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INTERNATIONAL CENTRE FOR THEORETICAL PHYSICS
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UNITED NATIONS INDUSTRIAL DEVELOPMENT ORGANIZATION

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SMR. 628 - 16

Research Workshop in Condensed Matter,
Atomic and Molecular Physics
(22 June - 11 September 1992)

Working Party on:
"Energy Transfer in Interactions with
Surfaces and Adsorbates"
(31 August - 11 September 1992)

"Quantum Versus Semiclassical Regimes of Sticking"

PART I

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Quantum versus semiclassical regimes of sticking

1) Quantum effects in substrate { phonons, e-h-pairs

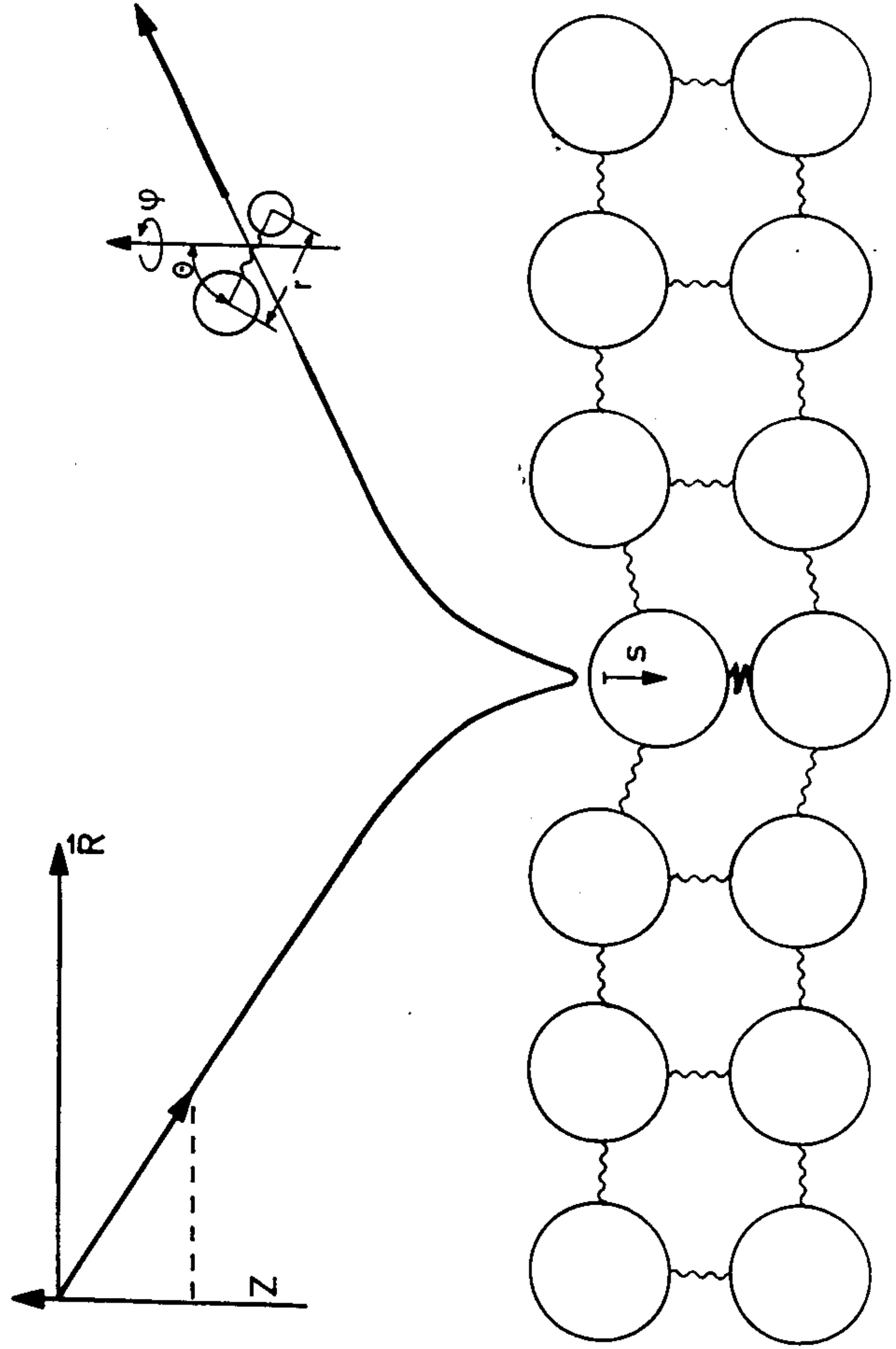
Coworkers:
Böheim, Brunner

Experiments:
Schlichting, Menzel; Brunner, B.
J. Chem. Phys. 97, 1('92)

2) Quantum effects in adsorbate

Coworkers:
Böheim, Stutzki, Russ

Experiments:
J. Doyle, Kleppner, Greytak
P.R.L. 67, 603 ('91)
(Therry: P.R.L. 68 412 ('92)
Carraro, Cole P.R. B 45 12930 ('92)



1) Single Quantum Oscillator driven by classical force $f(t)$:

Goldmann, Krivchenkov, Kogan, Galitzkii: "Selected probl. in Q.M." (ter Haar ed. 1964); Feynman (1948?)

Average energy transfer

$$\Delta\varepsilon = \frac{1}{2m}\,|f(\omega)|^2 \qquad \omega = \text{osc. freq.}$$

$$c = \Delta\varepsilon/\hbar\omega$$

Energy distribution of oscill. (initially at rest)

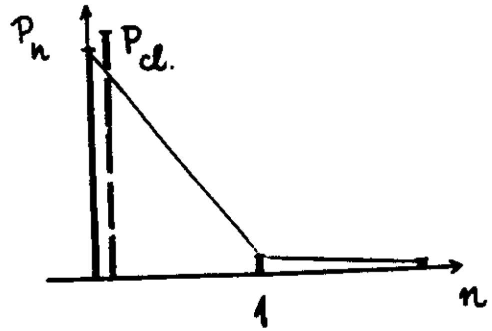
$$P_n = \frac{c^n}{n!}\,e^{-c} \qquad \text{(Poisson distr.)}$$

in n-th quantum state

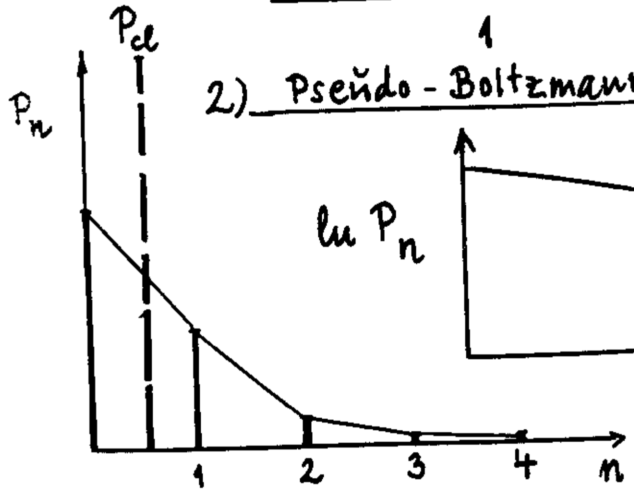
$$P_0 = e^{-\Delta\varepsilon/\hbar\omega} = \text{Debye-Waller factor}$$

