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# Oscillatory coupling between interfaces in metallic multilayers

E Bruno† and B L Gyorffy

H H Wills Physics Laboratory, University of Bristol, Royal Fort, Tyndall Avenue, Bristol BS8 1TL, UK

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**Abstract.** We have developed a theory that views the total energy of an array of multilayers as a weighted average of bulk contributions corrected by a sum of single-interface and interface–interface interactions terms. We show that for metallic systems these interactions oscillate as functions of the appropriate interface–interface separations. Furthermore, we demonstrate that they are due to the same quantum-mechanical property of a degenerate Fermi liquid in a localized external potential as that which gives rise to the Friedel charge and magnetic oscillation around an impurity and the corresponding Ruderman–Kittel–Kasuya–Yoshida (RKKY) interactions. We argue that the recently discovered oscillatory magnetic coupling between magnetic layers separated by paramagnetic metallic layers should be accompanied by elastic interactions that give rise to periodic strain modulations,  $\Delta a/a$ , in the spacer layers. We illustrate the foregoing discussion by explicit calculation on an inhomogeneous jellium model.

## 1. Introduction

The advent of high-quality metallic multilayers [1, 2] provides a new context in which to study the metallic state. Indeed, one of the more striking features of these structures, namely the oscillatory coupling between magnetic layers in arrays where they are separated by non-magnetic metals, relies for its existence on the electrons forming a degenerate Fermi liquid [3]. In this paper we wish to contribute to the understanding of these very interesting phenomena by developing a new point of view for its description.

The systems we shall have in mind consist of alternate layers of two pure metals (A and B), which may be both paramagnetic, as illustrated in figure 1, or one paramagnetic (B) and the other ferromagnetic (A), as depicted in figure 2. Experimentally well studied examples are Fe/Cr, Co/Cr, Co/Ru and Fe/Cu [2], to mention but a few. As was discovered by a number of groups [4–6], at temperatures where each ferromagnetic layer is spontaneously magnetized, the relative orientation of layers may be parallel or antiparallel. In fact it is found [7–10] that the coupling between the magnetic moments of the individual layers oscillates from ferromagnetic to antiferromagnetic as the thickness of the non-magnetic (spacer) layer is varied. It is the physics of these oscillations that we shall be concerned with here. A very

† Permanent address: Dipartimento di Fisica, Università di Messina, Casella Postale 50, Villaggio S Agata, I-98166 Messina, Italy.

general explanation of these phenomena is provided by the tight-binding model of Edwards *et al* [11] and by the perturbative schemes of P Bruno and Chappert [12] and Coehoorn [13]. Although we agree with these interpretations, we shall rederive their basic conclusions by employing a different set of arguments. These, in our view, amplify certain important points and shed new light on the subject.

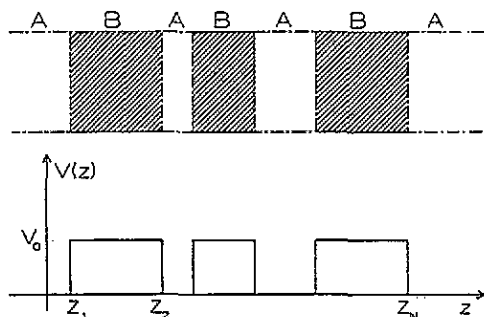


Figure 1. Model for a non-magnetic metallic multilayer. The volumes occupied by the metals A and B are bounded by  $n$  planar interfaces located at the positions  $\{Z_i\}$ . Corresponding to each interface, the potential varies by an amount  $V_0$ .

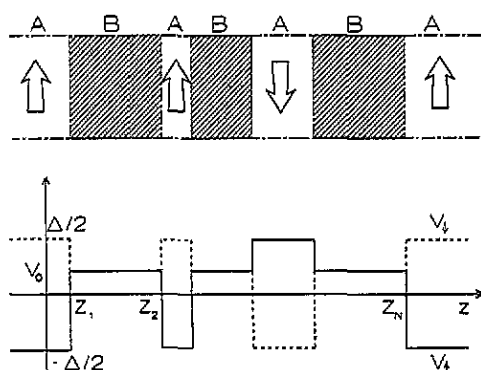


Figure 2. Model for a magnetic metallic multilayer. The metals A and B are bounded by  $n$  planar interfaces located at the positions  $\{Z_i\}$ . A magnetic moment pointing up or down is associated with each layer of A.  $V_1(z)$  (full line) and  $V_2(z)$  (broken line) are the potentials felt by majority and minority electrons,  $\Delta$  is the magnetic exchange splitting of A. Note that both the potentials have the common value  $V_0$  within the non-magnetic layers B.

The approach we wish to develop rests on regarding the above structures as systems of interacting interfaces. The thermodynamics of these are described in section 2. Our central result, there, will be the introduction of an interfacial grand potential  $\gamma$  and a set of thermodynamic forces  $\{f_i\}$ , analogous to the solvation forces between surfaces immersed in liquids [14, 15], conjugate to the positions of the interfaces  $\{Z_i\}$ . In section 3 we calculate these forces for simple models of two interfaces and show that they oscillate as a function of the separation. These oscillations will be seen as a consequence of the well known charge oscillation [16] in an electron gas at a sudden potential jump such as occurs naturally at impurities, surfaces and interfaces. Furthermore, as shown in section 4, they can be decomposed into magnetic and non-magnetic components, each of which oscillates. We estimate the size of the total forces and conclude that it should lead to observable strain,  $\Delta a/a$ , oscillations in the spacer layers. In the final section 5, we summarize our results and comment on the salient features of the problem.

## 2. The thermodynamics of interface–interface interactions

We model a multilayer as alternate slabs, of A and B metals, each slab having a thickness of several monolayers (figure 1). We assume the interfaces to be planar, but not necessarily sharp, each one located at the positions  $Z_i$  ( $i = 1, \dots, n$ ). The planes  $z = Z_i$  are, then, Gibbs dividing surfaces for the multilayer [17]. In the

following we consider the effect of 'moving' these interfaces: in the present context this has to be intended as the simple replacement of one kind of atom with the other, without any strain. We consider two different geometries, the periodic geometry (PG), in which the sequence  $Z_1, \dots, Z_n$  of the interfaces repeats periodically along the  $z$  direction, and the non-periodic one (NPG), in which one of the metals extends throughout the half-spaces, both on the left of  $Z_1$  and on the right of  $Z_n$ . Whereas an epitaxial multilayer is periodic in  $x$  and  $y$  directions, with the same unit translation vectors as the bulk metals A or B, the periodicity along  $z$  is very different in PG (with unit translation vectors of many atomic lengths), and there is no periodicity at all in NPG. We consider a multilayer in thermal contact with a 'reservoir of electrons' at fixed temperature  $T$  and chemical potential  $\mu$ ; consequently, the appropriate thermodynamic potential is the grand potential  $\Omega$ . Evidently, in the present case, it also depends on the areas  $\{A_i\}$  and locations  $\{Z_i\}$  of the interfaces. Namely,

$$\Omega = \Omega(T, V, \mu, \{A_i\}, \{Z_i\}) \quad (2.1a)$$

and the second law of thermodynamics takes the following form:

$$d\Omega = S dT - p dV - N d\mu + \sum_i \sigma_i dA_i - \sum_i (f_i A_i) dZ_i \quad (2.2)$$

where  $S$  is the entropy,  $p$  the pressure,  $V$  the total volume and  $N$  the total number of electrons. Clearly, the thermodynamic forces conjugate to  $\{A_i\}$  and  $\{Z_i\}$  are the surface tensions

$$\sigma_i = (d\Omega/dA_i)_{T, V, \mu, \{A_j, j \neq i\}, \{Z_i\}} \quad (2.3)$$

associated with each interface, and, respectively, the force on each interface

$$-(f_i A_i) = (d\Omega/dZ_i)_{T, V, \mu, \{A_i\}, \{Z_j, j \neq i\}} \quad (2.4)$$

where the notation, using the force per unit area  $f_i$ , explicitly indicates that these forces are proportional to  $A$ . We note that only  $n - 1$  of the locations of the interfaces are truly independent, because the grand potential has to be invariant under the translation  $Z_i \rightarrow Z_i + \alpha$ . Thus,

$$(\partial\Omega/\partial\alpha) = \sum_i (\partial\Omega/\partial Z_i) = - \sum_i (f_i A_i) = 0. \quad (2.1b)$$

In other words, we will use the grand potential (2.1a) together with the constraint (2.1b) that will ensure that all the relations we are going to derive are invariant with respect to translations of the origin.

Clearly, in (2.2), the term  $-(f_i A_i) dZ_i$  corresponds to the work done when the  $i$ th interface is displaced by an infinitesimal amount  $dZ_i$ , keeping temperature, chemical potential, interface areas and the positions of all the other interfaces fixed; it is then due only to the interactions between interfaces, so we may regard them as the resultant of interface-interface (II) forces [16]. It is interesting to note that these forces are the extension to a quantum system with an arbitrary number of permeable interfaces of the concept of 'solvation force', already introduced in the study of adsorption for classical fluids confined between two impenetrable walls [14, 15].

As is customary in the thermodynamic treatment of inhomogeneous materials, we define the excess grand potential [18]

$$\Omega_{\text{exc}} = \Omega - \Omega_{\text{bulk}} = \Omega - (V_A \omega_A + V_B \omega_B) \quad (2.5)$$

as the difference between the grand potential of the multilayer and  $\Omega_{\text{bulk}}$ . This latter quantity is defined as the grand potential of a homogeneous system (i.e. without interfaces) in which the metals A and B occupy the same volumes  $V_A$  and  $V_B = V - V_A$  as in the multilayer and have the same grand potential densities,  $\omega_A = -p_A$  and  $\omega_B = -p_B$ , as in the homogeneous metal. The differential of the grand potential excess can then be expressed in terms of surface excess quantities,

$$d\Omega_{\text{exc}} = S_{\text{exc}} dT - N_{\text{exc}} d\mu + \sum_i \sigma_i dA_i - \sum_i (f_i A_i) dZ_i \quad (2.6)$$

where, with obvious notation, we introduced the entropy and the electron excesses

$$S_{\text{exc}} = S - S_{\text{bulk}} = S - (V_A s_A + V_B s_B) \quad (2.7a)$$

$$N_{\text{exc}} = N - N_{\text{bulk}} = N - (V_A n_A + V_B n_B). \quad (2.7b)$$

We now observe that the thermodynamic potential

$$W = U + \sum_i (A_i f_i) Z_i$$

( $U$  is the internal energy) is a first-order homogeneous function of the extensive variables  $S$ ,  $V$ ,  $N$ ,  $\{A_i\}$  and  $\{A_i f_i\}$ , and hence it is straightforward to show [14, 19] that the following generalized equation of state [20] holds:

$$W = TS - pV + \mu N + \sum_i \sigma_i A_i + \sum_i (A_i f_i) Z_i.$$

Furthermore, the Legendre transformations from  $W$  to  $U$ ,  $\Omega$  and  $\Omega_{\text{exc}}$  lead to the following strikingly simple relations:

