Energy spectrum of electrons above a thin liquid helium film in a clamping electric field

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The spectrum of electrons above a thin helium film on a metal substrate is calculated. The dependences of the transition frequencies between the levels on the intensity of the clamping electric field and on the film thickness are also calculated. It is shown that the quasiclassical approximation is applicable over a broad range of clamping-field intensities and helium film thicknesses.

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1. INTRODUCTION

The energy spectrum of electrons, localized above the surface of liquid helium under the action of electrostatic image forces and an external clamping electric field \mathscr{C}_1 , have been studied in a number of researches (see the reviews by Shikin and Monarkhal and by Edel'man^2 . Of important significance for the theoretical calculations of the spectrum is the correct choice of potential $W(x)$, which should not have infinite discontinuities and jumps at the surface. 3.4 As shown previously,⁵ this can be done by taking consistent account of the spatial dispersion of the permittivity of the helium $\varepsilon(\mathbf{k})$. It turns out here that $W(x)$ can be approximated with good accuracy $\langle 3\% \rangle$ by a simple model potential of the form^{3,4}

$$
W_{\mathfrak{o}}(x) = -\frac{e^{z}(\epsilon - 1)}{4(x \dot{+} x_{\mathfrak{o}})(\epsilon + 1)},
$$
\n(1)

where ϵ is the static permittivity of helium in a uniform field $(\epsilon - 1 \ll 1)$, and the parameter $x_0 \approx 1$ Å.

In addition, there is interest in the investigation of the spectrum of the electrons above a thin film of helium, when the dominant role is played by the image forces of the substrate (see Ref. 2). Since the thickness of the wetting superfluid $He⁴$ film generally amounts to tens and hundreds of angstroms, $^{1)}$ effects of spatial dispersion in the substrate can be neglected, so that the image-force potential is equal to

$$
W_1(x) = -\frac{e^2(e_1 - 1)}{4(x+d)(e_1 + 1)},
$$
\n(2)

where d is the film thickness, and ε_1 is the permittivity of the substrate.

The spectrum of the electrons above a thin film of helium on metal is calculated in the present work in Sec. 2 (as ε_1 + ∞), and it is shown that it can differ markedly from the quasi-hydrogenlike spectrum of electrons above the surface of a deep helium bath. The effect of a clamping electric field \mathscr{C}_1 on the one-electron spectrum is considered in Sec. 3 both for sufficiently thick films with thickness $d \gg \langle x \rangle_n$ (where $\langle x \rangle_n$ is the mean distance of the electron from the helium surface in the n -th quantum state), when the effect of the substrate reduces to renormalization of the external field, and also for comparatively thin films with $d \ll \langle x \rangle_n (\varepsilon - 1)^{-1}$, when we can neglect the image forces in the helium film. It is shown that the quasiclassical approximation is valid with sufficient accuracy over a wide range of fields and thicknesses.

2. IMAGE FORCES AND THE SPECTRUM OF ELECTRONS ABOVE A HELIUM FILM

The image-force potential of a charge located at a distance x from the surface of a helium film²⁾ on a metal substrate can be represented according to (1) and (2) in the form

$$
W(x) = -\frac{Z'e^2}{x+x_0} - \frac{e^2}{4(x+d)}, \quad Z \approx \frac{1}{8} (e-1) \ll 1, \quad x_0 \ll d \tag{3}
$$

with good accuracy. Small terms of the order of *Z** and x_0/d are discarded.

For thick films of helium, with thickness

 $d \gg \langle x \rangle_n (\varepsilon - 1)^{-1}.$

the small corrections $\Delta E_n(d)$ to the energy spectrum, due to the effect of the substrate, can be calculated in first-order perturbation theory with hydrogenlike functions. Taking into account the relations7

$$
\int_{0}^{\infty} \frac{x^{y}e^{-\mu x}}{x+d} dx = d^{y}e^{\mu t} \Gamma(\nu+1) \Gamma(-\nu, \mu d),
$$
\n
$$
\Gamma(-\nu, y) = \frac{(-1)^{\nu+1}}{\nu!} \left[\text{Ei}(-y) + e^{-y} \sum_{m=0}^{\nu-1} (-1)^{m} \frac{m!}{y^{m+1}} \right],
$$
\n(4)

where $\Gamma(\nu,\nu)$ is the incomplete gamma function and Ei(y) is the integral exponential function, we get, with accuracy to terms of first order in the small parameter $a_0/d(\epsilon - 1) \ll 1$ (where a_0 is the Bohr radius of the electron)

$$
\Delta E_n(d) = -\frac{e^2}{4d} \left[1 - \frac{3}{2} \frac{n^2 a_0}{Z d} \right], \quad n = 1, 2, 3, \dots \tag{5}
$$

We note that the expression (5) is identical with the correction to the spectrum of electrons above the surface of helium in a weak clamping field \mathscr{E}_{μ} (the linear Stark effect),^{1,2} if we discard the insignificant constant $-e^2/4d$ and introduce the effective field $\tilde{\mathscr{E}}_1 = e/4d^2$ (see below).

