

Inelastic neutron scattering as an indication of a new type of gapped surface excitations in liquid helium

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Plan of the talk:

1. **Introduction.** Puzzle of strong temperature dependence of surface tension. Surfons.
2. Microscopic model of surfons as surface bound states of atoms or molecules [1,4].
3. Experimental indications of surfon existence: temperature dependence of surface tension [4], mobility of electrons on the surface of He and H₂ [2], temperature shift and broadening of electron transition line on the surface of helium and hydrogen [3].
4. **Inelastic neutron scattering as a possible direct observation of surfons**

[1] A.D. Grigoriev, A.M. Dyugaev, P.D. Grigoriev, J. Low Temp. Phys. 163, 131–147 (2011)

[2] P.D. Grigoriev, A.M. Dyugaev, E.V. Lebedeva, JETP 104 (1), p. 1-10 (2008)

[3] P. D. Grigor'ev, A. M. Dyugaev and E. V. Lebedeva, JETP Lett. 87, 106 (2008)

[4] A.M. Dyugaev, P.D. Grigoriev, JETP Lett.78(7), 466 (2003).

[5] A.D. Grigoriev, P.D. Grigoriev, A.M. Dyugaev, A.F. Krutov, Low Temp. Phys. 38, 1005-1012 (2012)

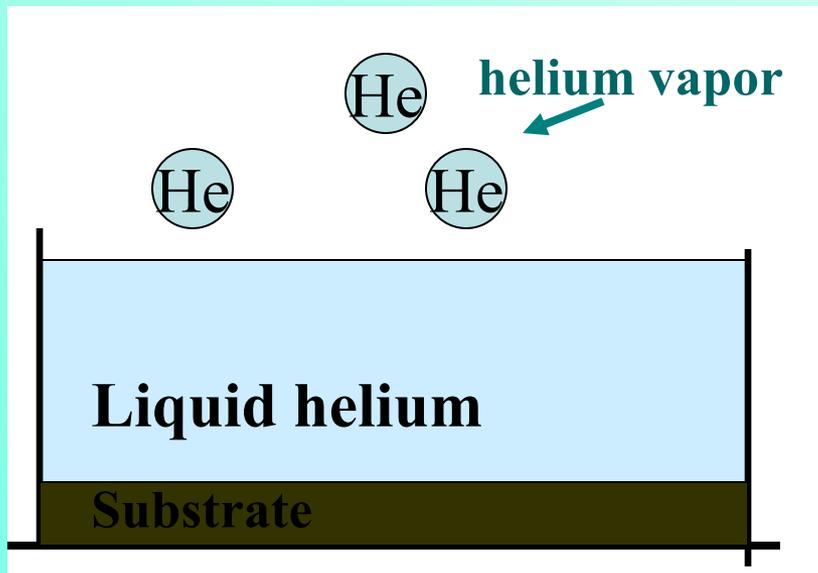
[6] P.D.Grigoriev, A.D.Grigoriev, A.M.Dyugaev, JETP 128, 297 (2019) [ZhETF 155, 338 (2019)]

Liquid helium

Helium is the only liquid at $T=0$. Boiling temperatures of ${}^4\text{He}$ $T_4=4.2\text{K}$, ${}^3\text{He}$ $T_3=3.2\text{K}$. It is the most clean liquid, because all impurities are stick to the substrate and walls by the van der Waals force. Only quanta of surface waves (rippions) and helium vapor are present.

The concentration of He vapor is very small at low temperature:

$$N(T) \propto \exp\left(-\frac{\Delta E}{k_B T}\right), \Delta E = 7.17\text{K}.$$



The dispersion relation of ripples

$$\omega_q^2 = \frac{\sigma_{He}}{\rho_{He}} q(q^2 + \kappa^2) \tanh(qd),$$

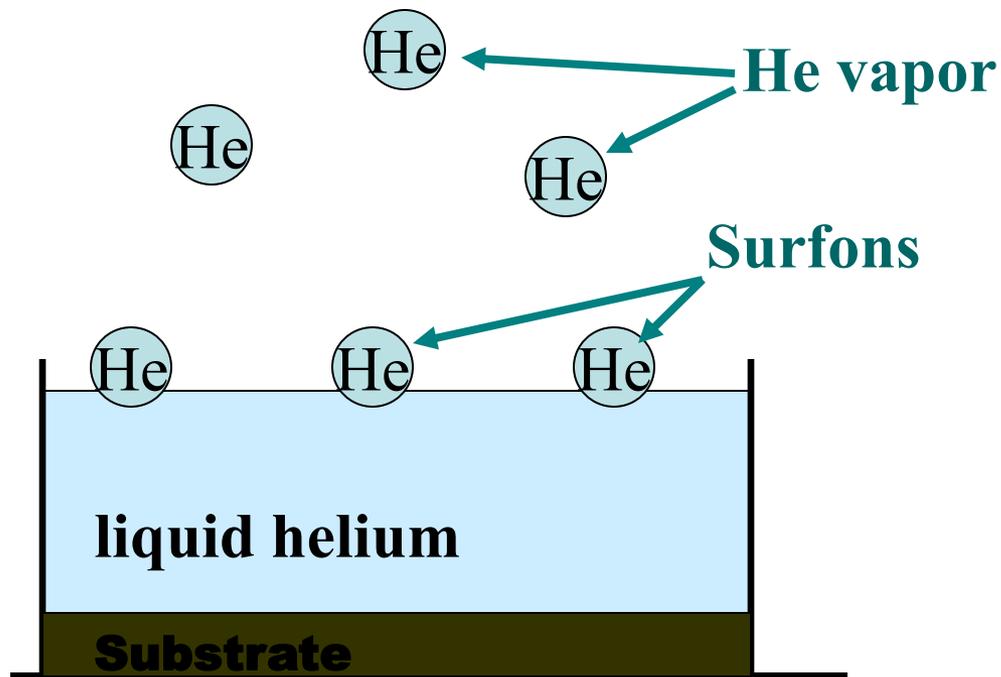
where density of He4: $\rho_{He4} = 0.147\text{g/cm}^3$,

d – liquid depth (width of He film),

surface tension coefficient

$$\sigma_{He4} = 0.36\text{dyn/cm}, \quad \kappa = 20\text{cm}^{-1}.$$

Atoms on the surface quantum level (surfons)



The temperature dependence of the surface tension $\Delta\sigma(T)$ of both He isotopes can be explained if introduce a new type of excitations – surfons.

Surfons appear because there is a bound state of an atom at the surface, which has lower energy than He vapor and can propagate along the surface

A.M. Dyugaev and P.D. Grigoriev, JETP Lett.78, 466 (2003).

A.D. Grigoriev, P.D. Grigoriev, A.M. Dyugaev, J. Low Temp. Phys. 163, 131–147 (2011)

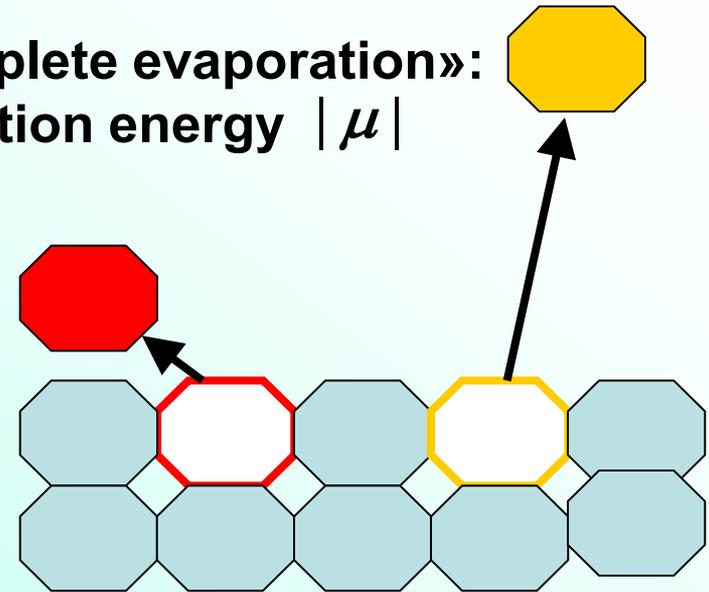
Surfon idea is not entirely new

Solid-vapor equilibrium

«Incomplete evaporation»:
activation energy $\Delta < |\mu|$

J. I. Frenkel, Z. Phys. 26, 37 (1924).

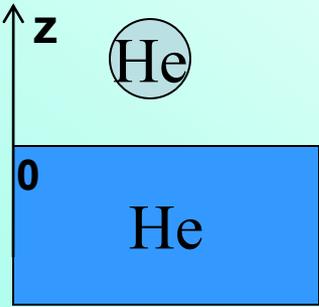
«Complete evaporation»:
activation energy $|\mu|$



Surfons also resemble the Andreev states of ^3He atoms on the surface of ^4He [A.F. Andreev, JETP 23, 939 (1966)], introduced to explain surface tension of ^3He - ^4He solution

Introduction

Estimate of surfon energy.

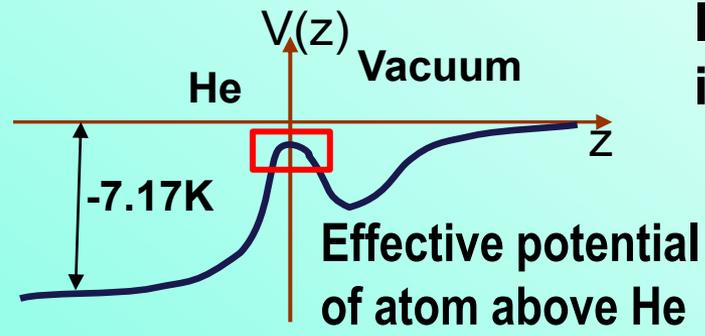


The Lennard-Jones potential between two He atoms

$$v(r) = 4\epsilon_0 \left[\left(\frac{\sigma_0}{r} \right)^{12} - \left(\frac{\sigma_0}{r} \right)^6 \right] \quad (1)$$

For He4 $V_0 = 140K$ $\sigma_{0*} = 2.09A$

Integration of (1) over half-space $z < 0$ gives the interaction of He atoms with flat liquid surface



$$V(z) = V_0 \left[\left(\frac{\sigma_{0*}}{z} \right)^9 - \left(\frac{\sigma_{0*}}{z} \right)^3 \right] \quad (2)$$

The solution of 1D Schrodinger equation $H\Psi = E\Psi$ with potential (2) gives the energy E of a bound state of He atom at the surface.

This energy for two He isotopes is

$$E_{s0}^{He4} \approx -1.24K, \text{ и } E_{s0}^{He3} \approx -0.342K.$$

More accurate (but semi-phenomenological) estimates of surfon energy with surface dimple give lower values but still in the interval $-7.17K < E_s < 0$

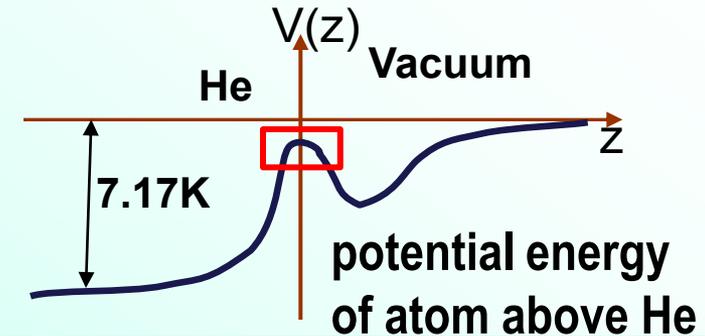
This proves the existence of surfons as quasi-stationary states

Is surfon a quasiparticle?

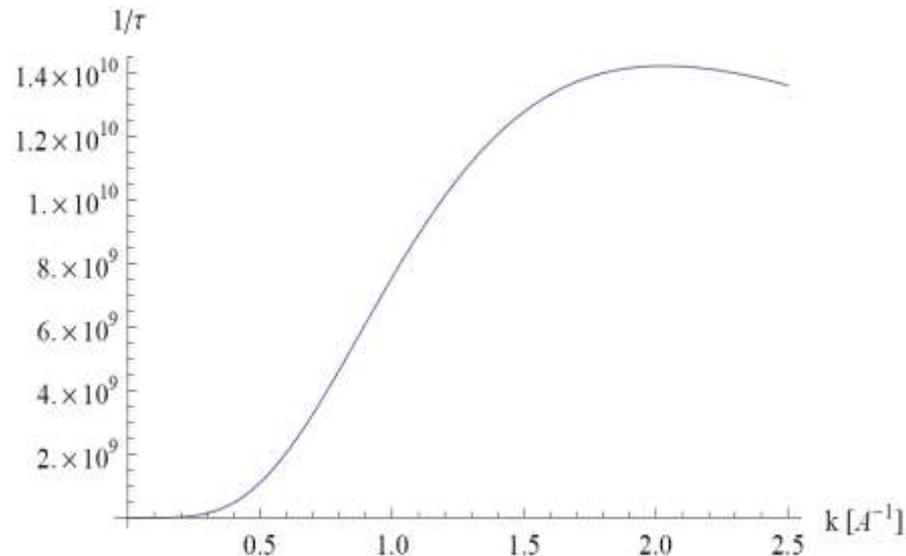
Two channels limit the surfon-quasiparticle lifetime:
(1) merging into the liquid and (2) evaporation.

1) to merge into the bulk the surfon must
(a) overcome the potential barrier;
(b) push away other atoms.

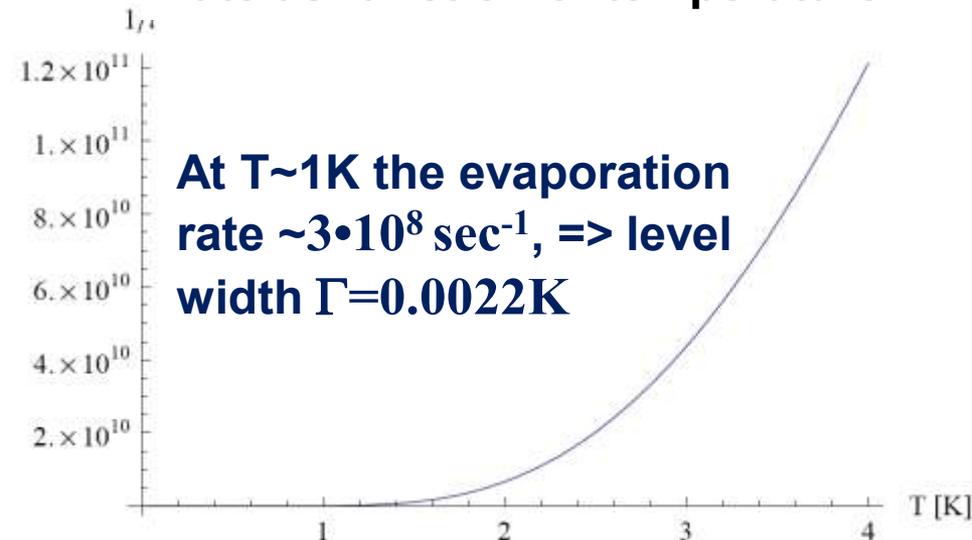
Therefore, the surfon level width $\Gamma \ll E$.