$$U = TS - pV + \mu N + \sum_i \sigma_i A_i \quad (2.8a)$$

$$\Omega = -pV + \sum_i \sigma_i A_i \quad (2.8b)$$

$$\Omega_{\text{exc}} = \sum_i \sigma_i A_i. \quad (2.8c)$$

Moreover, from (2.6) and (2.8c), using  $A_i = A$  for all  $i$ , we find the Gibbs equation

$$d\gamma = -(S_{\text{exc}}/A) dT - (N_{\text{exc}}/A) d\mu + \sum_i f_i dZ_i \quad (2.9)$$

where  $\gamma = \sum_i \sigma_i$ . The total surface tension  $\gamma$  turns out to be the most natural thermodynamic potential for the problem. Evidently it is closely related to the  $\Pi$  forces. In fact, it follows from (2.9) that

$$f_i = -(d\gamma/dZ_i)_{T, \mu, A, \{Z_j, j \neq i\}}. \quad (2.10)$$

To proceed further with our analysis for our inhomogeneous jellium model, we move all the interfaces according to the scaling relation

$$Z_i = \lambda \xi_i. \quad (2.11a)$$

Note that this procedure leaves unchanged all the areas, while the volumes occupied by the two metals change according to

$$V_A = \lambda V_A^0 \quad V_B = \lambda V_B^0 \quad (2.11b)$$

for the PG, and

$$V_B = \lambda V_B^0 \quad V_A = V - V_B = V - \lambda V_B^0 \quad (2.11c)$$

for the NPG (in the latter case the total volume  $V$  remains unchanged). As a consequence, from equations (2.5) and (2.8) we have

$$\Omega(\lambda) = A\gamma(\lambda) + [p_A V_A(\lambda) + p_B V_B(\lambda)] \quad (2.12)$$

and, making use of (2.11), we find, in the case of PG,

$$p^+(\lambda) = \sum_i f_i \xi_i + \frac{1}{A\lambda} [p_A V_A(\lambda) + p_B V_B(\lambda)] = -\frac{d\gamma}{d\lambda} + \frac{1}{A\lambda} [p_A V_A(\lambda) + p_B V_B(\lambda)] \quad (2.13a)$$

and, in the case of NPG,

$$p^+(\lambda) = \sum_i f_i \xi_i + \frac{1}{A\lambda} (p_B - p_A) V_B(\lambda) = -\frac{d\gamma}{d\lambda} + \frac{1}{A\lambda} (p_B - p_A) V_B(\lambda) \quad (2.13b)$$

where

$$p^+(\lambda) = -(1/A)(d\Omega/d\lambda) \quad (2.14)$$

is the pressure of the multilayered systems. Clearly, the term  $\sum_i f_i \xi_i = -(d\gamma/d\lambda)$  should be regarded as an extra pressure due to the interface-interface interactions (III).

After an integration over  $\lambda$ , we obtain the central result of this paper, for the PG and the NPG, respectively

$$\gamma(\lambda) - \gamma(\lambda_0) = \int_{\lambda_0}^{\lambda} [-p^+(\lambda') + (p_B V_B^0 + p_A V_A^0)/A] d\lambda' \quad (2.15a)$$

$$\gamma(\lambda) - \gamma(\lambda_0) = \int_{\lambda_0}^{\lambda} [-p^+(\lambda') + (p_B - p_A) V_B^0/A] d\lambda'. \quad (2.15b)$$

It can be easily seen, by taking the  $\lambda \rightarrow \infty$  limit, that the excess grand potential per unit area  $\gamma(\lambda)$  is the sum of two different contributions,  $\gamma(\infty)$ , the total surface tension when all the interfaces are infinitely apart from one another, and an excess term, which we attribute to the III. Naturally this latter contribution goes to zero when  $\lambda$  goes to infinity. Consequently we can define the III part of the grand potential as

$$\Omega_{\text{III}}(\lambda) = A[\gamma(\lambda) - \gamma(\infty)]. \quad (2.16)$$

As we shall see in the following sections,  $\Omega_{\text{III}}$  contains all the contributions that give rise to the much studied oscillatory coupling in multilayers. In summary

$$\Omega = \Omega_{\text{bulk}} + A\gamma(\infty) + \Omega_{\text{III}}. \quad (2.17)$$

An important aspect of the above conceptual framework is that it leads to some remarkably simple and suggestive formulae even for a fully interacting electron system. Using the language of the density-functional theory [21] we show, in appendix 1, that

$$-p^+(\lambda) = \frac{1}{A} \int d\mathbf{r} n(\mathbf{r}) \frac{\partial V(\mathbf{r})}{\partial \lambda} \quad (2.18)$$

where  $n(\mathbf{r})$  is the charge density in the inhomogeneous ground state. Moreover, in the very important case in which the external potential  $V(\mathbf{r})$  has a finite range and all the distances between the interfaces are larger than this, an increase of  $\lambda$  results only in a solid shift of the external potential, whose  $\lambda$  derivative can then be easily expressed in terms of the  $z$  derivative. We merely quote here, as an example, the result for a two-interface system (note that the origin of the  $z$  axis is chosen, as in figure 3, to be halfway between the interfaces)

$$p^+(\lambda) = \frac{1}{2} \int dz \operatorname{sgn}(z) n(z) \frac{dV(z)}{d\lambda}. \quad (2.19)$$

Since the derivative of the external potential is large only in the interfacial regions, (2.19), as well as (2.18), consists mainly of contributions from the charge density in these regions. Clearly this is fully consistent with our interpretation of  $p^+(\lambda)$  as due to interface–interface interactions.

### 3. Simple two-interface models

#### 3.1. One-dimensional model

Before embarking on realistic first-principles calculations of the  $\Pi$  forces introduced above, it is worth while to explore, in detail, the oscillatory coupling between layers by explicit calculations for simple inhomogeneous jellium models.

The simplest possible model of interest is that in which the junction between the metals A and B is described as a finite discontinuity of the external potential (figure 3), which we chose to be the difference between the work functions of the two metals. That is to say

$$V(z) = \begin{cases} 0 & \text{for } -t < z < -L/2, L/2 < z < t \\ V_0 & \text{for } -L/2 < z < L/2. \end{cases} \quad (3.1)$$

Namely, the metal A occupies the regions in which the external potential is zero, and B that in which the potential has the constant value  $V_0$ . All the space is filled by a one-dimensional (1D) electron gas, in thermal contact with a reservoir of electrons (see section 2). Later we will take the limit  $t \rightarrow \infty$ . Note that the present sandwich model is formally equivalent to considering the metal B as a single (atomic) ‘impurity’ in the infinite medium A. Most of the results of the present section may easily be generalized to the case of an arbitrary number of such ‘impurities’ described by finite-ranged potentials. Clearly, a periodic arrangement of these can be thought of as a model for multilayers.

Following the well known idea of Friedel [22] we will consider the difference, for each spin, between the grand potential  $\Omega$  of the sandwich and  $\Omega_0$ , which corresponds to a reference system without impurities, i.e. the pure A metal. At zero temperature, we find that

$$\Omega - \Omega_0 = \int_0^{E_F} dE (E - E_F) \int_{-t}^t dz [n(E, z) - n_0(E)] + \Omega_{\text{bound}} + \Omega' - \Omega'_0 \quad (3.2)$$

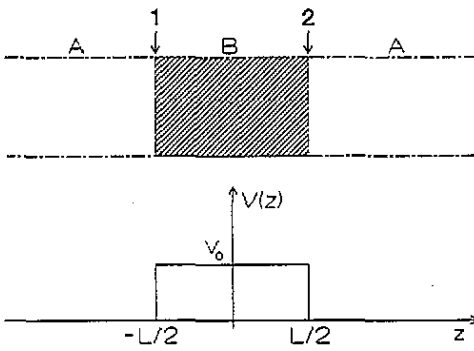


Figure 3. Model for a two-interface non-magnetic metallic multilayer. The interfaces are marked by 1 and 2 and located at  $z = \pm L/2$ . Corresponding to each interface, the external potential  $V(z)$  varies by an amount  $V_0$ .

where  $n_0(E) = (2\pi\sqrt{E})^{-1}$  is the 1D free-electron density of states (DOS),  $E_F$  the Fermi energy and

$$n(E, z) = -(1/\pi)\text{Im Tr}\{G(E, z, z)\} \quad (3.3)$$

is the diagonal part of the electronic density matrix for the multilayer and is given in terms of the one-electron Green function  $G(E, z, z')$ . In equation (3.2)  $\Omega_{\text{bound}}$  takes into account possible contributions from bound states,  $\Omega'$  and  $\Omega'_0$  are, for the multilayer and the homogeneous metal A, the interacting part of the grand potential for the multilayer and the homogeneous metal A, respectively. To find  $G(E, z, z')$  we use the one-dimensional scattering theory of Butler [23, 24]. In this the wavefunction is expanded in terms of its symmetric and antisymmetric components labelled by the pseudo-angular-momentum index  $l = 0$  and  $l = 1$  respectively. Then the solutions of the Schrödinger equation are described by the pseudo-radial functions  $R_l(z, E)$  and the corresponding phase shifts  $\delta_l(E)$ . For further details the reader is referred to [23] and [24] and to appendix 2. The main result from the above theory is

$$n(E, z) = \sum_l n_l(E, z) = \sum_l \frac{R_l^2(z, E) Y_l^2(z)}{\pi\sqrt{E}}. \quad (3.4)$$

Using theorem A2.2 ( $A_l^2(E) = (\pi\sqrt{E})^{-1}$ ), we find that

$$\int_{-t_m}^{t_m} [n_l(E, z) - \frac{1}{2}n_0(E)] dz = \frac{1}{\pi} \frac{d}{dE} \delta_l(E). \quad (3.5)$$

If now we choose, for each value of  $l$  and  $E$ ,  $t_m$  large enough to have  $n(E, Z) - n_0(E) = 0$ , for  $|z| > t_m$ , equation (3.2) can be rewritten as

$$\begin{aligned} \Omega - \Omega_0 &= \frac{1}{\pi} \sum_l \int_0^{E_F} dE (E - E_F) \frac{d}{dE} \delta_l(E) + \Omega_{\text{bound}} + \Omega' - \Omega'_0 \\ &= \frac{1}{\pi} \sum_l \int_0^{E_F} dE [\delta_l(0) - \delta_l(E)] + \Omega_{\text{bound}} + \Omega' - \Omega'_0 \end{aligned} \quad (3.6)$$

where for  $V_0 < 0$  the contribution from bound states,  $\Omega_{\text{bound}}$ , also has to be considered. In that case it turns out to be

$$\Omega_{\text{bound}} = \sum_{k=1}^{n_{\text{max}}} E_k - (E_F - V_0)n_{\text{max}} \quad (3.7)$$



with

$$n_{\max} = 1 + \text{Int}(L\sqrt{-V_0}).$$

Evidently the above result is the one-dimensional analogue of the celebrated Friedel sum [25].