In the case of sufficiently thin films, when

$$
d{\ll}\langle x\rangle_n\;(\epsilon{-}1)^{-i}
$$

(but $d \gg a_0$), the potential energy of the electrons, in accord with (3) and with accuracy to terms $\sqrt{\epsilon - 1}d$ $\langle x \rangle_n$, is determined by the image forces of the substrate, so that the Schrödinger equation for the electron wave function Ψ in the region $x > 0$ reduces to a special case of the Whittaker equation⁸:

$$
\frac{d^2\Psi}{dy^2} + \left(-\frac{1}{4} + \frac{\kappa_n}{y}\right)\Psi(y) = 0, \tag{6}
$$

$$
y = \frac{x+d}{2a_0x_n}, \quad x_n = \frac{e^2}{4\hbar} \left(\frac{m}{2|E_n|}\right)^{\nu_n}.
$$
 (7)

The solution of Eq. (6), which falls off exponentially at infinity, has the following form in the case of arbitrary (non-integer) values of the parameter κ .

$$
\Psi(y) \sim y e^{-y/2} U(1-\kappa_n, 2; y), \qquad (8)
$$

where U is the Kummer confluent hypergeometric function.³⁾ If we take into account the finiteness of the repulsive potential $(V_0>0)$ on the helium surface, then the wave function inside the film $(-d \le x \le 0)$ has the $form⁴$

$$
\Psi(x) \sim \exp(-k_n|x|), \quad k_n = \left[\frac{2m}{\hbar^2}(V_0 + |E_n|)\right]^{1/2}.
$$
 (9)

From the conditions of continuity of Ψ and $d\Psi/dx$ at the point $x = 0$, we obtain a transcendental equation for the determination of the eigenvalues E_n (cf. Ref. 4):

$$
k_n dU (1 - \varkappa_n, 2; y_n) = (1 - y_n/2) U (1 - \varkappa_n, 2; y_n)
$$

$$
- \frac{\Gamma(2 - \varkappa_n)}{\Gamma(1 - \varkappa_n)} y_n U (2 - \varkappa_n, 3; y_n), \quad y_n = \frac{d}{2a_0 \varkappa_n}.
$$
 (10)

At $V_0 \rightarrow \infty$, Eq. (10) reduces to the condition

$$
U(1-\kappa_n, 2; y_n)=0 \tag{11}
$$

and the spectrum of the eigenfunctions is determined by the zeros of the function U (see Ref. 9):

$$
y_n \approx \pi^2 (n- \nu/4 + \nu_n)^2 / 4 \nu_n. \tag{12}
$$

Equation (12) is valid only at large values of the argument $y_n \gg 1$, i.e., at $d/a_0 \gg \kappa_n \gg 1$. With account of **(7),** we then obtain

$$
|E_n| \approx \frac{e^2}{32a_0} \left[n - \frac{3}{4} + \frac{1}{\pi} \left(\frac{2d}{a_0} \right)^{1/2} \right]^{-2}, \quad n = 1, 2, 3, \tag{13}
$$

Thus the spectrum of electrons above a thin film of liquid helium differs markedly from the hydrogen-like spectrum

 $|E_n^{\circ}| = (Z^*e)^2/2a_0n^2$

above the surface of a deep helium bath. The dependences of the transition frequencies between the levels f_{12} and f_{13} on the dimensionless thickness of the film d/a_0 , calculated for the spectrum (13) are shown dashed in Fig. I.

FIG. 1. Dependence of the transition frequencies f_{12} and f_{13} on d/a_0 for the spectrum (13) (dashed line) and in the quasiclassical approximation (solid curves).

3. EFFECT OF THE CLAMPING FIELD ON THE SPECTRUM OF ELECTRONS ABOVE A HELIUM FILM

The spectrum of electrons above the surface of liquid helium in a clamping electric field \mathscr{C}_1 is usually calculated with the help of variational numerical methods, $3,4$ since perturbation theory does not allow us to move into the region of strong fields. However, the effect of the clamping field on the spectrum of the electrons can be taken into account by a much simpler method if we use the quasiclassical approximation, which in a number of cases, as will be seen from what follows, is in good agreement with the exact solution of the problem.

