$

It corresponds to a variation of kinetic energy (in unit volume per electron) of

$$\Delta\tilde{\varepsilon}_C \approx \frac{\varepsilon_F^{\uparrow} - \varepsilon_F^{\downarrow}}{2} = \frac{\Delta\varepsilon_S}{2} \quad (30)$$

At polarization ξ, the variation of the total energy of the system (in unit volume) will be

$$\Delta\varepsilon = \frac{n}{2}\xi\Delta\tilde{\varepsilon}_C - \frac{nU}{4}\xi^2 = \frac{n}{2}\xi\frac{\Delta\varepsilon_S}{2} - \frac{nU}{4}\xi^4 \quad (31)$$

We use eq. (6), approximated as:

$$n\xi = \int_{\varepsilon_F^{\downarrow}}^{\varepsilon_F^{\uparrow}} g(\varepsilon)\, d\varepsilon \approx g(\varepsilon_F)\left[\varepsilon_F^{\uparrow}(\xi) - \varepsilon_F^{\downarrow}(\xi)\right] = g(\varepsilon_F)\Delta\varepsilon_S(\xi) \quad (32)$$

Replacing in eq. (31), one obtains the variation of energy per unit volume at realization of ferromagnetic ordering:

$$\Delta\varepsilon = \frac{(n\xi)^2}{4g(\varepsilon_F)} - \frac{nU}{4}\xi^2 = \frac{n\xi^2}{4}\left[\frac{n}{g(\varepsilon_F)} - U\right] \quad (33)$$

Thus, the condition that the ferromagnetic state must be more stable than the paramagnetic state ($\Delta\varepsilon < 0$) results in the Stoner criterion [eqs. (27)-(28)].

Although the intuitive deduction of Stoner criterion presented in this section is simpler, the derivation of the Stoner instability from Sec. 5 is much more general and does not contain approximations except the assumption of rigid displacement of energy bands [formula (1)]. It also allows the computation of the energy dependence $\Delta w(\xi)$ and of the equilibrium polarization of the electron gas, whenever the ferromagnetic state is the most stable one.

6. Determinations of magnetic polarizations ξ for several dependencies of the density of states $g(\varepsilon)$

6.1. General considerations

An important observation is that in eq. (33) (the intuitive deduction of Stoner criterion) a series of approximations have been made in order to obtain the simple quadratic dependence of energy between the ferromagnetic state and the paramagnetic one: $\Delta\varepsilon = a\xi^2$.

$a > 0 \longrightarrow$ paramagnetic stability

$a < 0 \longrightarrow$ paramagnetic instabity (Stoner criterion)

The two situations are depicted in Fig. 2 by curves (1) and (2) respectively. Meanwhile, we may guess that the real situation could be reprezented by a curve (3) in Fig. 2, where $\Delta\varepsilon(\xi)$ has an (unstable) maximum in $\xi = 0$ and a minimum for a value $\xi_0 \neq 0$. The aim of the present section is to determine the value of band polarization, once the density of states $g(\varepsilon)$ is known. This will be possible by performing the next two steps:

i) Determination of the $\varepsilon_F^{\uparrow}(\xi)$ şi $\varepsilon_F^{\downarrow}(\xi)$ dependences starting from eqs. (5).

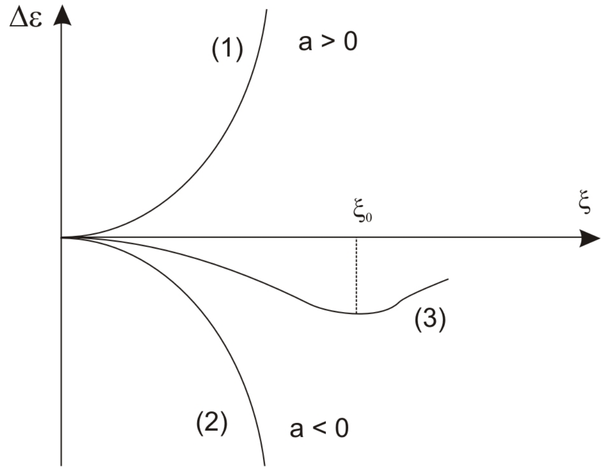

Fig. 2. Energy variation as function on the asymmetry parameter ξ. Case (1): stable paramagnetic state; case (2): unstable paramagnetic state; case (3) situation where the minimum energy is obtained for a given value of asymmetry ξ_0.

