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FINITE SIZE EFFECTS ON THE PLASMA FREQUENCY IN LAYERED ELECTRON GAS*

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ABSTRACT

The equation-of-motion technique is used within the random-phase-approximation to calculate the plasma frequency of the electron gas at zero temperature in the finite layered model of Visscher and Falicov with free surfaces. The plasma frequencies are given by the eigenvalues of a Toeplitz matrix. This matrix describes the Coulomb coupling between the plasmons which propagate in different layers. It is shown that this matrix splits into two parts: one which corresponds to the cyclic boundary condition imposed on the system of double thickness and the other due to the presence of the surfaces. The first contribution can be exactly diagonalized and the other one can be treated as a small perturbation for a sufficiently large number of layers. The first-order perturbation theory is applied to obtain finite-size corrections to the plasma frequency.

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1. INTRODUCTION

In the last years there has been a great deal of interest in the properties of the electron gas in layered structures, such as transition metal dichalcogenides[1-4]. In these anisotropic compounds the electrons are relatively restricted to move along a regular array of parallel planes, the tunneling between planes being more or less forbidden. The analysis of the inelastic scattering of fast electrons shows that the electron energy losses are directly related to the frequency of the plasma oscillations[5,6]. It is then important to study the dispersion relation for plasmons in these structures. On the other hand, these systems offer the possibility of investigating the finite-size effects and the role of the altered dimensionality in plasma oscillations.

A simple model of layered electron gas was proposed by Visscher and Falicov[7]. The model consists of a succession of parallel equally spaced planes. The electrons are free to move within the planes in a neutralizing rigid background of positive charge, which is also confined to the layers. The tunneling between layers is completely forbidden. A hydrodynamic model has been used by Fetter[8] to study the electrodynamic effects in this system, such as screening and plasma oscillations. In this very intuitive approach the electrons are approximated by a charged fluid which is characterized by a local density and velocity. Many-body techniques were used to describe the collective behaviour of the electron gas in the layered model of Visscher and Falicov. The Bohm-Pines canonical transformation method has been applied to calculate the plasma frequency in this system with cyclic boundary condition[9]. Further generalizations of the model have been performed[10,11], including the tunneling between layers in the tight-binding approximation.

The aim of this paper is to investigate the finite-size effects on the plasma frequency in the layered model of Visscher and Falicov. The system consists of a finite number of planes with free surfaces, that is no special condition is imposed on the boundary planes. In Sec.2 the model is presented. The unperturbed single-particle wave-functions correspond to a free electron motion within each layer, any motion of the electrons along the direction perpendicular to the plane of the layers being forbidden. The equation-of-motion method is applied in the RPA for the operator of an electron-hole pair. A set of coupled algebraic equations is obtained. This set of equations reflects the Coulomb coupling between the plasmons which propagate in different layers. The finding of the plasma frequency amounts to calculate the eigenvalues of the coupling matrix. This is a Toeplitz matrix that cannot be exactly diagonalized. The properties of the coupling matrix are discussed in Sec.3. It is shown that this matrix has a double representation as a sum of two matrices. The first matrix (in both representations) corresponds to the case when cyclic boundary condition is imposed on the system of double thickness. Its eigenvalues are exactly known. The second matrix (in both representations) represents the influence of the free surfaces and it can be treated as a small perturbation for a sufficiently large number of layers. The first-order perturbation theory is applied to calculate the finite-size corrections to the plasma frequency. A discussion of the results is given in Sec.4.

2. EQUATION OF MOTION METHOD

The Visscher and Falicov model with free surfaces consists of a succession of N parallel planes, the distance between two neighbouring planes being taken equal to unit. The electrons are strictly confined to the layers along which they are moving freely in a rigid neutralizing background of positive charge. The planes are labeled by m, $m=0,1,\ldots,N-1$. The single-particle wave-functions of the unperturbed eigenstates of the electrons are given by