Using the explicit expression for the phase shifts given in appendix 3 and identifying the terms proportional to  $\omega_A$  and  $\omega_B$ , the excess grand potential (2.5) for the present model may be written as

$$\Omega_{\text{ex}}(L) = -\frac{2}{\pi} \int_0^{k_F} dk kg(k) + \Delta\Omega_{\text{bound}}(L) + \Delta\Omega'(L) \quad (3.8)$$

where  $k_F$  is the Fermi momentum, and

$$g(k) = \tan^{-1} \left[ \left( \frac{k}{2\sqrt{(k^2 - V_0)}} + \frac{\sqrt{(k^2 - V_0)}}{2k} \right) \tan[L\sqrt{(k^2 - V_0)}] \right] - L\sqrt{(k^2 - V_0)} + m\pi \quad (3.9)$$

is the sum over the phase shifts with a contribution linear in  $L$  subtracted. We stress that  $g(k)$  is a continuous function of both  $k$  and  $L$  and the integer  $m$  is chosen to ensure continuity (see appendix 3). Under these conditions

$$\Delta\Omega_{\text{bound}}(L) = \left( \sum_{k=1}^{n_{\max}} E_k + \frac{2L}{3\pi} (-V_0)^{3/2} \right) \Theta(-V_0) \quad (3.10)$$

where  $\Theta$  is the Heaviside function and

$$\Delta\Omega'(L) = \Omega'(L) - \Omega'_{\text{bulk}}. \quad (3.11)$$

### 3.2. A three-dimensional inhomogeneous jellium model

We note with considerable interest that this model is no more difficult to solve, either analytically or computationally, than the genuinely one-dimensional model described above. It consists of a three-dimensional external potential, which is, however, a constant in the  $x$  and  $y$  directions. Such a model, already investigated extensively for the single-interface problem [16, 26, 27], will be referred to in the following as the inhomogeneous jellium.

For the time being we neglect the electron-electron interactions and study the solutions of the Schrödinger equation

$$-\nabla^2\psi(x, y, z) = [E - V(z)]\psi(x, y, z). \quad (3.12)$$

Evidently, the eigenvalues are

$$E_{Q, k_z} = Q^2 + k_z^2 \quad (3.13)$$

and the corresponding eigenfunctions may be factorized as

$$\psi_{Q, k_z}(x, y, z) = \chi_Q(\mathbf{R})\phi_{k_z}(z) \quad (3.14)$$

where for the two-dimensional wavevector  $Q = (k_x, k_y)$  and position vector  $\mathbf{R} = (x, y)$

$$\chi_Q(\mathbf{R}) = e^{i\mathbf{Q}\cdot\mathbf{R}} \quad (3.15)$$

and  $\phi_{k_z}(z)$  is the solution of the 1D Schrödinger equation

$$-\frac{d^2}{dz^2}\phi_{k_z}(z) = [k_z^2 - V(z)]\phi_{k_z}(z). \quad (3.16)$$

For the present model the density matrix can be evaluated through the formula

$$n(E, z) = \frac{1}{4\pi} \int dQ \left( \frac{1}{2\pi} \int dk_z + \Theta(-V_0) \sum_m \right) |\phi_{k_z}(z)|^2 \delta(E - (Q^2 + k_z^2)) \quad (3.17)$$

where  $m$  is the quantum number labelling, when appropriate, states bounded along the  $z$  direction. After integration over  $Q$ , using the fact that the external potential is an even function of  $z$ , we find

$$n(E, z) = \frac{1}{4\pi^2} \int_0^{\sqrt{E}} dk |\phi_k(z)|^2 + \Theta(-V_0) n_{\text{bound}}(E, z) \quad (3.18a)$$

where the contribution coming from bound states is given by

$$n_{\text{bound}}(E, z) = \frac{1}{4\pi} \sum_m |\phi_{k_z, m}(z)|^2 \Theta(E - k_{z, m}^2). \quad (3.18b)$$

On integration over the energy, one is led to the expression for the charge density, which for metallic systems ( $E_F > 0$ ) is

$$n(z) = \frac{1}{4\pi^2} \int_0^{k_F} dk (k_F^2 - k^2) |\phi_{k_z}(z)|^2 + \Theta(-V_0) n_{\text{bound}}(z) \quad (3.19a)$$

$$n_{\text{bound}}(z) = \frac{1}{4\pi} \sum_m |\phi_{k_z, m}(z)|^2 (k_F^2 - k_{z, m}^2). \quad (3.19b)$$

Then, using the density of states per unit volume

$$n(E) = \frac{1}{2t} \int dz n(E, z) \quad (3.20)$$

the  $T = 0$  grand potential is

$$\Omega = A \int dE (E_F - E) \Theta(E_F - E) \int dz n(E, z) + \Omega' \quad (3.21)$$

where  $A$  is the area normal to the  $z$  direction. Following the 1D discussion in the previous subsection, the difference between  $\Omega$  and the grand potential of the homogeneous metal  $A$  may be written as

$$\Omega - \Omega_0 = A \int_0^{E_F} dz [n_v(E, z) - n_0(E)] + \Omega_{\text{bound}} + \Omega' - \Omega'_0 \quad (3.22)$$

where  $n_v(E, z)$  is the valence part of the density matrix, i.e.  $n(E, z) - n_{\text{bound}}(E, z)$ . Since the relevant solutions for  $|z| > L/2$  of equation (3.16) can be written in terms of the 1D solutions of the previous subsection, namely

$$\phi_{k_z}(z) = \sqrt{2} \sum_l R_l \left( \sqrt{E}|z| \right) Y_l(z)$$

and the homogeneous DOS  $n_0(E) = \sqrt{E}/(4\pi)$ , we may again use theorem A2.2 to write

$$\int_{-t_m}^{t_m} dt [n_v(E, z) - n_0(E)] = \frac{1}{4\pi^2} \int_0^{\sqrt{E}} dk \frac{d}{dk} \delta_l(k^2).$$

Thus the 3D analogue of the result in equation (3.6) is

$$\Omega - \Omega_0 = \frac{A}{4\pi^2} \int_0^{E_F} dE (E - E_F) \sum_l [\delta_l(E) - \delta_l(0)] + \Omega_{\text{bound}} + \Omega' - \Omega'_0. \quad (3.23)$$

As in section 3.1, equation (A3.3) implies

$$\Omega_{\text{exc}} = \frac{A}{2\pi^2} \int_0^{k_F} dk (k^2 - k_F^2) kg(k) + \Delta\Omega_{\text{bound}} + \Delta\Omega' \quad (3.24)$$

where

$$\Delta\Omega_{\text{bound}} = \left( -\frac{1}{8\pi} \sum_{k=1}^{n_{\text{max}}} (E_k - E_F)^2 + \frac{1}{30\pi^2} [5(-V_0)^{3/2} E_F - 2V_0] L \right) \Theta(-V_0). \quad (3.25)$$

This is the principal result of this section. Clearly it is an easy-to-evaluate, exact, non-interacting-electron formula for investigating the  $\Pi$  interactions defined in section 2. It should be noted that equation (3.24), as opposed to equation (3.6), was not derived for a single 'impurity' but for a 'plane of impurities', and, therefore, it is a non-trivial generalization of the Friedel theorem. An expression similar to (3.24) was already derived by Sugiyama [28] for the excess energy in a single-interface model within a Thomas-Fermi approximation.

## 4. The oscillatory contribution to the interface-interface forces

### 4.1. General considerations

An interesting confirmation of the very general, thermodynamic, arguments in section 2 follows from explicit calculations for the simple models of two interfaces discussed in section 3. Evidently, without any reference to section 2, the total force per unit area between two interfaces, separated by a spacer layer of thickness  $L$ , may be defined as

$$p_{12}^+(L) = -(1/A)(\partial\Omega/\partial L). \quad (4.1)$$

For the two models of interest, using equations (3.6) and (3.23), for the case in which  $V_0 > 0$ , we find

$$p_{12}^+(L) = \frac{1}{\pi} \int_0^{E_F} dE \frac{\partial}{\partial L} \sum_l \delta_l(E) \quad (4.2)$$

and

$$p_{12}^+(L) = -\frac{1}{4\pi^2} \int_0^{E_F} dE (E - E_F) \frac{\partial}{\partial L} \sum_l \delta_l(E) \quad (4.3)$$

respectively. We now note, and show in appendix 4, that

$$\frac{\partial}{\partial L} \sum_i \delta_i(E) = \sum_i \int_{-\infty}^{+\infty} dz \frac{R_i^2(E)}{\sqrt{E}} \frac{\partial V(z)}{\partial L} \quad (4.4)$$

and hence for the one-dimensional as well as the three-dimensional jellium

$$p_{12}^+(L) = - \int_{-\infty}^{+\infty} dz n(z; L) \frac{\partial V(z)}{\partial L} \quad (4.5)$$

where we have used equations (3.4), (3.19a) and (3.22) to identify the appropriate expressions for the charge density  $n(z)$ , which clearly depends parametrically on the separation  $L$ .

That the present, explicitly quantum-mechanical, derivation yields equation (4.5) in agreement with equation (2.18) supports our conclusions in section 2 concerning the generality of equation (2.18). Namely, the simple classical solvation force formula [15], equation (2.18), remains valid in a fully quantum-mechanical treatment of the electron liquid.

Although equation (4.5) is valid for arbitrarily shaped potentials  $V(z)$ , we now wish to focus on rigidly shifting sudden jumps as in figure 3. In this case equation (4.5) further simplifies to

$$p_{12}^+(L) = -\frac{1}{2} V_0 [n(-L/2) + n(L/2)] = -V_0 n(L/2) \quad (4.6)$$

where we used the symmetry that  $n(z) = n(-z)$ . Recalling that the charge density at a semi-infinite jellium undergoes characteristic Friedel oscillation at the surface [25, 26], it is tempting to suppose that the total force  $p^+(L)$  in equation (4.6) should oscillate as a function of  $L$ . In short, the oscillations originating at one interface reach the other with a phase that varies with  $L$ . As we shall now demonstrate, this effect gives rise to a variety of interesting oscillatory phenomena.

