Ripplonic Lamb Shift for Electrons on Liquid Helium

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We study the shift of the energy levels of electrons on a helium surface due to the coupling to the quantum field of surface vibrations. As in quantum electrodynamics, the coupling is known, and it is known to lead to an ultraviolet divergence of the level shifts. We show that there are diverging terms of different nature and use the Bethe-type approach to show that they cancel each other, to leading order. This resolves the long-standing theoretical controversy and explains the existing experiments. The results allow us to study the temperature dependence of the level shift. The predictions are in good agreement with the experimental data, with no adjustable parameters.

Electrons above the surface of liquid helium were one of the first observed two-dimensional electron systems (2DESs) [1–4]. In this system, the conceptual simplicity is combined with far-from-trivial behavior, which allows studying many-body effects in a well-characterized setting. The system displays the highest mobility known for 2DESs, exceeding 2×10^8 cm^2/(V s) [5] and can be exquisitely well controlled [6]. The electron-electron interaction is typically strong, so that the electrons can form a Wigner solid [7,8] or a strongly correlated liquid with unusual transport properties [9,10]. A number of new many-electron phenomena have been found recently [11–15].

An advantageous feature of the system is the simple shape of the confining potential. It is formed by the high Pauli barrier at the helium surface and the image potential. One then expects the electron energy spectrum to be well understood. Indeed, already the first experiment on transitions between the sub-bands of quantized motion normal to the surface showed a good, albeit imperfect, agreement with the model [4]. Much work has been done on improving, sometimes empirically, the form of the confining potential, cf. Refs. [4,16–19]. On the other hand, it has been known that the electrons are also coupled to a bosonic field, the capillary waves on the surface of helium (ripplons), and that this coupling affects the electron energy spectrum [20–24]. The importance of this effect was demonstrated in explaining the Wigner crystallization [8,25] and through cyclotron resonance measurements [26].

In terms of the coupling to a bosonic field, electrons on helium are a condensed-matter analog of systems studied in quantum electrodynamics (QED). In both cases, the spectrum of the bosons and the coupling are known [27], and for electrons on helium the coupling can be controlled. The parallel with QED is further strengthened by the full

quantitative agreement between a large number of experiments on electron transport and the parameter-free theory that accounts for the coupling and also for many-electron effects. This is why the difference between the theoretical and experimental values of the sub-band energy spacing has been a concern. Another significant concern is the well-established experimentally slow energy relaxation of strongly excited electrons. So far, it has been described either phenomenologically or by modifying the form of the electron-ripplon coupling [28].

A major and largely overlooked problem closely related to those mentioned above is the ripplon-induced shift of the energy levels of electron motion normal to the surface. The lowest-order expression is ultraviolet divergent. The divergence is strong, as a high power of the wave number. Unless one deals with it carefully, the resulting level shifts become comparable to the "bare" electron binding energy already for a short-wavelength cutoff approaching twice the interatomic distance. Besides electrons on helium, a similar problem emerges in the analysis of 2D electron systems in semiconductor heterostructures and metal-oxide-semiconductor systems with short-range correlated interface roughness and a high surface barrier. The approach developed below can be extended to such systems as well.

In this Letter, we show that, in fact, the level shift due to the coupling to ripplons is small. The situation is reminiscent of the Lamb shift in QED. In the spirit of the Bethe approach to the analysis of the Lamb shift, we show that, in the electron-ripplon problem, there are two groups of diverging terms, and the leading ultraviolet-divergent terms are compensated in the overall level shift. Interestingly, the remaining correction still displays a power-law ultraviolet divergence, but with a smaller exponent. This shows the nontrivial nature of the compensation. Once account is taken of the natural cutoff at the interatomic distance, the

correction becomes small. It describes a Lamb-shift-type deviation from the energy spectrum in the absence of the electron-ripplon coupling.