ii) Estimation of energy dependence on polarization and expressing the possibility of ferromagnetic ordering realization; finding the equilibrum value ξ_0:

$$\Delta w(\xi) = \int_{\varepsilon_F}^{\varepsilon_F^{\uparrow}(\xi)} \varepsilon g(\varepsilon) d\varepsilon - \int_{\varepsilon_F^{\downarrow}(\xi)}^{\varepsilon_F} \varepsilon g(\varepsilon) d\varepsilon - \frac{nU}{4}\xi^2 \tag{34a}$$

$$\left(\frac{\partial \Delta w}{\partial \xi}\right)_{\xi=\xi_0} = 0\,; \tag{34b}$$

$$\left(\frac{\partial^2 \Delta w}{\partial \xi^2}\right)_{\xi=\xi_0} > 0 \tag{34c}$$

6.2 The two-dimensional (2D) case. The case of constant density of states

This is the simplest case because $g(\varepsilon) = \text{const.} = g_0$ (see Fig. 3).

It may be easily seen that

$$2g_0\varepsilon_F = n \quad \Rightarrow \quad g_0 = \frac{n}{2\varepsilon_F} \tag{35}$$

The Stoner criterion thus becomes

$$g_0 U > n \quad \Rightarrow \quad U > 2g_0 \tag{36}$$

The Fermi levels shift themselves obeing Eqs. (5).
After introduction of the DOS (35), we obtain

$$\varepsilon_F^{\uparrow}(\xi) = \varepsilon_F(1+\xi); \quad \varepsilon_F^{\downarrow}(\xi) = \varepsilon_F(1-\xi) \tag{37}$$

The splitting is $2\varepsilon_F\xi$ and it does not depend on U. The energy has the parabolic dependence:

$$\Delta w(\xi) = \frac{n}{2}\left(\varepsilon_F - \frac{U}{2}\right)\xi^2 \tag{38}$$

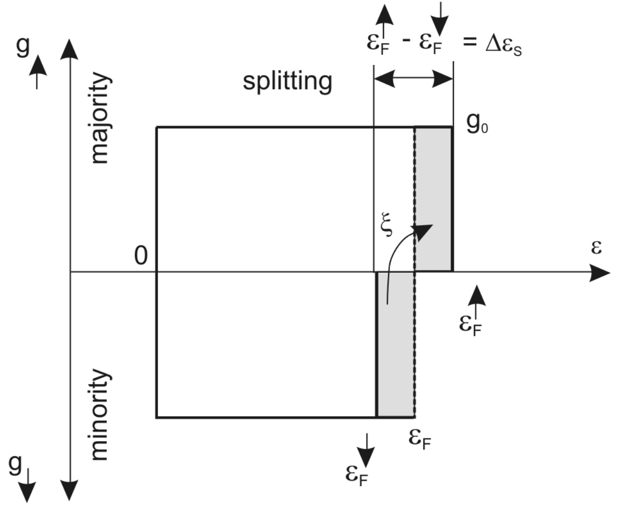

Fig. 3. Spin-resolved density of states for the ideal 2D case.

In the case of satisfaction of Stoner criterion, $U > 2\varepsilon_F$, $\Delta w(\xi)$ is a concave parabola (curve (2) in Fig. 2). There is no second minimum. The system develop by itself to the maximum admitted value of ξ which is $\xi = 1$ and it corresponds to the 100% polarization. In fact, the system envolves towards the maximum magnetic moment per atom allowed by Hund's rules. It is known that, for example, in the case of Fe surfaces, the atomic magnetic moment increases to 2.8-2.9 μ_B [14], which is near of maximum admitted value of 3 μ_B for the $4s^1 3d^7$ configuration of Fe. Also, even when deposited on a semiconductor and taking into account the presence of magnetically dead or of low magnetic moments at the interface [15], the Fe magnetic momentum of the thin Fe layers (2.7-2.8 μ_B; see [16]) exceeds largely the bulk value (2.2 μ_B). In fact, the realization of pure 2D magnetic system is rather difficult in practice. Other experimental cases illustrating at least qualitatively these findings are the induction of a net ferromagnetic ordering of manganese in Mn layers deposited on InAs(100), where Mn magnetic moments of 0.7 μ_B [17,18] have been reported, although the Mn ion itself is not in a very high spin state (only 2.4 μ_B; see [18]) and despite the strong interdifussion observed of the Mn into the semiconductor. An even higher Mn magnetic moment (1.6 μ_B) was reported in pure 2D Mn layers c(2×2) reconstructed on Cu(001) [19]. On contrary, in the case of semi-Heusler alloy trilayers NiMnSb/MgO/NiMnSb, a Mn ferromagnetic ordering with a magnetic moment as high as 3.9 μ_B (very close to the theoretical value of 4 Bohr magnetons, for the manganese $4s^1 3d^6$ configuration) was recently reported [20]. All these experimental findings may be summarized as in the following sentence: whenever intermixing, contamination or other interface effects may be minimized [21], a 2D system evolves alone towards the maximum polarization allowed by the Hund's rules. We add here that this is a just a consequence of the specific shape of the DOS (almost constant) for a 2D system.

