Plasma Frequency of the Electron Gas in Layered Structures

D. Grecu*
Physics Division, Euratom, Ispra (Varese), Italy
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The approximations necessary to obtain the model of Visscher and Falicov for a layered electron gas are discussed. The Hamiltonian is generalized to include the electron tunneling between adjacent planes. The plasma frequency is calculated in the random-phase approximation using the equation-of-motion method. It shows a very strong anisotropy, depending on the angle between the wave vector of the plasma wave and the perpendicular direction to the layered structure.

I. INTRODUCTION

Recently, Visscher and Falicov[1] proposed a simple but very anisotropic model for an electron gas in a layered structure. The system consists of electrons constrained to move on parallel equally spaced planes, any motion in the perpendicular direction being completely forbidden. The static-screening properties have been calculated. It is the aim of this paper to make a detailed analysis of the approximations involved in this model and to calculate the plasma frequency of the system in the random-phase approximation (RPA).

The second-quantization formalism is used. The single-particle wave functions are given by the product of a plane wave, describing the free motion of an electron in the plane, and the exact solution of the Schrödinger equation with a periodic potential in the perpendicular direction. In order to obtain a localized picture, Wannier functions will be used. The very localized model of Visscher and Falicov is obtained in the tight-binding approximation when any overlap of the wave functions on neighboring sites is completely neglected. This analysis leads naturally to the introduction of an additional term in the Hamiltonian, which permits electron tunneling between adjacent planes. It is of the form used some years ago by different authors[2] to describe the electron tunneling between two metals separated by an insulating film, especially when one of the metals is in the superconducting state. Given this Hamiltonian, the plasma frequency is calculated in the RPA using the equation-of-motion method.[3] It shows a very strong anisotropy, depending on the angle between the perpendicular to the plane motion and the propagation direction of the plasma wave. It vanishes in the model of Visscher and Falicov when the propagation is along the perpendicular direction, but is finite everywhere when the tunneling is included, as is expected from the three-dimensional character of the motion.

II. MODEL HAMILTONIAN

Let us denote the exact single-particle wave function by $\psi_{\vec{K}}(\vec{R})$. It is given by

$$\psi_{\vec{K}}(\vec{R}) = \frac{1}{\sqrt{A}}\, e^{i\vec{k}\cdot\vec{r}}\, \varphi_{\kappa\nu}(z)\ . \tag{1}$$

The condensed notations $\vec{R} = (\vec{r}, z)$, $\vec{K} = (\vec{k}, \kappa)$ will be used throughout. Here $\vec{r}$ is the position variable in the plane, A is the area of the region in which the motion is confined, and $\vec{k}$ is the corresponding two-dimensional wave vector. The Bloch functions $\varphi_{\kappa\nu}(z)$, describing the electron motion in the perpendicular direction (z axis), are solutions of the Schrödinger equation with a periodic potential, written as a superposition of deep and narrow potential wells on each lattice site, $\sum_m U(z - mc)$ (c is the lattice constant in z direction). The eigenfunctions of an isolated potential well $U(z)$ are denoted by $\varphi_\nu(z)$. One will assume that $U(z)$ is arbitrarily deep and narrow, so that only the lowest band is occupied; consequently, the band index ν will be omitted.

In order to obtain a localized picture, we shall, instead of working with the Bloch states $\varphi_\kappa(z)$, use the Wannier functions defined by[4,5]

$$a(z - mc) = \frac{1}{\sqrt{N}} \sum_\kappa \varphi_\kappa(z)\, e^{-i\kappa mc}\ , \tag{2}$$

where N is the number of the planes in z direction, and the summation is restricted to the first Brillouin zone. In the Wannier representation the single-particle wave function becomes

$$\psi_{\vec{k}m}(\vec{R}) = \frac{1}{\sqrt{A}}\, e^{i\vec{k}\cdot\vec{r}}\, a(z - mc)\ . \tag{1'}$$

Now we can define the field operator

$$\Psi(\vec{R}) = \sum_{\vec{k}m} \psi_{\vec{k}m}(\vec{R})\, \alpha_{\vec{k}m}\ , \tag{3}$$