Indeed, coming back to the theory of section 2, from equation (2.13b) putting  $\xi_1 = -1/2$  and  $\xi_2 = 1/2$ , since  $f_1 + f_2 = 0$ , we obtain

$$p_{12}^+(L) = -f_1(L) + (p_B - p_A) = f_2(L) + (p_B - p_A). \quad (4.7)$$

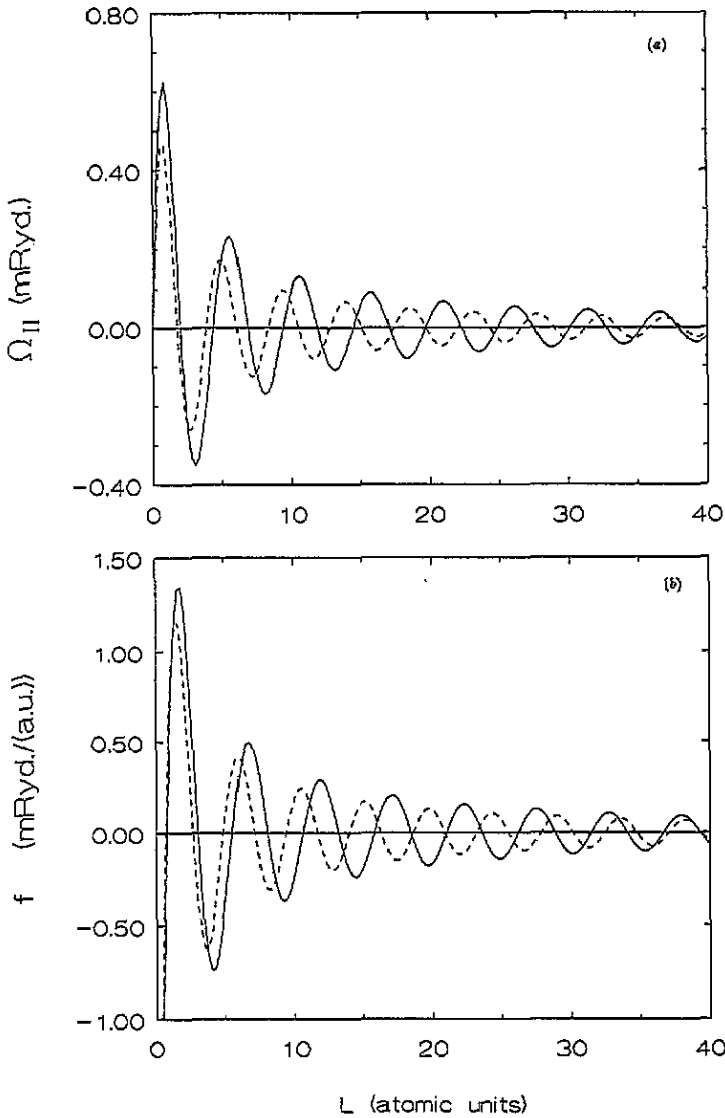
Therefore, we can distinguish in  $p^+(L)$  an  $L$ -independent contribution, which is the difference between the pressures of the B and A bulk metals, and an  $L$ -dependent term, the interface-interface force, which is, thus, responsible for the oscillations.

#### 4.2. Oscillatory contributions to the excess grand potential

We have evaluated the formula for  $\Omega_{\text{exc}}(L)$  given in equations (3.8), for a one-dimensional, and (3.24), for the three-dimensional non-interacting jellium models. The results are shown in figures 4(a) and 5(a) as functions of  $L$ . To emphasize their principal feature, namely the oscillations, we have subtracted from each curve a constant background, which, according to equation (2.16), is proportional to the sum of the surface tensions when  $L \rightarrow \infty$ .

Evidently the above  $L$ -dependent contribution can be regarded as the interface-interface interaction grand potential  $\Omega_{\Pi}(L)$  given in equation (2.16). Indeed, if we evaluate  $\Omega_{\Pi}(L)$  using

$$\Omega_{\Pi}(L) = A \int_{-\infty}^L d\lambda f_1(\lambda) \quad (4.8)$$



**Figure 4.** (a) Interface-interface interactions  $\Omega_{II}(L)$  for the 1D model with  $V_0 = -0.08$  Ryd and spacer Fermi energies  $E_F = 0.36$  Ryd (full curve) and  $E_F = 0.46$  Ryd (broken curve). The abscissa shows the spacer thickness  $L$ . (b) The  $\Pi$  force  $f$  for the same systems as in (a). All the quantities are plotted in atomic units.

and the formulae for the  $\Pi$  forces, given in equations (4.6) and (4.7), we find perfect agreement with the above direct calculations using the appropriate forms of the Friedel sum. This demonstrates that, as expected, the oscillatory contribution in the grand potential is due to the  $\Pi$  force  $f(L)$ . These are displayed in figures 4(b) and 5(b).

To sum up, the interaction grand potential, as well as the force between the interfaces,  $p^+(L)$  or  $f(L)$ , oscillates because the charge densities at the interface  $n(L/2) = n(-L/2)$  oscillate. The origin of the latter is the same as that of Friedel

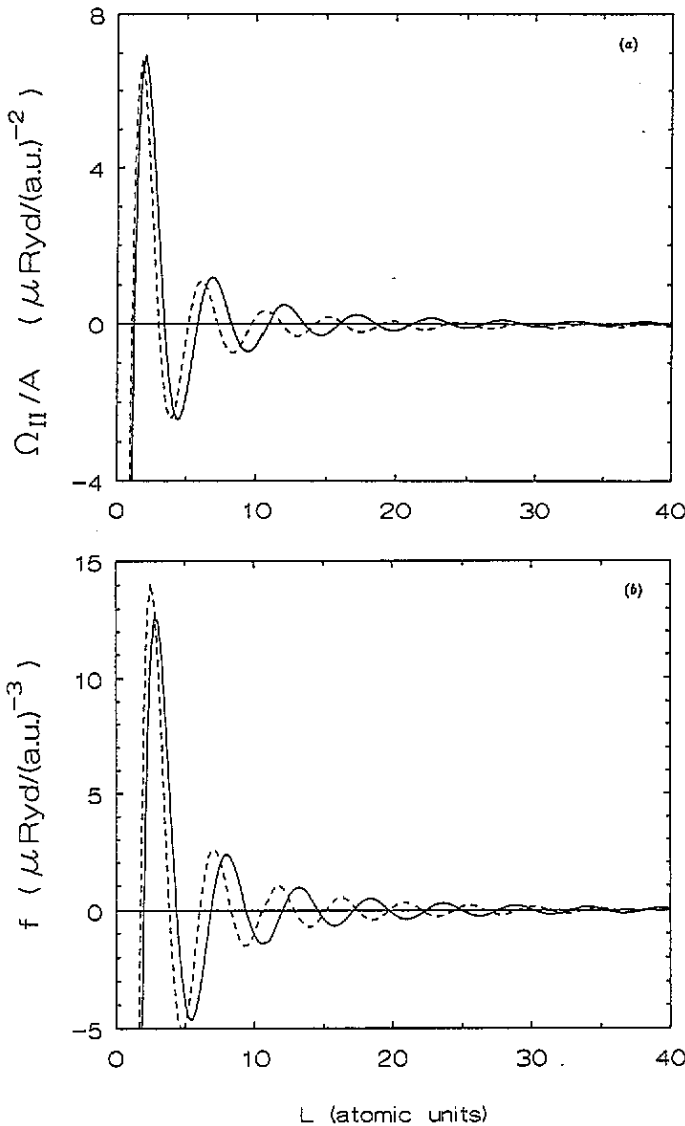


Figure 5. (a) Interface-interface interactions per unit area  $\Omega_{II}(L)/A$  for the inhomogeneous jellium model with  $V_0 = -0.08$  Ryd and  $E_F = 0.36$  Ryd (full curve) and  $E_F = 0.46$  Ryd (broken curve). The abscissa shows the spacer thickness  $L$ . (b) The  $\parallel$  force  $f$  for the same multilayers as in (a). Observe the faster decrease of both the grand potential excess and the  $\parallel$  force with respect to the 1D model (figure 4).

oscillations of charge around an impurity. To make this point quite explicitly, we studied the asymptotic forms of the integral representations

$$n^{1D}(L/2) = \frac{1}{\pi} \int_0^{k_F} dk [\cos^2(kL/2 + \delta_0) + \cos^2(kL/2 - \pi/2 + \delta_1)] \quad (4.9a)$$

$$n^{3D}(L/2) = \frac{1}{4\pi^2} \int_0^{k_F} dk (k_F^2 - k^2) [\cos^2(kL/2 + \delta_0) + \cos^2(kL/2 - \pi/2 + \delta_1)]$$

(4.9b)

that follow from equations (3.4) and (3.19). For the sake of clarity, here and in the following subsection, we measure the energy from the spacer potential level  $V_0$ . The asymptotic behaviours of equations (4.9) are dominated by the presence of an upper limit of integration,  $k_F$ . We omit the algebra, which follows closely the straightforward method by Lighthill [29], and merely quote the results for the  $L$ -dependent parts of  $n^{1D}(L/2)$  and  $n^{3D}(L/2)$ :

$$\Delta n^{1D}(L/2) = \frac{1}{\pi L} \sum_l (-1)^l \sin \delta_l(E_F) \cos \left( 2k_F \frac{L}{2} + \delta_l(E_F) \right) + O(L^{-2}) \quad (4.10a)$$

$$\Delta n^{3D}(L/2) = \frac{k_F}{\pi^2 L^2} \sum_l (-1)^l \sin \delta_l(E_F) \sin \left( 2k_F \frac{L}{2} + \delta_l(E_F) \right) + O(L^{-3}). \quad (4.10b)$$

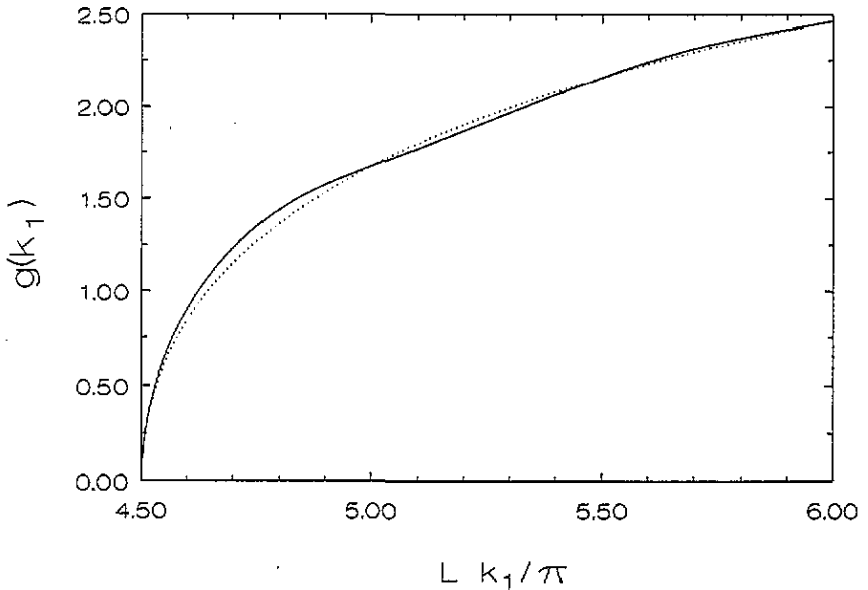
Allowing for the difference between the geometries of planar interfaces and point defects respectively, these charge densities are very similar to those one finds in standard discussions [30] of Friedel oscillations about impurities in metals. The main difference is the  $L$  dependence of the phase shifts, i.e. the 'size' of a defect, which is not a point. This connection clarifies that the origin of the oscillations is the nature of the Fermi sea; namely, the existence of a Fermi surface that separates occupied and unoccupied plane-wave states. That is to say, they are a fundamental property of the metallic state. An interesting version of similar oscillations in semiconductors has been studied by Cheng *et al* [31].