2) Surfon evaporation rate as function of its in-plane momentum at $T = 1.5\text{K}$



Average surfon evaporation rate as function of temperature



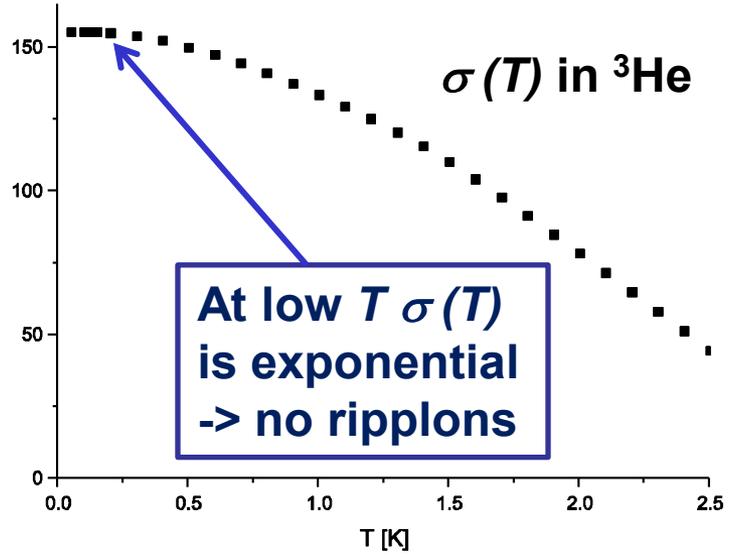
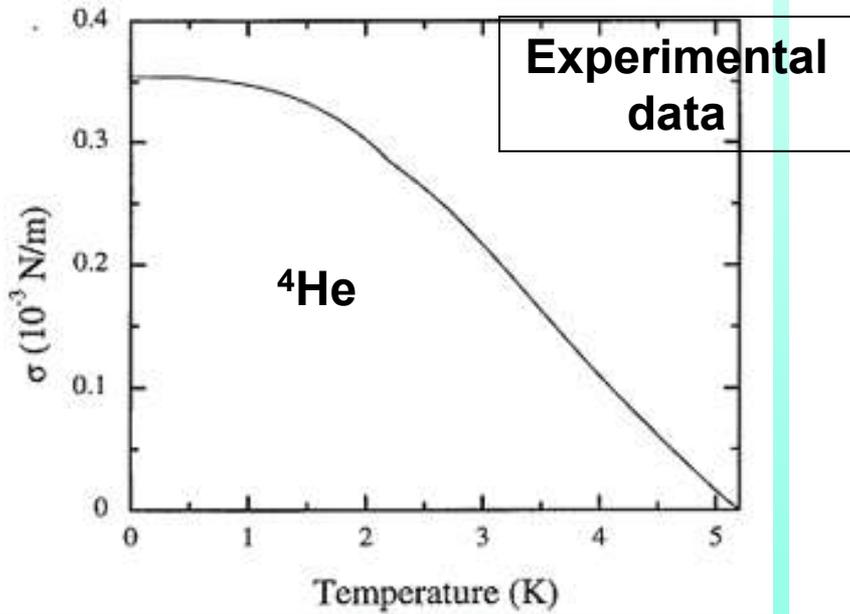
At $T \sim 1\text{K}$ the evaporation rate $\sim 3 \cdot 10^8 \text{ sec}^{-1}$, \Rightarrow level width $\Gamma = 0.0022\text{K}$

Experimental evidences of surfons are all indirect

- temperature dependence of surface tension,
- temperature shift and broadening of electron transition line on the surface of helium and hydrogen
- mobility of electrons on the surface of He

The puzzle of too strong temperature dependence of surface tension of both liquid helium isotopes

Surface tension coefficient σ is the free energy of unit area of liquid surface. At low temperature $T \rightarrow 0$ $\sigma = \sigma_0 > 0$, and the difference $\Delta\sigma(T) = \sigma(0) - \sigma(T)$ comes from all types of surface excitations. In ^4He there are ripples (surface waves), but their contribution is not enough to explain the observed $\Delta\sigma(T)$. In ^3He the large viscosity at low T damps the short-wave-length ripplon (which make the main contribution to the free energy because of large phase volume).



Origin of the temperature dependence of surface tension coefficient

The thermodynamic potential $\Omega = F - PV$ (free energy at fixed pressure). The contribution coming from surface excitations with dispersion $\varepsilon(p)$ is given by the integral over all quantum states (all momenta)

$$\Delta\Omega(T) = -T \sum_{excit} \int \frac{d^2 p}{h^2} \ln \left[1 - \exp \left(\frac{\mu - \varepsilon(p)}{T} \right) \right].$$

It turns out that in liquid He in both isotopes the known excitations are not sufficient to explain the observed strong temperature dependence of $\Delta\sigma(T)$.

Temperature dependence of surface tension of liquid ^4He & ^3He

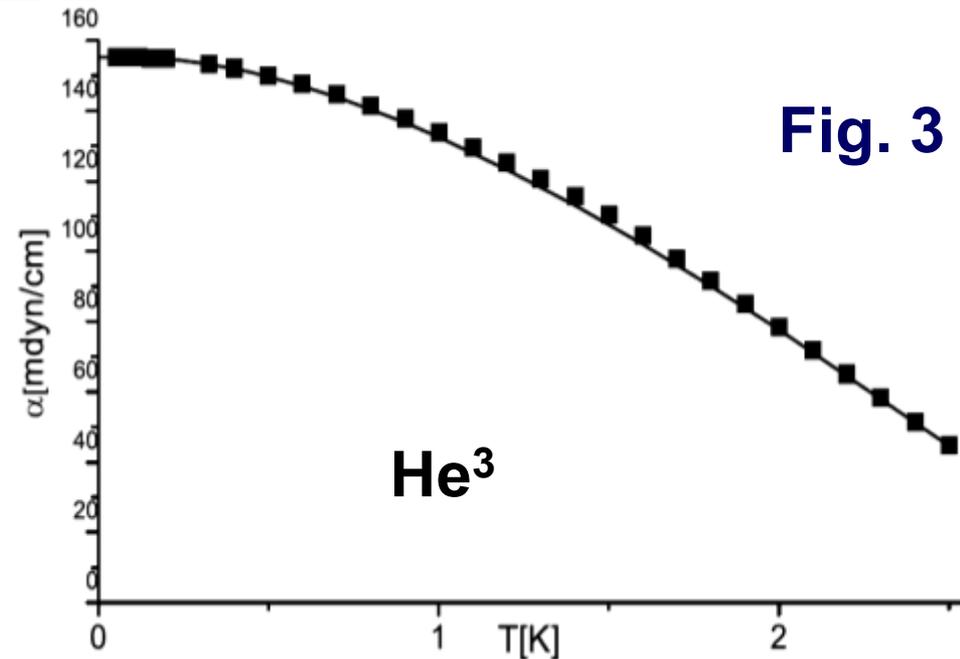
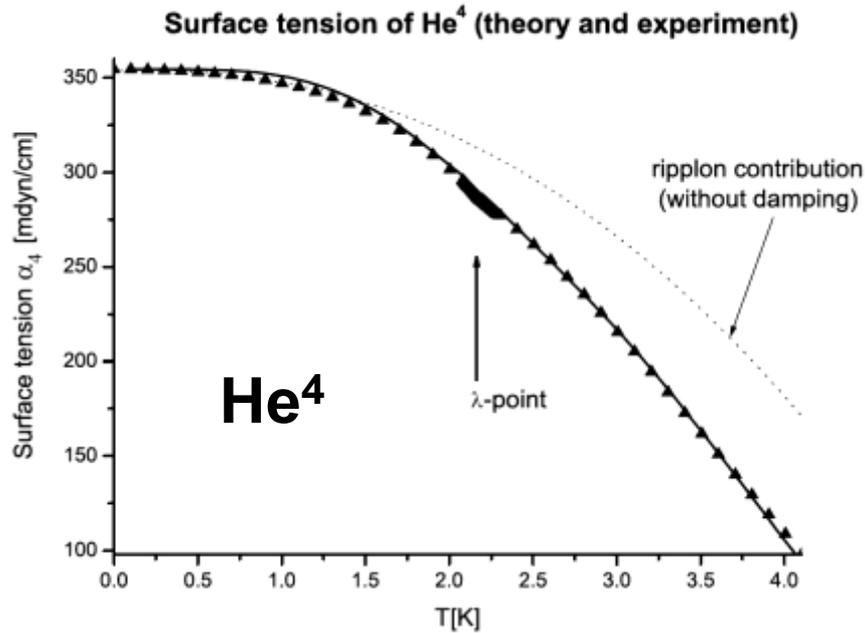


Fig. 3

There is one fitting parameter – the effective mass of surfons, which determines their DOS. Another fitting parameter – activation energy Δ is fixed by low-T part.

The best fit gives the effective masses $M_4^* \approx 2.65 M_4^0$, and $M_3^* \approx 2.25 M_3^0$,

Estimated surfon effective mass is slightly less than that giving the best fit.

$$M_4^* \approx M_4^0 (1 + n_{\text{He}4} V_{4d}) = 2.04 M_4^0.$$

$$M_3^* \approx M_3^0 (1 + n_{\text{He}3} V_{3d}) = 2.23 M_3^0.$$

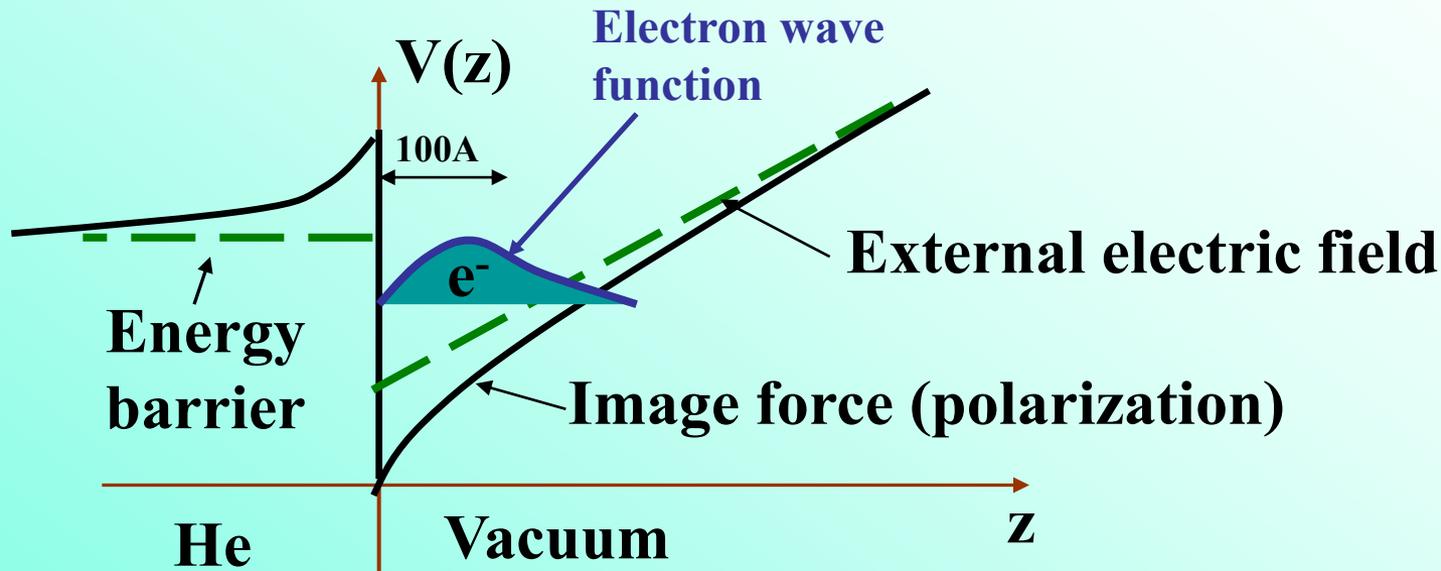
A.D. Grigoriev, A.M. Dyugaev, P.D. Grigoriev, J. Low Temp. Phys. 163, 131–147 (2011).

**Other experiments,
indicating the existence of surfons**

**Surface electrons
and their interaction with surfons**

Surface electrons

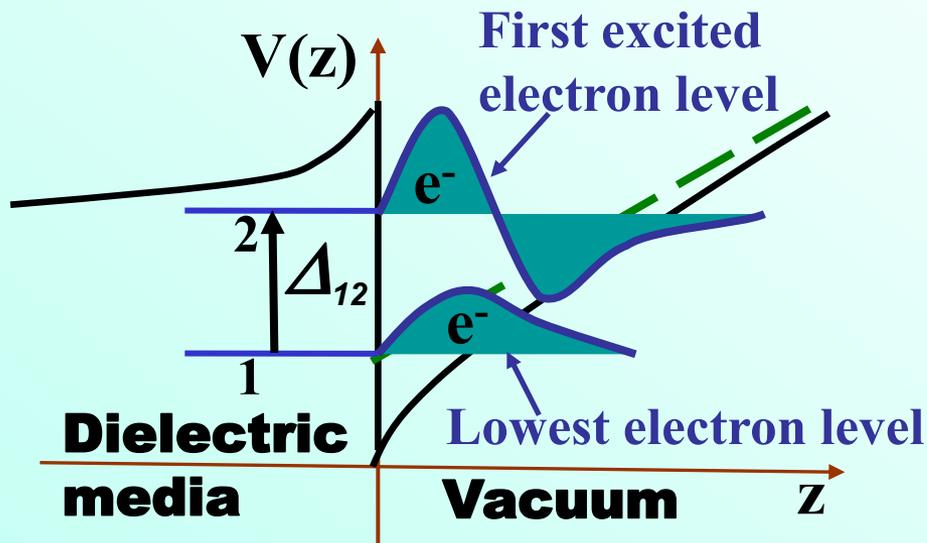
Potential energy of an electron as function of the distance to the surface:



Electrons are clamped to the surface by external electric field and by image forces.

A 2D electron gas is formed on the surface

Various energy levels of surface electrons



Electron transitions between different energy levels above the surface can be measured.

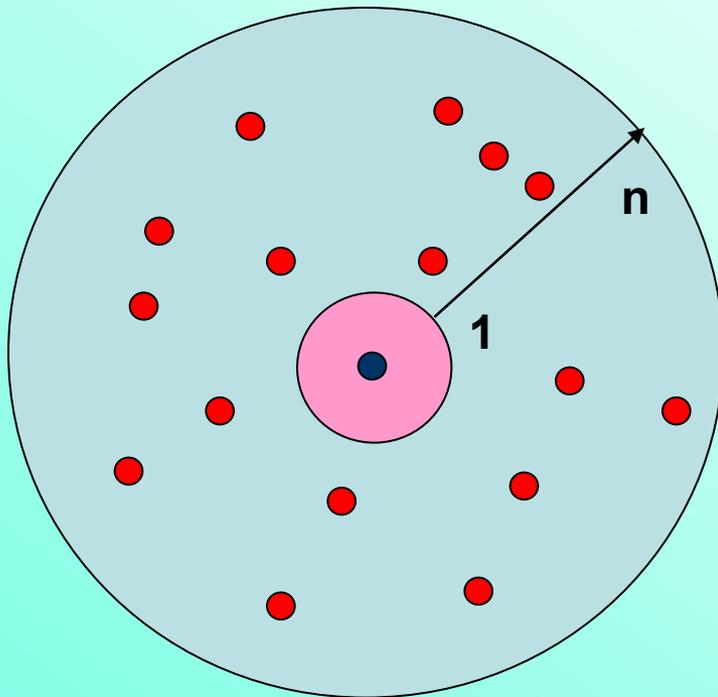
The position and width of the transition line depends on T!

