Dilution refrigerators - concepts

Concept of Landau's Fermi liquid theory

elementary excitations of interacting Fermions are described by almost independent fermionic quasiparticles

state of Fermi liquid described simply by quasiparticle distribution

Phenomenological Theory by Landau

energy functional:

$$E = E_0 + \sum_{\vec{k},\sigma} \epsilon_{\sigma}(\vec{k}) \delta n_{\sigma}(\vec{k}) + \frac{1}{2\Omega} \sum_{\vec{k},\vec{k}'} \sum_{\sigma,\sigma'} f_{\sigma\sigma'}(\vec{k},\vec{k}') \delta n_{\sigma}(\vec{k}) \delta n_{\sigma'}(\vec{k}')$$

deviation from ground state

ground state distribution

$$\delta n_\sigma(\,ec{k}\,) = n_\sigma(\,ec{k}\,) - n^{(0)}_\sigma(\,ec{k}\,)$$

spin index $\,\sigma=\pm 1\,$

 $n^{(0)}_{\sigma}(\,ec{k}\,) = \Theta(k_F - |\,ec{k}\,|)$

filled Fermi sea

Fermi liquid theory

$$E = E_0 + \sum_{\vec{k},\sigma} \epsilon_{\sigma}(\vec{k}) \delta n_{\sigma}(\vec{k}) + \frac{1}{2\Omega} \sum_{\vec{k},\vec{k}'} \sum_{\sigma,\sigma'} f_{\sigma\sigma'}(\vec{k},\vec{k}') \delta n_{\sigma}(\vec{k}) \delta n_{\sigma'}(\vec{k}')$$

effective quasiparticle spectrum:

$$\tilde{\epsilon}_{\sigma}(\vec{k}) = \frac{\delta E}{\delta n_{\sigma}(\vec{k})} = \epsilon_{\sigma}(\vec{k}) + \frac{1}{\Omega} \sum_{\vec{k}',\sigma'} f_{\sigma\sigma'}(\vec{k}, \vec{k}') \delta n_{\sigma'}(\vec{k}')$$

bare quasiparticle spectrum:

Fermi velocity:

$$\epsilon_{\sigma}(\vec{k}) = \frac{\hbar^2 \vec{k}^2}{2m^{**}} \leq \frac{\text{effective}}{\text{mass}} \left. \frac{1}{\hbar} \vec{\nabla}_{\vec{k}} \epsilon_{\sigma}(\vec{k}) \right|_{k_F} = \vec{v}_F = \frac{\hbar \vec{k}_F}{m^*}$$

density of states at ϵ_F :

$$N(\epsilon_F) = \frac{1}{\Omega} \sum_{\vec{k},\sigma} \delta(\epsilon_{\sigma}(\vec{k}) - \epsilon_F) = \frac{k_F^2}{\pi^2 \hbar v_F} = \frac{m^* k_F}{\pi^2 \hbar^2}$$