We also study the temperature-dependent shift of the energy levels. We find a good agreement with the experimental results on the spectra of inter-sub-band transitions [29,30] obtained by studying resonant microwave absorption in a broad range of temperatures and electron densities. The theory also explains the long lifetime of the electron states, which is critical for the potential implementation of a quantum computer based on electrons on helium [31–33]. The proposed divergence compensation mechanism is fairly general for quasi-two-dimensional systems.

The contributions to the level shift due to the warping of the helium surface, i.e., to the ripplons, can be separated into three parts. One is kinematic and comes from the electron kinetic energy over a warped surface. Another is electrostatic and comes from the difference of the electron potential energies above the plane and a warped surface. The third comes from the inertia of the surface waves. The key point of the analysis of these contributions is that we have to go beyond the standardly used polaron theory in which the electron system is assumed to be two dimensional. It is necessary to take into account the ripplon-induced mixing of the different states of motion normal to the surface.

The kinematic level shift is the major one. To see how it comes about, we choose $\mathbf{r} = (x, y)$ and $\mathbf{p} = (p_x, p_y)$ as the two-dimensional coordinate and momentum of motion parallel to the surface; the coordinate z is normal to the surface, and the electron motion along z is quantized; see Fig. 1(a). Qualitatively, one can think that the nonuniform surface displacement $\xi(\mathbf{r})$ changes the effective width of the potential well in the z direction. This changes the
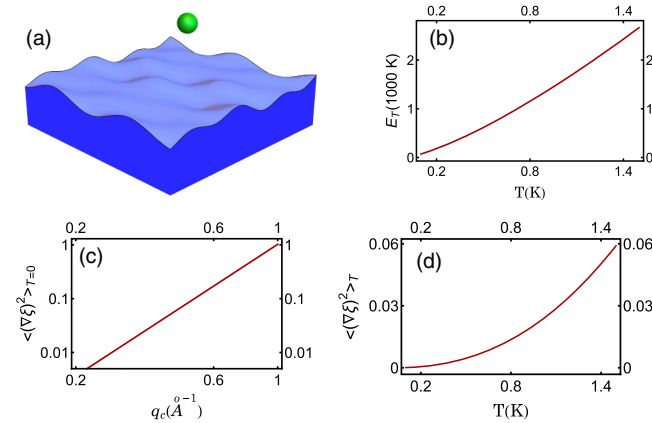

FIG. 1. (a) Sketch of an electron above a warped helium surface. (b) Electron energy $E_T = \hbar^2 q_T^2 / 2m$ for a thermal ripplon wave number q_T, $\hbar\omega_{q_T} = k_B T$. (c) The relative correction to the reciprocal electron mass due to the direct kinematic two-ripplon coupling for $T = 0$ as function of the short-wavelength cutoff q_c. (d) Same as in (c), but when only the thermal contribution is taken into account.

kinetic energy of the confined motion. Since $\langle \xi(\mathbf{r}) \rangle = 0$, the change is quadratic in ξ—and in fact, in $\nabla\xi$—because a uniform surface displacement does not change the energy. The energy change is positive, since the kinetic energy scales as the squared reciprocal confinement length.

On the other hand, the motions parallel and normal to the surface are mixed by the warping. In the second order (again, quadratic in ξ), the mixing leads to shifts of the levels of motion normal to the surface, which are negative for low-lying levels, as expected from the standard perturbation theory. It turns out that, taken separately, both mechanisms display strong ultraviolet divergences of the energy shift, which partly compensate each other.

The warping-induced change of the electrostatic energy has linear and quadratic in $\xi(\mathbf{r})$ terms as well. The shift of the electron energy levels due to the quadratic term also displays an ultraviolet divergence, but it is weaker than for the kinematic mechanism. This is because the image potential is much less sensitive to short-wavelength fluctuations of the helium surface. The divergence is partly compensated by the single-ripplon processes, as for the kinematic coupling. The analysis is similar to the analysis of the kinematic effect given below and is provided in the Supplemental Material [34,35].