Given the above compelling simple picture of how the oscillations arise, it is surprising to find that there is an alternative, equally simple, description of them. For clarity we shall deal with the three-dimensional case only. Using the result, derived in appendix 3, that for our model the Friedel sum is given by

$$\delta_0(E) + \delta_1(E) = -L\sqrt{(E + V_0)} + \tan^{-1} \left[ \left( \frac{\sqrt{E}}{2\sqrt{(E + V_0)}} + \frac{\sqrt{(E + V_0)}}{2\sqrt{E}} \right) \tan(L\sqrt{E}) \right] + m\pi \quad (4.11)$$

we note that the integrand in equation (3.23) oscillates about the difference between the free-electron integrated density of states of the two metals  $N_B(E) - N_A(E)$ . Following the argument in appendix 3, the root cause of these oscillations is simply the resonant increase of the phase shifts,  $\delta_0(k)$  and  $\delta_1(k)$ , through the resonant energies given by  $kL = (n + 1/2)\pi$ . These moving ahead and then falling behind at each resonant energy, as shown in figure 6, can be identified as the cause of the oscillatory contribution  $\Omega_{\Pi}(L)$ . Although this explanation appears to be quite different from our previous arguments, which involved the interface–interface forces, clearly, it describes the same effect. For clarity and latter reference we add that, in order to have visible differences, in figure 6 we used a potential well with a step more than one order of magnitude larger than any realistic one. The point is that interface–interface forces are a subtle effect coming from tiny differences from the bulk behaviour. That is to say, their energy scale is much smaller than the scale of bulk energies and even of the surface tensions in the  $L \rightarrow \infty$  limit.

In the next subsection, where we shall study a magnetic version of the present model, the above two faces of the same coin will turn out to be very useful in making contact with results by other authors on the same subject.



**Figure 6.** Plot of the Friedel sum  $g(k_1) = [\delta_0(k_1) + \delta_1(k_1) - \delta_0(k_1 = 0) - \delta_1(k_1 = 0)]/\pi$  (full curve) versus  $Lk_1/\pi$ , where  $k_1 = \sqrt{E - V_0}$  is the spacer momentum and  $L$  the separation between the interfaces, for a non-magnetic monolayer with two interfaces.  $V_0$  is chosen to be 2 Ryd, the separation between the interfaces is  $L = 10$  au. The dotted curve represents the bulk contribution  $[f(k_1) - f(k_1 = 0)]/\pi$  after equation (A3.5). Note that in order to have a visible difference between the two curves we have chosen a very high value for  $V_0$ .

Before moving on to discussions of magnetic models of experimental interest, we wish to make one final remark concerning the interface–interface force point of view developed in this section. Because  $p^+(L)$  is actually a uniaxial pressure in the spacer layer, it is reasonable to assume that an oscillating pressure will lead to systematic variation with  $L$  of the lattice parameter  $a$ . Namely we expect that the strain,  $\Delta a/a$ , will oscillate according to the formula

$$\Delta a/a = [1/(3B)]V_0n(L/2) \tag{4.12}$$

where  $B$  is the uniaxial elastic (bulk) modulus. In the next subsection we shall give quantitative estimates of this effect.

### 4.3. Magnetic coupling across a non-magnetic spacer layer

Interestingly, some useful insights into the origin of magnetic coupling in metallic multilayers can be obtained from a simple modification of the above calculations. We imagine that, in the magnetic layers, the majority and minority electrons feel different, exchange-split potentials, while in the non-magnetic spacer layer  $B$  they move in the same potential  $V_0$  as depicted in figure 7. Namely,

$$V_1(z) = \begin{cases} -\Delta/2 \\ V_0 \end{cases} \quad V_1(z) = \begin{cases} \Delta/2 & \text{for } -t < z < -L/2, L/2 < z < t \\ V_0 & \text{for } -L/2 < z < L/2 \end{cases} \tag{4.13}$$

where  $\Delta$  is the magnetic exchange splitting of the metal A and  $V_0$  is the ‘spin-averaged jump’, which can be assumed equal to the difference between the work functions of A and B ( $W_A - W_B$ ).



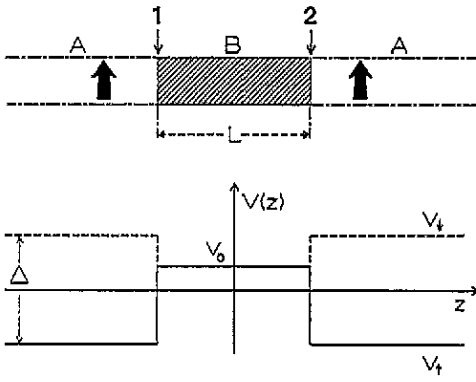


Figure 7. Model for a two-interface magnetic metallic multilayer, in the configuration in which the magnetic moments are parallel. The interfaces are marked by 1 and 2 and located at  $z = \pm L/2$ .  $V_1(z)$  (full line) and  $V_1(z)$  (broken line) have the same meaning as in figure 2,  $\Delta$  is the magnetic exchange splitting of A. Note that both the potentials have the common value  $V_0$  within the non-magnetic layer B.

In a one-electron theory, the grand potential of the multilayer is simply the sum of those of the two non-interacting subsystems of spin-up and spin-down electrons:

$$\Omega = \Omega_{\uparrow} + \Omega_{\downarrow}. \tag{4.14}$$

Each subsystem is described by the potential well in equation (3.1) and we can use all the results of the previous sections. In particular, from equations (4.6) and (4.7) we obtain that the  $\Pi$  forces, forgetting about non-oscillatory terms, are given by

$$\begin{aligned} f_1 = -f_2 &= (1/A)(\partial\Omega/\partial L) \simeq (V_0 - \Delta/2)n_{\uparrow}(L/2) + (V_0 + \Delta/2)n_{\downarrow}(L/2) \\ &= V_0 n(L/2) - (\Delta/2)m(L/2) \end{aligned} \tag{4.15}$$

where  $n_{\uparrow(\downarrow)}(L/2)$  is the spin-up (down) electronic density at the interfaces and

$$n(L/2) = n_{\uparrow}(L/2) + n_{\downarrow}(L/2) \tag{4.16}$$

$$m(L/2) = n_{\uparrow}(L/2) - n_{\downarrow}(L/2). \tag{4.17}$$

Integrating the force in equation (4.15) over  $L$ , we obtain

$$\Omega_{\Pi}(L) = \Omega_{\Pi}^{(es)}(L) + \Omega_{\Pi}^{(m)}(L) \tag{4.18a}$$

where we have distinguished charge (es) and magnetic (m) contributions to be calculated using

$$\frac{1}{A}\Omega_{\Pi}^{(es)}(L) = V_0 \int_{-\infty}^L d\lambda n(\lambda/2) \tag{4.18b}$$

$$\frac{1}{A}\Omega_{\Pi}^{(m)}(L) = -\frac{\Delta}{2} \int_{-\infty}^L d\lambda m(\lambda/2). \tag{4.18c}$$

The argument for this, rather natural, separation of what appear to be electrostatic and magnetic contributions, and which is illustrated in figure 8, becomes even more compelling if we realize that, in the limit where the interfaces are infinitely far apart, equation (4.18c) agrees with the conventional definition of magnetic coupling [11] for the geometry considered here. The latter is the difference between the grand potential in the configuration in which the magnetic moments of the adjacent magnetic layers are parallel ( $\uparrow\uparrow$ ) as in figure 7 and that in which they are antiparallel ( $\uparrow\downarrow$ ) as in figure 9. Namely, it is defined to be [11]

$$AJ = \Omega_{(\uparrow\uparrow)} - \Omega_{(\uparrow\downarrow)}.$$

The important point we wish to make is that, as shown in appendix 5, the difference

$$2\Omega_{\text{II}}^{(m)}(L) - [\Omega_{(\uparrow\uparrow)}(L) - \Omega_{(\uparrow\downarrow)}(L)] \tag{4.19}$$

vanishes exponentially as the spacer thickness increases. Moreover, we note that this difference is exactly zero in the special cases in which  $\Delta = 0$  or  $V_0 = 0$ . Note that, while for transition-metal pairs  $V_0$  is small ( $\lesssim 1$  eV), for transition-metal/noble-metal pairs it is comparable with  $\Delta$  [32].

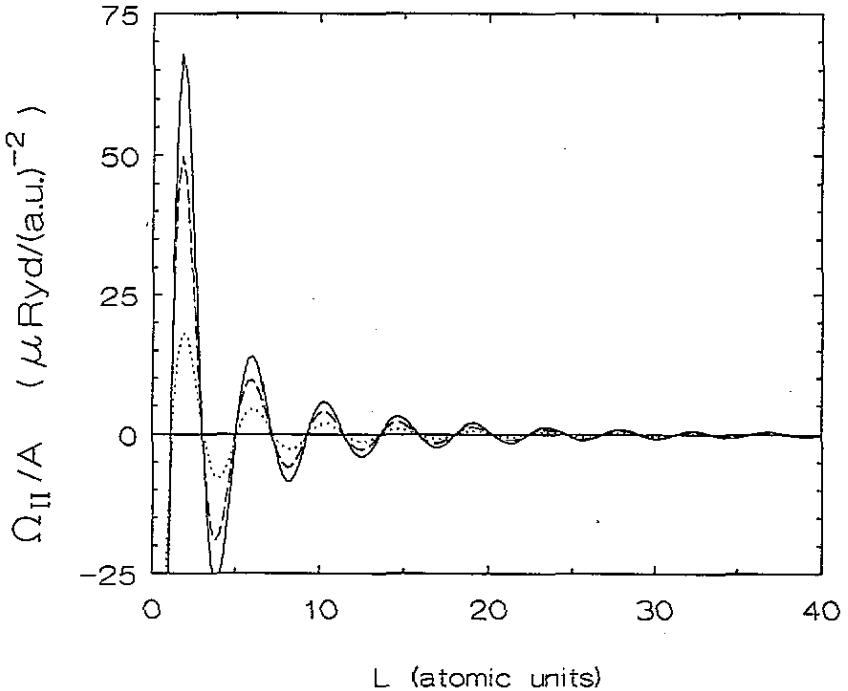


Figure 8. According to equation (4.18), the total III per unit area  $\Omega_{\text{II}}(L)/A$  (full curve) is decomposed into electrostatic  $\Omega_{\text{II}}^{(es)}(L)/A$  (broken curve) and magnetic  $\Omega_{\text{II}}^{(m)}(L)/A$  (dotted curve) contributions, for the inhomogeneous jellium magnetic model, with  $V_0 = -0.22$  Ryd,  $\Delta = 0.16$  Ryd, spacer  $E_F = 0.50$  Ryd. The abscissa shows the spacer thickness  $L$ .

Thus  $\Omega_{\text{II}}^{(m)}$  in equation (4.18c) describes an oscillatory coupling between two semi-infinite magnetic metals separated by a non-magnetic spacer layer. The spatial periodicity of this coupling is  $k_F^{-1}$ , which is not what is observed in experiments on magnetic multilayers where  $k_F^{-1} \simeq a$ , the lattice parameter. However, this discrepancy is a simple artifact of our model, which replaces the ions by uniform positively charged backgrounds. In short, we suggest that equation (4.18) correctly identifies the mechanism of oscillatory magnetic coupling, and if implemented for a more realistic lattice model it would give oscillations of the observed period [13]. Moreover, we conclude that in general there will be both electrostatic and magnetic forces communicated across the spacer layers and they both oscillate.