$\alpha_{\vec{k}m}$ ($\alpha^\dagger_{\vec{k}m}$) being the annihilation (creation) operator of an electron with the wave vector $\vec{k}$ localized mainly in the plane m. Then the single-particle part of the Hamiltonian becomes

$$\sum_{\vec{k}m} \frac{\hbar^2 k^2}{2m_e}\, \alpha^\dagger_{\vec{k}m}\, \alpha_{\vec{k}m} + \sum_{\vec{k}mm'} E_{|m-m'|}\, \alpha^\dagger_{\vec{k}m}\, \alpha_{\vec{k}m'}\ , \tag{4}$$

where $E_{|m-m'|}$ is the overlap integral,

$$E_{|m-m'|}=\int dz\, a^*(z-mc)\left(-\frac{\hbar^2}{2m_e}\frac{\partial^2}{\partial z^2}+\sum_{m_1} U(z-m_1c)\right)a(z-m'c)\,. \qquad (4')$$

Up to now these relations are exact. To go further one must make several approximations. The first is the tight-binding approximation in which the exact Bloch state is written as a superposition of eigenfunctions of the isolated potential wells,

$$\varphi_\kappa(z)\cong\frac{1}{\sqrt{N}}\sum_m e^{i\kappa mc}\,\varphi(z-mc)\,; \qquad (5)$$

then $a(z-mc)=\varphi(z-mc)$ as is easily seen from (2). Assuming a very small overlap between two wave functions φ in two neighboring lattice sites, the various overlap integrals (4′) have different orders of magnitude. We shall neglect all of them except the energy contribution E_0 of the lowest state of the isolated potential well and of the integral

$$T=\int dz\,\varphi^*(z)\,U(z)\,\varphi(z\pm c) \qquad (6)$$

(nearest-neighbor approximation). By a suitable choice of the energy scale we can take $E_0=0$ and then the single-particle Hamiltonian (4) becomes

$$\sum_{\xi m}\frac{\hbar^2k^2}{2m_e}\,\alpha^\dagger_{\xi m}\,\alpha_{\xi m}+T\sum_{\xi m}(\alpha^\dagger_{\xi m}\,\alpha_{\xi m-1}+\alpha^\dagger_{\xi m}\,\alpha_{\xi m+1})\,. \qquad (7)$$

The first term in the Hamiltonian (7) describes a system of free electrons in each plane, and the second term describes electrons which tunnel from one plane to its nearest-neighbor plane with the tunneling constant T. If T is also neglected, the very localized model of Visscher and Falicov is obtained.

The expression (7) can be diagonalized by the unitary transformation

$$\alpha_{\xi m}=\frac{1}{\sqrt{N}}\sum_\kappa e^{-i\kappa mc}\,\alpha_{\xi\kappa}\,, \qquad (8)$$

with the result

$$\sum_{\vec{k}}\xi_{\vec{k}}\,\alpha^\dagger_{\vec{k}}\alpha_{\vec{k}}\,, \qquad (9)$$

where

$$\xi_{\vec{k}}=\hbar^2k^2/2m_e+2T\cos\kappa c\,. \qquad (10)$$

Expression (9) could be obtained directly, starting with the single-particle representation (1) and using the tight-binding and nearest-neighbor approximation. Indeed, the transformation (8) brings back a Wannier function into a Bloch state, and $\xi_{\vec{k}}$ is easily recognized to be the approximate eigenvalue of the tight-binding method.[4] But we have preferred to use the Wannier representation in order to arrive in a natural way at the model of Visscher and Falicov and to have the possibility to generalize this model by including a tunneling term in the Hamiltonian.