[V.V. Zav'yalov, I.I. Smol'yaninov, JETP Lett. 44, 182 (1986) (electrons on H_2)]

Effect of quantum refraction

E. Fermi, Nuovo Cim. 11, 157 (1934).

Shift of electron atomic high levels in the atmosphere



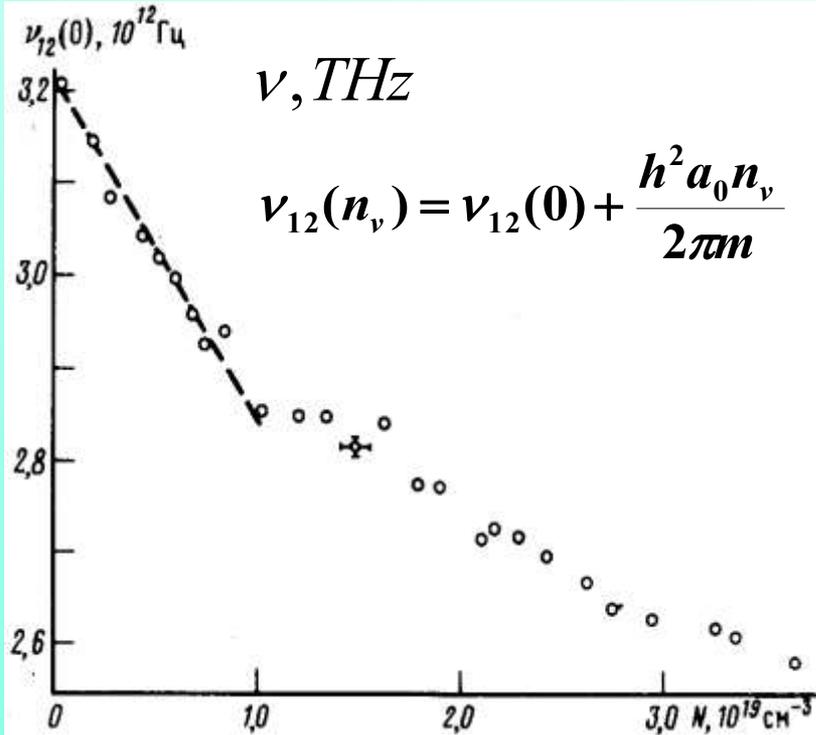
- - atom of outer gas
- - electron localization region in the ground state
- - electron localization region in the excited state

$$\Delta E_n = \frac{h^2 a_0}{2\pi m} n_V \quad \begin{array}{l} a_0 - \text{scattering amplitude} \\ n_V - \text{density of the gas} \end{array}$$

The volume of the n -th excited electron state is $V_{ne} \sim r_B^3 n^6$

$$\begin{array}{l} V_{1e} n_V \ll 1 \\ V_{ne} n_V \gg 1 \end{array}$$

Quantum transitions of electrons on the surface



Experimental data from : V.V. Zav'yalov, I.I. Smol'yaninov, JETP Lett. 44, 182 (1986); Sov. Phys. JETP 65, 194 (1987); ibid 67, 171 (1988).

The temperature dependence of the shift of the frequency of electron transition 1→2 plotted as function of the vapor density N_V

$$N(T) \propto \exp(-|\mu| / k_B T), |\mu| \approx 92K$$

does not fit the line as suggested by theoretical prediction

$$\Delta E_n = \frac{2\pi\hbar^2}{m} a_0 n_v$$

If one applies the linear fit of $\nu_{12}(n_V)$ the obtained value

$$a_0 = -1.4 \text{ \AA}$$

The accepted value, obtained from atomic scattering experiments, has different sign:

$$a_0 = 0.67 \text{ \AA}$$

This disagreement suggests that the hydrogen vapor only does not explain of the temperature shift of transition frequency

2D electrons above solid hydrogen

[V. V. Zav'yalov and I. I. Smol'yaninov, Sov. Phys. JETP 65, 194 (1987)]

● - vapor hydrogen

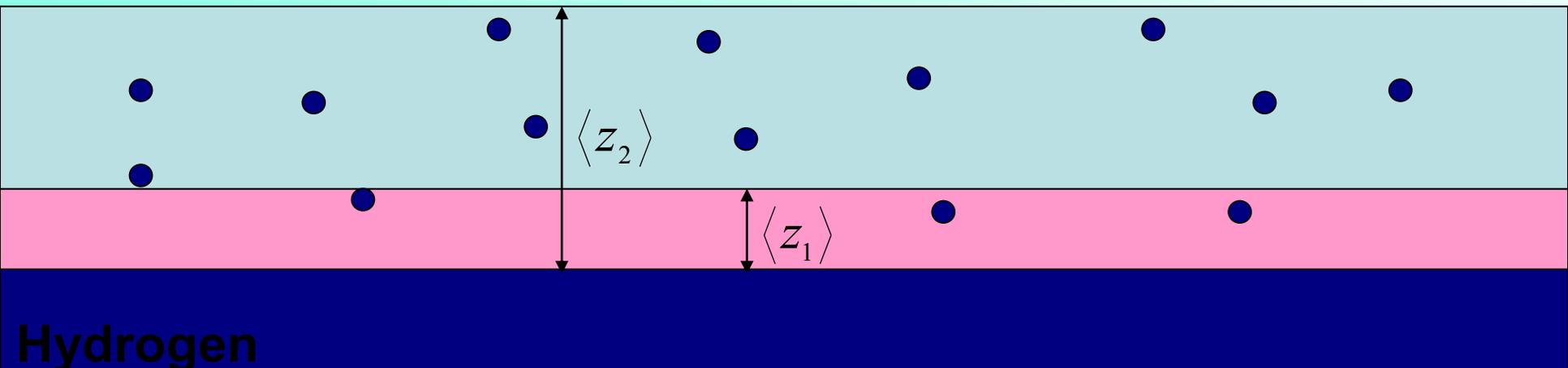
■ - solid hydrogen

■ - electron localization region in the ground state

■ - electron localization region in the 1st excited state

$$\begin{aligned} \langle z_1 \rangle &= 25A & \langle z_2 \rangle &= 101A \\ \langle R \rangle_{H_2} &= n_v^{-1/3} = 30 - 170A \\ \nu_{12}(n_v) &= \nu_{12}(0) + \hbar a_0 n_v / 2\pi m \end{aligned}$$

$$\begin{aligned} \Psi(z, \rho) &= \varphi_n(z) \exp(i\rho\rho) \\ V_{ne} &\equiv \infty \end{aligned}$$



Calculation of the shift of electron energy levels due to electron interaction with surfons

$$E_n = E_{n0} + \Delta E_n, \quad \Delta E_n = \frac{\hbar^2 a_0}{2\pi m} \int_0^\infty \varphi_{n0}^2(z) n_v(z) dz, \quad \int_0^\infty \varphi_{n0}^2 dz = 1$$

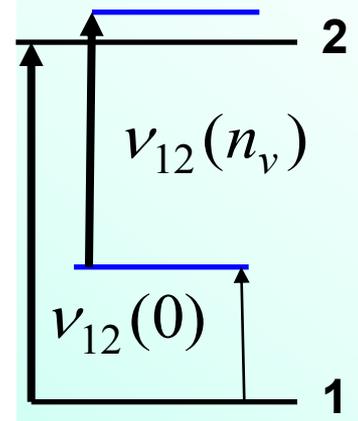
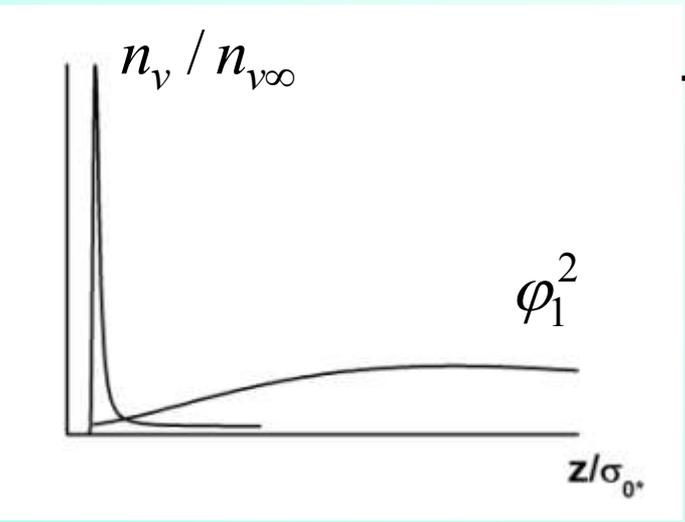
$$E_n - E_1 = E_{n0} - E_{10} - \frac{\hbar^2 a_0}{2\pi m} \int_0^\infty (\varphi_{10}^2 - \varphi_{n0}^2)(n_v(z) - n_{v\infty}) dz$$

$$E_n - E_1 = E_{n0} - E_{10} - \delta n_s$$

$$\delta = \frac{\hbar^2 a_0}{2\pi m} [\varphi_{10}^2(z_0) - \varphi_{n0}^2(z_0)]$$

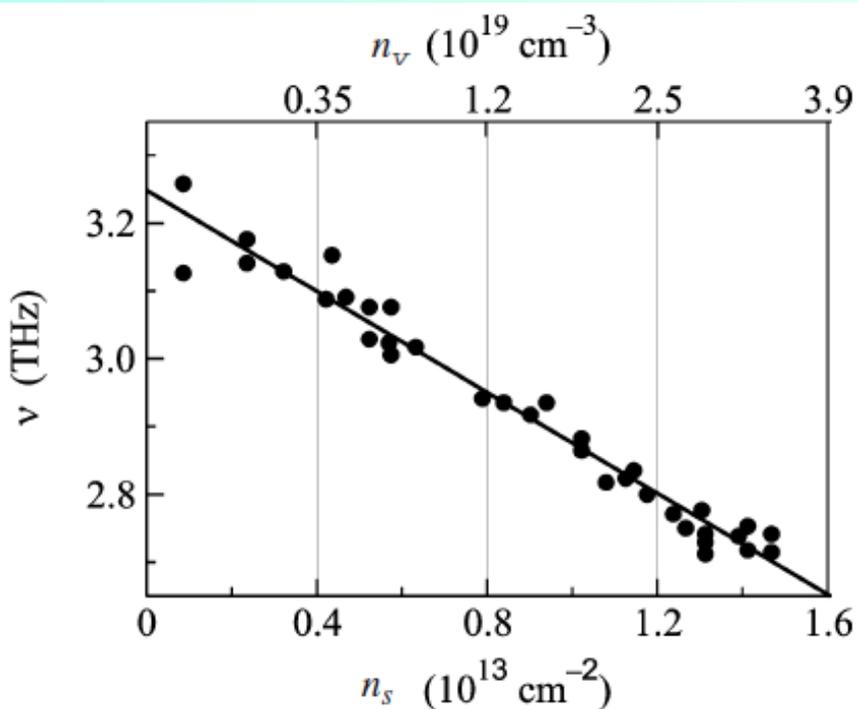
$$\varphi_{10}^2 = \frac{4z^2}{r_e^3} \exp(-2z/r_e)$$

$$\varphi_{20}^2 = \frac{z^2}{2r_e^3} \exp(-z/r_e) \left(1 - \frac{z}{2r_e}\right)$$



Now the ground state is shifted stronger \Rightarrow scattering length has positive sign $a_0 > 0$

Quantum transitions of electrons on the surface shifted due to the interaction with surfons. The shift depends on T



Experimental data from : V.V. Zav'yalov, I.I. Smol'yaninov, JETP Lett. 44, 182 (1986); Sov. Phys. JETP 65, 194 (1987); ibid 67, 171 (1988).

The shift of electron transition line is linear as function of surfon density n_s

P. D. Grigor'ev, A. M. Dyugaev and E. V. Lebedeva, JETP Lett. 87, 106 (2008).

This shift gives a strong proof of the existence of surfons !

The surfons exist not only on helium or hydrogen surfaces, but in all cryogenic liquids.

Contribution of surface level atoms to the electron scattering

Inverse scattering time is related to the He atom density $n(z)$ above the surface

$$\frac{1}{\tau} = \int dz \Psi_e^4(z) n_{He}(z) \frac{Ah}{\pi m}.$$

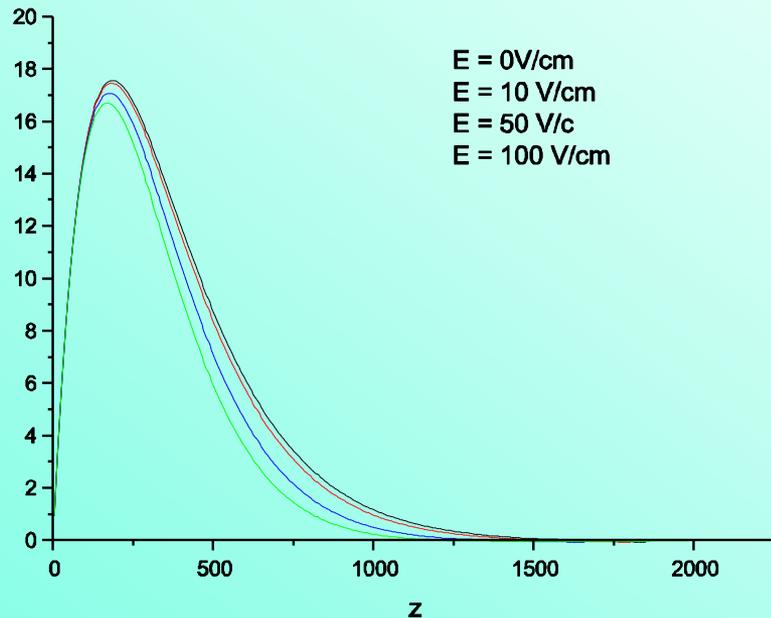
where A is the scattering amplitude.

This He atom density $n(z)$ contains vapor atoms and surfons.

Electron mobility $\eta = \tau / m.$

One can expect to answer another long-standing question: why the electron mobility on liquid helium surface is ~2 times smaller than the theoretical prediction.

Effective interaction between electrons and surfons on liquid helium is small.



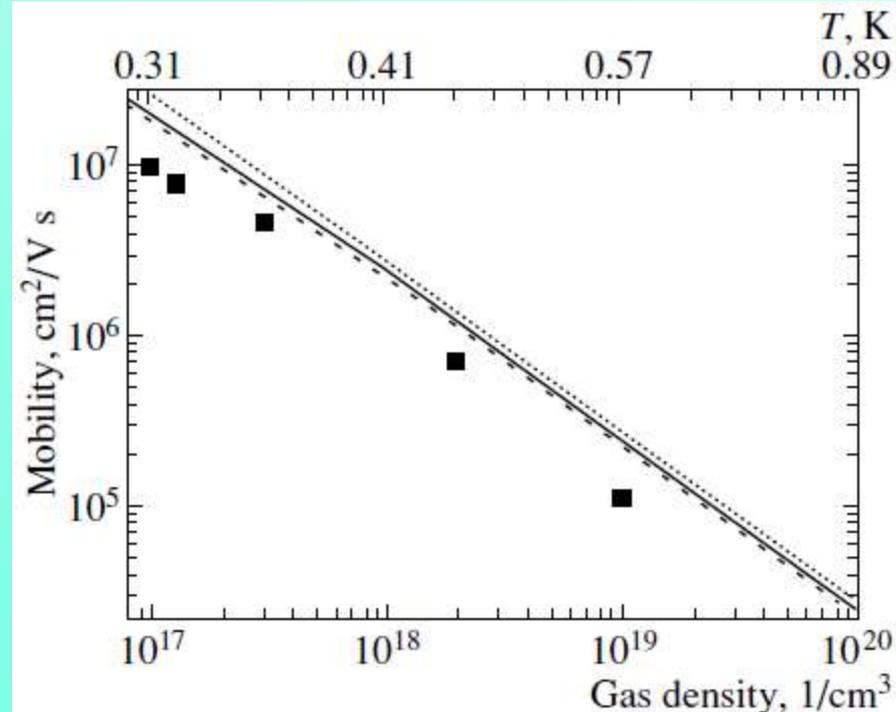
Electron wave function above helium surface for several values of clamping field (numerical calculation).

Contribution from surfons to the electron scattering above liquid helium is small because of small overlap of their wave functions: the electron wave function is localized on a distance $\sim 100\text{ \AA}$, while the helium atom is localized on a distance $\sim 5\text{ \AA}$

Other experiments, indicating the existence of surfons

Mobility of surface electrons above liquid helium.

The scattering rate of surface electrons on He vapor is not enough (~ 2 times) to explain the low electron mobility. The scattering by surfons considerably improves the agreement (by a factor of ~ 1.5) but still not enough.