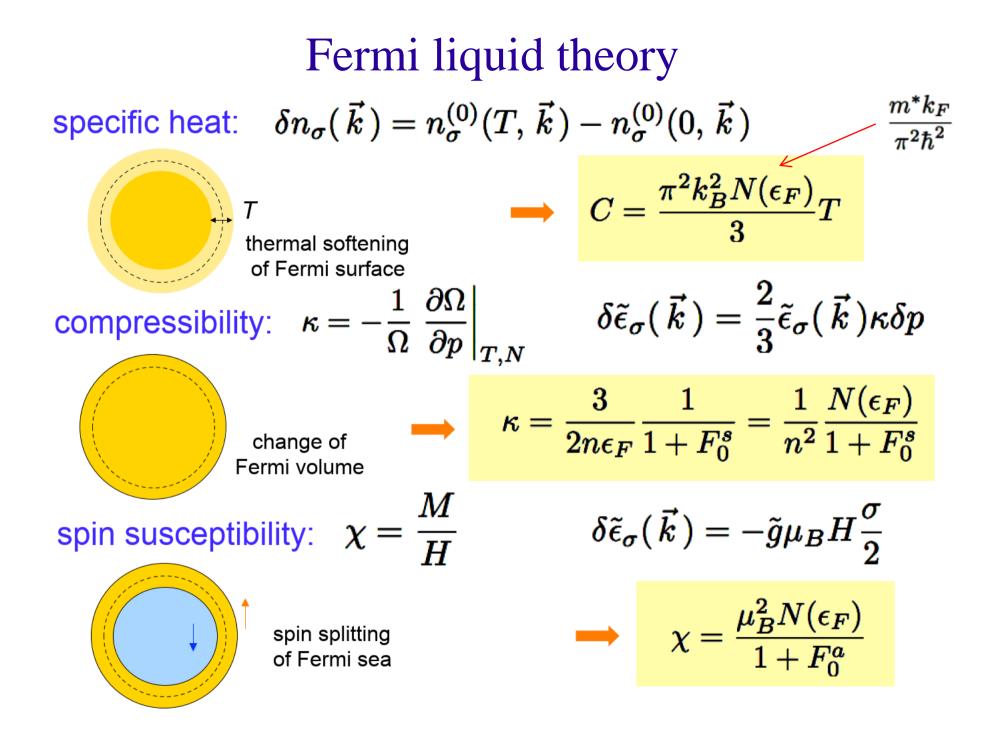
Fermi volume conserved

$$k_F = (3\pi^2 n)^{1/3}$$

Fermi liquid theory

$$\begin{split} E &= E_0 + \sum_{\vec{k},\sigma} \epsilon_{\sigma}(\vec{k}) \delta n_{\sigma}(\vec{k}) + \frac{1}{2\Omega} \sum_{\vec{k},\vec{k}'} \sum_{\sigma,\sigma'} f_{\sigma\sigma'}(\vec{k},\vec{k}') \delta n_{\sigma}(\vec{k}) \delta n_{\sigma'}(\vec{k}') \\ \text{couplings:} \quad & f_{\sigma\sigma'}(\vec{k},\vec{k}') = f^s(\hat{k},\hat{k}') + \sigma\sigma'f^a(\hat{k},\hat{k}') \\ & \text{symmetric} \\ \text{(charge)} & \text{antisymmetric} \\ \text{(spin)} \\ \end{split}$$

$$\begin{aligned} \text{spherical symmetry:} \quad f^{s,a}(\hat{k},\hat{k}') = \sum_{l=0}^{\infty} f_l^{s,a} P_l(\cos\theta_{\hat{k},\hat{k}'}) & \text{Legendre Polynomials} \\ \text{Landau parameters:} & \int_{-1}^{+1} dz \ P_l(z) \ P_{l'}(z) = \frac{2\delta_{ll'}}{2l+1} \\ & \prod_{|\mathbf{x}-\mathbf{x}'|} = \frac{1}{\sqrt{r^2 + r'^2 - 2rr'\cos\gamma}} = \sum_{\ell=0}^{\infty} \frac{r'^{\ell}}{r^{\ell+1}} P_{\ell}(\cos\gamma), \\ P_0(x) = 1, \ P_1(x) = x, \ P_2(x) = (3x^2 - 1)/2, \ \text{etc.} \\ \end{aligned}$$



Let us now compute the *compressibility*:

$$\begin{bmatrix} V = N/n \\ Vdp = dE, \ dE/N = d\mu \end{bmatrix}$$

$$\kappa = -\frac{1}{V}\frac{\partial V}{\partial P} = \frac{1}{n^2}\frac{\partial n}{\partial \mu} \qquad \delta n = \frac{1}{V}\sum_{p,\sigma}\delta n_{p,\sigma} \qquad (5.38)$$

where P is the pressure. At T = 0,

$$\delta n_{\sigma}(\vec{p}) = \frac{\partial n_{\sigma}(\vec{p})}{\partial \varepsilon_{\sigma}(\vec{p})} (\delta \varepsilon_{\sigma}(\vec{p}) - \delta \mu) \qquad \delta \varepsilon_{p,\sigma} = \frac{1}{V} \sum_{p'\sigma'} f_{p\sigma,p'\sigma'} \delta n_{p'\sigma'}$$