In order to calculate the Coulomb potential energy we start from its expression in terms of the pair-distribution function, namely

$$V_{\mathrm{Coul}}=\frac{1}{2\Omega}\sum_{\vec{Q}}\frac{4\pi e^2}{Q^2}\,S(\vec{Q})\,, \qquad (11)$$

where $\vec{Q}=(\vec{q},\theta)$, Ω is the volume of the crystal and $S(\vec{Q})$ is the Fourier transform of the pair-distribution operator

$$S(\vec{R}_1,\vec{R}_2)=\Psi^+(\vec{R}_1)\,\Psi^+(\vec{R}_2)\,\Psi(\vec{R}_2)\,\Psi(\vec{R}_1)\,. \qquad (12)$$

We shall calculate $S(\vec{Q})$ directly in the single-particle representation (1), using the tight-binding approximation (5) for the Bloch states. Neglecting overlap integrals [no enhancement factor like $U(z)$ exists now, and we shall retain only the integrals with wave functions belonging to the same lattice site] after straightforward calculations one gets

$$V_{\mathrm{Coul}}=\frac{1}{2\Omega}\sum_{\vec{k}\vec{k}'\vec{Q}}\frac{4\pi e^2}{Q^2}\,|g(\theta)|^2\,\alpha^\dagger_{\vec{k}+\vec{Q}}\,\alpha^\dagger_{\vec{k}'-\vec{Q}}\,\alpha_{\vec{k}'}\,\alpha_{\vec{k}}\,, \qquad (11')$$

where

$$g(\theta)=\int dz\,e^{-i\theta z}|\varphi(z)|^2\,. \qquad (13)$$

For a δ-function behavior of $|\varphi(z)|^2$ (approximation used by Visscher and Falicov), $g(\theta)=1$. This is not an essential approximation for calculating the plasma frequency, however, because in the long-wavelength limit we have $g(\theta)\to 1$; for simplicity we shall take $g(\theta)=1$. Then the model Hamiltonian which will be used in further calculations is given by the expression

$$H=\sum_{\vec{k}}\xi_{\vec{k}}\,\alpha^\dagger_{\vec{k}}+\frac{1}{2\Omega}\sum_{\vec{k}\vec{k}'\vec{Q}}V(Q)\,\alpha^\dagger_{\vec{k}+\vec{Q}}\,\alpha^\dagger_{\vec{k}'-\vec{Q}}\,\alpha_{\vec{k}'}\,\alpha_{\vec{k}}\,, \qquad (14)$$

where $V(\vec{Q})=4\pi e^2/Q^2$ and $\xi_{\vec{k}}$ is given by (9). The strong anisotropy of the system is reflected in the dispersion relation (9) for the single-particle energy and in the shape of the Fermi surface, which now has a cylindrical symmetry and a cosine profile along the z axis. It can be shown that the value of the Fermi energy of the independent electron system is not influenced by the presence of the electron tunneling, and is given by[1] $\mu=\hbar^2k_F^2/2m_e$, $k_F^2=2\pi n_s$, n_s being the electron density per unit area.[6]

III. PLASMA FREQUENCY

In order to determine the plasma frequency the motion of an electron-hole pair of momentum $(-\vec{Q})$ will be followed. Using the equation-of-motion method in the RPA, the calculations are standard, and one obtains

$$1 = V(Q)\frac{1}{\Omega}\sum_{\vec{K}}\frac{f(\vec{K}+\vec{Q})-f(\vec{K})}{\xi_{\vec{K}+\vec{Q}}-\xi_{\vec{K}}-\hbar\omega}\ , \tag{15}$$

where $f(\vec{K})$ is the Fermi–Dirac distribution function corresponding to the energy $\xi_{\vec{K}}$. Formally, (15) has the same form as for a homogeneous three-dimensional electron gas. The differences consist in a new form of the Fermi surface and in the dispersion relation (9), and their effect on the plasma frequency is notable.

Let us consider first the case of the very localized model of Visscher and Falicov. Then the sum over κ is straightforward, and we remain with an integral over the two-dimensional wave vector $\vec{k}$. In the long-wavelength limit the calculations are easily done, and one obtains

$$\omega_{pl}^2 = \omega_0^2 \sin^2 p\ , \tag{16}$$

where $\omega_0^2 = 4\pi e^2 n/m_e$, and p is the angle between the wave vector $\vec{Q}$ of the plasma wave and z direction. It shows a very strong anisotropy, and its vanishing when $p \to 0$ is related to the rigidity of the model with respect to the motion in the perpendicular direction. It is expected that in the extended model when the electron tunneling is taken into account it will be finite everywhere.