For comparatively thick films, with $d \gg \langle x \rangle_n$, when the effect of the substrate reduces to renormalization of the external field \mathscr{C}_1 , the quasiclassical Bohr-Sommerfeld quantization condition is of the form

$$
\int_{c}^{L(E_n)} dx \left\{ 2m \left[\mathcal{U} - |E_n| \right] \right\}-\epsilon \tilde{\mathcal{E}}_{\perp} x + \frac{Z^* e^z}{x} \right\}^{V_n} = \pi \hbar n,
$$
\n
$$
L(E_n) = \frac{l}{2} \left\{ 1 - \frac{|E_n|}{\mathcal{U}} \right\}-\left[\left(1 - \frac{|E_n|}{\mathcal{U}} \right)^2 + \frac{4Z^* e^z}{\mathcal{U}} \right]^{V_n} \right\},
$$
\n(15)

where *I* is the distance from the helium surface to the negative electrode. It is taken into account in (14) that $l \gg x_0$, and that as $\tilde{\mathscr{C}}_1 \rightarrow 0$, the electron spectrum should be identical with the hydrogenlike spectrum E_n^0 .

The integral in (14) is expressed in terms of the complete elliptic integrals of the first, *K(p),* and second, $E(p)$, kind, so that Eq. (14) takes the following form in the special case $l = d$ (the helium level is the midplane of a parallel-plate capacitor):

$$
\frac{2}{3} \left(\beta_n + \alpha \right)^{\frac{1}{2}} \left\{ \frac{1}{2} \left[\left(\beta_n + \alpha \right)^{\frac{1}{2}} - \beta_n^{\frac{1}{2}} \right] K(p_n) - \beta_n^{\frac{1}{2}} E(p_n) \right\} \tag{16}
$$
\n
$$
= \frac{\pi \hbar n}{l(2mU_t)^{\frac{1}{2}}}, \quad C_i = e \tilde{\mathcal{E}}_{\perp} l,
$$
\n
$$
p_n^2 = \frac{(\beta_n + \alpha)^{\frac{1}{2}} + \beta_n^{\frac{1}{2}}}{2(\beta_n + \alpha)^{\frac{1}{2}}}, \quad \beta_n = \left(1 - \frac{|E_n|}{U_t} \right)^2, \quad \alpha = \frac{4Z^* e^2}{lU_t}. \tag{17}
$$

In the region of weak fields, we find from (16) the corrections to the electron spectrum that are linear and quadratic in $\tilde{\mathscr{E}}_1$:⁵⁾

$$
E_n(\tilde{\mathscr{E}}_{\perp}) = E_n^0 - U_l + \frac{3}{2} n^2 \frac{e \tilde{\mathscr{E}}_{\perp} a_0}{Z} - \frac{7}{16} \frac{n^6}{|E_n^0|} \left(\frac{e \tilde{\mathscr{E}}_{\perp} a_0}{Z^*}\right)^2.
$$
 (18)

Estimates show that the expression (18) is valid, in the case of $n \sim 1$, at field intensities $\tilde{\mathscr{E}}_1 \ll 10 \text{ V/cm}$.

In the limit of infinitely strong fields $(\tilde{\mathscr{E}}_1 \rightarrow \infty)$ we get formally from (16) :⁶⁾

$$
|E_n(\tilde{\mathscr{E}}_\perp)| = U_l - \left(\frac{3\pi n}{2}\right)^{\eta_l} \left(\frac{\hbar^2}{2m}\right)^{\eta_2} \left(e^{\tilde{\mathscr{E}}_\perp}\right)^{\eta_2}.\tag{19}
$$

On the other hand, in a strong clamping field, when we can neglect the weak (of the order of $\varepsilon - 1$) image forces in the helium, the problem can be solved exactly, since the Schrödinger equation reduces to the Airy equation¹⁰

$$
\frac{d^2\Psi}{d\xi^2} - \xi \Psi(\xi) = 0,\tag{20}
$$

where

$$
\xi(x) = (2me\widetilde{\mathscr{E}}_{\perp}/\hbar)^{\nu_i}(x-\widetilde{x}_n), \quad \widetilde{x}_n = (U_i - |E_n|)/e\widetilde{\mathscr{E}}_{\perp}
$$

the solution of which inside a triangular potential well in the region $0 \le x \le x_n$, when $\xi(x) \le 0$, has the form