The quasiparticle energy $\varepsilon_{\sigma}(\vec{p})$ depends on μ only through its dependence on $\delta n_{\sigma'}(\vec{p'})$ (*i.e.*, quasiparticle interactions, see Eq. 5.9). As $T \to 0$ both $\frac{\partial n}{\partial \varepsilon}$ and $\delta n_{\sigma}(p)$ vanish unless all momenta are *at* the Fermi-surface.

$$\delta \varepsilon_{\sigma}(p) = f_0^S \frac{1}{V} \sum_{\sigma', \vec{p}'} \delta n_{\sigma'}(\vec{p}') \equiv f_0^S \delta n \tag{5.40}$$

G. Baym and C. Pethick, Landau Fermi liquid theory where f_0^S is Landau parameter with with l = 0. Hence, we have

$$\delta n_{\sigma}(\vec{p}) = \frac{\partial n_{\sigma}(\vec{p})}{\partial \varepsilon_{\sigma}(\vec{p})} (f_0^S \delta n - \delta \mu)$$
(5.41)

and

$$\delta n = \frac{1}{V} \sum_{\sigma, \vec{p}} \delta n_{\sigma}(\vec{p}) = \frac{1}{V} \sum_{\sigma, \vec{p}} \frac{\partial n_{\sigma}(\vec{p})}{\partial \varepsilon_{\sigma}(\vec{p})} (f_0^S \delta n - \delta \mu)$$
(5.42)

Similarly,

$$\frac{\partial n_{\sigma}(\vec{p})}{\partial \varepsilon_{\sigma}(\vec{p})} \longrightarrow -\delta(|p| - p_F) \text{ for } T \to 0$$
(5.43)