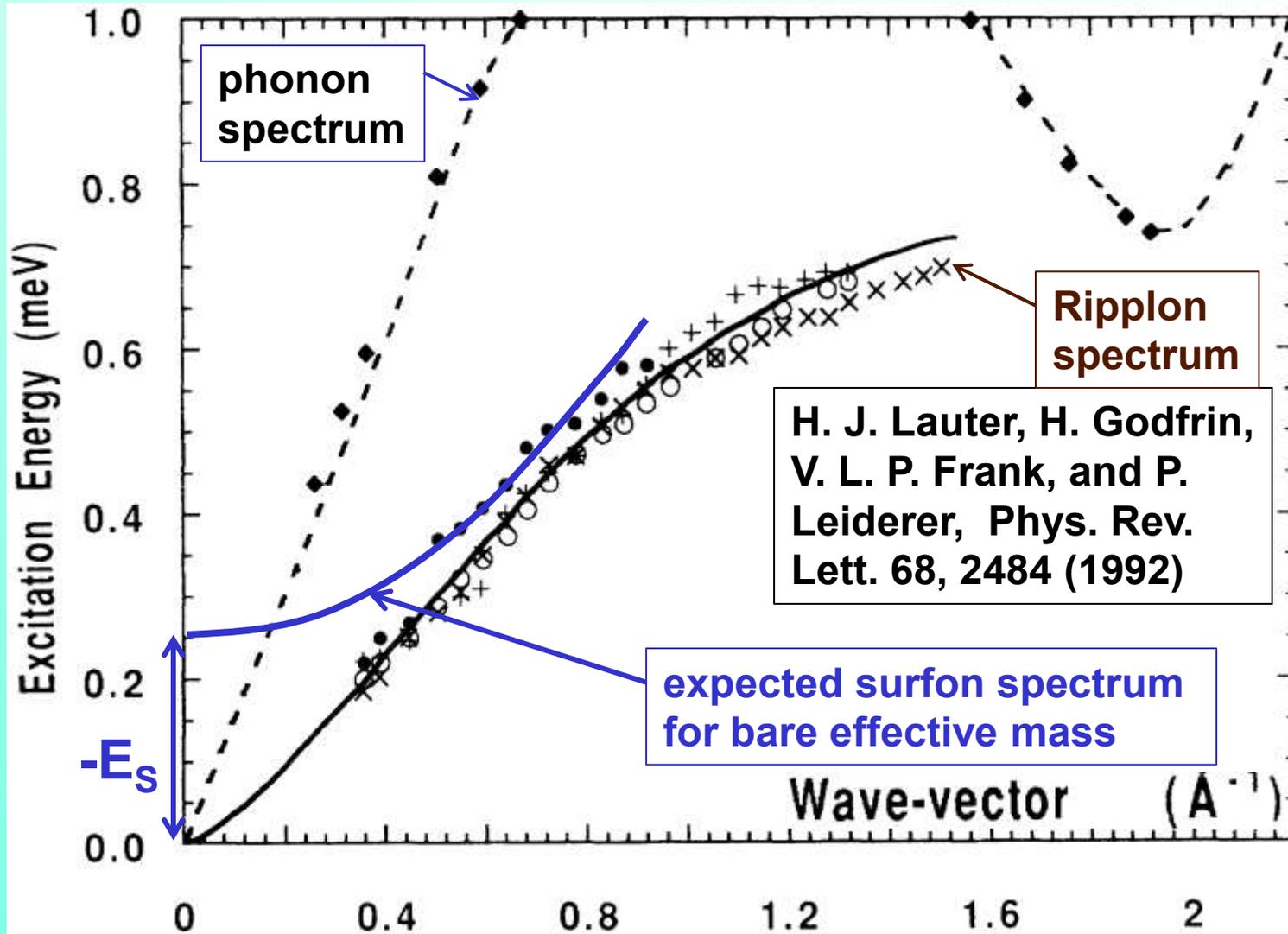
The electron-surfon interaction above liquid helium is weak because of small overlap of electron and surfon wave functions. The result is very sensitive to the exact shape of surfon $\Psi(z)$.

[P.D. Grigoriev, A.M. Dyugaev, E.V. Lebedeva, "Electron mobility on a surface of dielectric media: influence of surface level atoms", JETP 104 (1), p. 1-10 (2008);]

Experimental evidences of surfons are all indirect

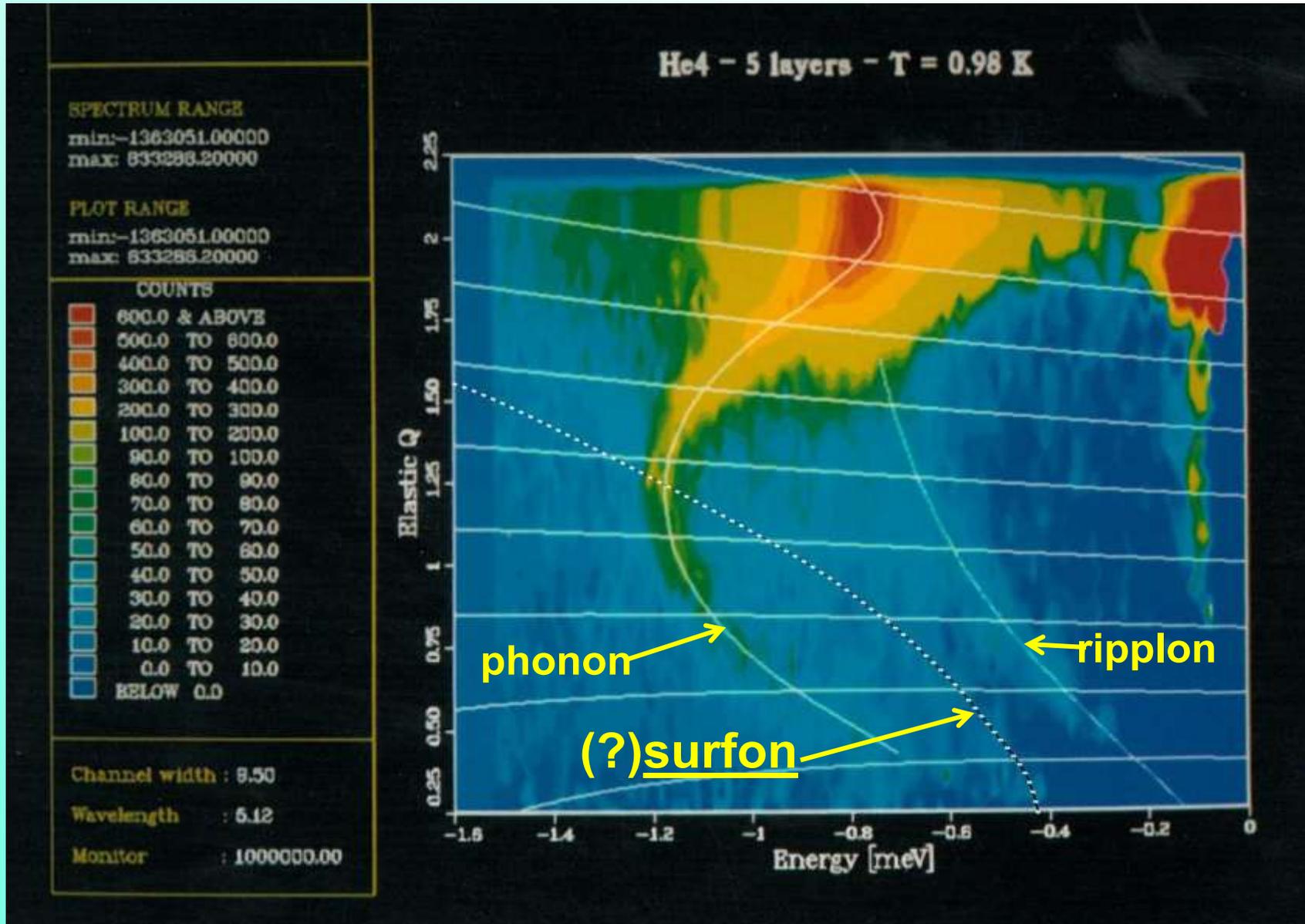
Suggestions for direct observation of surfons by inelastic neutron scattering

The measured ripplon spectrum by neutrons

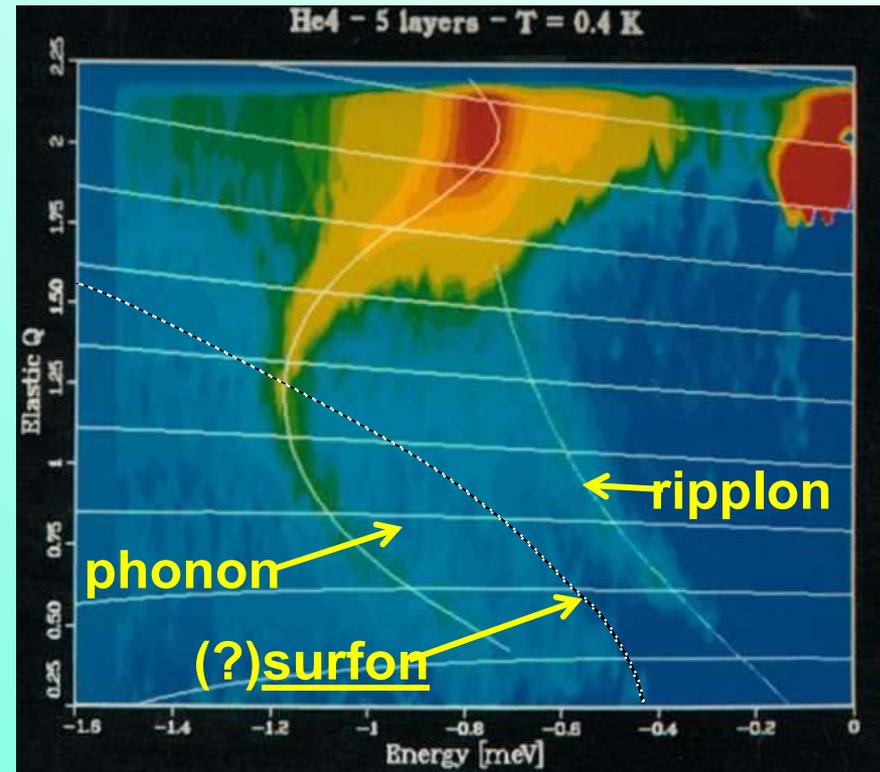


Experiments show that the ripplon spectrum softens only at $E \sim 10\text{K}$, which does not affect the surface tension at low temperature.

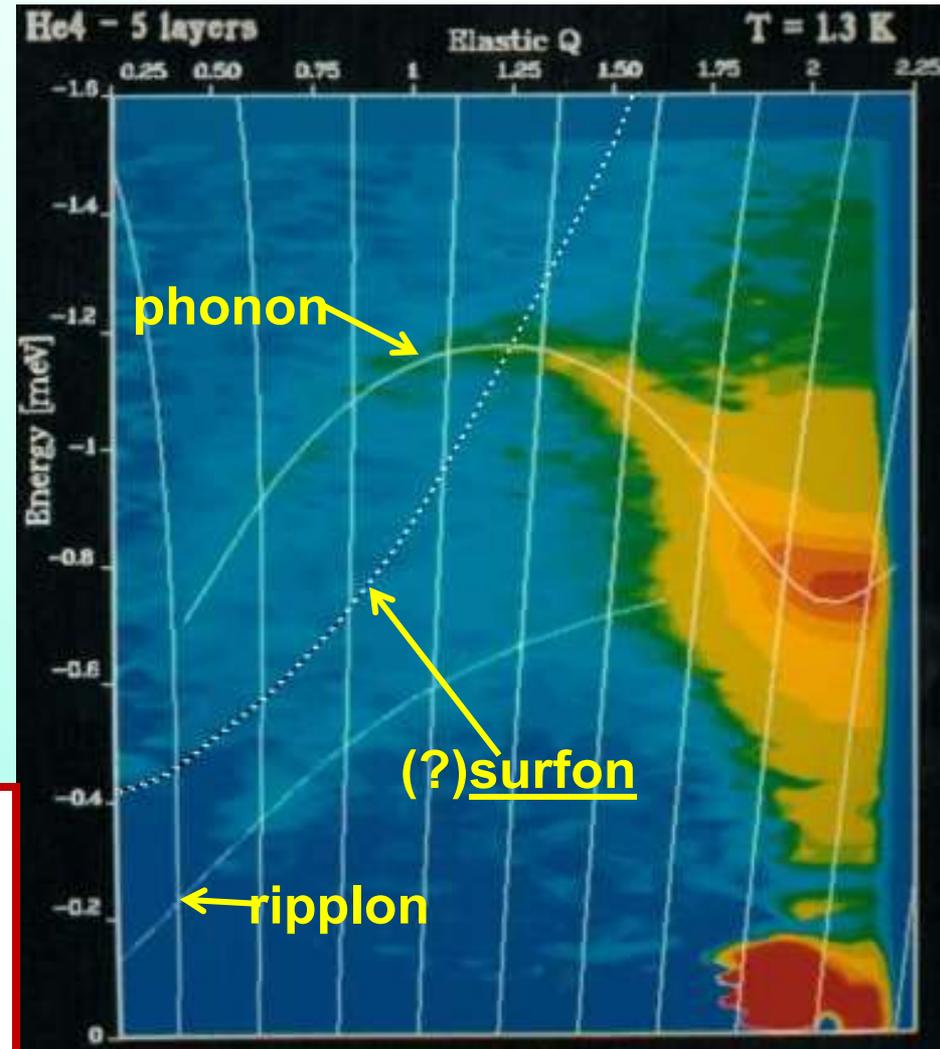
Experimental data by H.J. Lauter & H. Godfrin, and their analysis



Experimental data and their analysis



Third branch of excitations with activation energy $\sim 0.4 \text{ meV} = 4.7 \text{ K}$ resembles much the surfon



This is probably the first direct observation of surfons

Comparison with first-principle calculations ²⁶

The microscopic numerical calculations also propose several types of surface excitations in addition to ripples,[16–22] both on thick and thin He films

- [14] E. Feenberg, Theory of Quantum Fluids (Academic, New York, 1969).
- [15] G. Mahan, Many-Particle Physics, 2nd ed. (Plenum Press, New York, 1990), Ch. 10.
- [16] E. Krotscheck, Phys. Rev. B 31, 4258 (1985).
- [17] E. Krotscheck, Phys. Rev. B 32, 5713 (1985).
- [18] E. Krotscheck and C. J. Tymczak, Phys. Rev. B 45, 217 (1992).
- [19] B. E. Clements, E. Krotscheck, and C. J. Tymczak, Phys. Rev. B 53, 12253 (1996).
- [20] K.A. Gernoth, J.W. Clark, G. Senger and M.L. Ristig, Phys. Rev. B 49, 15836 (1994).
- [21] K. A. Gernoth and M. L. Ristig, Phys. Rev. B 45, 2969 (1992).
- [22] B. E. Clements, E. Krotscheck, C. J. Tymczak, J. Low Temp. Phys. 107, 387 (1997).
- [23] H.-J. Lauter, in Excitations in Two-Dimensional and Three Dimensional -Quantum Fluids, NATO ASI series, edited by A. F. G.Wyatt aud H. J. Lauter (Plenum, New York, 1991).

But their structure and energy spectrum does not correspond to surfons. Additional excitations in He films [18,19] are quantized (along z) phonons (so-called “breathing modes”, or standing waves propagating along surface).

Although numerical calculations of excitations in He describe well the ripplon and phonon branches, they do not support surfons [E. Krotscheck, private communication]

Approximations in first-principle calculations 27

The numerical calculations [16–22] of surface excitations in He apply the correlated basis function (CBF) method [15] for inhomogeneous liquid, using additional approximations: (1) The CBF calculations are based on the Feenberg wave function with only pair correlations and performed in the hypernetted-chain approximation. (2) Assume that only one-body component of the Feenberg function is affected by excitations and by external perturbations.[16] **This assumption of static two-body correlations limits the regime of validity of this theory to wavelengths longer than the average distance between two particles. It also may restrict the theory to small deviations from the equilibrium (ground-state) density of liquid He.** (3) The backflow effects [14] are also ignored. Therefore, the obtained theoretical excitation energies calculated at large wave numbers are substantially higher than the experimental results.[20] (4) Since the applied Jastrow variational treatment of the bulk liquid does not produce a self-bound system at saturation He density, an external potential is introduced phenomenologically in these numerical calculations to stabilize the surface.[20, 21] The strength of this additional phenomenological potential is adjusted so that the calculated chemical potential matches the experimental saturation value.[20] (5) The standard numerical computations assume that surface excitations do not violate the translational symmetry along the surface, which may not describe the case of a single surfon with zero in-plane momentum. (6) **Vapor states above He are usually ignored**

Resonant interface state (almost localized superposition of vapor quantum states)

[K.A. Gernoth and M.L. Ristig, Phys. Rev. B 45, 2969 (1992).]