Our starting point is the electron Hamiltonian for a flat helium surface,

$$H_0 = \hat{T} + V(z), \qquad \hat{T} = (2m)^{-1}(\mathbf{p}^2 + p_z^2), \quad (1)$$

where the potential energy $V(z)$ for $z > 0$ comes from the image force and from the electric field $E_\perp$ usually applied to press the electrons to the surface [27,28]. The potential has an atomically steep repulsive barrier at $z = 0$ with height > 1 eV formed by helium atoms. The eigenstates of Hamiltonian (1) are products of the wave functions of quantized motion normal to the surface $|n\rangle \equiv \psi_n(z)$ and the plane waves of lateral motion $\propto \exp(i\mathbf{p}\mathbf{r}/\hbar)$. The energies of the normal and lateral motions are E_n and $E_\mathbf{p}$, and the total energy is $E_{n\mathbf{p}} = E_n + E_\mathbf{p}$.

The ripplon Hamiltonian and the ripplon-induced surface displacement are

$$\hat{H}_r = \hbar \sum_\mathbf{q} \omega_q b_\mathbf{q}^\dagger b_\mathbf{q}, \quad \xi(\mathbf{r}) = \sum_\mathbf{q} Q_q e^{i\mathbf{q}\mathbf{r}}(b_\mathbf{q} + b_{-\mathbf{q}}^\dagger). \quad (2)$$

Here, $b_\mathbf{q}$ is the annihilation operator of a ripplon with the wave number $\mathbf{q}$ and frequency ω_q; $Q_q = (\hbar q / 2\rho\omega_q S)^{1/2}$, where ρ is the helium density and S is the area [36].

The effect of the ripplon-induced curvature of the electron barrier at the helium surface can be taken into account in a standard way [37] by making a canonical transformation $U = \exp[-i\xi(\mathbf{r})p_z/\hbar]$, which shifts the electron z coordinate so that it is counted off from the local position of the surface, $z \to z - \xi(\mathbf{r})$ [21]. The transformed electron kinetic energy and the ripplon energy

$U^\dagger(\hat{T} + \hat{H}_r)U$ is the sum $\hat{T} + \hat{H}_r + \hat{H}_i^{(1)} + \hat{H}_i^{(2)}$, where $\hat{H}_i^{(1)}$ and $\hat{H}_i^{(2)}$ describe the linear and quadratic in $\xi(\mathbf{r})$ kinematic electron-ripplon couplings:

$$\hat{H}_i^{(1)} = -\frac{1}{2m}p_z\{\mathbf{p}, \boldsymbol{\nabla}\xi(\mathbf{r})\}_+ + i\hbar^{-1}p_z[\xi(\mathbf{r}), H_r],$$

$$\hat{H}_i^{(2)} = p_z^2 h_i^{(2)}, \quad h_i^{(2)} = \frac{1}{2m}(\boldsymbol{\nabla}\xi)^2 + \sum_{\mathbf{q}}\omega_q|Q_q|^2/\hbar, \quad (3)$$

$[\{A, B\}_+ = AB + BA, \boldsymbol{\nabla} = (\partial_x, \partial_y)].$

The term $\hat{H}_i^{(2)}$ averaged over the thermal distribution of ripplons gives a correction to the electron energy E_n already in the first order,

$$\Delta E_n^{(2)} = \langle n|p_z^2|n\rangle\langle h_i^{(2)}\rangle. \quad (4)$$

This correction is just a renormalization of the electron mass for motion normal to the surface $m^{-1} \to m^{-1} + \sum_{\mathbf{q}}|Q_q|^2[2m\omega_q + \hbar q^2(2\bar{n}_q + 1)]/m\hbar$, where $\bar{n}_q = [\exp(\hbar\omega_q/k_BT) - 1]^{-1}$ is the Planck number. Since $\omega_q \propto q^{3/2}$ for large q [38], the sum over $\mathbf{q}$ diverges as $q_c^{7/2}$ for $T = 0$, where q_c is the short-wavelength cutoff. Figure 1(c) shows the factor $\langle(\boldsymbol{\nabla}\xi)^2\rangle$ as a function of the short-wavelength cutoff q_c. If we set $q_c = 1$ Å^{-1} [39], the energy shift (4) is $\Delta E_n^{(2)} \sim E_n$, indicating the breakdown of the perturbation theory.