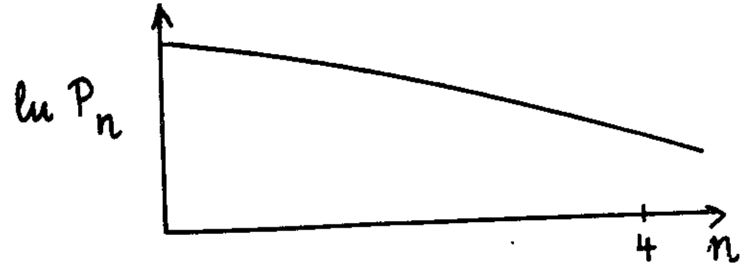
Three typical cases $\quad c = \frac{\Delta}{\hbar\omega}$

1) One Phonon case $c \ll 1$

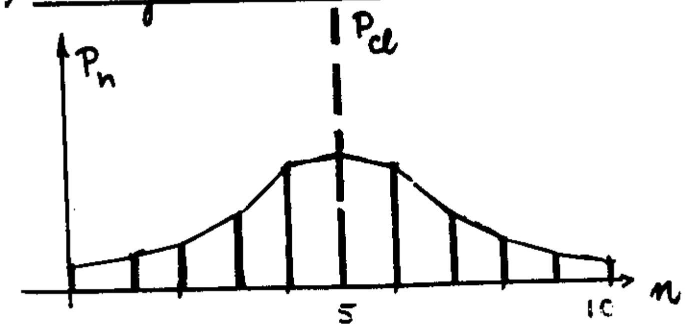


2) Pseudo-Boltzmann case $c \approx 0.5$





3) Many Phonon case $c \gg 1$



note: $\langle \Delta\varepsilon^2 \rangle = \hbar\omega \langle \Delta\varepsilon \rangle$

2) Generalization to surface physics:

a) fixed force $f(t) = f(z(t))$

$z(t)$ trajectory in static potential $V(z)$.

Kaplan, Drakoglis; Surf. Sci 36 1('73)

Garibaldi, Levi, Spadacini, Tommei; " 48 649 ('75)

b) With recoil + quantum effects of adsorbate in cumulant expansion (exponentiated perturbation theory)

B.: Z. Phys B 36 81('79)

Böheim, B: " B41 ('81)

In both a) and b) a system of quantum oscil. at $T \neq 0$ is considered.

Energy distr. $p(\varepsilon, \varepsilon')$ of particle after "1st roundtrip"

$$\mathcal{U} = T e^{i\int V(t)\,dt} \quad ; \quad V = V_0 - \langle V_0 \rangle$$

$$p(\varepsilon', \varepsilon) = \int \langle \varepsilon | \mathcal{U}^+ e^{iH_s\tau} \mathcal{U} e^{-iH_s\tau} | \varepsilon \rangle e^{-i\varepsilon'\tau} d\tau$$

$$\Rightarrow \int e^{\langle \varepsilon | \int V(t)(V_\tau(t') - V(t'))\,dt\,dt' | \varepsilon \rangle - i\varepsilon'\tau} d\tau$$

(2nd order cumul.)

$$\boxed{p = \int d\tau\, e^{-i\varepsilon'\tau} \exp \int |f_\varepsilon(\omega)|^2 s(\omega)(e^{i\omega\tau} - 1)\,d\omega}$$

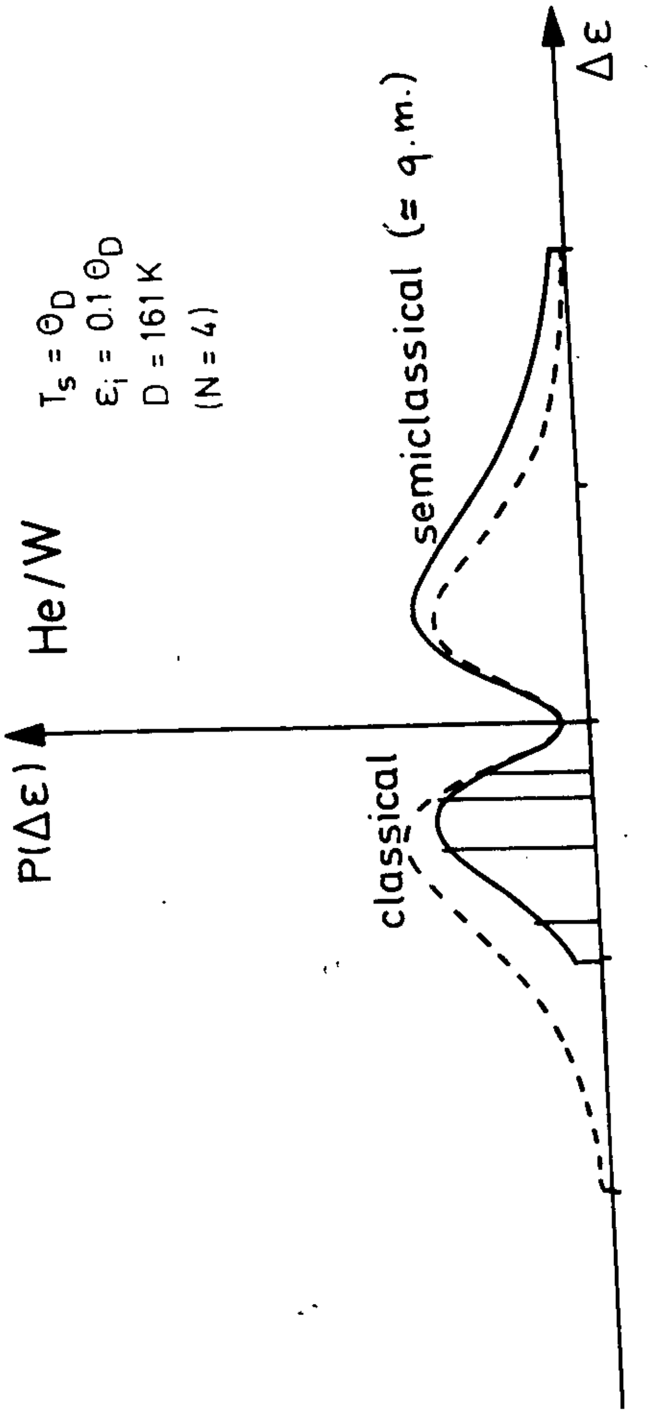
$$f_\varepsilon(\omega) = \langle \varepsilon | \frac{\partial V}{\partial z} | \varepsilon + \omega \rangle$$