Thus,

$$\delta n = -N(0)(f_0^S \delta n - \delta \mu) \tag{5.44}$$

and

$$\delta n[1 + N(0)f_0^S] = N(0)\delta\mu \tag{5.45}$$

using the expression for the (s-wave) symmetric (singlet) Landau parameter,

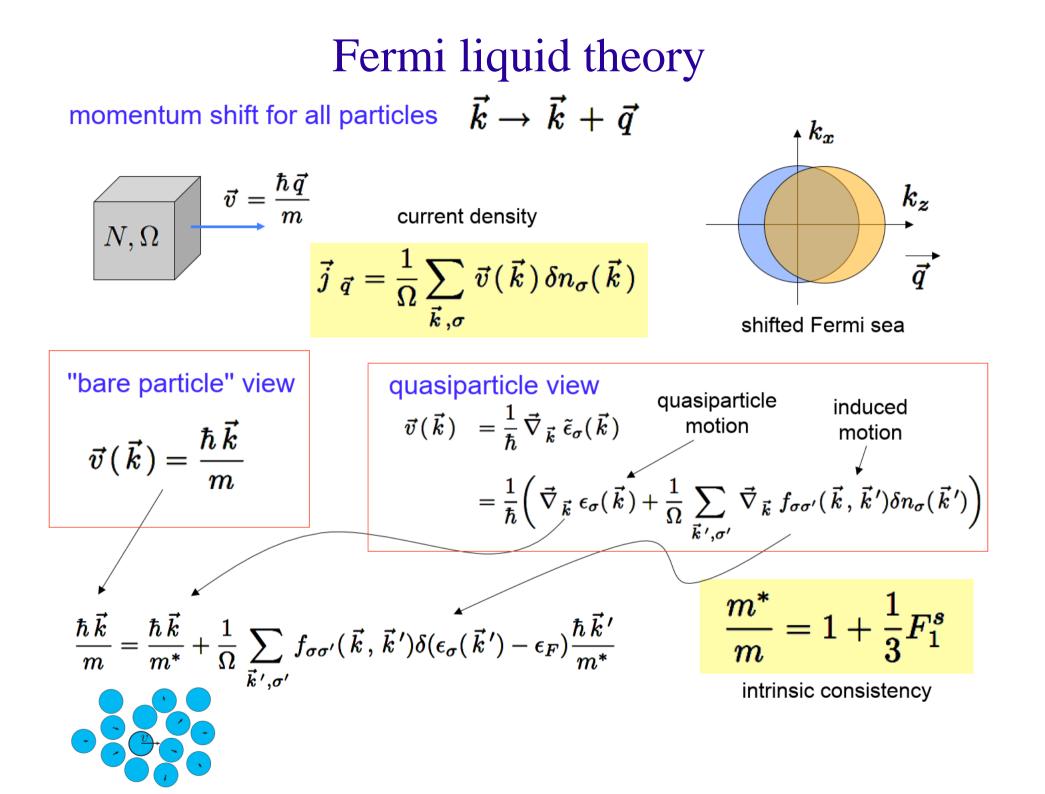
$$F_0^S = N(0)f_0^S (5.46)$$

we can write

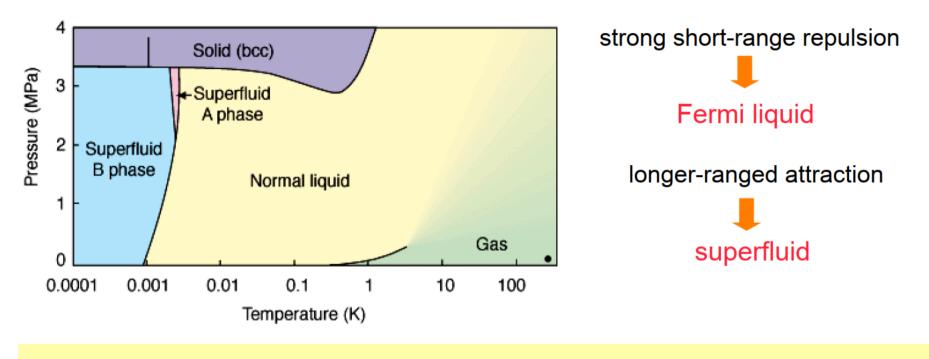
$$\frac{\partial n}{\partial \mu} = \frac{N(0)}{1 + F_0^S} \tag{5.47}$$

which leads to an expression for the *compressibility* κ :

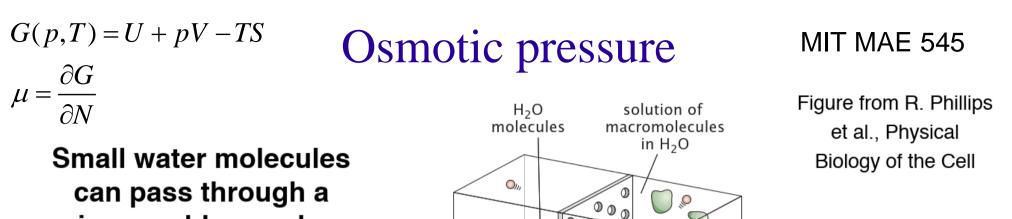
$$\kappa = \frac{1}{n^2} \frac{N(0)}{1 + F_0^S} \qquad \begin{array}{l} F_0^s > 10, \text{ "less} \\ \text{compressible"} \end{array} \tag{5.48}$$



³He as an example Fermi liquid



| | pressure | m^*/m | F_0^s | F_0^a | F_1^s | κ/κ_0 | χ/χ_0 |
|------------------------------|----------------------|---|--------------|---------|-----------------|-------------------|---------------|
| | 0 | 3.0 | 10.1 | -0.52 | 6.0 | 0.27 | 6.3 |
| | $< p_c$ | 6.2 | 94 | -0.74 | 15.7 | 0.065 | 24 |
| enhanced diminished enhanced | | | | | | | |
| $\frac{m^*}{m}$ = | $=1+rac{1}{3}F_1^s$ | $\frac{1+\frac{1}{3}F_{1}^{s}}{Speed of (first) sound} c = v_{F}\sqrt{\frac{1}{3}(1+F_{0}^{s})(1+\frac{1}{3}F_{1}^{s})}.$ $c_{0} = v_{F}\sqrt{\frac{F_{0}^{(s)}}{3}} Speed of zero sound$ | | | | | |
| | | Spe | ed of (first | c_0 | $-v_F \sqrt{3}$ | zero sour | |



щО

semipermeable membrane, which blocks large solute macromolecules.