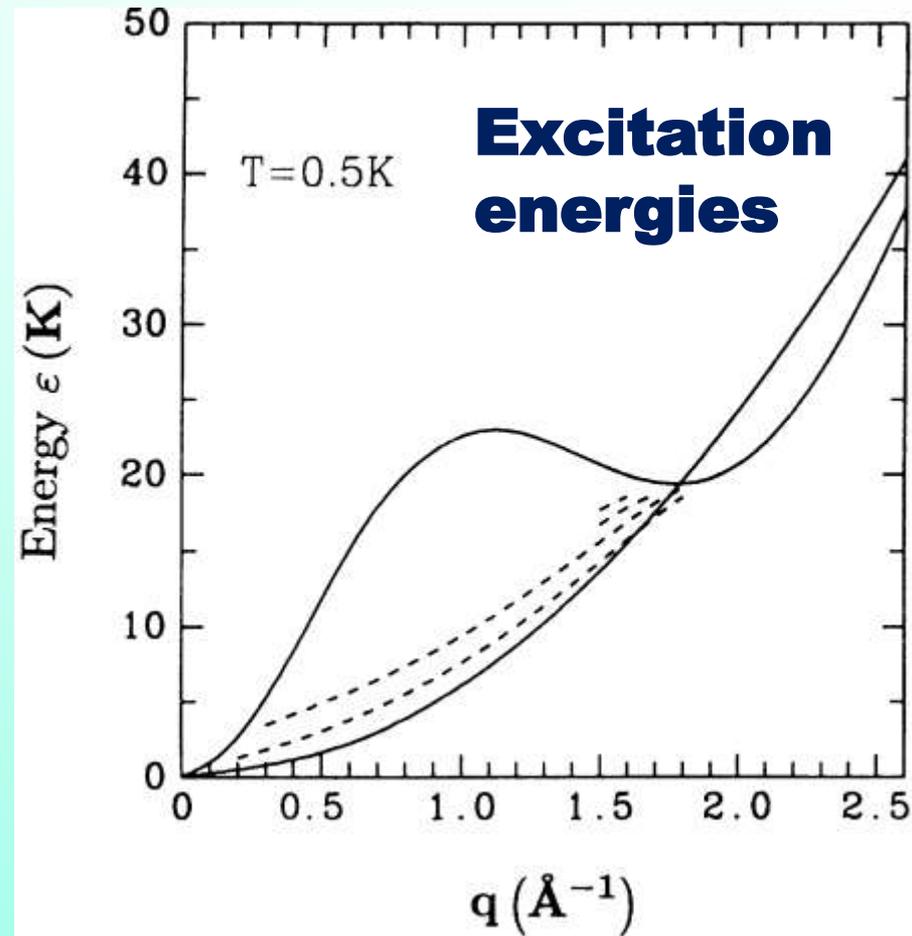
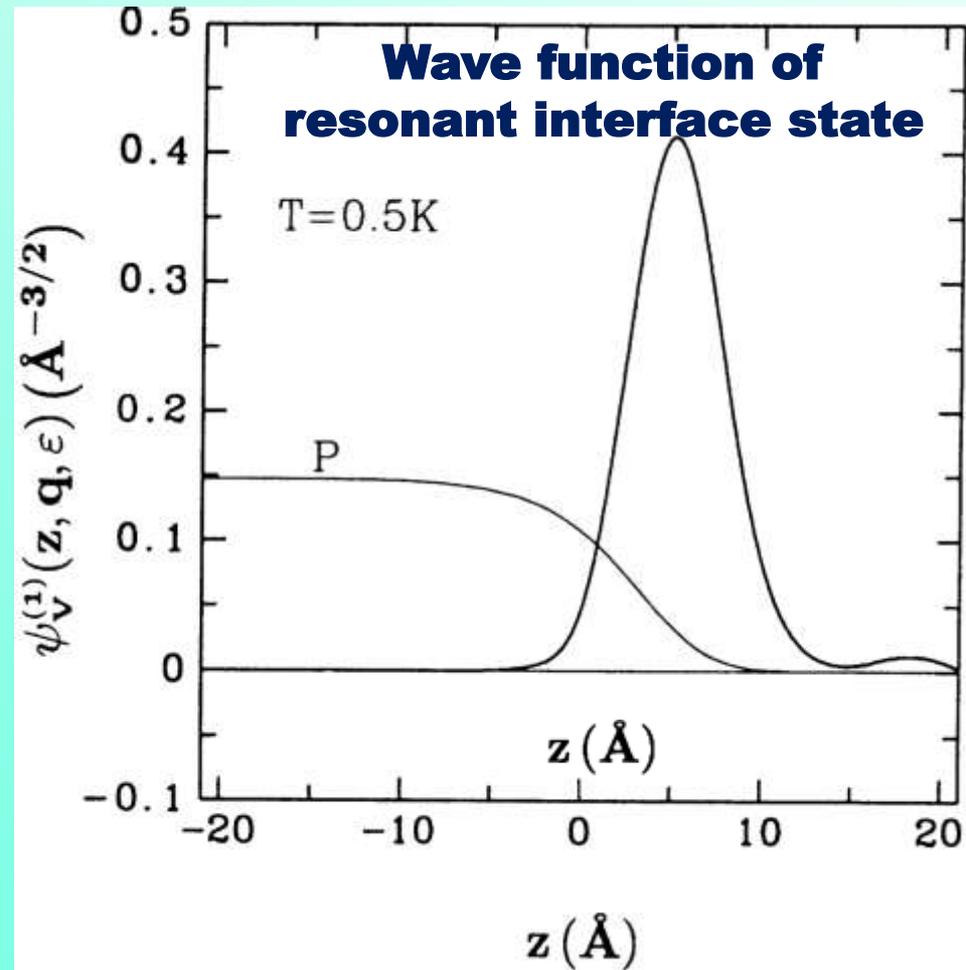


FIG. 6. Wave function $\psi_V^{(1)}(z, q, \epsilon)$ of a resonant interface state with wave number $q = 1.0 \text{\AA}^{-1}$ and energy $\epsilon = \epsilon^{(1)} = 7.6 \text{ K}$,

Resemble very much the surfons!

Summary

- **Surfons – the interface localized states of atoms (molecules) – have been proposed theoretically earlier but they received only indirect experimental substantiation from temperature dependence of surface tension and from the interaction of surfons with surface electrons (by shifting the electron transition line and decreasing their mobility).**
- **Here we analyze experimental data on the inelastic scattering of neutrons by thin helium films. It suggests (in addition to phonons and ripplons) the gapped surface excitations with activation energy $\sim 0.4\text{meV} \approx 4.5\text{K}$, which may be a direct observation of surfons.**
- **We review the previous numerical calculations of surface excitations in liquid He and argue that the excitations as surfons were obtained only in those calculations which include vapor states in the model.**

Appendices

Surface tension of ^4He

Density of surfons increase with temperature:

$$n_4(T) = \int \frac{d^2 p}{(2\pi)^2} \left[\exp\left(\frac{\varepsilon_4(p) - \mu_4}{T}\right) - 1 \right]^{-1}, \quad \varepsilon_4(p) = \Delta_4 + \frac{p^2}{2m_4}$$

The surface tension $\sigma_4(T) = \sigma_4(0) + \sigma_4^R(T) + \Omega_4^S(\mu_4, T)$,

where ripplon contribution $\sigma_4^R(T) = -AT^{7/3}$ (Atkins, 1953)

and the surface atoms contribution

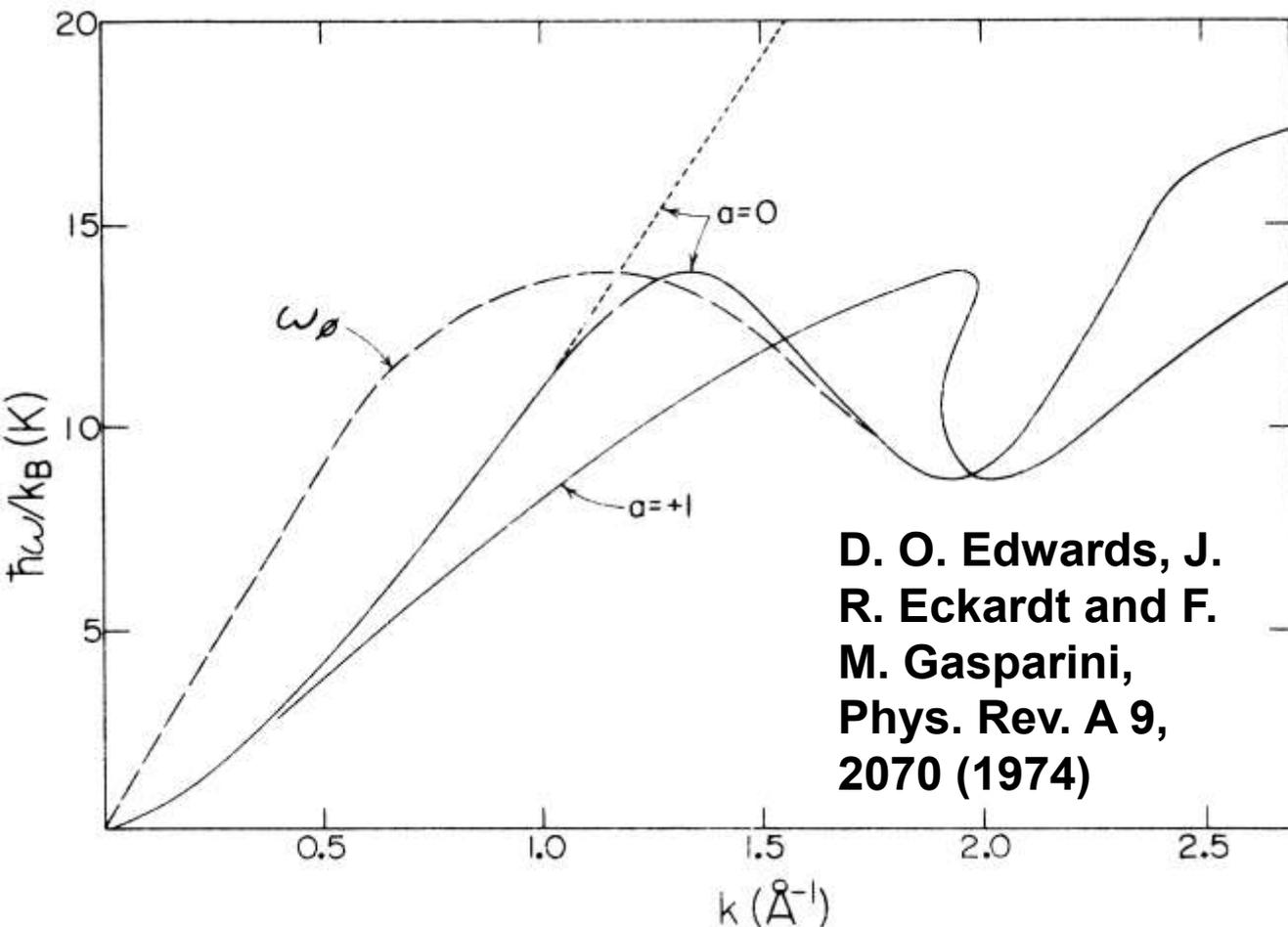
$$\Omega_4(T) = -T \int \frac{d^2 p}{h^2} \ln \left[1 - \exp\left(\frac{\mu_4 - \varepsilon_4(p)}{T}\right) \right] = -\frac{4\pi M_4 T^2}{h^2} \int_0^\infty \frac{x dx}{\exp\left(x + \frac{\Delta_4}{T}\right) - 1}.$$

A.M. Dyugaev, P.D. Grigoriev, JETP Lett.78(7), 466 (2003).

Ripplon spectrum softening at large momentum

32

does not help to explain strong temperature dependence of $\sigma(T)$



D. O. Edwards, J. R. Eckardt and F. M. Gasparini, Phys. Rev. A 9, 2070 (1974)

FIG. 1. Ripplon frequency expressed in degrees, $\hbar\omega/k_B$ vs the horizontal wave vector k . The empirical phonon spectrum $\omega_\phi(k)$ is shown for comparison. The other curves are short-dashed, calculated using $\omega = cq$ (constant velocity of sound); solid, calculated using the empirical phonon spectrum $\omega = \omega_\phi(q)$. The values of a are in \AA^2 ; δ was assumed to be zero.

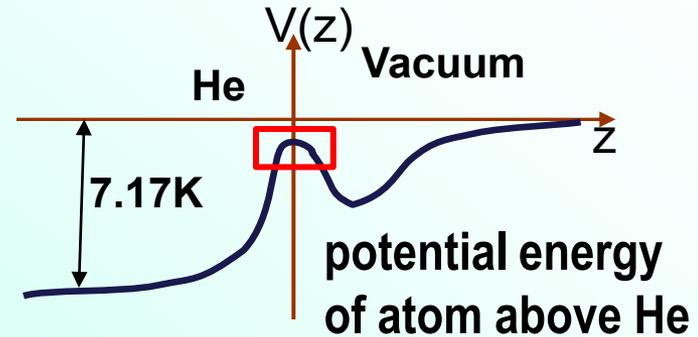
The predicted softening of the ripplon spectrum happens at energy $E \sim 10\text{K}$ and does not change the surface tension at low temperature.

A second ripplon branch (still debated) was proposed to fit $\sigma(T)$ [L. Pricapenko and J. Treiner, J. Low Temp. Phys. 101, 809 (1995)]

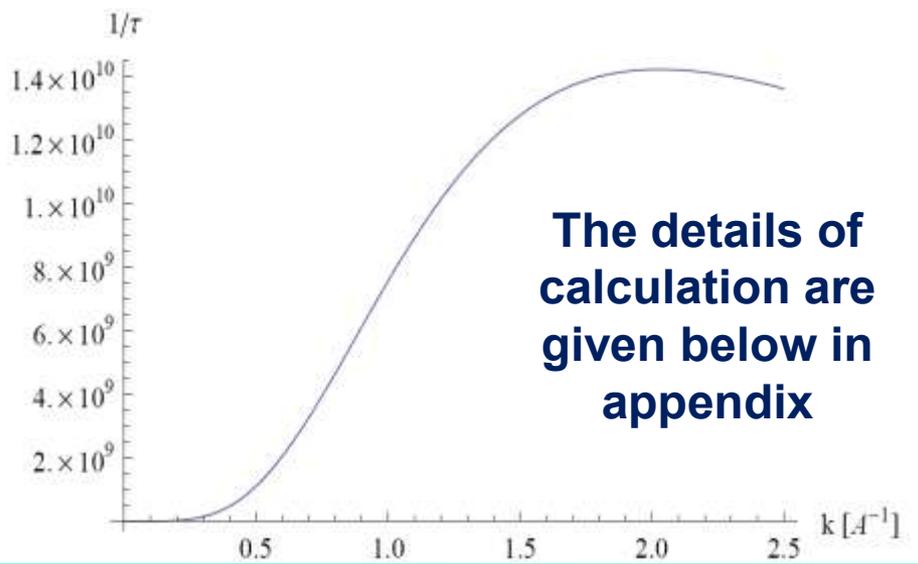
Estimates of surfon lifetime

Two channels limit the surfon-quasiparticle lifetime: (1) merging into the liquid and (2) evaporation.