$$\varphi_{mk}(x,r) = e^{ikr}\chi(x-m) \quad , \tag{1}$$

where spin is disregarded for simplicity. The wave-vector of an electron state is denoted by k, the in-plane vector r is confined to a region of unit area and x represents the coordinate along the direction perpendicular to the plane of the layers. A number of n_s electrons is assumed on the in-plane unit area. These electrons interact through Coulomb potential. The function $\chi(x-m)$ is arbitrarily highly localized on the m-th plane and it is effectively the square root of a $\delta(x-m)$ function[7]. Using the wave-functions (1) the Hamiltonian of the system may be written in the second quantization as follows:

$$H = \sum_{mk} \varepsilon_k c^+_{mk} c_{mk} + \sum_{\substack{k_1 k_2 k \\ (k\neq 0)}} \sum_{m,n} \frac{\pi e^2}{k} A_{mn} c^+_{mk_1+k} c^+_{nk_2-k} c_{nk_2} c_{mk_1} \, , \tag{2}$$

where $\varepsilon_k = \hbar^2 k^2/2M$ is the kinetic energy of an electron (e and M denote the electron charge and mass, respectively), c^+_{mk} (c_{mk}) is the creation (annihilation) operator of an electron state localized on the m-th

plane with the wave-vector k and $A=(A_{mn})$, $m,n=0,1,\ldots,N-1$,

$$A_{mn} = e^{-k|m-n|} \tag{3}$$

is the matrix that comes from the in-plane Fourier transform of the Coulomb potential. The equation-of-motion method will be applied in the random-phase-approximation (RPA) for the operator $\rho_m(k_1-k,k_1) = c^+_{mk_1-k} c_{mk_1}$ where $\rho_m(k) = \sum_{k_1} \rho_m(k_1-k,k_1)$ is the Fourier transform of the electron density in the m-th plane. The calculations are standard and one obtains

$$\rho_m(k) = \frac{\omega_0^2}{2k} D(\omega,k) \sum_n A_{mn} \rho_n(k) \quad , \tag{4}$$

where $\omega_0 = (4\pi n_s e^2/M)^{1/2}$ is the well known bulk plasma frequency and

$$D(\omega,k) = \frac{M}{n_s} \sum_{k_1} \frac{n_{k_1} - n_{k_1+k}}{\hbar\omega + \varepsilon_{k_1} - \varepsilon_{k_1+k}} \tag{5}$$

In (5) n_k is the Fermi distribution at zero temperature for the two-dimensional electron gas in each plane. For sufficiently small k, $D(\omega,k)$ may be approximated by k^2/ω^2, so that the plasma frequency is given by

$$\omega^2 = \frac{\omega_0^2 k}{2} \lambda \tag{6}$$

where λ denotes the eigenvalues of the matrix A. These eigenvalues will be calculated in the next Section.

If we impose the cyclic condition on the boundaries of the system and use the following representation for the function $\delta(x-m) = |\chi(x-m)|^2$,

$$\delta(x-m) = N^{-1} \sum_{\varkappa} e^{i\varkappa(x-m)} \quad ,$$

where $\varkappa = 2p\pi/N$ with p running over all integers, then the matrix A is replaced by $A^{cyc} = (A^{cyc}_{mn})$, $m,n = 0,1,\ldots,N-1$,

$$A^{cyc}_{mn} = 2kN^{-1} \sum_{\varkappa} (\varkappa^2 + k^2)^{-1} e^{-i\varkappa(m-n)} = 2kN^{-1} \sum_{\varkappa\in B} \sum_{G} \left[(\varkappa+G)^2 + k^2\right]^{-1} e^{-i\varkappa(m-n)} \, ,$$

where $\varkappa \in B$ means that $\varkappa$ is restricted to the first Brillouin zone ($-\pi < \varkappa \leq \pi$) and $G = 2p\pi$ with p any integer, are vectors in the reciprocal lattice. The summation over G is easily to perform. We get[12]

$$A^{cyc}_{mn} = N^{-1} \sum_{\varkappa\in B} f(\varkappa)\, e^{-i\varkappa(m-n)} \quad , \tag{7}$$

where

$$f(\varkappa) = \cosh k\,(\sinh k - \cos\varkappa)^{-1} \, . \tag{8}$$

The eigenvectors of A^{cyc} are $N^{-1/2} e^{-i\varkappa m}$ and its eigenvalues are $f(\varkappa)$. The plasma frequency for cyclic boundary condition is

$$\omega^2 = \frac{\omega_0^2 k}{2} f(\varkappa) \quad ,$$

a result which has been previously obtained[8,9].