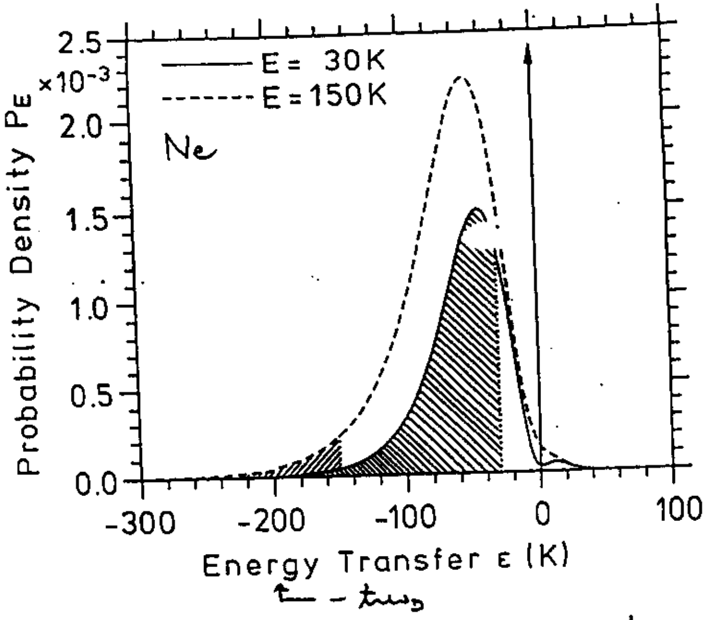
$$\Rightarrow \int f^{(t)}_{\varepsilon + \hbar\omega/2} e^{i\omega t} dt \quad \text{(semiclass. trajectory)}$$

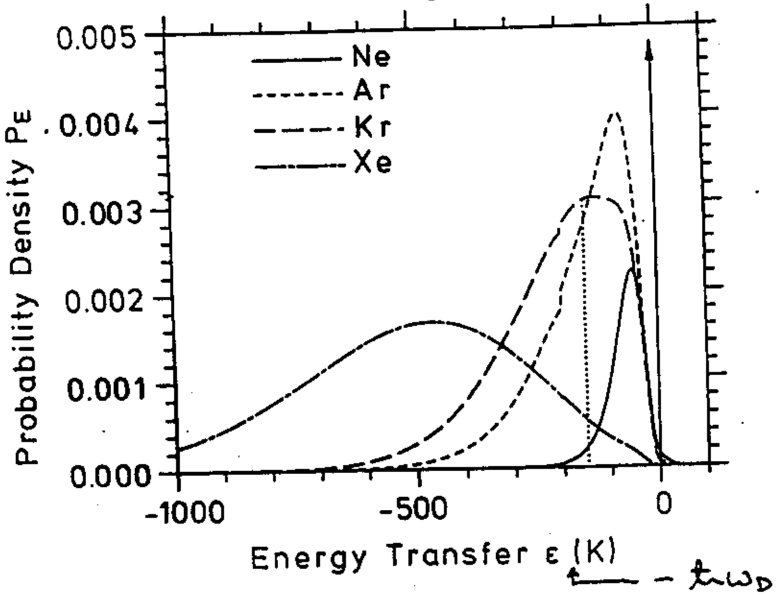
$$s(\omega) = \chi''(\omega)\,(1 + n(\omega, T_s)) \quad \text{(displacement corr.}$$

FT of $\langle x(t)\, x(0) \rangle$

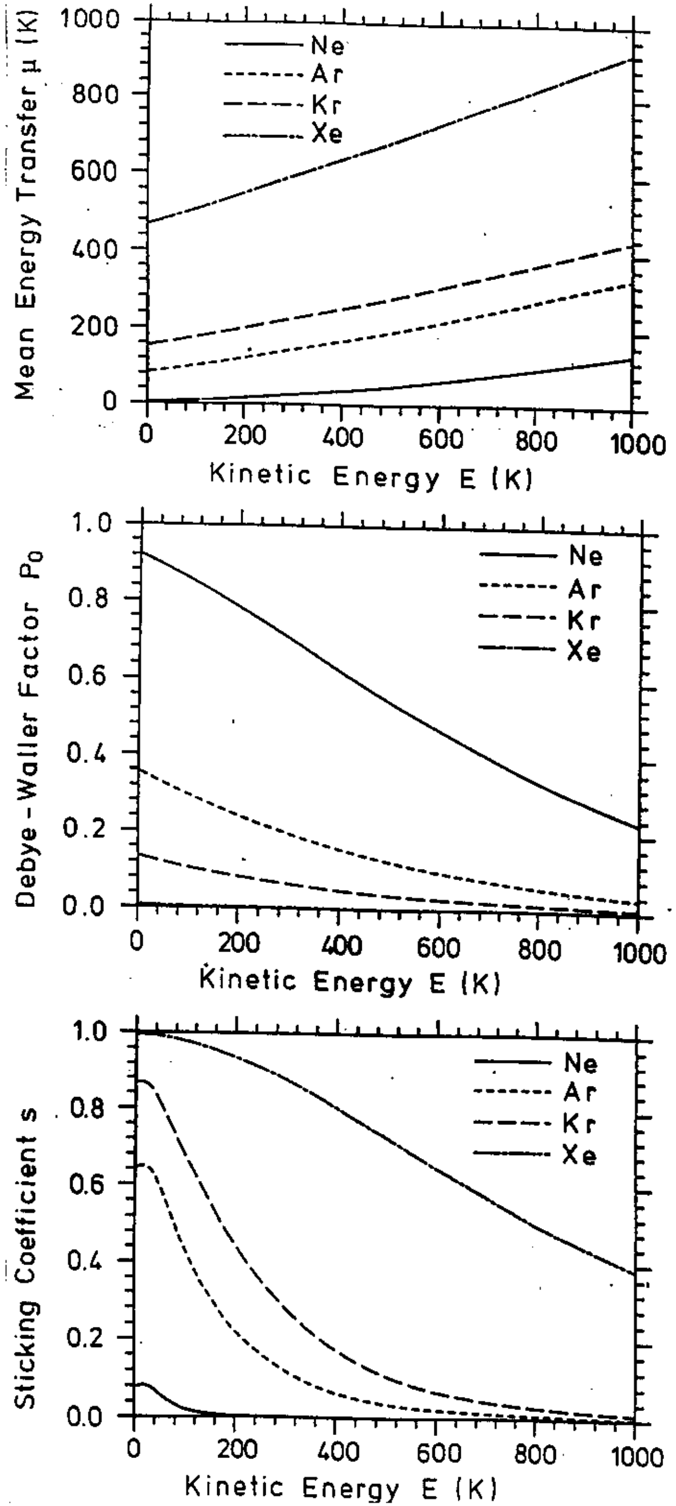
$$\chi'' = \omega \begin{cases} \dfrac{1}{M\omega_D^3} & ; \ |\omega| \leq \omega_D \ ; \text{ phonon} \\ \dfrac{1}{m_{el}\,\omega_f^3} & ; \ |\omega| \lesssim v_f \bar{k} \ ; \text{ electron-hole pair} \end{cases}$$