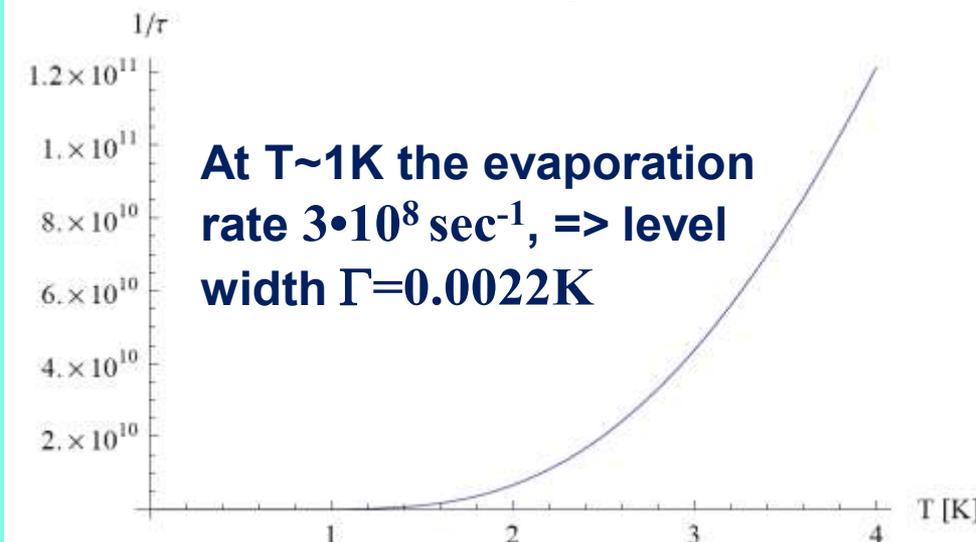
1) to merge into the bulk the surfon must (a) overcome the potential barrier; (b) push away other atoms. Therefore, the surfon level width $\Gamma \ll E$.



2) Surfon evaporation rate as function of its in-plane momentum

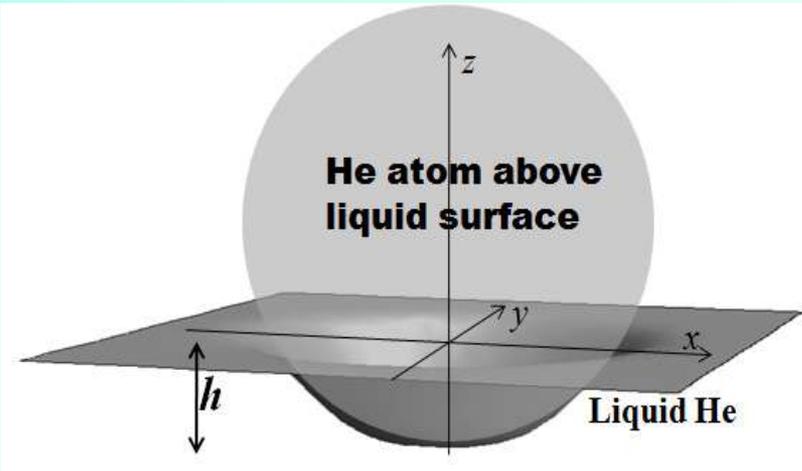


Surfon evaporation rate as function of temperature



Model **Rough model of the surface adjustment.**

Dimple under the surfon (ripplonic polaron).



The total energy of the dimple is

$$E_{tot} = E_{surf} + E_W + E_k$$

The energy of the surface tension is

$$E_{surf} = \alpha \int_0^\infty \pi \rho [\xi'(\rho)]^2 d\rho$$

The energy gain from interaction with other atoms in the liquid is

$$E_W = \int d^3 \mathbf{r} \Delta n_b(\mathbf{r}) V_{LJ}(\mathbf{r} - \mathbf{r}'),$$

Quantum kinetic energy (the minimal estimate) is

$$E_k = \frac{\hbar^2 \alpha_1^2}{2M^0 R^2} \approx \frac{2.88 \hbar^2}{M^0 R^2},$$

Minimization of the total energy gives the energy of the dimple:

$$E_{4tot \min} \approx -2.7 \text{ K}$$

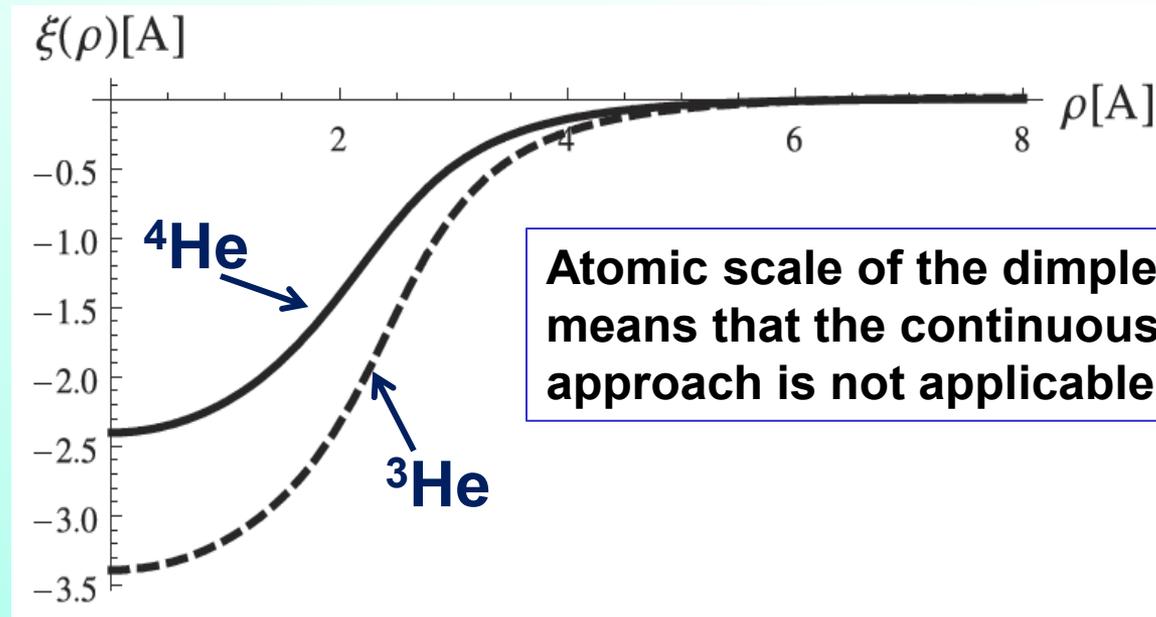
$$E_{3tot \min} \approx -1.85(\pm 0.1) \text{ K},$$

This energy reduces the surfon activation energy.

Estimate of surfon effective mass due to dimple

Estimated dimple profile and depth:

This dimple depth is overestimated, because the quantum kinetic energy is taken smaller than it is.



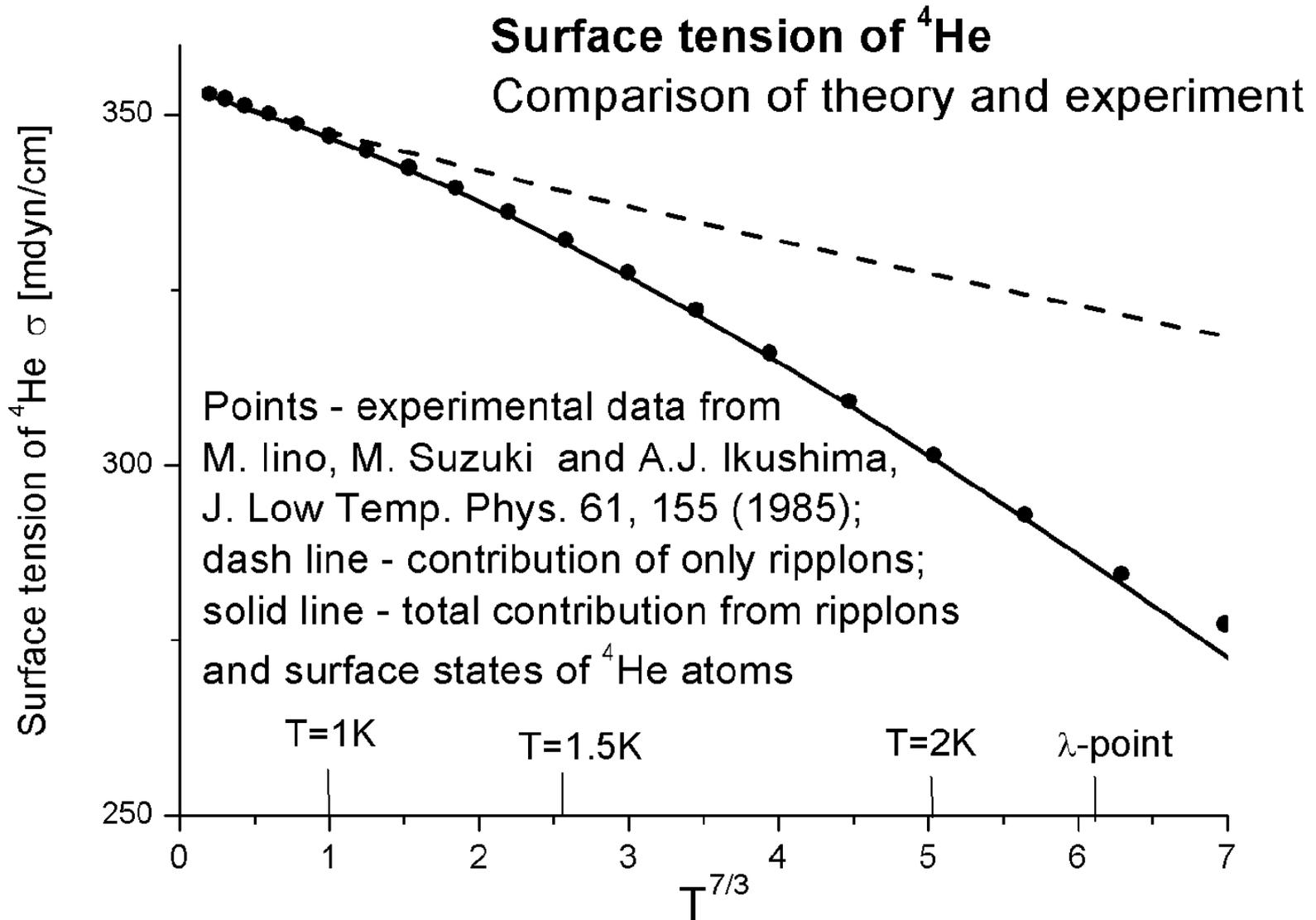
The surfon effective mass is roughly given by the dimple volume:

$$M_4^* \approx M_4^0 (1 + n_{\text{He}4} V_{4d}) = 2.04 M_4^0.$$

$$M_3^* \approx M_3^0 (1 + n_{\text{He}3} V_{3d}) = 2.23 M_3^0.$$

This rough calculation shows that there is an optimal dimple depth: the atom does not completely merge into the liquid but stop sinking at some optimal dimple depth. Hence, there is a potential barrier between surface and bulk; => a quasi-stable state of an atom at the He surface.

Fig. 3



Surface tension of ^3He

Density of surfons increase with temperature:

$$n_3(T) = 2 \int \frac{d^2 p}{h^2} \left[\exp\left(\frac{\varepsilon_3(p) - \mu_3}{T}\right) + 1 \right]^{-1}, \quad \varepsilon_3(p) = \Delta_3 + \frac{p^2}{2m_3}$$

The surface tension $\sigma_3(T) = \sigma_3(0) + \Omega_3(\mu_3, T)$,

where
$$\Omega_3(T) = -2T \int \frac{d^2 p}{(2\pi)^2} \ln \left[1 + \exp\left(\frac{\mu_3 - \varepsilon_3(p)}{T}\right) \right].$$

$$\sigma_3(T) = 155.3 - 61.2T^2 \int_0^{\infty} \frac{x dx}{\exp\left(x + \frac{\Delta_3}{T}\right) + 1}$$

A.M. Dyugaev, P.D. Grigoriev, JETP Lett.78(7), 466 (2003).