In the case of Fe thin films, the value of the polarization may be calculated as follows: in paramagnetic state, $n = 7\ e^{-}/\mathrm{atom}$ and each sub-band is occupied with $n = 3.5\ e^{-}/\mathrm{atom}$. In the case of the configuration

↑↓	↑↓	↑	↑	↑

$n_{\uparrow} = 5\ e^{-}/\mathrm{atom}$ and $n_{\downarrow} = 2\ e^{-}/\mathrm{atom}$, then $1.5\ e^{-}$ are transfered from sub-band ↓ into sub-band ↑. Aplication of eqs. (5) or (44) consists in calculation of ξ as follows:

$$\frac{n}{2}\xi = 3.5\xi = 1.5 \ \Rightarrow\ \xi = \frac{1.5}{3.5} = 42.86\%$$

This is the maximum allowed polarization. There are no more electrons to can pass from sub-band ↓ into sub-band ↑ because there are no more free states.

In the case of solid iron (3*d*) the atomic magnetic moment is sensible considerably lower, being 2.2 μ_B/atom. It means that

$$n_{\uparrow} - n_{\downarrow} = 2.2,$$

thus $n_{\uparrow} = 3.5 + 1.1 = 4.6$ and $n_{\downarrow} = 3.5 - 1.1 = 2.4$, which means $\xi = 1.1/3.5 = 31.43\%$. We anticipate a result for the case 3D, namely, at a given moment the 3D dimensionality conditions will produce an energy curve of type (3) in Fig. 2, with a energy defined minimum for a value $\xi \notin (0, 1)$.

6.3 Density of states in the free electron aproximation for systems of variable dimensionality

The DOS is written as power function:

$$g(\varepsilon) = g_0^{(r)}\varepsilon^{r} \tag{39}$$

$$\text{with}\quad \begin{array}{ll} r = -\dfrac{1}{2} & (1\mathrm{D}) \\ r = \ \ 0 & (2\mathrm{D}) \\ r = +\dfrac{1}{2} & (3\mathrm{D}) \end{array} \tag{40}$$

$$\text{Generally,}\quad r = \frac{m}{2} - 1, \tag{41}$$

with m being the dimensionality of the system.

We have to keep in mind that g reffers to the sub-band DOS, and n reffers to the total electron density, that is, in the case of paramagnetism,

$$\int_0^{\varepsilon_F} g(\varepsilon)\, d\varepsilon = \frac{n}{2}\,. \tag{42}$$

The value of the constant $g_0^{(r)}$ may be easily deduced from eq. (42), being:

$$g_0^{(r)} = \frac{(r+1)n}{2\varepsilon_F^{r+1}} = \frac{mn}{4\varepsilon_F^{m/2}} \tag{43}$$

The position of Fermi energies in the two sub-bands ↑ and ↓ are derived from eqs. (5):

$$\varepsilon_F^{\uparrow}(\xi) = \varepsilon_F \cdot (1+\xi)^{\frac{1}{r+1}} = \varepsilon_F \cdot (1+\xi)^{\frac{2}{m}} \tag{44a}$$

$$\varepsilon_F^{\downarrow}(\xi) = \varepsilon_F \cdot (1-\xi)^{\frac{1}{r+1}} = \varepsilon_F \cdot (1-\xi)^{\frac{2}{m}} \tag{44b}$$