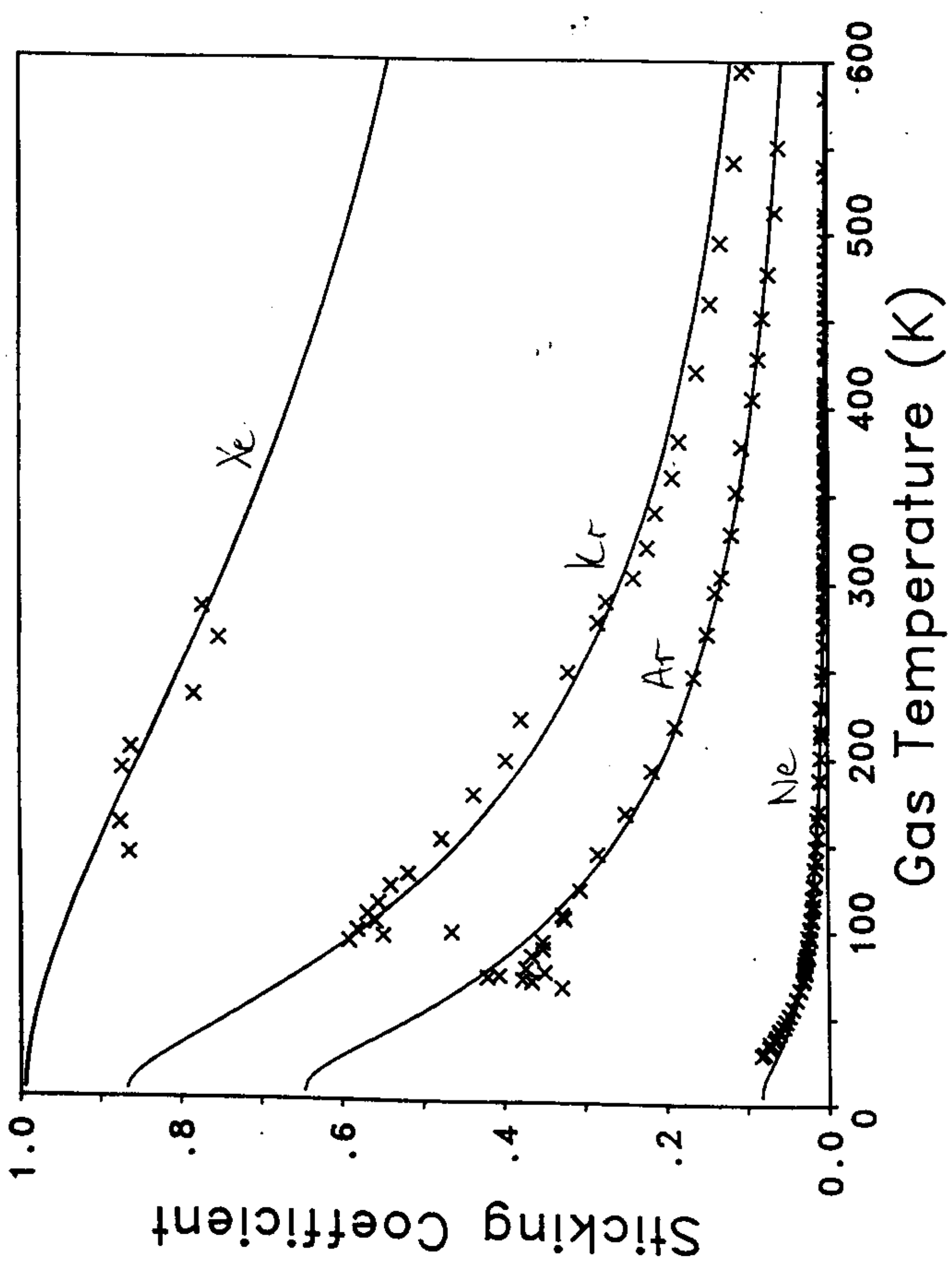






Schlichting, Menzel, Brunner, Brenig, JCP, in print


Mean Energy Transfer μ (K)
Kinetic Energy E (K)
Ne
Ar
Kr
Xe
Debye-Waller Factor P_0
Kinetic Energy E (K)
Ne
Ar
Kr
Xe
Sticking Coefficient s
Kinetic Energy E (K)
Ne
Ar
Kr
Xe


Sticking Coefficient
Gas Temperature (K)
Xe
Kr
Ar
Ne

2) Quantum reflection of adsorbate at low energy.