Operator $\hat{H}_i^{(1)}$ contributes to the level shift in the second order. For a state with given quantum numbers n and $\mathbf{p}$, the shift is

$$\Delta E_{n\mathbf{p}}^{(1)} = \sum_{n',\mathbf{q}}\sum_{\alpha=\pm1}\mathcal{N}_{q\alpha}\frac{|\langle n|p_z|n'\rangle|^2\Delta_{\mathbf{p},\mathbf{q},\alpha}^2}{E_n - E_{n'} - \Delta_{\mathbf{p},\mathbf{q},\alpha}}. \quad (5)$$

Here, α allows for the processes with virtual emission ($\alpha = 1$) or absorption ($\alpha = -1$) of a ripplon, $\Delta_{\mathbf{p},\mathbf{q},\alpha} = E_{\mathbf{p}+\hbar\mathbf{q}} - E_\mathbf{p} + \alpha\hbar\omega_q$ is the difference of the energies of the in-plane motion in the initial and intermediate electron states with the added or subtracted ripplon energy, and $\mathcal{N}_{q\alpha} = |Q_q|^2[\bar{n}_q + (1 + \alpha)/2]/\hbar^2$.

We find the overall kinematic energy shift $\Delta E_{n\mathbf{p}}^{\mathrm{kin}} = \Delta E_{n\mathbf{p}}^{(1)} + \Delta E_n^{(2)}$ by rewriting in Eq. (5)

$$\frac{\Delta_{\mathbf{p},\mathbf{q},\alpha}}{E_n - E_{n'} - \Delta_{\mathbf{p},\mathbf{q},\alpha}} = \frac{E_n - E_{n'}}{E_n - E_{n'} - \Delta_{\mathbf{p},\mathbf{q},\alpha}} - 1. \quad (6)$$

The last term here exactly cancels $\Delta E_n^{(2)}$ in $\Delta E_{n\mathbf{p}}^{\mathrm{kin}}$, since $\sum_{n'}|\langle n|p_z|n'\rangle|^2 = \langle n|p_z^2|n\rangle$ and Q_q, ω_q are independent of the direction of $\mathbf{q}$. Then

$$\Delta E_{n\mathbf{p}}^{\mathrm{kin}} = \sum_{n',\mathbf{q},\alpha}\mathcal{N}_{q\alpha}\frac{\Delta_{\mathbf{p},\mathbf{q},\alpha}|\langle n|p_z|n'\rangle|^2(E_n - E_{n'})}{E_n - E_{n'} - \Delta_{\mathbf{p},\mathbf{q},\alpha}}. \quad (7)$$

We will be interested in $\Delta E_{n\mathbf{p}}^{\mathrm{kin}}$ for low-lying out-of-plane states, $n \sim 1$, and for small momenta $p \lesssim (mk_BT)^{1/2}$.

The unperturbed energies E_n, $E_{n'}$ and the matrix elements $\langle n'|p_z|n\rangle$ in Eq. (7) can be found from the one-dimensional Schrödinger equation for an electron above the flat helium surface. To get an analytic insight, we note that the main contribution to Eq. (7) comes from large in-plane wave numbers q compared to the reciprocal out-of-plane localization length r_B^{-1} in the ground state $n = 1$ ($r_B \lesssim 100$ Å [36,40]). The energy $\Delta_{\mathbf{p},\mathbf{q},\alpha}$ largely exceeds k_BT and $|E_n|$ with $n \sim 1$. Then, of primary importance is the contribution to Eq. (7) of highly excited states with $n' \gg 1$. Such states are semiclassical. They correspond to electron motion in an almost triangular potential well formed by the barrier at $z = 0$ and the field $eE_\perp$ that presses the electrons against the surface. The Wentzel-Kramers-Brillouin (WKB) approximation gives $E_{n'} \approx [3\pi\hbar eE_\perp(n' - 1/4)/2\sqrt{2m}]^{2/3}$ for $n' \gg 1$ (a better approximation is based on matching the WKB and the small-z wave functions [35]). Since the large-n' wave functions are fast oscillating on length r_B, one can show that $|\langle n'|p_z|n\rangle|^2 \approx (\hbar^4 eE_\perp/2mE_{n'}^2)|\partial_z\psi_n|_{z=0}^2$.