The energy [eq. (34a)] becomes:

$$\begin{aligned} \Delta w(\xi) &= \frac{n}{2}\frac{r+1}{r+2}\varepsilon_F\left[(1+\xi)^{\frac{r+2}{r+1}} + (1-\xi)^{\frac{r+2}{r+1}} - 2\right] - \frac{nU}{4}\xi^2 \\ &= \frac{n}{2}\frac{m}{m+2}\varepsilon_F\left[(1+\xi)^{1+\frac{2}{m}} + (1-\xi)^{1+\frac{2}{m}} - 2\right] - \frac{nU}{4}\xi^2 \end{aligned} \tag{45}$$

The Stoner criterion [eq. (34c)] easily results:

$$U > \frac{2\varepsilon_F}{r+1} \quad \text{or} \quad U > \frac{4\varepsilon_F}{m} \tag{46}$$

Particular cases:

a) One-dimensional systems, $m = 1$,

$$\varepsilon_F^{\uparrow}(\xi) = \varepsilon_F(1+\xi)^2 \tag{47a}$$

$$\varepsilon_F^{\downarrow}(\xi) = \varepsilon_F(1-\xi)^2 \tag{47b}$$

$$\Delta\varepsilon_S(\xi) = 4\varepsilon_F\xi \tag{47c}$$

$$\Delta w(\xi) = n\left(\varepsilon_F - \frac{U}{4}\right)\xi^2 \tag{47d}$$

result which confirms what we announced in the previous section. The Stoner criterion, $U > 4\varepsilon_F$, result in a shape of $\Delta w(\xi)$ as a concave parabola, i.e. curve (2) in Fig. 2.

b) Two-dimensional systems, $m = 2$. We obtain the same results as in the previous section:

$$\varepsilon_F^{\uparrow}(\xi) = \varepsilon_F(1+\xi) \tag{48a}$$

$$\varepsilon_F^{\downarrow}(\xi) = \varepsilon_F(1-\xi) \tag{48b}$$

$$\Delta\varepsilon_S(\xi) = 2\varepsilon_F\xi \tag{48c}$$

$$\Delta w(\xi) = \frac{n}{2}\left(\varepsilon_F - \frac{U}{2}\right)\xi^2 \tag{48d}$$

The Stoner criterion:

$$U > 2\varepsilon_F \tag{48e}$$

As a consequence, both in the 1D and 2D cases, whenever the Stoner criterion is satisfied, the most stable state is that with the maximum allowed polarization $\xi = 1$ or $\xi = \xi_{max}$, allowed by Hund's rules.

c) Three-dimensional systems, $m = 3$. The situation is to a certain extent more complicated and we will bring up numerical estimations (by using nowadays computing possibilities, which were not available when Stoner published his first work in 1938 [1] - this is a reason for which most part of this work is dedicated to the treatment of several approximations, series developments, etc.):

$$\varepsilon_F^{\uparrow}(\xi) = \varepsilon_F (1+\xi)^{2/3} \tag{49a}$$

$$\varepsilon_F^{\downarrow}(\xi) = \varepsilon_F (1-\xi)^{2/3} \tag{49b}$$

The energy is given by:

$$\Delta w(\xi) = \frac{n}{2}\left\{ \frac{3}{5}\varepsilon_F \left[(1+\xi)^{5/3} + (1-\xi)^{5/3} - 2 \right] - \frac{U}{2}\xi^2 \right\} \tag{50}$$

The Stoner criterion results as

$$U > \frac{4}{3}\varepsilon_F \tag{51}$$

In the 3D case, for certain values of the fraction $\frac{U}{\varepsilon_F}$ (between $4/3$ and $3/2$, result reported also by Stoner [1]) local minima of energy are obtained in the range $\xi \in (0,1)$. The $\Delta w(\xi)$ curves as eq. (50) are plotted in Fig. 4. It may be observed that the equilibrium assymetry value ξ_0 strongly varies as function of parameter $u = U/\varepsilon_F$.

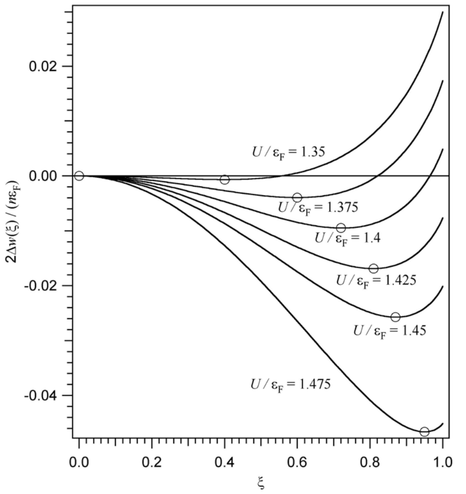

Fig. 4. Energy dependence on the asymmetry parameter, for the 3D case.