I Short range potential

a) Lennard-Jones, Strachan: Proc. Roy. Soc. A150 442 ('35) (DWBA)

b) B. Z. Phys. B36 227 ('80) (non perturb.)

$$\left[\psi(z \approx 0)/\psi(\infty)\right]_{WKB} \approx \frac{E^{1/4}}{\sqrt{v(0)}} = \left(\frac{E}{D}\right)^{1/4}$$

$$\left[\psi(z \approx 0)/\psi(\infty)\right]_{QM} \approx \left(\frac{E}{D}\right)^{1/2}$$

$$\psi_{E=0}(z) \Rightarrow z - a \quad (z \to \infty) \to$$

$$s(E) \propto |\langle \psi_E | H_{int} | \psi_b \rangle|^2 / \sqrt{E} \propto \sqrt{E}$$

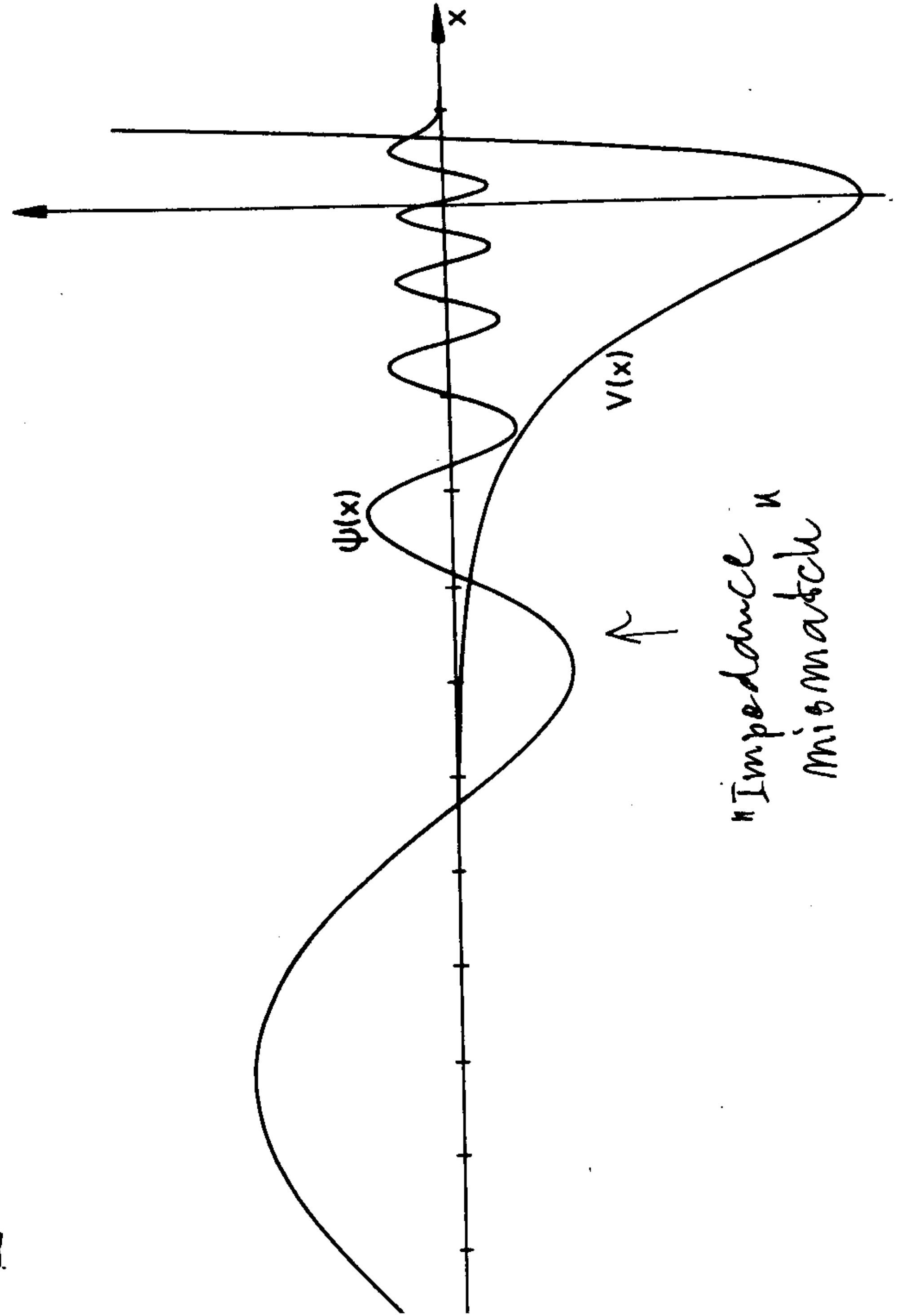
II Van der Waals potential

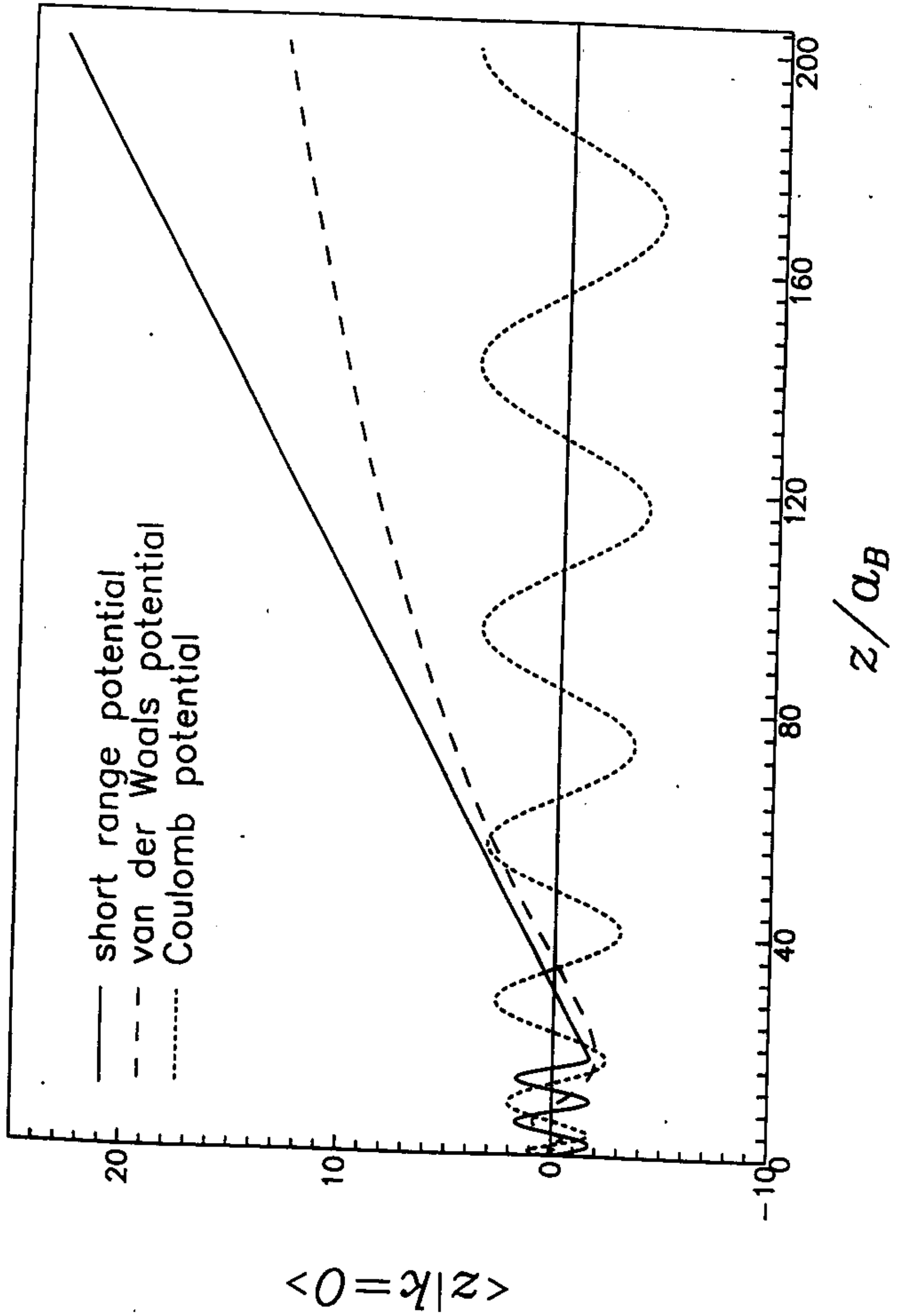
$$\psi_{E=0}(z) \Rightarrow z - a \ln z$$