Although the above model is very crude, it has a number of interesting quantitative consequences. Using the work-function difference  $W_{\text{Fe}} - W_{\text{Cu}} \simeq 0$  [32] and the

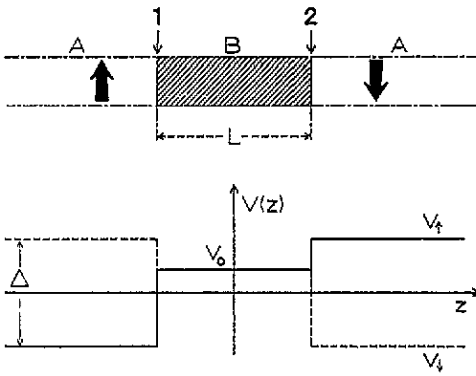


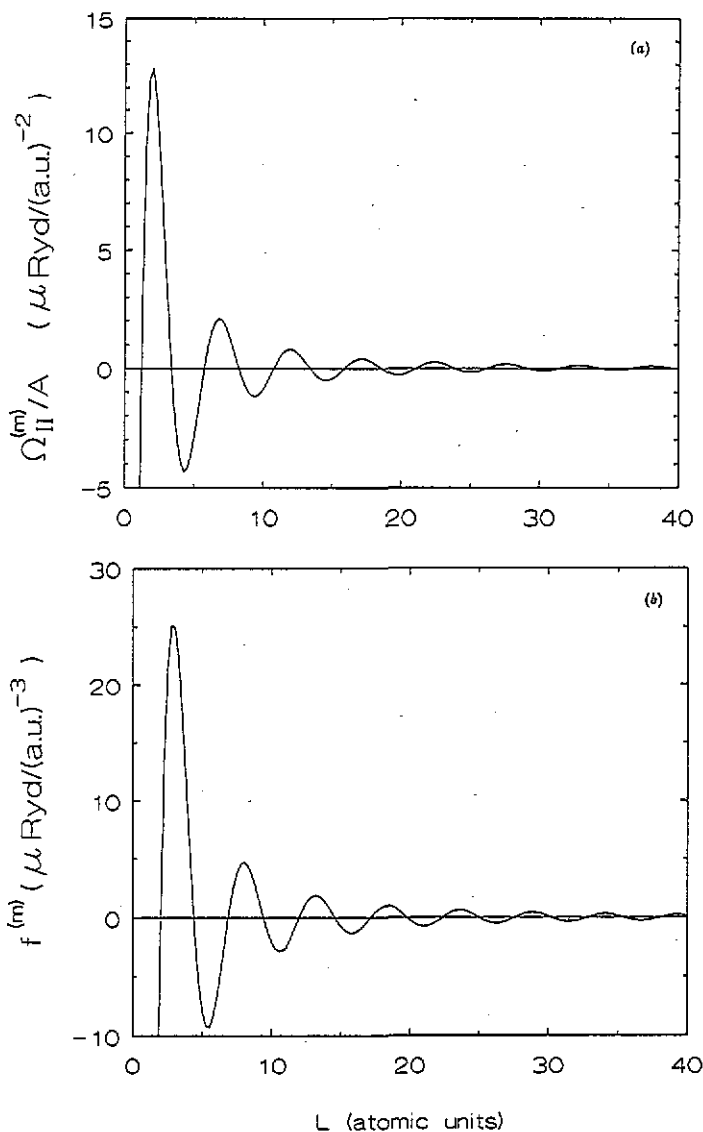
Figure 9. Model for a two-interface magnetic metallic multilayer, in the configuration in which the magnetic moments are antiparallel (11). The interfaces are marked by 1 and 2.  $V_1(z)$  (full line),  $V_2(z)$  (broken line),  $V_0$  and  $\Delta$  have the same meanings as in figure 2.

exchange splitting of Fe ( $\Delta \simeq 2$  eV) we have calculated  $\Omega_{\Pi}^{(m)}$  and the  $\Pi$  force for the  $(\text{Fe})_m(\text{Cu})_n$  system, studied by Heinrich *et al* [8]. The results are shown in figure 10. Surprisingly, for a separation of about 10 monolayers (or 26.5 au) we obtain a peak in  $\Omega_{\Pi}^{(m)}/A$  whose height is  $\simeq 0.12$  erg  $\text{cm}^{-2}$ , which is to be compared with the experimental value [8] of  $0.22$  erg  $\text{cm}^{-2}$ . The encouraging results emboldened us to estimate the total force, which we find to be  $f \simeq 6 \times 10^7$  erg  $\text{cm}^{-3}$ , and the strain  $\Delta a/a$ . This latter works out to be  $2 \times 10^{-5}$ . Much larger effects should occur at smaller separations; for instance, at 3.4 au we find  $f \simeq 1.7 \times 10^{-5}$  Ryd  $\text{au}^{-2}$ , which corresponds to  $\Delta a/a \simeq 6 \times 10^{-4}$ ; such a large deformation should be observable in high-quality x-ray measurements.

## 5. Conclusions and comments

We have introduced the idea of interface–interface forces to describe that part of the grand potential of a multilayer system which depends on the layer thicknesses. We showed that, just as the Friedel oscillations of charge and magnetization around an impurity give rise to Ruderman–Kittel–Kasuya–Yoshida (RKKY) forces that oscillate as a function of the spatial separation between the impurities, the well known [16, 26] analogous charge and magnetization oscillations due to surfaces and interfaces give rise to oscillatory coupling between interfaces. We demonstrated by explicit calculations for the two interfaces marked by 1 and 2 in figure 3 that the oscillatory contribution to the total free energy comes from  $\Omega_{\Pi}(L)$ , in equation (2.16), which is, as shown by equation (2.15a), the work done against the interface–interface force  $f(L)$  in bringing the interfaces 1 and 2 together from infinite separation ( $L = \infty$ ) to a finite distance  $L$ .

As shown in section 4.2 the above view leads naturally to an explanation for the oscillatory magnetic coupling in systems of alternating magnetic and non-magnetic layers. Although the physical mechanism of this interaction is the same as has been identified in the theories of Edwards and Mathon [3, 11] or in that of Coehoorn [13], the present formalism highlights a feature of the problem that has not been commented upon before. Namely, it makes it clear that, in general, the oscillatory magnetic coupling is accompanied by similar oscillatory electrostatic interactions and that the two together will lead to uniaxial strains that oscillate with the thicknesses of the spacer layers. We estimated the amplitude of these oscillation to be observable:  $\Delta a/a \simeq 2 \times 10^{-5}$  in the Cu spacer layer of the  $(\text{Cu})_m(\text{Fe})_n$  multilayer system. Evidently, observations of this



**Figure 10.** (a) Magnetic II per unit area  $\Omega_{II}^{(m)}(L)/A$  plotted versus the spacer thickness  $L$  for the inhomogeneous jellium model of Fe/Cu multilayer ( $V_0 = 0$ ,  $\Delta = 0.16$  Ryd, spacer  $E_F = 0.36$  Ryd). Note that, because the 'averaged jump' is actually zero, according to equation (4.18) there is now no electrostatic contribution to the II interactions and forces. (b) The II force  $f$  versus spacer thickness  $L$  for the same Fe/Cu model as in (a).

effect would be a strong confirmation of the general mechanism described in this paper, and therefore it is very tempting to interpret the recent experimental results by Kyuno *et al* [33] as evidence for such strain oscillations. It is also very interesting to observe here that the strains in Si-C polytypes were explained by Cheng *et al* [31] in terms of quantum forces between the atoms of the chain, in a manner very similar to the II forces in the above discussion. Remarkably these forces are due, in part, to the 'remnant' of Friedel oscillations that is known to occur in semiconductors [34].

In addition to the above brief summary of the main results, a number of comments are in order:

(i) The forces defined in equation (2.18) are a simple example of the more general quantum-mechanical stress tensor discussed by Nielsen and Martin [35]. In this connection we observe that the calculation of these forces and their integration, as in equation (2.15), to find the interface–interface grand potential  $\Omega_{\Pi}$  is numerically much more stable than the direct evaluation of  $\Omega$ , because of the different scales of energies that are involved, as we remarked towards the end of section 4.2. This suggests that in realistic, first-principles calculations for multilayer systems, the present interface–interface approach may have considerable advantages over the standard total energy methods [21].

(ii) Another point of general interest concerns the insensitivity of the oscillatory results to the shape of the interface potential  $V(z)$  in equation (2.18). As shown in appendix 6 the force  $(d\Omega/dL)/A$  depends only on the wavefunction and its derivative at the geometrical centre of the interface,  $\phi(0)$  and  $\phi'(0)$ , regardless of the functional dependence of the potential on  $z$ . This feature of the results suggests that the experimentally unavoidable roughness of the interfaces need not have deleterious effects on the oscillatory coupling between them, provided they remain reasonably well localized.

(iii) Somewhat similar remarks apply to the consequences of taking into account the electron–electron interactions. As is well known charge oscillations at the surfaces of a jellium model are generic features of a fully interacting electronic system [16, 26]. The other central result on which our arguments were based, namely the Friedel sum, also remains valid in a strongly interacting electron liquid [25]. Indeed, following the arguments leading to the local Fermi liquid theory of Nozières [36], it is easy to envision that the oscillatory coupling we have discussed would be generic properties of the metallic state.

(iv) Finally we must refer to the problem of generalizing the approach in order to treat proper atomistic models. Evidently the conceptual framework of the interface–interface forces can be readily developed in the language of density-functional theory and their calculation may be implemented within the local-density approximation using ‘band-theory’ methods such as Korringa–Kohn–Rostoker (KKR), linear muffin-tin orbitals (LMTO), etc. We hope to present such calculations in the not-too-distant future. However, there is a qualitative new feature of this model that may be mentioned here. This arises because the interface–interface distance  $L$  can change only in steps governed by the addition of monolayers. As is clear from figure 5 for the inhomogeneous jellium model, the periodicity of the  $\Pi$  interactions is  $k_F^{-1}$  and this is inconsistent with the much larger periods ( $\simeq 12 \text{ \AA}$ ) observed in the experiments [3, 8]. However, the entry of step size, the lattice parameter  $a$ , into the problem suggests a simple way out of the dilemma. By evaluating the rapidly varying function  $\Omega_{\Pi}(L)$  in equation (4.1) at discrete distances  $L = na$ , where  $a$  is incommensurate with  $k_F^{-1}$ , it returns to its former value only after a change  $\Delta L \simeq (a^{-1} - k_F)^{-1}$ , which can easily be as large as  $5a$  or  $10a$ . This argument has been developed in quantitative details by Coehoorn [13] and hence we need not dwell on it further.

### Acknowledgments

We would like to acknowledge some helpful discussions with Professor R Evans. This work was supported by the British SERC (Grant G/50780).