Fig. 5 represents the numerical solutions of the transcendent equation for the energy minimum condition:

$$\frac{2}{n\varepsilon_F}\left(\frac{\partial \Delta w}{\partial \xi}\right)_{\xi_0} = 0 \;\Rightarrow\; (1+\xi_0)^{2/3} - (1-\xi_0)^{2/3} = u\xi_0 \tag{52}$$

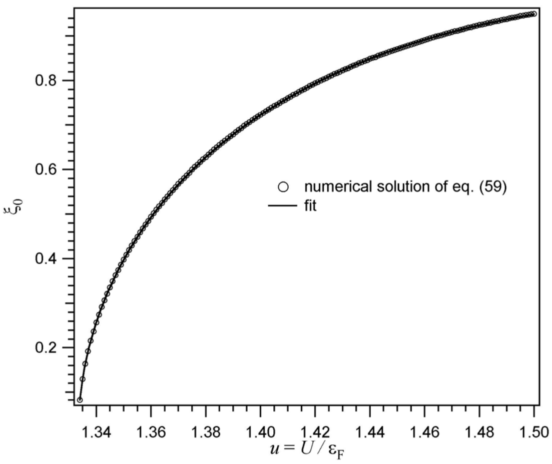

Fig. 5. Variation of the equilibrium asymmetry parameter ξ_0, as function on the ratio between the Hubbard term U and the Fermi energy ε_F.

The numerical solutions are well approximated by the following empirical formula:

$$\xi_0 \approx 3.28\left(u - \frac{4}{3}\right)^{0.504} - 3.93\left(u - \frac{4}{3}\right)^{1.304} \tag{53}$$

which is also represented in Fig. 5. In the following we will apply these computations to the case of ferromagnetic Fe and Ni.

Finally, Fig. 6 represents the energy variation of the minimum corresponding to the ferromagnetic ordering: $\Delta w(\xi_0) = f(u)$. It is obvious that for $u \gtrsim 1.5$ a 100% (or a maximum admitted) polarization is obtained using the previous considerations.

Back to the case of three-dimensional *bcc* iron, we have previous infered a 31.43% polarization. By analizing the numerical solutions (Fig. 5) we obtain $U/\varepsilon_F \approx 1.344$. Now, from band structure calculations, $\varepsilon_F \approx 6$ eV [5,22]. This results in a value of the Hubbard parameter of about 8 eV. Recently, in Ref. [22] a value of the Hubbard energy of 2.0 eV was theoretically computed by local spin density approximation (LSDA) calculations.

But, in the definition of the Hubbard energy of the above reference a further $1/2$ factor appears before the sum. On the other hand, in eq. (3) from [22] the sum was not performed over all nearest neighbours. To translate the Hubbard energy from Ref. [22] to the value derived in this work one has first to divide by 2, then to multiply by the number of nearest neighbors, which is 8 for *bcc* Fe.

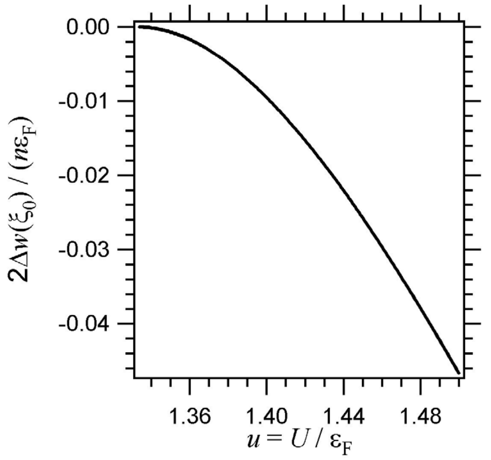

Fig. 6. Energy gain of the ferromagnetic state, as function on the ratio between the Hubbard term U and the Fermi energy ε_F.

This yields 8.0 eV, in agreement with the simple derivation of states (which is roughly the same, whatever computation method). Hence, the present theory offers a hint for the derivation of a parameter of fundamental importance (the Hubbard energy) from the magnetic properties in the band magnetism model.