Changing from summation over n' in Eq. (7) to integration, we obtain from the above estimate

$$\Delta E_{n\mathbf{p}}^{\mathrm{kin}} \approx \frac{\hbar}{\sqrt{2m}}|\partial_z\psi_n|_{z=0}^2\sum_{\mathbf{q}}|Q_q|^2(2\bar{n}_q + 1)\Delta_{\mathbf{p},\mathbf{q},1}^{1/2}. \quad (8)$$

We disregarded here the contribution of the states with energies $E_{n'} \lesssim E_n$. We also disregarded the ripplon energy $\hbar\omega_q$ compared to the in-plane electron energy $E_{\hbar\mathbf{q}}$. This is a good approximation, because ripplons are slow, $\omega_q \approx (\sigma/\rho)^{1/2}q^{3/2}$, where σ is the helium surface tension. Therefore, their thermal momentum q_T given by condition $\hbar\omega_{q_T} = k_BT$ corresponds to the electron energy $E_{\hbar q_T}$ varying from ≈ 73 K to $\approx 1.6 \times 10^3$ K for T varying from 0.1 to 1 K; see Fig. 1(b).

The $T = 0$ term in Eq. (8) still has an ultraviolet divergence. It scales with the short-wavelength cutoff q_c as $q_c^{5/2}$. This is a much weaker divergence than that of $\Delta E_n^{(2)}$. Overall, the $T = 0$ term is $\sim\Delta E_n^{(2)}/(q_cr_B)$. For the cutoff $q_c \sim 1$ Å^{-1}, this term is on the order of a few percent of the electron binding energy $|E_1| = \hbar^2/2mr_B^2 \sim 8$ K. Its dependence on the control parameter $E_\perp$ is described by Eq. (7). There are also other contributions to the $T = 0$ level shift. They include the effect of the electron correlations [11,41], the intraband polaronic shift [22], and, last but not least, the finite steepness and height of the barrier for electron penetration into the liquid helium [4,16–19]. It is important that, as it follows from the previous work and

from Eq. (8), all contributions to the $T = 0$ level shift are small.

Understanding the experiment requires finding the T-dependent part of the shift of the electron energy levels. For $T \gtrsim 10$ mK, the kinematic contribution (7) is the dominating part of this shift. From Fig. 1(d), this shift is small even before the renormalization. However, it is directly observable. In the approximation (8), the T-dependent part $\Delta E_{n\mathbf{p}T}^{\mathrm{kin}}$ of the kinematic level shift is

$$\Delta E_{n\mathbf{p}T}^{\mathrm{kin}} \approx A_n^{\mathrm{kin}} (k_B T)^{5/3}. \qquad (9)$$

The coefficient $A_n^{\mathrm{kin}} \approx c^{\mathrm{kin}} |\partial_z \psi_n|_{z=0}^2 (\hbar^4 \rho / \sigma^4)^{1/3} / m$ [$c^{\mathrm{kin}} = \Gamma(5/3)\zeta(5/3) \approx 0.1$] sensitively depends on the electron state n. Equation (9) predicts a power-law dependence of the level shift on T with exponent $5/3$.

The dependence of the energy renormalization on the level number n leads to a temperature-dependent shift of the peaks of microwave absorption due to $n \to n'$ transitions. Since the characteristic ripplon momenta $\hbar q$ in Eq. (7) largely exceed the thermal electron momentum, the level shift is essentially independent of the electron momentum $\mathbf{p}$.