### Appendix 1. The derivative of the grand potential with respect to the external potential

In the context of density-functional theory we can evaluate the functional derivative of the grand potential with respect to the external potential  $V(\mathbf{r})$  as follows:

$$\frac{\delta\Omega}{\delta V(\mathbf{r})} = \left( \frac{\delta\Omega}{\delta V(\mathbf{r})} \right)_{n(\mathbf{r})=\bar{n}(\mathbf{r})} + \int d\mathbf{r}' \frac{\delta\Omega}{\delta n(\mathbf{r}')} \frac{\delta n(\mathbf{r}')}{\delta V(\mathbf{r})} \quad (\text{A1.1})$$

where the first derivative on the RHS is evaluated keeping the charge density fixed at the value  $\bar{n}(\mathbf{r})$  for which the unique functional  $\Omega[n]$  is minimum. Corresponding to this minimum

$$\delta\Omega/\delta n(\mathbf{r}) = 0$$

and hence the integral on the RHS of (A1.1) vanishes. Now we note that

$$\Omega = \int d\mathbf{r} [V(\mathbf{r}) - \mu]n(\mathbf{r}) + F[n(\mathbf{r})] \quad (\text{A1.2})$$

where  $F[n]$  is a universal functional of  $n(\mathbf{r})$ . Thus

$$\delta\Omega/\delta V(\mathbf{r}) = n(\mathbf{r}) \quad (\text{A1.3})$$

as in classical density-functional theories for liquids [37].

### Appendix 2. Some exact results on one-dimensional wavefunctions

These are the one-dimensional analogues of those by Friedel [22]. In short, we shall consider two theorems satisfied by any solution  $\Phi_E(x)$  of the 1D Schrödinger equation

$$-\frac{d^2}{dx^2}\Phi_E(x) = [E - V(x)]\Phi_E(x) \quad (\text{A2.1})$$

with a potential satisfying the muffin-tin condition

$$V(x) = 0 \quad \text{for } |x| > x_{\text{MT}}. \quad (\text{A2.2})$$

It will be convenient if we think of  $\Phi_E(x)$  as given by the linear combination [23] of its symmetric,  $R_0(x, E)$ , and antisymmetric,  $R_1(x, E)$ , parts as follows:

$$\Phi_E(x) = \sum_l A_l(E) R_l(x, E) Y_l(x) \quad (\text{A2.3})$$

where

$$R_l(x, E) = \cos[\sqrt{E}|x| - l\pi/2 + \delta_l(E)] \quad (\text{A2.4})$$

$$Y_l(x) = (1/\sqrt{2})(\text{sgn } x)^l \quad (\text{A2.5})$$

and  $\delta_l(E)$  are the phase shifts.

**Theorem A2.1.** Let  $\Phi_E(x)$  be any real eigenfunction of the Schrödinger equation (A2.1) whose eigenvalue  $E$  lies in the continuum spectrum. It follows (here and in the following we denote with a prime the differentiation with respect to  $x$ ) that

$$\int_a^b dx \Phi_E^2(x) = - \left[ \Phi_E^2(x) \frac{d}{dE} \frac{\Phi'_E(x)}{\Phi_E(x)} \right]_a^b. \quad (\text{A2.6})$$

*Proof.* Consider the solutions corresponding to the two eigenvalues  $E \pm \Delta E$

$$- \Phi''_{E \pm \Delta E}(x) = [E \pm \Delta E - V(x)] \Phi_{E \pm \Delta E}(x).$$

Multiplying the equation corresponding to  $E + \Delta E$  by  $\Phi_{E - \Delta E}(x)$  and that corresponding to  $E - \Delta E$  by  $\Phi_{E + \Delta E}(x)$ , subtracting one from the other, and integrating from  $a$  to  $b$  one has

$$\begin{aligned} - \int_a^b dx [\Phi_{E - \Delta E}(x) \Phi''_{E + \Delta E}(x) - \Phi_{E + \Delta E}(x) \Phi''_{E - \Delta E}(x)] \\ = 2\Delta E \int_a^b dx \Phi_{E + \Delta E}(x) \Phi_{E - \Delta E}(x). \end{aligned} \quad (\text{A2.7})$$

The theorem follows on integrating by parts the LHS, then expanding in Taylor series around  $E$ , dividing by  $\Delta E$  and taking the  $\Delta E \rightarrow 0$  limit.

**Theorem A2.2.** For any real solution

$$\Phi_{E,l}(x) = A_l(E) R_l(x, E) Y_l(x) \quad \text{for } |x| > x_{\text{MT}}$$

belonging to the continuum spectrum of the 1D Schrödinger equation (A2.1) with a potential satisfying the muffin-tin condition (A2.2), there exists an infinite number of lengths

$$t_m = [m\pi - 2\delta_l(E)] / (2\sqrt{E}) > x_{\text{MT}} \quad (\text{A2.8})$$

for which

$$\int_{-t_m}^{t_m} dx \Phi_{E,l}^2(x) = A_l^2(E) \left( \frac{t_m}{2} + \sqrt{E} \frac{d}{dE} \delta_l(E) \right). \quad (\text{A2.9})$$

*Proof.* Using theorem A2.1, equations (A2.4) and (A2.5), if  $t_m > x_{\text{MT}}$ , we have

$$\int_{-t_m}^{t_m} dx \Phi_{E,l}^2(x) = A_l^2(E) \left( \frac{t_m}{2} + \sqrt{E} \frac{d}{dE} \delta_l(E) + \frac{\sin[2t_m - l\pi + 2\delta_l(E)]}{4\sqrt{E}} \right)$$

and then if we chose  $t_m$  as in (A2.8) the sine term vanishes and the theorem follows.

### Appendix 3. The one-dimensional phase shifts

Analytical expressions for the one-dimensional phase shifts for the potential well (3.1) can be obtained by standard methods [24]. We merely quote here the results

$$e^{2i\delta_0} = a/(1 - ib) \quad e^{2i\delta_1} = a/(1 + ib) \quad (\text{A3.1})$$

where

$$\begin{aligned} a &= \frac{1}{2}e^{-ik_0L} [2 \cos(k_1L) + i\alpha \sin(k_1L)] & b &= \frac{1}{2}\beta \sin(k_1L) \\ \alpha &= k_1/k_0 + k_0/k_1 & \beta &= k_1/k_0 - k_0/k_1 \\ k_0 &= \sqrt{E} & k_1 &= \sqrt{(E - V_0)}. \end{aligned}$$

The expressions for the phase shifts are completely determined by the requirement of continuity and by the additional conditions that [23]

$$\lim_{E \rightarrow 0} \delta_0(E) = 0 \quad \lim_{E \rightarrow 0} \delta_1(E) = \pi/2 \quad (\text{A3.2a})$$

for  $V_0 > 0$  or, for  $V_0 < 0$ ,

$$\lim_{E \rightarrow 0} \delta_0(E) = 0 \quad \lim_{\substack{E \rightarrow 0 \\ V_0 \rightarrow 0}} \delta_1(E) = \pi/2. \quad (\text{A3.2b})$$

Furthermore, some straightforward algebraic manipulations yield the following form of the Friedel sum:

$$\delta_0(E) + \delta_1(E) = -k_0L + \tan^{-1}[\frac{1}{2}\alpha \tan(k_1L)] + m\pi/2 \quad (\text{A3.3})$$

where the integers  $m$  are completely determined by (A3.2) and the continuity condition. The behaviour of the Friedel sum is mainly determined by the jumps of  $\tan(k_1L)$ . These occur when

$$k_1L = (n + 1/2)\pi. \quad (\text{A3.4})$$

The important thing to note is that, owing to the continuity conditions, when (A3.4) is satisfied  $m$  in (A3.3) must be incremented by 1. Consequently, the Friedel sum oscillates around the function

$$f(E) = (k_1 - k_0)L + (m_0 + 1/2)\pi \quad (\text{A3.5})$$

( $m_0$  is an integer to be chosen according to the conditions (A3.2)) crossing it each time (A3.4) is satisfied. This behaviour is displayed in figure 6.

#### Appendix 4. An alternative derivation of equation (2.18) for the one-dimensional and the inhomogeneous jellium models

In section 2, using a very general procedure, we derived a relation, namely equation (2.18), that allows the practical calculation of  $\Pi$  forces, and, then, of  $\Pi$  interactions. We will show in this appendix that the same relation can also be obtained within the framework of the scattering theory, for both the 1D and the inhomogeneous jellium two-interface models, provided the external potential  $V(z)$  has finite range, i.e. satisfies the condition in equation (A2.2).

For the sake of simplicity, we suppose here that there are no states bounded in the  $z$  direction. The multilayer pressure, defined in equation (2.14), can be obtained by deriving equations (3.6) and (3.23) with respect to the spacer thickness  $L$ , for the 1D and the inhomogeneous jellium models, respectively. In fact, the homogeneous term  $\Omega_0$  does not depend on  $L$ , and, since in the present case the condition (A3.2a)



holds, the phase shifts at zero energy also do not depend on the spacer thickness and we obtain

$$p^+(L) = \frac{1}{\pi} \int_0^{E_F} dE \frac{\partial}{\partial L} \sum_l \delta_l(E) \quad (\text{A4.1})$$

for the 1D model, and

$$p^+(L) = -\frac{1}{4\pi^2} \int_0^{E_F} dE (E - E_F) \frac{\partial}{\partial L} \sum_l \delta_l(E) \quad (\text{A4.2})$$

for the inhomogeneous jellium. It is more convenient to rewrite the last equation as an integral over the momentum  $k = E^{1/2}$ , as follows:

$$p^+(L) = -\frac{1}{2\pi^2} \int_0^{E_F} dk (k^2 - k_F^2) \frac{\partial}{\partial L} \sum_l \delta_l(k^2). \quad (\text{A4.3})$$

In order to calculate the derivative of the phase shifts we use the explicit expression for the 1D  $t$  matrix given by Butler [23]

$$t_l = -\sqrt{E} e^{i\delta_l} \sin(\delta_l) \quad (\text{A4.4})$$

obtaining

$$\frac{\partial \delta_l}{\partial L} = \text{Im} \left( t_l \frac{\partial t_l^{-1}}{\partial L} \right) = \text{Im} \left( \int_0^\infty dz t_l \frac{\delta t_l^{-1}}{\delta V(z)} \frac{\partial V(z)}{\partial L} \right). \quad (\text{A4.5})$$

Now, we can use the expression of Johnson *et al* [38] to evaluate the functional derivative of  $t^{-1}$  with respect to the potential  $V(z)$ , namely

$$\frac{\delta t_l^{-1}}{\delta V(z)} = -Z_l^2(z, E) \quad (\text{A4.6})$$

or, using the explicit expression of  $Z_l(z, E)$  [24], for  $|z| > z_{MT}$ ,

$$\frac{\delta t_l^{-1}}{\delta V(z)} = \frac{R_l^2(z, E) Y_l^2(z)}{E \sin^2 \delta_l(E)} \quad (\text{A4.7})$$

and, then from (A4.5) and (A4.7)

$$\frac{\partial}{\partial L} \sum_l \delta_l = \sum_l \int_0^\infty dz \frac{R_l^2(z, E) Y_l^2(z)}{\sqrt{E}} \frac{\partial V(z)}{\partial L}. \quad (\text{A4.8})$$

Equation (2.18) can be obtained easily now. In the 1D case one has to divide both sides of (A4.8) by  $\pi$ , integrate over the energy up to the Fermi level and use the expression (3.4) for the charge density; for the inhomogeneous jellium it is necessary to put in (A4.8)  $E = k^2$ , multiply by the factor  $k(k_F^2 - k^2)/(2\pi)$ , integrate up to the Fermi momentum and use the expression (3.17) for the jellium charge density.