For the *fcc* Ni case, the "ideal" configuration is $4s^1 3d^9$, with *d* orbital occupancy represented below:

↑↓	↑↓	↑↓	↑↓	↑

Here the average occupancy number per sub-band is 4.5 and the maximum number of transfered electrons is 1. The maximum allowed polarization yields as $\xi = 1/4.5 \approx 22.2\%$. In fact, both for the case of 3D and 2D Ni layers, the experimental value obtained at very low temperatures is 0.6 μ_B [23]. This means, in fact, that only 0.3 electrons are transfered from the $\downarrow$ into sub-band $\uparrow$. The effective polarization in this case is $0.3/4.5 \approx 6.7\%$. In this case, the value of the ratio U/ε_F is fairly close to its lower limit of 4/3 for the occurence of ferromagnetism (Fig. 5). Now, taking into account band structure calculations [22], $\varepsilon_F \approx 10$ eV and that means that the Hubbard parameter is about 13.3 eV for *fcc* Ni. In Ref. [22] this energy (multiplied by two, but divided to the number of nearest neighbors, which are 12 for *fcc* structures) was reported from LSDA calculation as being 3.0 eV, whereas, according to our evaluations, it should be $13.3 \times 2/12 \approx 2.22$ eV. The agreement is again good enough, but not as fair as in case of *bcc* Fe. The reason for this disagreement could also be connected to the strong configuration interaction reported for nickel [24].

6.4. Parabolic density of states, half-filled at equilibrium

We treated this subject owing also to the following arguments: (i) $3d$ bands are often represented as parabolic-like DOSs [11]; (ii) also, often in the case of systems of reduced dimensionality, there is a considerable depletion of the DOS by electron transfer to the substrate [15-18,21]; (iii) the presence of surface defects, impurities, terraces, nanoparticles, colloids, often results in the narrowing of the $3d$ bands [25]; (iv) especially in the case of Ni, it was recognized from several decades ago that a symmetric narrow function could be very well simulate its DOS [26]; (v) recently, we computed density of states obtained from the dispersion laws $\varepsilon(\vec{k})$ computed in the tight-binding approximation. As an example, for a simple cubic lattice, $\varepsilon(\vec{k}) = \varepsilon_0 - 2\gamma(3 - \cos k_x a - \cos k_y a - \cos k_z a)$, etc. [27]. The result $g(\varepsilon)$ may be approximated in some cases with a parabola. This work is still in progress and will be detailed in a future paper.

In this paragraph we shall treat only the case of half-filled parabolic DOS, since this may be useful for practical cases, when maximum polarization is aimed from a finite size (or nanostructured) system. The case of arbitrary band filling will be detailed also in a future communication.

The density of states is described, in this case, by:

$$g(\varepsilon) = g_0 - \alpha(\varepsilon - \varepsilon_F)^2 \tag{54}$$

and lies in the energy range $\varepsilon_- \leqslant \varepsilon \leqslant \varepsilon_+$, where:

$$\varepsilon_\pm = \varepsilon_F \pm \sqrt{\frac{g_0}{\alpha}} = \varepsilon_F \pm \delta \tag{55}$$

See figure 7. The parameters g_0 and α may be written as functions of the half width δ and of the (total) electron density n:

$$\int_{\varepsilon_-}^{\varepsilon_F} g(\varepsilon)\, d\varepsilon = \int_{\varepsilon_F}^{\varepsilon_+} g(\varepsilon)\, d\varepsilon = \frac{n}{2} \tag{56}$$

This means that

$$\alpha = \frac{3n}{4\delta^3};\ g_0 = \frac{3n}{4\delta} \tag{57}$$

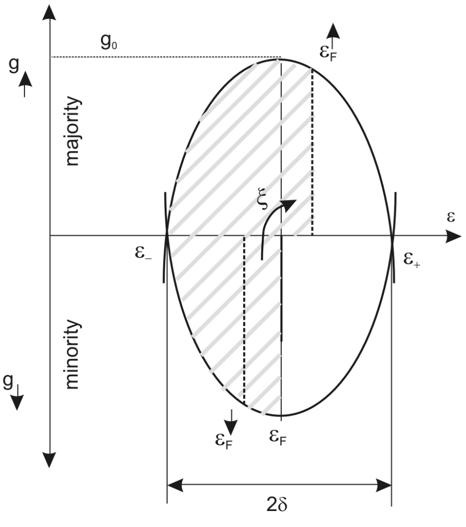

Fig. 7. The parabolic density of states, half-filled for zero polarization.