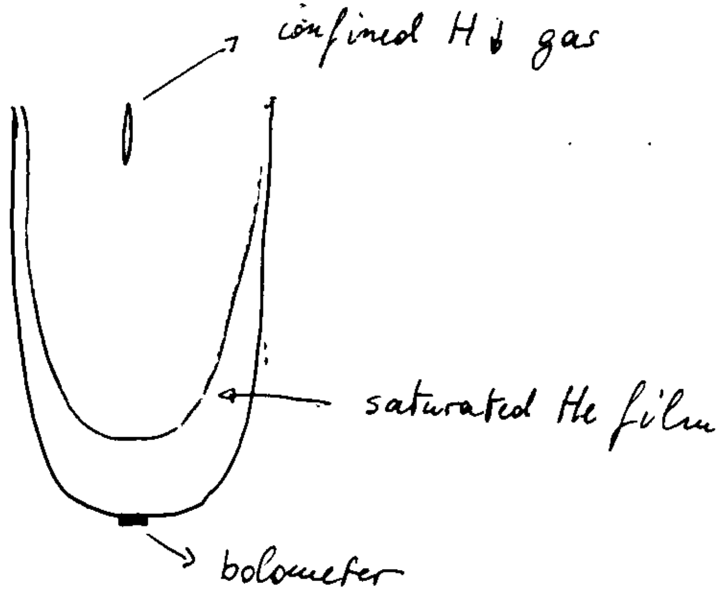
still: $s(E) \to \sqrt{E}$ but turnover from classical to q.m. at much lower E (for He at about 10^{-4} K)

Lit. B., Böheim, Stützki: Z.Ph. B48 48 ('8

Caravo Cole: P.R.L. 68 412 ('92)