Indeed, in the limit $|\vec{Q}| \to 0$ one can write (15) in the form

$$1 = V(Q)\frac{1}{\Omega}\sum_{\vec{K}}\frac{\vec{v}_{\vec{K}}\cdot\vec{Q}}{\omega-\vec{v}_{\vec{K}}\cdot\vec{Q}}\,\delta(\xi_{\vec{K}}-\mu)\ , \tag{15'}$$

where $\vec{v}_{\vec{K}} = (1/\hbar)\nabla_{\vec{K}}\xi_{\vec{K}}$ is the group velocity. A series expansion of the denominator gives

$$1 = V(Q)\frac{1}{\Omega}\sum_{\vec{K}}\frac{\vec{v}_{\vec{K}}\cdot\vec{Q}}{\omega}\left(1+\frac{\vec{v}_{\vec{K}}\cdot\vec{Q}}{\omega}+\cdots\right)\delta(\xi_{\vec{K}}-\mu)\ , \tag{15''}$$

and the integrals are now straightforward. Introducing the tunneling velocity $v_T = Tc/\hbar$ and the velocity at the Fermi surface $v_F = (2\mu/m_e)^{1/2}$, the final result can be written

$$\omega_{pl}^2 = \omega_0^2\left(\sin^2 p + 4\,\frac{v_T^2}{v_F^2}\cos^2 p\right). \tag{17}$$

In contrast with (16), the plasma frequency given by (17) is everywhere finite, but its strong anisotropy is still present. This is the expected result because when the tunneling is also included the electron motion has a three-dimensional character and the energy of the collective excitation has to be finite. The obtained result shows the qualitative difference between the layered-electron model and a two-dimensional electron gas. Remember that for a two-dimensional system, the plasma frequency vanishes in the long-wavelength limit.[7]

The same result can be found from the dielectric constant. In the RPA (or in the self-consistent approximation[8]) this is given by

$$\epsilon(\vec{Q},\omega) = 1 - V(Q)\frac{1}{\Omega}\sum_{\vec{K}}\frac{f(\vec{K}+\vec{Q})-f(\vec{K})}{\xi_{\vec{K}+\vec{Q}}-\xi_{\vec{K}}-\hbar\omega-i\hbar\alpha}\ . \tag{18}$$

One can compare (18) with the expression of $\epsilon(\vec{Q},\omega)$ calculated using the Bloch states of the crystal.[8] If the periodic structure in the x, y plane is neglected, we have to calculate the matrix elements of $\rho_\theta = e^{-i\theta z}$ between two Bloch states $\varphi_{\kappa\nu}(z)$. Because the separation between two energy levels of the isolated potential well $U(z)$ is assumed very great, all the interband transitions can be neglected. Then in the tight-binding approximation the matrix elements $\langle \kappa|\rho_\theta|\kappa+\theta\rangle$ becomes equal with $g(\theta)$ defined by (13), and taking $g(\theta)=1$ the expression (18) is found. In order to improve the the model, the first step would be to take into account the periodic structure in the x, y plane. In the present form we have a simple but very anisotropic model, for which simple results are easily obtained, and which is opposite to the free-electron model.

From the experimental point of view the strong anisotropy of the plasma frequency (17) could be observed in energy-loss experiments. As it is known for a high-energy-electron beam the transferred momentum is very nearly perpendicular to the incident direction. Then varying the direction of the incident beam one can vary the propagation direction of the plasma wave and consequently, the peak position in the characteristic energy-loss spectra.

Note added in proof. A strong anisotropy of electron energy losses in graphite was first found by K. Zeppenfeld [Z. Physik 211, 391 (1968)] and a theoretical interpretation was given by E. Tosatti and F. Bassani [Nuovo Cimento 65, 161 (1970)] and J. Cazaux [Opt. Commun. 3, 225 (1971)].

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