The Stoner criterion (27) would be in this case written by linking the Hubbard interaction with the sub-band width:

$$U > \frac{4}{3}\delta \tag{58}$$

The following substitution will be further employed:

$$\frac{\varepsilon_F^{\uparrow} - \varepsilon_F}{\delta} = \frac{\varepsilon_F - \varepsilon_F^{\downarrow}}{\delta} = x \tag{59}$$

The first equality results from obvious considerations of symmetry. Further, we deduce the variation of the shift of sub-bands Fermi levels as function of polarization degree $x(\xi)$ starting from eqs. (5):

$$\frac{n\xi}{2} = \int_{\varepsilon_F}^{\varepsilon_F^{\uparrow}(\xi)} g(\varepsilon)\, d\varepsilon = \frac{\varepsilon_F^{\uparrow}(\xi) - \varepsilon_F}{4\delta}\left[3 - \frac{(\varepsilon_F^{\uparrow}(\xi) - \varepsilon_F)^2}{\delta^2}\right]$$
$$\Rightarrow\ x(3 - x^2) = 2\xi \tag{60}$$

This is an algebraic cubic equation:

$$x^3 - 3x + 2\xi = 0 \tag{61}$$

with the only acceptable solution

$$x_0(\xi) = 2\cos\left[\frac{1}{3}(\pi + \cos^{-1}\xi)\right] \tag{62}$$

This function is plotted in Fig. 8.

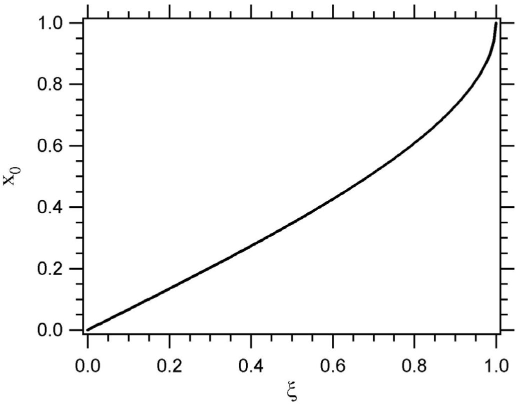

Fig. 8. Solutions (62) of equation (61).

Further, the energy may be easily estimated:

$$\frac{1}{n\delta}\Delta w(\xi) = 3\cos^2\left[\frac{1}{3}\left(\pi + \cos^{-1}\xi\right)\right] \times$$
$$\times \left\{1 - 2\cos^2\left[\frac{1}{3}\left(\pi + \cos^{-1}\xi\right)\right]\right\} -$$
$$- \frac{1}{4}\left(\frac{U}{\delta}\right)\xi^2 \tag{63}$$

The energy dependence as function of asymmetry parameter ξ is plotted in Fig. 9 for various values of the fraction U/δ. The result obtained in this case is qualitatively the same as for 3D free electron DOS, but in this case the Hubbard energy U has to be compared with the band half-width δ, whereas the absolute value of the Fermi energy does not occur in any equation. Further, by making the substitution $y = x_0^2$, the energy may be written as:

$$\frac{1}{n\delta}\Delta w(y) = \frac{3}{4}y\left(1 - \frac{y}{2}\right) - \frac{u}{16}y(3 - y)^2 \tag{64}$$

This allows the analytical computation of the minimum of the energy curves:

$$\frac{1}{n\delta}\left(\frac{\partial \Delta w}{\partial y}\right)_{y_{\min}} = 0 \implies y_{\min} = 3 - \frac{4}{u}$$
$$\implies \xi_{\min} = \frac{2}{u}\left(3 - \frac{4}{u}\right)^{1/2} \tag{65}$$

This also represents an alternative proof of the Stoner criterion, since there is no real value of ξ minimizing the function (70) if the Stoner criterion is not fulfilled, i.e. $u > 4/3$.