### Appendix 5. The magnetic coupling in the parallel and antiparallel configurations

In a quite general way the configurations of the two-interface models in which the magnetic moments of two adjacent layers are parallel ( $\uparrow\uparrow$ ) or antiparallel ( $\uparrow\downarrow$ ), as shown in figures 7 and 9, respectively, can be described by the potentials

$$V_{\uparrow}^{(\uparrow\uparrow)}(x) = V(x) - \Delta V^{(\uparrow\uparrow)}(x) \quad (\text{A5.1a})$$

$$V_{\downarrow}^{(\uparrow\uparrow)}(x) = V(x) + \Delta V^{(\uparrow\uparrow)}(x) \quad (\text{A5.1b})$$

$$V_{\uparrow}^{(\uparrow\downarrow)}(x) = V(x) - \Delta V^{(\uparrow\downarrow)}(x) \quad (\text{A5.1c})$$

$$V_{\downarrow}^{(\uparrow\downarrow)}(x) = V(x) + \Delta V^{(\uparrow\downarrow)}(x) \quad (\text{A5.1d})$$

$$\Delta V^{(\uparrow\uparrow)}(x) = f(x) \quad (\text{A5.1e})$$

$$\Delta V^{(\uparrow\downarrow)}(x) = f(x)[\Theta(-x) - \Theta(x)] \quad (\text{A5.1f})$$

where, for instance,  $V_{\uparrow}^{(\uparrow\downarrow)}(x)$  means the spin-up potential in the antiparallel configuration,  $V(z)$  is the average between up and down potentials (these must have the same average in both configurations) and  $f(x)$  is an even function. In order to obtain the potential wells defined in section 4.2, and plotted, respectively, in figures 7 and 9, we must put

$$V(x) = V_0\Theta(x + L/2)\Theta(L/2 - x) \quad (\text{A5.2})$$

$$f(x) = \frac{1}{2}\Delta[\Theta(-x - L/2) + \Theta(x - L/2)]. \quad (\text{A5.3})$$

We develop a perturbative theory in which the system with the averaged potential  $V(x)$  is the reference and  $\Delta V^{(\uparrow\uparrow)}(x)$  or  $\Delta V^{(\uparrow\downarrow)}(x)$  is the perturbation. In order to do this we start from the Lippman-Schwinger equation for the unperturbed Green function  $G$  [39]

$$G = G_0 + G_0 V G \quad (\text{A5.4})$$

( $G_0$  is, as usual, the free-electron Green function). After some straightforward algebra, it is possible to obtain an exact expression for  $\Delta G$ , the variation of the Green function when the external potential is increased by  $\Delta V$

$$\Delta G = (1 - G\Delta V)^{-1}G\Delta V G = \sum_{n=1}^{\infty} (G\Delta V)^n G. \quad (\text{A5.5})$$

From this equation we can write the expressions for the variations of both the spin-up and spin-down Green functions

$$\Delta G_{\uparrow} = \sum_{n=1}^{\infty} (-B)^n G \quad (\text{A5.6a})$$

$$\Delta G_{\downarrow} = \sum_{n=1}^{\infty} B^n G \quad (\text{A5.6b})$$

where  $B$  is  $G\Delta V^{(11)}$  or  $G\Delta V^{(1)}$ , depending on the configuration considered. From (A5.6), the following operators related to the charge density and the magnetization can be obtained:

$$\Delta N = \Delta G_{\uparrow} + \Delta G_{\downarrow} = G_{\uparrow} + G_{\downarrow} - 2G = 2 \sum_{n=1}^{\infty} B^{2n} G \quad (\text{A5.7})$$

$$M = \Delta G_{\uparrow} - \Delta G_{\downarrow} = G_{\uparrow} - G_{\downarrow} = -2 \sum_{n=0}^{\infty} B^{2n+1} G. \quad (\text{A5.8})$$

Using the force rule (4.5) as we did in section 4, we obtain for the potentials given by (A5.2) and (A5.3) the total  $\Pi$  force in the antiparallel configuration,

$$f^{(11)} = \frac{1}{2} V_0 [n^{(11)}(-L/2) + n^{(11)}(L/2)] - \frac{1}{4} \Delta [-m^{(11)}(-L/2) + m^{(11)}(L/2)]$$

where  $L$  is the separation between the interfaces. Now, because the symmetry requires  $n^{(11)}(z)$  to be an even function and  $m^{(11)}(z)$  odd,

$$f^{(11)} = V_0 n^{(11)}(L/2) - \frac{1}{2} \Delta m^{(11)}(L/2) \quad (\text{A5.9})$$

so, using (4.15), we have for the difference between the forces in the two configurations

$$f^{(11)} - f^{(1)} = V_0 [n^{(11)}(L/2) - n^{(1)}(L/2)] - \frac{1}{2} \Delta [m^{(11)}(L/2) - m^{(1)}(L/2)]. \quad (\text{A5.10})$$

In the  $L \rightarrow \infty$  limit  $n^{(11)}(L/2) = n^{(1)}(L/2)$  and  $m^{(11)}(L/2) = -m^{(1)}(L/2)$ , so that  $g(L/2)$  is zero. We have to study the asymptotic behaviour for large separations of  $g(L/2)$ . Let us start by considering the operator  $\Delta N^{(11)} - \Delta N^{(1)}$ . From (A5.7) we find that

$$\Delta N^{(11)} - \Delta N^{(1)} = 2 \sum_{n=1}^{\infty} [(G\Delta V^{(11)})^{2n} - (G\Delta V^{(1)})^{2n}] G. \quad (\text{A5.11})$$

The  $n$ th term on the RHS of equation (A5.11) is proportional to  $\Delta^{2n}$  and, at  $L/2$ , has the integral representation

$$\begin{aligned} & \int dx_1 \dots \int dx_{2n} G(L/2, x_1) [\Delta V^{(11)}(x_1) G(x_1, x_2) \Delta V^{(11)}(x_2) \dots G(x_{2n-1}, x_{2n}) \\ & \quad \times \Delta V^{(11)}(x_{2n}) - \Delta V^{(11)}(x_1) G(x_1, x_2) \Delta V^{(11)}(x_2) \dots G(x_{2n-1}, x_{2n}) \\ & \quad \times \Delta V^{(11)}(x_{2n})] G(x_{2n}, L/2). \end{aligned} \quad (\text{A5.12})$$

The effect of the operators  $\Delta V$  is to break up the domain of integration into two subsets, the intervals  $(-\infty, -L/2)$  and  $(L/2, +\infty)$ . From a careful analysis we see that only those integrals for which an odd number of  $x_i$  run in the interval  $(-\infty, -L/2)$  do not cancel each other. Thus each addendum involves at least two Green functions  $G(x_i, x_{i+1})$  with  $|x_{i+1} - x_i| > L$  throughout all the domain of integration, and, because

$$G(x_1, x_2) \simeq \exp(\sqrt{E}|x_1 - x_2|)$$

all the contribution to  $\Delta N^{(\uparrow\uparrow)} - \Delta N^{(\uparrow\downarrow)}$  must go to zero as  $\exp(-2\sqrt{E}L)$  when  $L \rightarrow \infty$ . Using the same analyses, we find that also the operator  $M^{(\uparrow\uparrow)} - M^{(\uparrow\downarrow)}$  has the above exponential asymptotic behaviour. Now, since  $g(L/2)$  is given by

$$g(L/2) = \int dE q(E, E_F) \left\{ V_0 [\Delta N^{(\uparrow\uparrow)}(L/2) - \Delta N^{(\uparrow\downarrow)}(L/2)] + \frac{1}{2} \Delta [M^{(\uparrow\uparrow)}(L/2) - M^{(\uparrow\downarrow)}(L/2)] \right\} \quad (\text{A5.13})$$

(the kernel  $q(E, E_F)$  has to be chosen according to the theory developed in section 3), it follows that  $g(L/2)$  must exhibit the same exponential decay, and from equation (A5.10) we have a relation involving the difference between the forces in the parallel and antiparallel configurations,

$$f^{(\uparrow\uparrow)}(L) - f^{(\uparrow\downarrow)}(L) + \Delta m^{(\uparrow\uparrow)}(L) \simeq \exp(-2\sqrt{E}L). \quad (\text{A5.14})$$

After integrating over  $L$  equation (A5.14), using equations (4.8) and (4.18c), we find finally

$$\Omega_{\text{II}}^{(\uparrow\uparrow)}(L) - \Omega_{\text{II}}^{(\uparrow\downarrow)}(L) - 2\Omega_{\text{II}}^{(m)}(L) \simeq \exp(-2\sqrt{E}L). \quad (\text{A5.15})$$

We note that even in the case of smooth potentials, like those described in appendix 6, for large enough separations, the operators  $\Delta V$  are zero in a region of length  $L - t$ ,  $t$  being the range of the potential, and then all the results quoted above remain valid if  $L$  is substituted by  $L - t$ .

#### Appendix 6. Interface-interface forces for two interfaces with smooth external potential

If the distance between the interfaces is large enough, the only effect of an increase of the separation  $L$  is a rigid shift of the potential, i.e.

$$V(x, y, z; L + \Delta L) = V(x, y, z - \text{sgn}(z) \cdot \Delta L/2; L). \quad (\text{A6.1})$$

In this case it is easy to show, expanding in Taylor series around  $(r; L)$ , that

$$\partial V(r)/\partial L = -\frac{1}{2} \text{sgn}(z) dV(r)/dz. \quad (\text{A6.2})$$

If we further assume that the potential depends only on  $z$  and is symmetric with respect to  $z = 0$ , from (2.19) and (3.18) it is possible to obtain a simple expression for the variation of the valence part of the grand potential. In fact, we have, for the three-dimensional jellium,

$$-\frac{1}{A} \frac{\partial \Omega_{\text{val}}}{\partial L} = \frac{1}{4\pi^2} \int_0^{k_F} dk (k^2 - k_F^2) \int_0^\infty dz |\Phi_k(z)|^2 V'(z). \quad (\text{A6.3})$$

The inner integral can be evaluated by differentiating with respect to  $z$  the Schrödinger equation (3.16), multiplying it by the wavefunction and integrating over  $z$  from 0 to  $\infty$ , obtaining, after integration by parts and reinsertion of the same Schrödinger equation,

$$\int_0^\infty dz |\Phi_k(z)|^2 V'(z) = [V(0) - k^2] |\Phi_k(0)|^2 - |\Phi'_k(0)|^2 \quad (\text{A6.4})$$

and then

$$-\frac{1}{A} \frac{\partial \Omega_{\text{val}}}{\partial L} = \frac{1}{4\pi^2} \int_0^{k_F} dk (k^2 - k_F^2) \{ [V(0) - k^2] |\Phi_k(0)|^2 - |\Phi'_k(0)|^2 \}. \quad (\text{A6.5})$$

*Note added in proof.* After we had submitted the present paper, we received a preprint by J Mathon, M Villeret and D M Edwards in which a formula for  $\partial\Omega/\partial L$  is derived and discussed for the 'sandwich' geometry. This quantity is proportional to the interface-interface interaction forces in our formalism (see equation (4.15)). The above paper has recently appeared [40].

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