3. PERTURBATION THEORY

The matrix A , given by (3), is a Toeplitz matrix[13]. It has the following integral representation:

$$A_{mn} = \frac{k}{\pi} \int_{-\infty}^{+\infty} dx' \, (x'^2 + k^2)^{-1} \, e^{-i x'(m-n)} ,$$

where x' is a continous variable and the integral is performed in the complex plane. By straightforward manipulations we get

$$A_{mn} = \frac{k}{\pi} \int_{-\pi}^{\pi} dx' \sum_G \left[(x'+G)^2 + k^2\right]^{-1} e^{-i x'(m-n)} = \frac{1}{2\pi} \int_{-\pi}^{\pi} dx' \, f(x') \, e^{-i x'(m-n)} , \quad (9)$$

a result which can be obtained from (7) by taking $N \to \infty$. The generating function of this matrix is just the function $f(x')$ given by (8), where x is replaced by the continuous variable x' . This function may be written as

$$f(x') = (1-r^2)(1 - 2r\cos x' + r^2)^{-1} , \quad (10)$$

where $r = e^{-k}$. In the limit of $N \to \infty$ the matrix A becomes a bounded self-adjoint Toeplitz operator ($f(x')$ is bounded, real and measurable on $(-\pi, \pi)$). According to a theorem of Hartman and Wintner[14] the continuous spectrum of this operator lies between $\min f(x') = f(\pi) = (1-r)(1+r)^{-1}$ and $\max f(x') = f(0) = (1+r)(1-r)^{-1}$. Therefore, in the thermodynamic limit ($N \to \infty$) the spectrum of A^{cyc} (system with cyclic boundary condition) coincides with that of A (system with free surfaces), a result which is expected because of the translational invariance of these systems.

Kac et al.[15] studied the eigenvalues of the matrix A using its characteristic equation. They found that these eigenvalues may be represented as

$$f\left(\frac{\pi p + \varepsilon(p,N)}{N+1}\right) , \quad p = 1, 2, \ldots , \quad (11)$$

where f is given by (10) and $\varepsilon(p,N) \to 0$ for fixed p and $N \to \infty$. The exact form of $\varepsilon(p,N)$ is unknown. A similar relation holds also for the eigenvalues placed at the bottom of the spectrum $f\left(\pi - \frac{\pi p - \varepsilon(p,N)}{N+1}\right)$, $p = 1, 2, \ldots$. From (11) we obtain that the eigenvalues may be represented also by $f\left(\frac{\pi p + \varepsilon'(p,N)}{N}\right)$ where $\varepsilon'(p,N) = [N\varepsilon(p,N) - p\pi]/(N+1)$. Our aim is to give approximate expressions for $\varepsilon'(p,N)$ for a sufficiently large N . These expressions represent precisely the finite-size corrections to the eigenvalues of the matrix A and, therefore, to the plasma frequency in the Visscher and Falicov model.

To this end, let us introduce the matrices $A_0^{\pm} = ((A_0^{\pm})_{mn})$, $m,n = 0, 1, \ldots, N-1$,

$$(A_0^{\pm})_{mn} = N^{-1} \sum_{x \in B} f(x) \, e^{-i x(m-n)} , \quad (12)$$

where $\pm$ correspond respectively to $x = 2p\pi/N$ and $x = (2p-1)\pi/N$, with p integers. One can see that A_0^{+} is just A^{cyc} . The matrices $A_0^{\pm}$ are cyclic matrices with the period $2N$. They correspond to the cyclic boundary condition imposed on the system of double thickness. The eigenvectors of these matrices are $N^{-1/2} e^{-ixm}$ and their eigenvalues are $f(x)$. From (12) we get[7]