Fig. 1

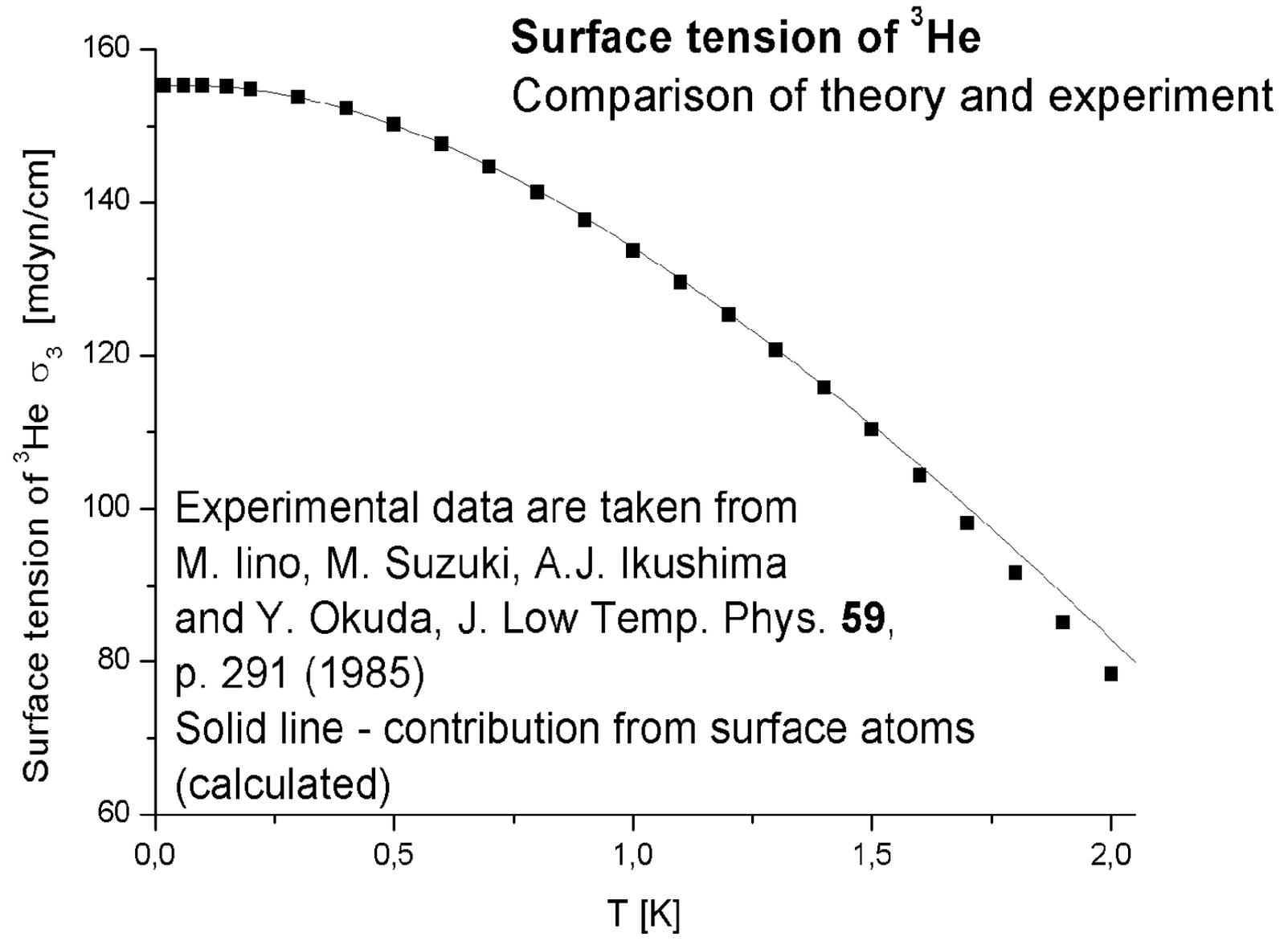


Fig. 2

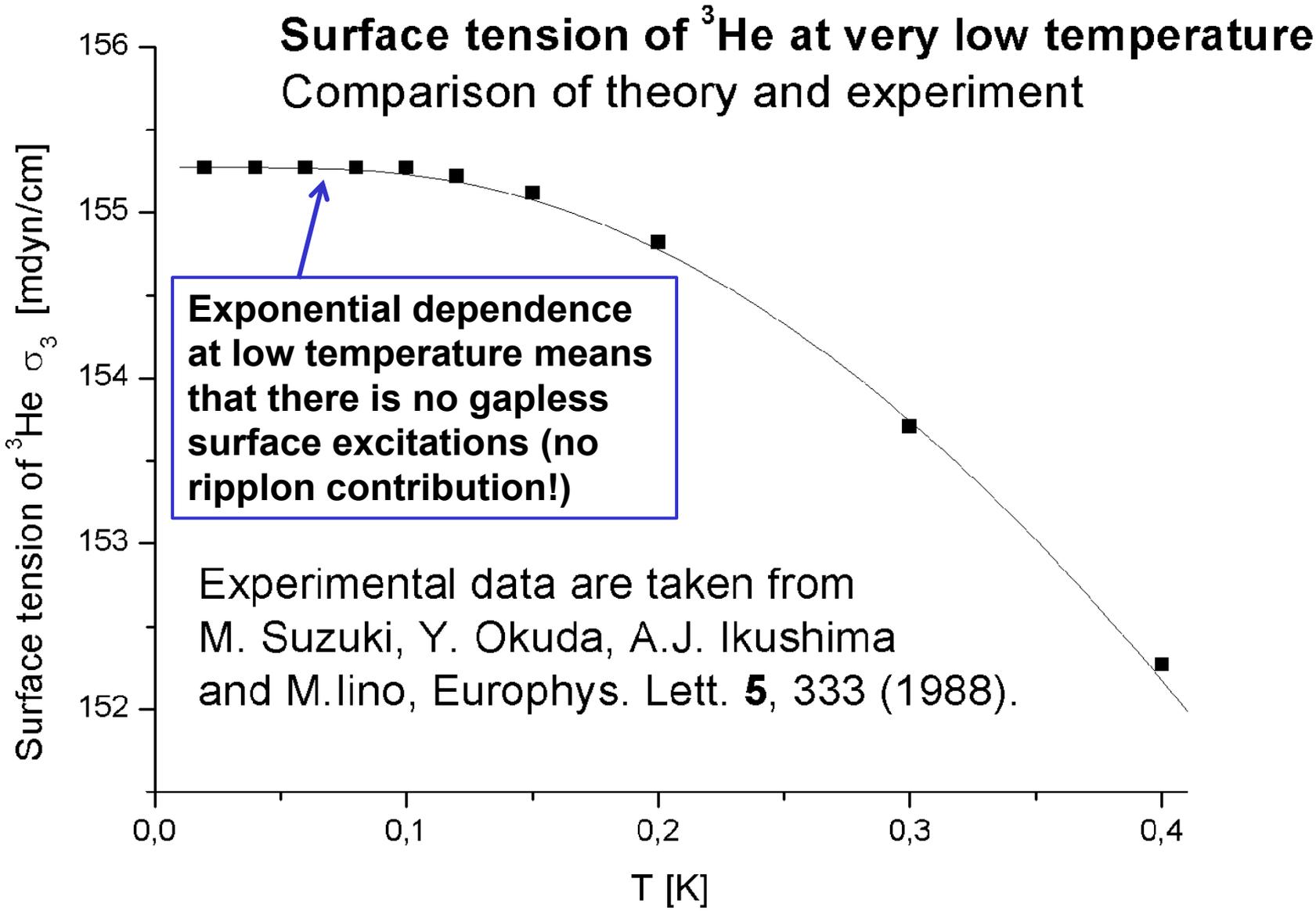


Fig. 6 The temperature dependence of the surface tension of ^4He , divided by T^2 . This graph shows the comparison between our theory and experiment in the coordinates where the difference can be visually observed. The low-temperature deviation at $T < T_\lambda$ is due to the ripplon contribution, while the deviation at $T > 4$ K is due to the proximity to the boiling point. The ripplon contribution at $T < T_\lambda$, extracted from this graph, is shown in Fig. 7

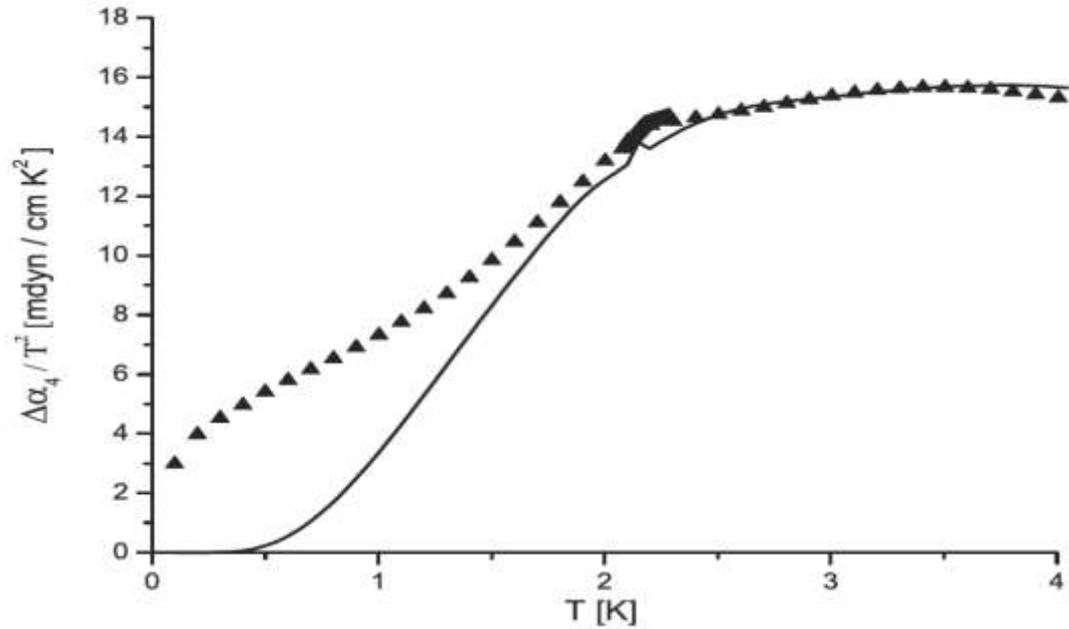
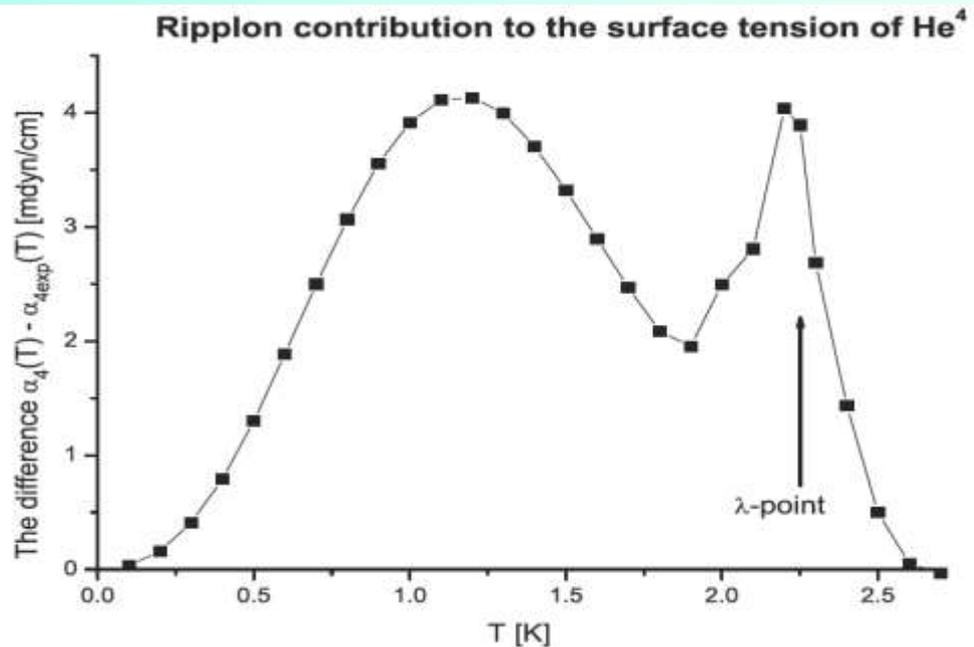


Fig. 7 The difference between the calculated temperature dependence of the surface tension $\alpha_4(T)$ of ^4He , where only the surfon contribution given by (28) is taken into account, and the experimental values $\alpha_{4\text{exp}}(T)$. The maximum at $T \approx 1.1$ K is due to the ripplon contribution. The second maximum at $T \approx 2.17$ K corresponds to phase transition at the λ -point of ^4He



Appendix 1:

The estimate of surfon lifetime due to evaporation

The probability of surfon evaporation per unit time is given by:

$$d\omega_{fi} = \frac{2\pi}{\hbar} |F_{fi}|^2 \delta(E_f - E_i^{(0)} - \hbar\omega) dv_f.$$

The main difficulty is to calculate the matrix elements F_{fi} of ripplon-surfon interaction.

Calculation of ripplon-surfon interaction Hamiltonian

In the adiabatic approximation (when the surfon has enough time to adjust to the time-dependent surface curvature) the surfon-ripplon interaction can be estimated in analogy to the electron-ripplon interaction:

$$\hat{V} = \delta H - \hbar k_z \dot{\xi}, \quad \text{where } k_z = -i\partial/\partial z,$$

$\delta H = \delta V_s + \delta K_s$ contains potential and kinetic energy:

Potential energy is

$$\delta V_s(\mathbf{r}, z) = n_{He} \int d^2\mathbf{r}_1 [\xi(\mathbf{r}_1) - \xi(\mathbf{r})] V_{LJ} \left(\sqrt{(\mathbf{r}_1 - \mathbf{r})^2 + z^2} \right).$$

where $\xi(\mathbf{r}_1) - \xi(\mathbf{r}) = \xi_0 \cos(\omega t) [\cos(\mathbf{q}\mathbf{r}_1) - 1]$.

The normalized ripplon amplitude (one ripplon per area S):

$$\xi_{0q} = \left(\frac{\hbar}{2S \sqrt{\rho_{He} \alpha q}} \right)^{1/2}$$

Quantum kinetic energy is $\delta K_s = -\frac{\hbar^2}{M_{He}} k_z (\mathbf{k} \nabla \xi),$

Calculation of ripplon-surfon interaction Hamiltonian (continuation 1).

Substitution of the L- J potential and the surface curvature after integration we obtain

$$\delta V_q(z) = 4\pi\epsilon_0 n_{He} \xi_0 \sigma_0^2 \cos(\omega t) f(q, z)$$

where

$$f(q, z) \equiv \left(\frac{q^5 K_5(qz)}{1920z^5} - \frac{1}{5z^{10}} \right) \sigma_0^{10} + \left(\frac{q^2 K_2(qz)}{4z^2} - \frac{1}{2z^4} \right) \sigma_0^4$$

and $K_n(qz)$ – modified Bessel's function of the n -th order.

The main divergent term at $z \rightarrow 0$ cancels.

For long-wavelength ripples

$$f_{qz \ll 1}(q, z) \approx q^2 \left[\frac{\sigma_0^{10}}{80z^8} - \frac{\sigma_0^4}{8z^2} \right].$$

At $qz \sim 1$ the function is finite:

$$f(q, z) \rightarrow \frac{5z^6 + 2\sigma_0^6}{10z^{10}} \sigma_0^4.$$

More precise approximation

$$f(q, z) \approx \left[\frac{\sigma_0^{10}}{80z^8} - \frac{\sigma_0^4}{8z^2} \right] \frac{q^2}{1 + q^2 z^3 / 8\sigma_0}.$$

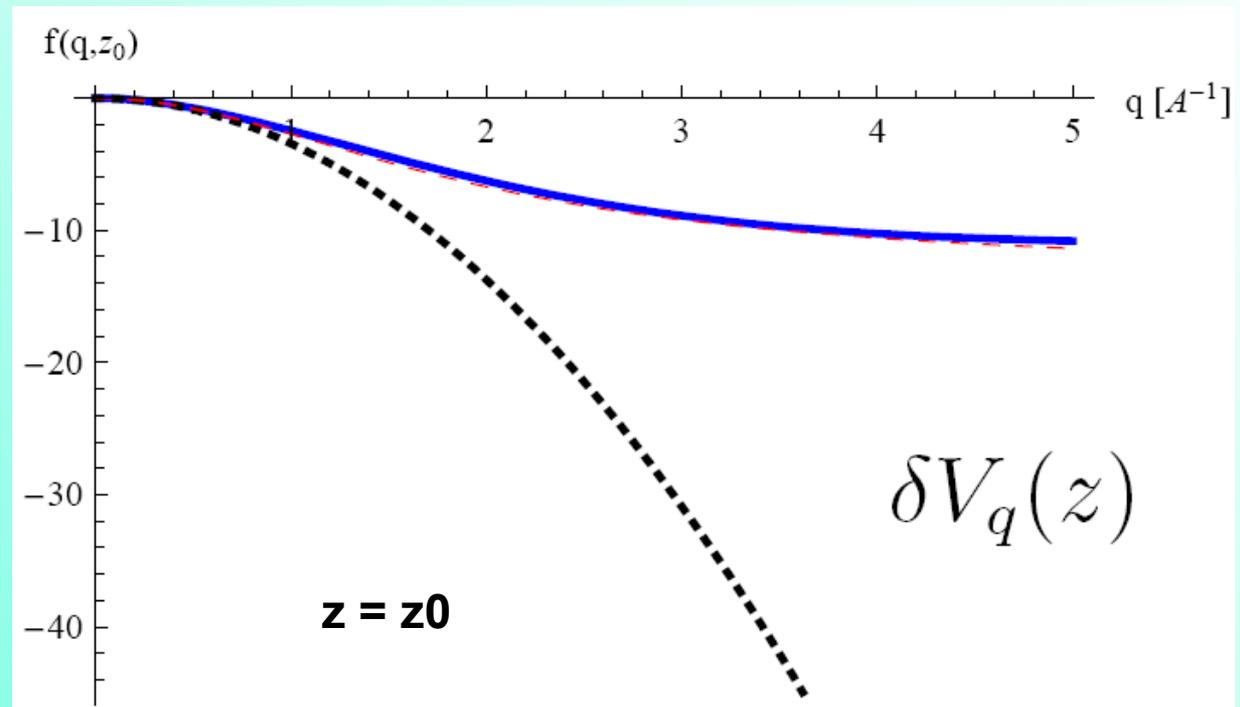
Ripplon-surfon interaction Hamiltonian (continuation 2).