In Fig. 2, we present the results for the thermally induced shift $\Delta \omega_{21}$ of the transition frequency $\omega_{21} = (E_2 - E_1)/\hbar$. It is calculated as a sum of the kinematic contribution (7) and the contribution from the electrostatic coupling given in the SM, keeping only the T-dependent terms in both expressions. To compare the theory with the experiment, the experimentally measured transition frequency [30] was extrapolated to $T = 0$, and the shift was counted off from the extrapolated value. The calculated T-dependent frequency shift is free from the ambiguity related to the form of the electron potential at the atomic distance from the helium surface.

The theoretical curve in Fig. 2 is in excellent agreement with the experimental data shown by squares, with no
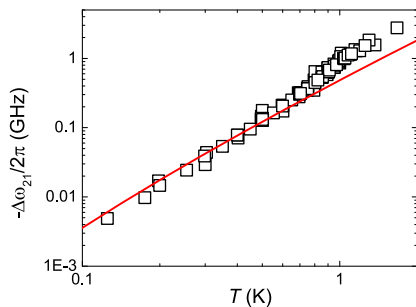

FIG. 2. Temperature dependence of the $1 \to 2$ transition frequency calculated for $E_\perp = 106$ V/cm (solid line). The squares are the experimental results [30]. In the studied density range 0.67×10^7–2.4×10^7 cm^{-2}, no dependence on the electron density was found, and the data for different densities are combined.

adjustable parameters. A deviation is observed only for $T \gtrsim 1$ K, where scattering by helium vapor atoms becomes substantial. The simple expression (9) gives $\Delta\omega_{21}$, which differs from the numerical result by a factor ~ 3.

Two-ripplon coupling has been attracting much interest as a mechanism of electron energy relaxation [28,32, 42–44]. Its importance is a consequence of the slowness of ripplons, which makes single-ripplon scattering essentially elastic. In contrast, for two-ripplon scattering, the total wave number of the participating ripplons $|\mathbf{q}_1 + \mathbf{q}_2|$ can be of the order of the reciprocal electron thermal wavelength or the reciprocal magnetic length, whereas the wave number of each ripplon $q_{1,2}$ can be much larger, so that the ripplon energies $\hbar\omega_{q_1} \approx \hbar\omega_{q_2}$ can be comparable to $k_B T$, the inter-sub-band energy gap $|E_n - E_{n'}|$, or the Landau level spacing.

From Eq. (3), the matrix element of an electron transition $|n, \mathbf{p}\rangle \to |n', \mathbf{p}'\rangle$ calculated for the direct kinematic two-ripplon coupling $\hat{H}_i^{(2)}$ is $\propto q_1 q_2 \approx -q_1^2$. It implies a high rate of deeply inelastic electron relaxation for large $q_{1,2}$. The single-electron kinematic coupling to ripplons $\hat{H}_i^{(1)}$ very strongly reduces the scattering rate. An approach similar to that used in the analysis of the level shifts allows one to show that the term $\propto q_1 q_2$ drops out from the transition matrix element calculated to the second order in $\hat{H}_i^{(1)}$. A similar cancellation occurs for the electrostatic electron-ripplon coupling [35]. This strongly reduces the energy relaxation rate, bringing it within the realm of the experiment. The full analysis of the electron energy relaxation requires also taking into account scattering by phonons in helium [32]. This analysis is beyond the scope of the present Letter.

In conclusion, we have shown that the system of electrons coupled to the quantum field of capillary waves on the helium surface enables studying a condensed-matter analog of the Lamb shift, which in this case is the shift of the sub-bands of the quantized motion transverse to the surface. As in the case of the Lamb shift, the matrix elements of the electron coupling to the quantum field are known, and there are terms in the expression for the shift that display an ultraviolet divergence. We have shown that the analysis may not be limited to the conventional intra-sub-band processes. We have revealed the diverging inter-sub-band terms and used the Bethe trick from the Lamb shift theory to demonstrate that different diverging terms cancel each other to the leading order, making the overall shift small. The considered system makes it possible to study the dependence of the level shift on temperature. Our theoretical results are in excellent agreement with the experimental observations.

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