$$(A_0^{\pm})_{mn} = 2kN^{-1} \sum_{x \in B} \sum_G \left[(x+G)^2 + k^2\right]^{-1} e^{-ix(m-n)} = 2kN^{-1} \sum_x (x^2+k^2)^{-1} e^{-ix(m-n)} , \quad (13)$$

in the last summation x being allowed to run over the whole space. We shall calculate $(A_0^{\pm})_{mn}$ given by (13) with a method of Lighthill[16]. Replacing $m-n$ by u , where $-N+1 \le u \le N-1$ we have to calculate

$$A_0^{\pm}(u) = 2kN^{-1} \sum_x (x^2+k^2)^{-1} e^{-ixu} .$$

Using the integral representation

$$2k(x^2+k^2)^{-1} = \int_{-\infty}^{+\infty} d\mu \, e^{ix\mu - k|\mu|}$$

and the relation

$$N^{-1} \sum_x e^{ix(\mu - u)} = \sum_{j=-\infty}^{+\infty} \delta(\mu - u + Nj) , \quad x = 2p\pi/N , \quad j, p \text{ integers},$$

the calculations are straightforward and one obtains

$$A_0^{+}(u) = \sum_{j=-\infty}^{+\infty} e^{-k|u-Nj|} = e^{-k|u|} + (e^{Nk}-1)^{-1}(e^{ku} + e^{-ku}) ,$$

$$A_0^{-}(u) = \sum_{j=-\infty}^{+\infty} e^{ij\pi} e^{-k|u-Nj|} = e^{-k|u|} - (e^{Nk}+1)^{-1}(e^{ku} + e^{-ku}) .$$

Therefore, the matrix A has the following double representation:

$$A = A_0^{\pm} + A_1^{\pm} , \quad (14)$$

where

$$(A_1^{\pm})_{mn} = r^N (r^N \mp 1)^{-1} (r^{m-n} + r^{n-m}) , \quad m,n = 0, 1, \ldots, N-1 , \quad (r = e^{-k}) \quad (15)$$

The matrices $A_1^{\pm}$ represent the influence of the surfaces and contain information about the finite-size effects. According to (11) and (12), for sufficiently large N the eigenvalues of A approach the eigenvalues of $A_0^{\pm}$. Consequently, the small deviations of these eigenvalues from those of $A_0^{\pm}$ can be calculated by means of the first-order perturbation theory, applied for the matrices $A_1^{\pm}$.

Let us take the eigenvalue $f(2p\pi/N)$ of the matrix A_0^{+}, $p = 1, 2, \ldots$ (p fixed). This eigenvalue is twofold degenerated since $f(x)$ is an even function (x is restricted to the first Brillouin zone, $-\pi < x \le \pi$). The first-order theoretical perturbation calculation is standard and one obtains

$$f(x) \pm 2r(1\mp r)^{2}(1\mp\cos x)(1-2r\cos x+r^{2})^{-2}N^{-1}\ , \quad x = 2p\pi/N\ , \qquad (16)$$

where terms of order $r^{N}N^{-1}$ were neglected. It is easily to see that, for large N ,the separation between two neighbouring eigenvalues of A_c^{+} (or A_c^{-}) is of the order of N^{-2} . For sufficiently large N the N^{-1} -term in (16) corresponding to the upper sign brings a contribution of order N^{-3} to $f(x)$. The expression corresponding to the lower sign in (16) brings a contribution of the same order to $f(x)$ with $x = \pi-(2p-1)\pi/N$ for N an odd integer and $x = \pi - 2p\pi/N$ for N an even integer ($p=1,2,\dots$). The calculation with the perturbation theory applied for the eigenvalues $f(x)$ corresponding to $x=\pi-(2p-1)\pi/N$ (which labels an eigenvalue of A_c^{-}) or $x=\pi-2p\pi/N$ will give a N^{-1} -term equal to that from (16). Therefore, a factor of two must be inserted in the N^{-1} -term given in (16). The perturbation theory can be applied in the same manner for the eigenvalues of A_0^{-} and the results are identical with those from (16). Therefore,the eigenvalues of A ,up to the first order of the perturbation theory, may be represented as

$$\lambda_x = f(x)+4r(1+r)^{2}(1-\cos x)(1-2r\cos x+r^{2})^{-2}N^{-1}\ , \quad x = p\pi/N\ , \qquad (17a)$$

$p=1,2,\dots$ (upper eigenvalues),

$$\lambda_x = f(\pi-x)-4r(1-r)^{2}(1-\cos x)(1+2r\cos x+r^{2})^{-2}N^{-1}\ , \quad x = p\pi/N\ , \qquad (17b)$$

$p=1,2,\dots$ (lower eigenvalues) .