Turn off confining fields

Measure decay rate of number of gas atoms

$$\frac{1}{\tau(E)} = \eta \left(\frac{2E}{MD^2}\right)^{1/2} s(E)$$

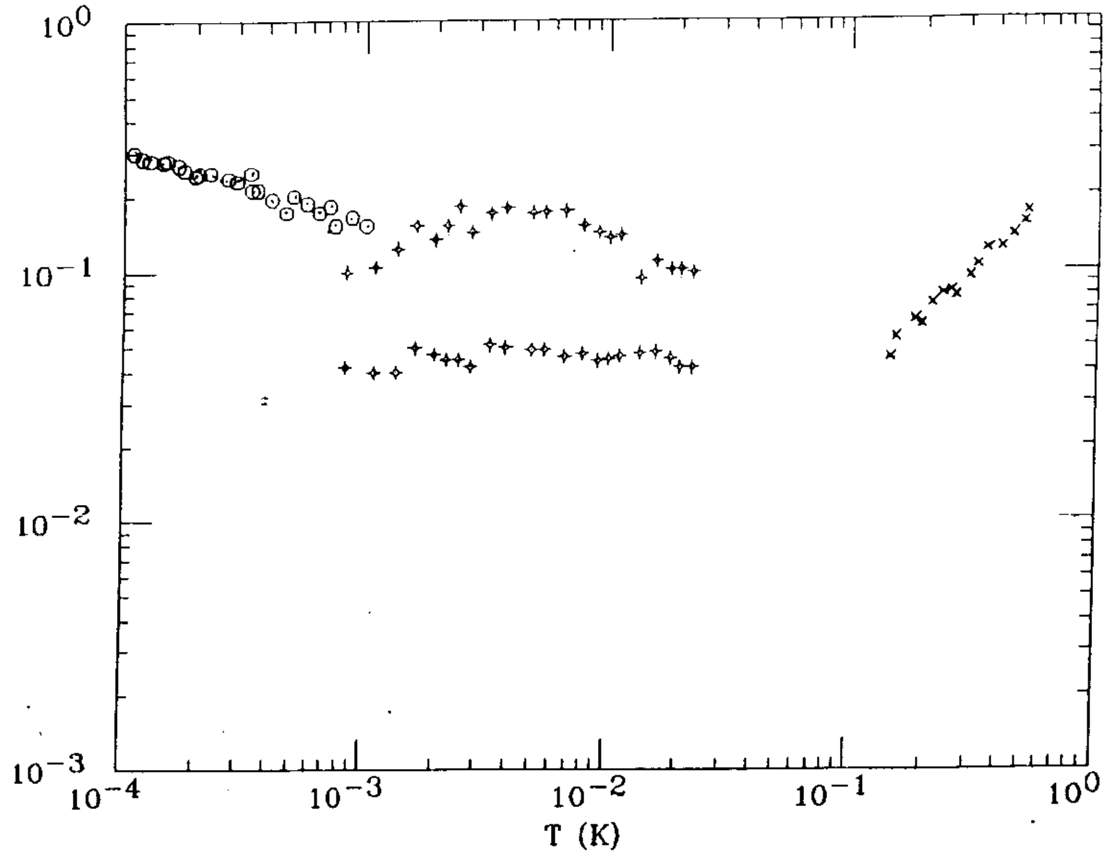
D = cell diameter

M = m_H

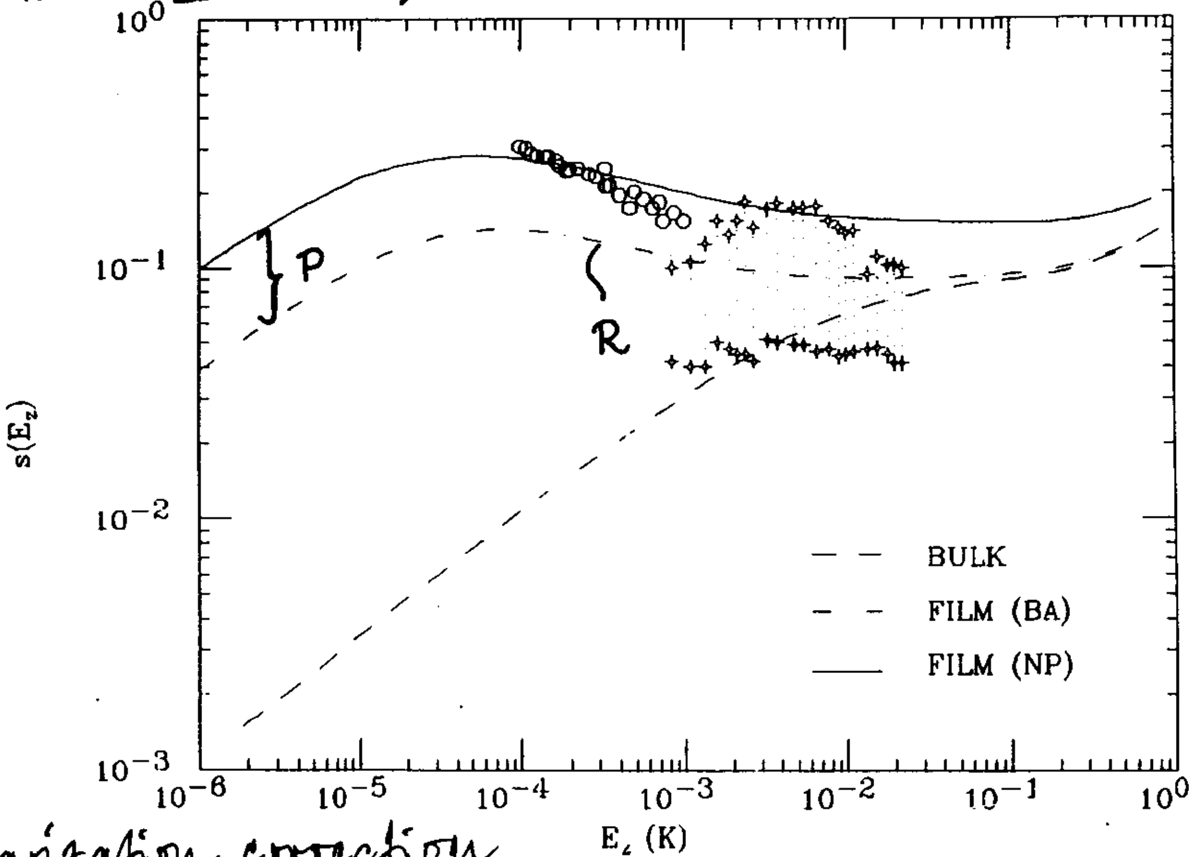
η = geometric factor

Berkhout et al., Phys. Rev. Lett. 57, 2387 (1986)

l on the
rface of
quid ^4He



R: Resonant increase of s(E) (bound state near E = 0)
(C.C. P.R.L. 68 412 ('92))



P: Polarization correction
to DWBA  (B.B.St. Z.Ph. B48 48 ('83))

III Coulomb forces (image force $\frac{e^2}{4\varepsilon \cdot z}$)

B., Rüss: Surf. Sci. ('92) in print

Main difference compared to I, II:

$\psi_{E=0}(z) \approx \psi_{WKB} \rightarrow$

classical result ≈ correct: $s(0) \neq 0$

in 1-phonon approximation

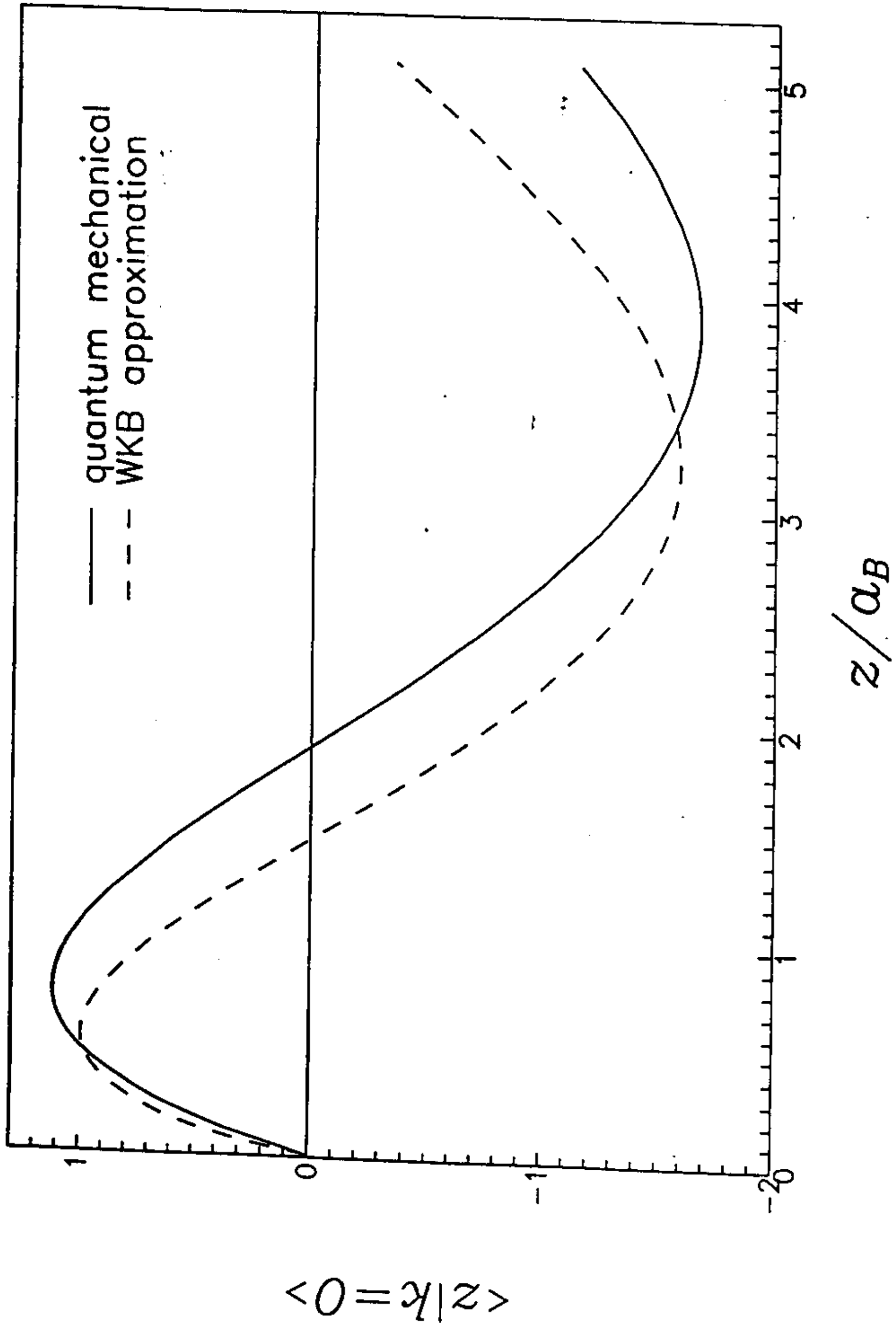
$$s(0) \approx \Delta E/\hbar\omega_D \approx \frac{4me^2}{M a_B^*}\frac{1}{\hbar\omega_D}$$