The surfon-ripplon interaction potential

$$\hat{V} = \sum_{\mathbf{q}} e^{i\mathbf{q}\mathbf{r} - i\omega_{\mathbf{q}}t} \left[\delta V_{\mathbf{q}}(z) - \xi_{0\mathbf{q}} \left(\frac{\hbar^2(\mathbf{k}_{\parallel}\mathbf{q})}{M_{He}} - \hbar\omega_{\mathbf{q}} \right) \frac{\partial}{\partial z} \right].$$

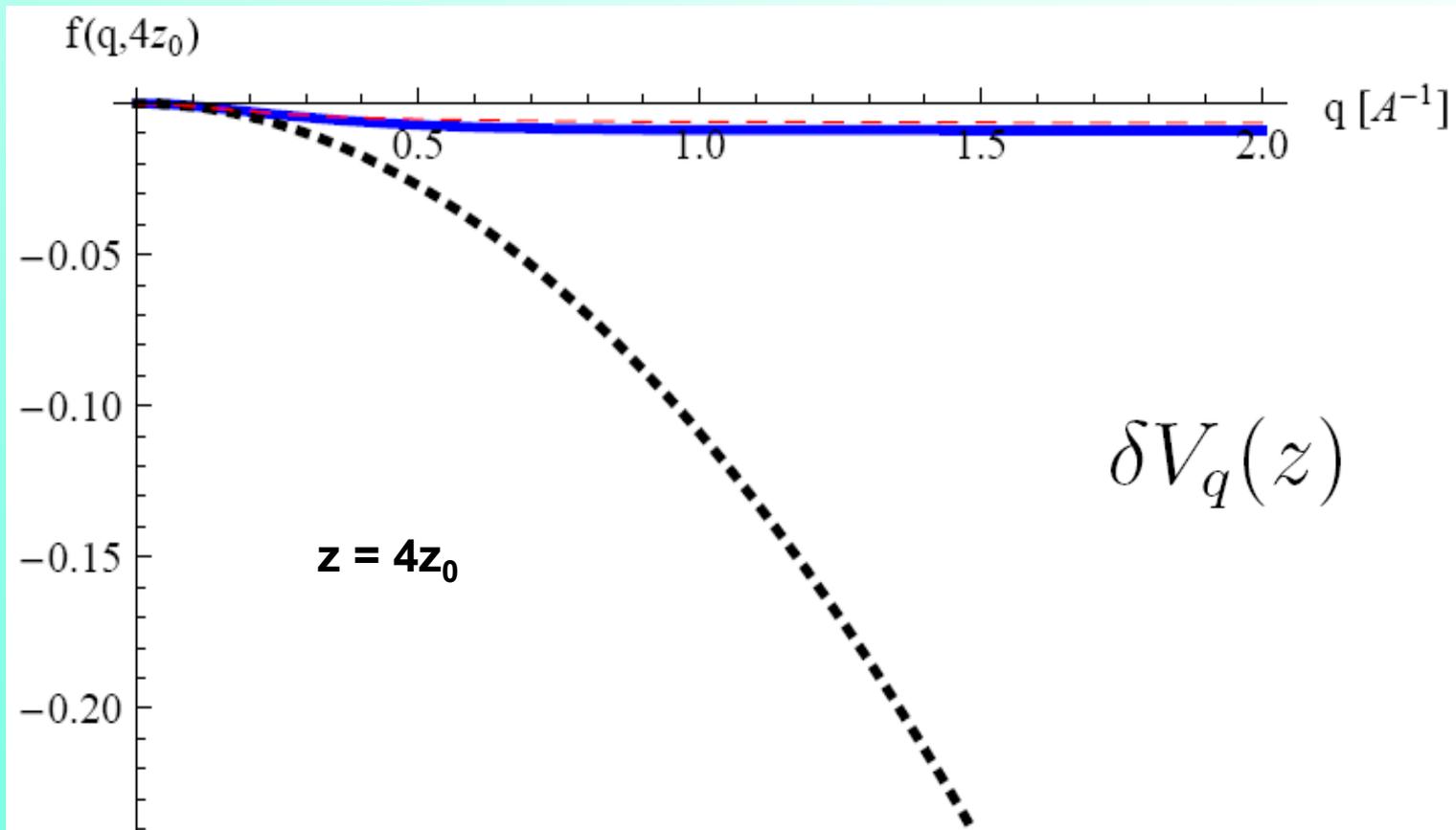
The main contribution (95%) comes from the potential $\square V_{\mathbf{q}}(z)$

Dependence $\square V_{\mathbf{q}}(z_0)$
(solid blue line), its
approximation at
 $qz \ll 1$ (dashed black),
and more precise
approximation
(dashed red line).



The interaction potential falls down rapidly with the increase of distance z

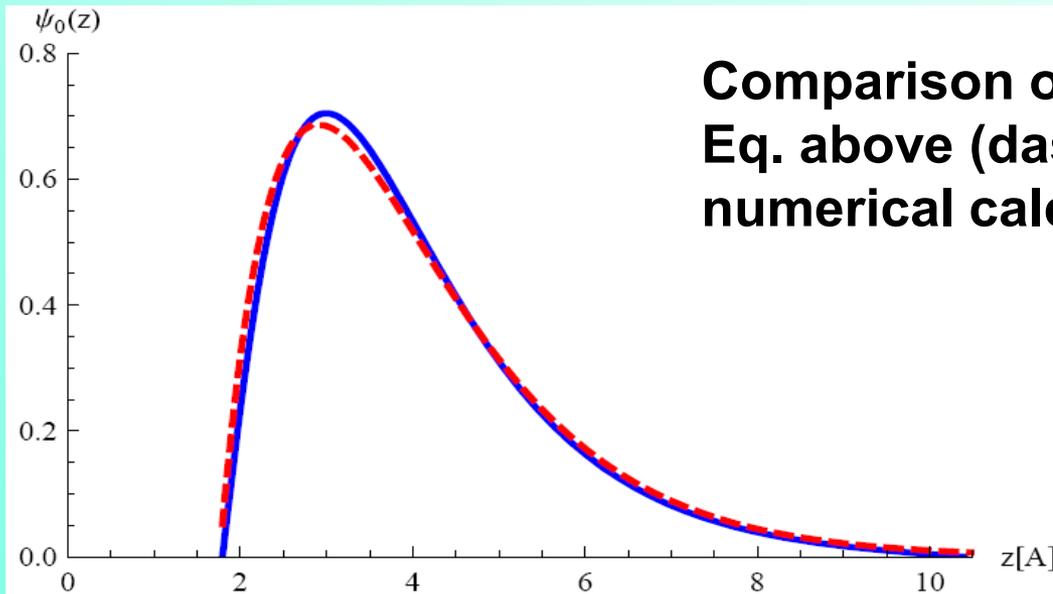
Dependence $\square V_q(4z_0)$ (solid blue line), its approximation at $qz \ll 1$ (dashed black), and used approximation (dashed red line).



Surfon wave function.

$$\psi_0(z) \approx \kappa^2(z - z_0) \exp(-\kappa(z - z_0)), \quad \kappa \equiv \sqrt{2M_{He}E_{s0}^{He}/\hbar^2}.$$

Here E_{s0}^{He} – the discrete energy, $z_0 \approx 1.75\text{\AA}$ – distance from the surface, where the surfon wave function is almost zero.



Comparison of surfon wave function from Eq. above (dashed red line) and from numerical calculation (blue solid line)

Matrix elements of the interaction potential (in the above approximation):

$$\delta V_{qk0} \approx 4\pi\epsilon_0 n_{He} \xi_{0q} \cos(\omega_q t) \int_{z_0}^{\infty} dz \psi_0^*(z) \psi_k(z) \left[\frac{\sigma_0^{12}}{80z^8} - \frac{\sigma_0^6}{8z^2} \right] \frac{q^2}{1 + q^2 z^3 / 8\sigma_0}$$

Wave functions of vapor atoms

Wave functions of continuous spectrum are zero at $z = z_0$ and orthogonal to $\Psi_0(z)$, being plane waves far from the surface:

$$\psi_k(z) = \sqrt{2/l} \sin[k(z - z_0)] - \lambda_k \psi_0(z),$$

where the factor λ_k is chosen to make ψ_k orthogonal to ψ_0 :

$$\begin{aligned} \lambda_k &= \int \psi_0^*(z) \psi_k(z) dz \\ &\approx \int_{z_0}^{\infty} \sqrt{2/l} \kappa^{3/2} \sin[k(z - z_0)] (z - z_0) \exp[-\kappa(z - z_0)] dz \\ &= \frac{2\sqrt{2/l} \kappa^{5/2} k}{(\kappa^2 + k^2)^2} \sim \frac{1}{\sqrt{\kappa l}} \ll 1. \end{aligned}$$

As $\lambda_k \ll 1$, all non-diagonal elements (except those, where the main term is zero) can be calculated using the simplified wave functions

$$\psi_k(z) = \sqrt{2/l} \sin[k(z - z_0)].$$

Along the surface, the wave functions of surfons and vapor atoms are plane waves:

$$\psi(\mathbf{r}_{\parallel}) = \exp(i\mathbf{k}_{\parallel} \mathbf{r}_{\parallel}).$$

Matrix elements of surfon-ripplon interaction (potential term)

The interaction term in the Hamiltonian

$$\hat{V} = \sum_{\mathbf{q}} e^{i\mathbf{q}\mathbf{r} - i\omega_q t} \left[\delta V_q(z) - \xi_{0q} \left(\frac{\hbar^2(\mathbf{k}_{\parallel}\mathbf{q})}{M_{He}} - \hbar\omega_q \right) \frac{\partial}{\partial z} \right].$$

Matrix elements of the interaction potential (in the above approximation):

$$\delta V_{qk0} \approx 4\pi\epsilon_0 n_{He} \xi_{0q} \cos(\omega_q t) \int_{z_0}^{\infty} dz \psi_0^*(z) \psi_k(z) \left[\frac{\sigma_0^{12}}{80z^8} - \frac{\sigma_0^6}{8z^2} \right] \frac{q^2}{1 + q^2 z^3 / 8\sigma_0}$$

Using $z^3 = \langle z^3 \rangle \approx 60\text{\AA}^3$ we obtain:

$$\delta V_{qk0} \approx V_{qt0} \int_0^{\infty} dz \sin[kz] z \exp[-\kappa z] \left[\frac{\sigma_0^6}{80(z+z_0)^8} - \frac{1}{8(z+z_0)^2} \right]$$

where

$$V_{qt0} \equiv 4\pi\epsilon_0 n_{He} \sigma_0^6 \kappa^{3/2} \sqrt{2/l} \cos(\omega_q t) \xi_{0q} \frac{q^2}{1 + q^2 \langle z^3 \rangle / 8\sigma_0}.$$

Matrix elements of surfon-ripplon interaction (kinetic term)

Matrix elements of momentum operator $\mathbf{p}_z = -i\hbar\partial/\partial z$ are

$$\int \psi_0^*(z) \frac{-i\hbar\partial}{\partial z} \psi_k(z) dz = \frac{2\sqrt{2/l}\hbar k \kappa^{3/2} (\kappa^2 - k^2)}{(\kappa^2 + k^2)^2}.$$

Matrix element of the kinetic part of surfon-ripplon interaction

$$K_{q0k} = \xi_{0q} \left(\frac{\hbar^2(\mathbf{k}_{\parallel}\mathbf{q})}{M_{He}} - \hbar\omega_q \right) \frac{\partial}{\partial z} \Big|_{q0k}$$

This matrix element is 10-100 times smaller than that of potential part and can be disregarded.

The evaporation rate estimate

The probability per unit time for a surfon to scatter to the continuous spectrum is

$$\begin{aligned}
 w_{if}(\mathbf{k}_{\parallel i}) &= \frac{2\pi}{\hbar} \int d\nu_i d\nu_f |V_{kq}|^2 \delta(E_f - E_i - \hbar\omega_q) \\
 &= \frac{2\pi}{\hbar} \int \frac{S d^2\mathbf{q}}{(2\pi)^2} n_B(\omega_q) \int \frac{ldk_z}{2\pi} |V_{kq}|^2 \\
 &\quad \times \delta\left(\frac{\hbar^2(\mathbf{k}_{\parallel i} + \mathbf{q})^2}{2M_4^0} + \frac{\hbar^2 k_z^2}{2M_4^0} - \frac{\hbar^2 \mathbf{k}_{\parallel i}^2}{2M_4^0} + |E_s^{He4}| - \hbar\sqrt{\frac{\alpha}{\rho}} q^{3/2}\right)
 \end{aligned}$$

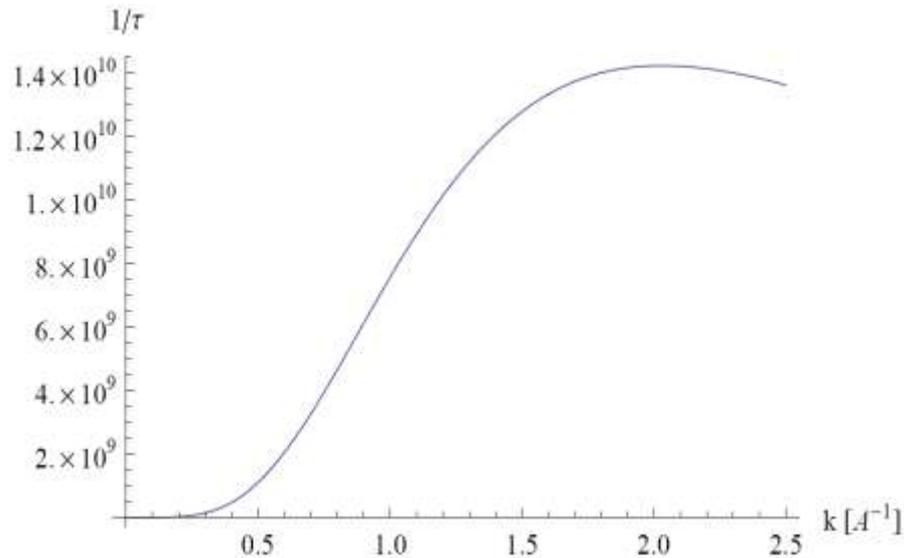
After integration over final states k_z and over ripplon momentum q , we obtain in polar coordinates, substituting the ${}^4\text{He}$ constants:

$$\begin{aligned}
 w_{if}^{He4}(k_{\parallel i}) &\approx 1.22 \cdot 10^{13} \int_0^\infty \frac{d(q^2)}{\exp\left(-\frac{11.8}{T} q^{3/2}\right) - 1} \frac{q^{7/2}}{(1 + 3q^2)^2} \\
 &\quad \times \frac{1}{\sqrt{(k_{\parallel i} q)^2 - \left(q^{3/2} - \frac{q^2}{2} - \frac{1}{2} - 0.615^2\right)^2}}
 \end{aligned}$$

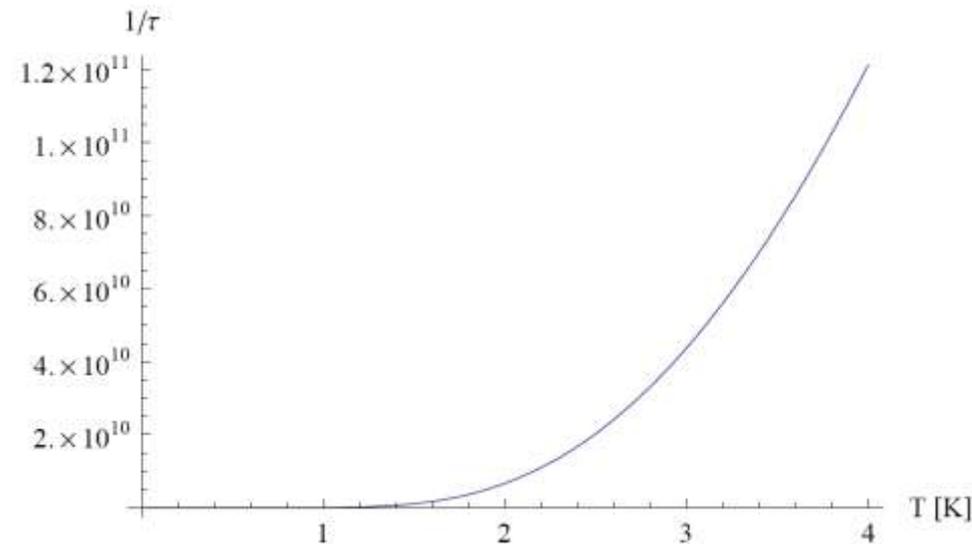
For surfons with energy $\sim T \sim 1\text{K}$ ($k_{\parallel i} \approx (3\text{\AA})^{-1}$) the evaporation rate $3 \cdot 10^8 \text{ sec}^{-1}$, that corresponds to the level width $\Gamma = 0.0022\text{K}$. This is much less than surfon energy \Rightarrow surfons are quasiparticles.

Surfon evaporation rate (results)

Surfon evaporation rate as function of its in-plane momentum



Surfon evaporation rate as function of temperature



With the increase of temperature and initial surfon momentum its evaporation rate rapidly increases (according to the physical intuition). However, even at $T=4$ K the surfon evaporation rate is less than its energy divided by \hbar . Hence, even at such high temperature, the surfons above liquid ^4He are quasi-particles.