Taking into account that $r = e^{-k}$ and using (6) these expressions give us the finite-size corrections to the plasma frequency in the layered model of Visscher and Falicov.

One can see from these results that the N^{-1} proves to be a good parameter of the perturbation theory. For sufficiently large N the perturbation theory produces the eigenvalues of the matrix A which are placed in the immediate neighbourhood of the eigenvalues of $A_0^{\pm}$ (the perturbation theory yields N^{-3} -terms while the separation between two neighbouring eigenvalues of $A_0^{\pm}$ is of the order of N^{-2}). This result is in agreement with the theorem (11) given by Kac et al.[15]. The matrices $A_0^{\pm}$ may have nondegenerate eigenvalues as $f(0)$ or $f(\pm\pi)$. The perturbation theory cannot be used in this case since the results do not agree with (11). For example,the perturbation calculation applied for the eigenvalue $f(0)$ brings a contribution $-2r(1-r)^{-2}N^{-1}$ which is much larger than the separation between two neighbouring unperturbed

eigenvalues.

4. DISCUSSION

It must be pointed out that the present method of perturbational treatment of the eigenvalues of the matrix A leads to a mathematical result established many years ago by Widom[17] for Toeplitz matrices. Widom shown that the eigenvalues placed at the top of the spectrum have the following asymptotic behaviour for large N :

$$\lambda_p = M - \frac{\sigma^2\pi^2p^2}{2N^2}\left(1+\frac{\alpha}{N}\right) + O(N^{-3})\ , \quad p = 1,2,\dots\ ,$$

where $M = \max f(x') = f(0) = (1+r)(1-r)^{-1}$, $\sigma^2 = -f''(0) = 2r(1+r)(1-r)^{-3}$

and

$$\alpha = \frac{1}{2\pi}\int_{-\pi}^{\pi} d\theta\, \mathrm{cosec}^2\frac{\theta}{2}\, \ln\left[\frac{M-f(\theta)}{2\sigma^2}\,\mathrm{cotan}^2\frac{\theta}{2}\right] .$$

This integral is easy to evaluate. We get[18] $\alpha = -2(1+r)(1-r)^{-1}$,so that

$$\lambda_p = \frac{1+r}{1-r}\left[1 - \frac{r\pi^2p^2}{(1-r)^2N^2}\left(1-2\frac{1+r}{1-r}N^{-1}\right)\right] + O(N^{-3})\ , \quad p=1,2,\dots .$$

The same result is obtained from (17a) by using an expansion of $\cos x$ in powers of x in the neighbourhood of $x = 0$. In the same manner we obtain from (17b) the asymptotic behaviour of the eigenvalues placed at the bottom of the spectrum

$$\lambda_p = \frac{1-r}{1+r}\left[1 + \frac{r\pi^2p^2}{(1+r)^2N^2}\left(1-2\frac{1-r}{1+r}N^{-1}\right)\right] + O(N^{-3})\ , \quad p=1,2,\dots .$$

Finally,we remark that the plasma frequency of the layered system with free surfaces (N layers) may be approximated (for sufficiently large N) by the plasma frequency corresponding to the system of double thickness with cyclic boundary condition. A similar result is well known for the system with short-range interactions (elementary excitations as phonons,magnons,etc.). The perturbational approach used in the present paper for calculating the plasma frequency in the finite layered model of Visscher and Falicov may be applied for evaluating the finite-size corrections to the thermodynamic quantities of this model. This problem will be treated in a forthcoming paper.

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