$$
\Psi\left(\xi\right) = \frac{(2m\hbar^2)^{\frac{n}{n}}|\xi|^{\frac{n}{n}}}{3\left(e\widetilde{\mathscr{E}}_\perp\right)^{\frac{n}{n}}}\left[J_{-\frac{n}{n}}\left(\frac{2}{3}|\xi|^{\frac{n}{n}}\right) + J_{\frac{n}{n}}\left(\frac{2}{3}|\xi|^{\frac{n}{n}}\right)\right],\tag{21}
$$

where $J_{\nu}(z)$ is a Bessel function of the first kind. In an infinite repulsive potential on the helium surface $(V₀)$ $\rightarrow \infty$), the eigenvalues E_n are determined from the boundary condition $\Psi(\xi) = 0$ at the point $x = 0$:

$$
|E_n(\tilde{\mathscr{E}}_\perp)| = U_i + \xi_n \left(\frac{\hbar^2}{2m}\right)^{\frac{1}{2}} \left(e\tilde{\mathscr{E}}_\perp\right)^{\frac{n}{2}},\tag{22}
$$

where ξ_n are the zeros of the Airy function (21), viz., $\xi_1 = -2.338$, $\xi_2 = -4.088$, $\xi_3 = -5.521$, ...

From a comparison of the expressions (19) and (22), we see that the spectrum agrees in the quasiclassical approximation with the exact electron spectrum in strong fields with satisfactory accuracy $(15\%$ for $n=1$, 8% for $n = 2$, and 5% for $n = 3$). However, strictly speaking, the quasiclassical approximation is valid only in comparatively weak electric fields satisfying the condition¹¹

$$
e\overline{\mathscr{E}}_{\perp}a_{\circ}^{\ast}\ll \mathrm{Ry}^{\ast} \equiv Z^{\ast}e^{2}/2a_{\circ}^{\ast}, \qquad a_{\circ}^{\ast} = a_{\circ}/Z^{\ast}.
$$
 (23)

For He⁴ with $Z^* = 6.95 \times 10^{-3}$, this corresponds to an effective clamping field $\mathscr{C}_1 \ll 1$ kV/cm.

On the other hand, in a comparatively strong electric field, instability of the charged helium surface develops relative to the buildup of gravitational-capillary waves (ripplons).^{12,13} Under the condition $d \ll (\sigma/\rho g)^{1/2}$, where ρ is the density, σ the coefficient of surface tension, and *g* the acceleration due to gravity, the limiting critical field and the maximum concentration of electrons at which electrocapillary instability of the helium film arises, are equal to

$$
\tilde{\mathscr{E}}_{c} = (\pi g \rho d)^{\nu_{c}}, \quad n_{c} = \tilde{\mathscr{E}}_{c} / 2\pi e. \tag{24}
$$

We note that the macroscopic approximation, which does not take into account the discreteness of the charge on the surface of the film, is valid in the case in which the mean distance between electrons $R_0 = (\pi n_c)^{-1/2} \ll d$, i.e., at sufficiently strong fields $\tilde{\mathscr{C}} > 7e/4d^2$. This condition, together with (24), leads to a lower bound on the thickness of the helium field $d > 6.8 \times 10^{-5}$ cm, and on the value of the effective critical field $\ddot{\mathscr{L}}_c > 50$ V/cm.

Thus, over the entire range of clamping fields

$$
7e/4d^2 < \mathcal{E}_1 < \max{\{\widetilde{\mathcal{E}}_c, (Z^*)^3e/2a_0^2\}}
$$

the quasiclassical approximation is valid. The solid lines in Fig. 2 represent the dependences of the frequencies of transition between the levels f_{1n} on \mathscr{C}_1 as d $-\infty$, calculated on the basis of the transcendental equation (16). The dashed curves are the results of the calculations of Ref. 4, which agree with the experimental data of Ref. 3 with good accuracy. **As** we see, the quasiclassical approximation, without a single adjusting parameter and at sufficiently deep potential approximation $(x_0-0, V_0-\infty)$ agree quite satisfactorily with the variational calculations of Ref. 4 and with the ex-

FIG. 2. Dependence of f_{12} and f_{13} on the clamping field ξ_1 **in** the quasiclassical approximation (solid curves) and by numerical results of Ref. 4 (dashed curves) as $d \rightarrow \infty$.

periment of Ref. 3. The displacement of the curves is easily eliminated if the quasiclassical functions $f_{1n}(\mathscr{C}_1)$ are made coincident with the experimental values of the frequencies at the point $\mathscr{C}_1= 0$, which corresponds to allowance for the finite values of x_0 and V_0 .