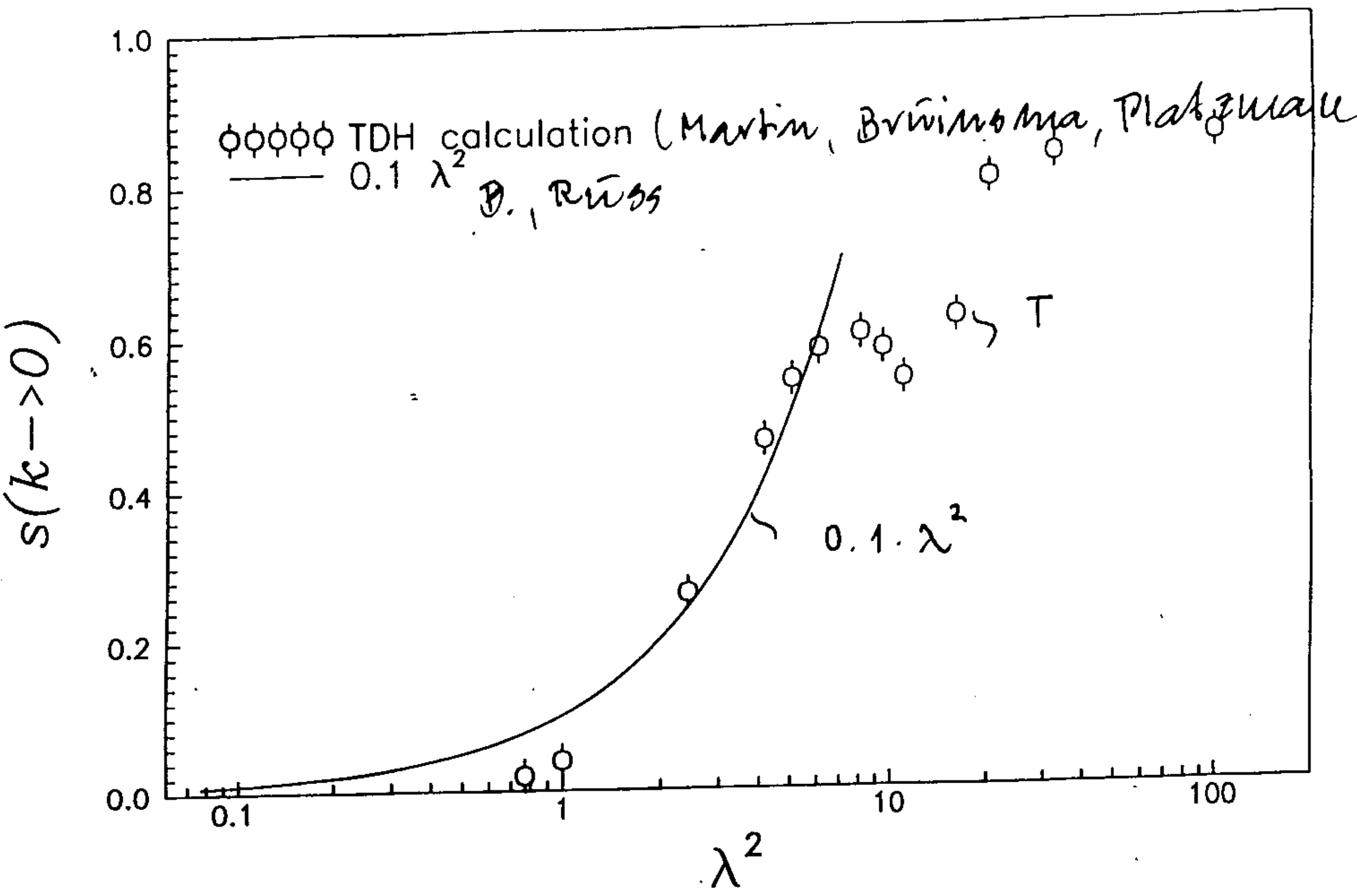
$$\approx \frac{\langle u^2 \rangle}{a_B^{*2}} = \lambda^2 \text{ (electrons/He)}$$

u mean square vib. ampl.

Due to (quantitative) deviations of WKB from QM: $\lambda^2 \rightarrow 0.1\lambda^2$

There is NO "destruction of quantum interference by dissipation" as claimed by Martin, Bruinsma and Platzman Phys Rev. B38 2257 ('88), B39 1241 ('89)




TDH calculation (Martin, Brüinsma, Platzman
0.1 λ²
B., Rüss
T
0.1 · λ²
s(k->0)
λ²
0.0
0.2
0.4
0.6
0.8
1.0
0.1
1
10
100