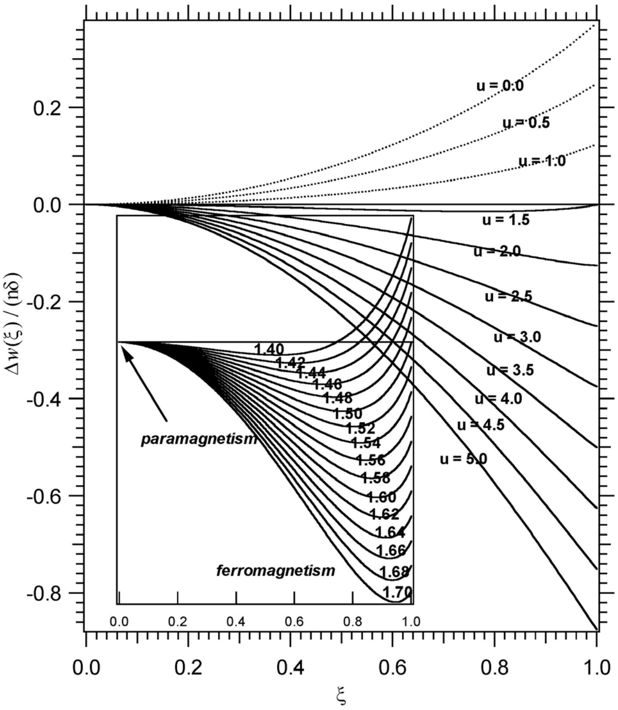

Fig. 9. Energy difference between the ferromagnetic and the paramagnetic state, as function on the asymmetry ξ, for several values of the parameter $u = U/\delta$ (ratio between Hubbard energy and DOS half-width). The dotted curves on the large graph represent cases where the Stoner criterion is not satisfied. The insert represents energy curves in the region of u yielding ferromagnetism with incomplete polarization.

Thus, the relevant cases are: (i) $u \leqslant 4/3$, the Stoner criterion is not satisfied and the paramagnetic state is the most stable one; (ii) $4/3 < u < 2$, a minimum occurs with incomplete polarization given by eq. (65); (iii) for $u \geqslant 2$, the system evolves towards maximum polarization $\xi = 1$, or the maximum allowed by other considerents, e.g. Hund's rules. We note here that the particular case considered of half-filled parabolic DOS does not imply automatically e.g. that the 3*d* bands are half-filled (i.e. a $3d^5$ configuration). The real DOS corresponding to all electronic states in the band may have low- (occupied) or high-energy (unoccupied) tails and behave as eq. (54) only locally, but for our model this is sufficient. It is clear that a generalization is needed, in the sense that one has to consider the band (54), represented in Fig. 7, but not necessarily half-filled. We intend to address this problem in a future work.

However, it is tempting to discuss the model presented in this Section at least to the case of Fe, where the computed DOS has some similarities with the half-filled parabolic DOS in absence of polarization [5,16,28]. Note also that in Ref. [28] tight-binding computations resulted in a similar model even for *constant* DOS. We noted in Sec. 6.3. that the Fe polarization is 31.4%, which in accordance with eq. (65) implies a value of $u \approx 1.354$. With a Hubbard energy of 8 eV (in our notation), this will imply that the band would have a half-width of 5.9 ≈ 6 eV, which is close to the theoretical computations [5]. (Remember that this was the starting value in order to derive the Hubbard energy in the 3D free electron DOS model.) We note here that it seems to be a striking equivalence between the parabolic DOS $g \sim g_0 - \alpha\varepsilon^2$ and the 3D free electron DOS $g \sim \varepsilon^{1/2}$, with some equivalence between the band half width δ with the Fermi energy of the second case. Moreover, similar equivalencies were reported in Ref. [28] even for the constant DOS which corresponds to the free electron 2D DOS. We suggest that more pure theoretical effort is needed to prove these equivalencies.

8. Conclusions

The Stoner model of band ferromagnetism at zero temperature is reviewed from the point of view of the Stoner instability of the paramagnetic state. Detailed calculations are presented for 1D, 2D and 3D systems, correlated with the available experimental data and making use of nowadays computing machines (necessary especially in the case of 3D systems). We obtain that for 1D and 2D systems, provided the Stoner criterion is satisfied, maximum polarization of the electrons occurs. Experimental available data support these assumptions. For the case of 3*d* electrons, we recover the original Stoner results, but in addition we exploit the numeric results in order to derive useful parameters, such as the Hubbard interaction energy. Examples are given related to detailed density of state calculations or to experimental results, mainly on metallic iron and nickel. Another special case treated in this paper is that of narrow, half-filled density of states, which may be approximated as parabolas near the Fermi level. It is shown that such systems behave in a very similar way with systems approximated by the three-dimensional free electron density of states. Further work will be devoted to (i) the non-zero temperature problem; (ii) to the case of a non-half-filled parabolic density of states; (iii) to adaptation of elements of Hirsch ferromagnetism in case of narrow density of states; (iv) to the application of Stoner theory to cases of DOS determined numerically by tight-binding theory.

Acknowledgements

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