For sufficiently thin helium films, when $d \ll \langle x \rangle_n \epsilon$ -1 ⁻¹, so that we can neglect image forces in the film, the quasiclassical quantization condition in an external clamping field $\mathscr{C}_1 \neq 0$ takes the form

$$
\int_{0}^{L_{\epsilon}(E_{n})} dx \left\{ \frac{[L_{+}(E_{n}) - x][x - L_{-}(E_{n})]}{x + d} \right\}^{1/2}
$$
\n
$$
= \frac{\pi \hbar n}{(2m e \mathcal{B}_{\perp})^{1/2}};
$$
\n(25)

$$
L_{\pm}(E_n) = \{U_i - |E_n| \pm ((U_i - |E_n|)^2
$$

+ $e\mathscr{E}_{\pm}d[e^2/d + 4(U_i - |E_n|)])^{\frac{1}{2}}/2e\mathscr{E}_{\pm},$

$$
U_i = e\mathscr{E}_{\pm}L.
$$
 (26)

The integral in (25) reduces to incomplete elliptic integrals of the first, $F(\varphi, p)$, and second, $E(\varphi, p)$, kind, and under the condition $d < |L_-|$, i.e., at $\mathscr{C}_1 < e/4d^2$, Eq. (25) reduces to the form⁷

$$
\frac{2}{3}(L_{+}+|L_{-}|)^{n_{1}}\{(L_{+}-|L_{-}|+2d)E(\varphi,p)+(|L_{-}|-d)F(\varphi,p)\}\qquad(27)
$$

$$
=\frac{2}{3}(L_{+}|L_{-}|d)^{n_{-}}=\frac{\pi\hbar n}{2}
$$

$$
\varphi = \arcsin\left(\frac{L_+}{L_++d}\right)^{\frac{1}{2}}, \quad p = \left(\frac{L_++d_-}{L_++L_-}\right)^{\frac{1}{2}}.\tag{28}
$$

In the absence of a clamping field (\mathscr{C}_1 = 0) Eq. (25) reduces to the following

$$
\sigma_n^{-\eta_i}\left[\frac{\pi}{2}-\arctg\left(\frac{\sigma_n}{1-\sigma_n}\right)^{\eta_i}\right]-(1-\sigma_n)^{\eta_i}=\frac{\gamma_2\pi\hbar n}{e\left(md\right)^{\eta_i}},\qquad (29)
$$

where $\sigma_n = 4d |E_n| / e^2$. As $d \rightarrow \infty$ the hydrogenlike spectrum $|E_n^0|$ is a solution of Eq. (29), while at $n \gg 1$, when $d/a_0n^2 \ll 1$ (but $d \gg a_0$), we get [cf. (13)]

$$
|E_n| \approx \frac{e^2}{32a_0 n^2} \left[1 - \frac{2}{\pi n} \left(\frac{2d}{a_0} \right)^{1/2} \right].
$$
 (30)

The solid curves in Fig. 1 represent the dependences of the frequencies of transition f_{12} and f_{13} on d/a_0 , calculated on the basis of (29). As we see, in the region of comparatively large thicknesses $(d/a_0 > 100)$ where the asymptotic expressions (12) and (13) are valid, the

quasiclassical functions f_1 **, (d)** are practically identical **with the exact solution of the problem (the dashed curves). It should be expected that in the case of helium films of small thickness, the quasiclassical approximation should give good results, as in the problem of** the clamping field (6) at $d \gg \langle x_{n} \rangle$.

At $\mathscr{C}_1 \neq 0$, calculations according to formulas (27) and (28) show that for films with $d \le 100 \text{ Å}$, the dependence of f_{2n} on \mathscr{E}_1 is sufficiently weak so that the effect of the compressing field under the condition $\mathscr{E} \ll \frac{e}{4d^2}$ can be **taken into account by means of perturbation theory.**

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- ¹⁾ Two-dimensional helium films condensed on Grafoil are an e^{α} ception. 6
- ²⁾ Everything that follows is valid for an arbitrary dielectric film with $\epsilon - 1 \ll 1$.
- ³⁾ Integer values of x_n correspond to the Coulomb problem; here U goes over into Laguerre polynomials.⁸
- ⁴⁾ It is assumed here that $k_n d \gg 1$, so that Ψ is exponentially small at $x = -d$.
- ⁵⁾ Compare this with the quadratic Stark effect for atomic hydrogen.¹⁰
- 6) The helium-surface instability that arises in sufficiently strong fields is not taken into account here (see below).
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