ഗര 0 pressure p_2 pressure semipermeable p_1 membrane

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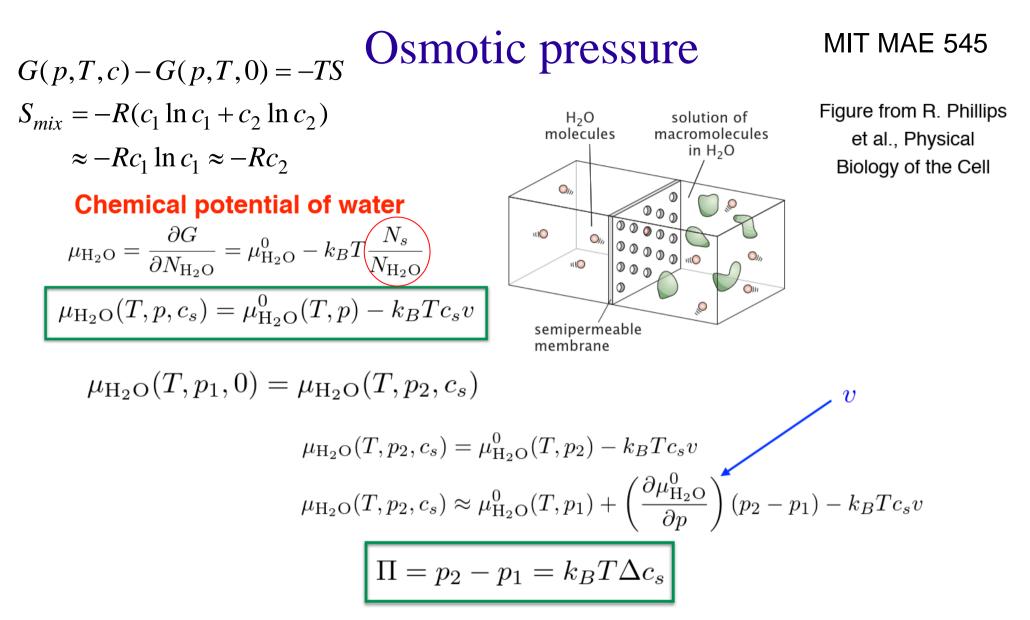
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 $G = N_1 \mu_{\rm H_2O}(T, p_1, 0) + N_2 \mu_{\rm H_2O}(T, p_2, c_s) + N_s \mu_s(T, p_2, c_s)$

In thermodynamic equilibrium the Gibbs free energy G is minimized, which means that chemical potentials of water are the same on both sides of the semipermeable membrane!

 $\mu_{\rm H_2O}(T, p_1, 0) = \mu_{\rm H_2O}(T, p_2, c_s)$

 $H_2O => {}^4He$ membrane => superleak



Osmotic pressure depends only on temperature and concentration difference across the membrane!

Dilution refrigerator

Only available CONTINUOUS cooling method below 0.3 K

- 1951 H. London proposed the operating principle at LT meeting in Oxford
- 1962 First practical concept about it (London, Clarke & Mendoza)
- -1965 First realization (Das, De Bruyn Ouboter & Taconis, Leiden, 0.22 K)
- 1966 Dubna, 25 mK
- 1999 Lancaster, 1.7 mK

Physical grounds: QUANTUM-effects make it possible !

- ³He atoms take more space than ⁴He (zp motion) => distinguishable
- ³He dissolves into ⁴He (Fermi/Bose systems) even at absolute zero temperature; $x_{3s}(T=0) = 6.6$ %
- molar entropy of ³He is higher in dilute mixture than in pure phase; heat of mixing: $L_{\rm m} = 84 \ T^2 \ J/({\rm mol} \ {\rm K}^2)$ $L=\Delta ST; \ \Delta S \sim T$
- -4He is superfluid and has no entropy (T < 0.5 K)
- osmotic pressure of ³He keeps balance against thermal gradient in mixture
- ³He vapor pressure higher than of ⁴He (possible to distill ³He out of ⁴He)

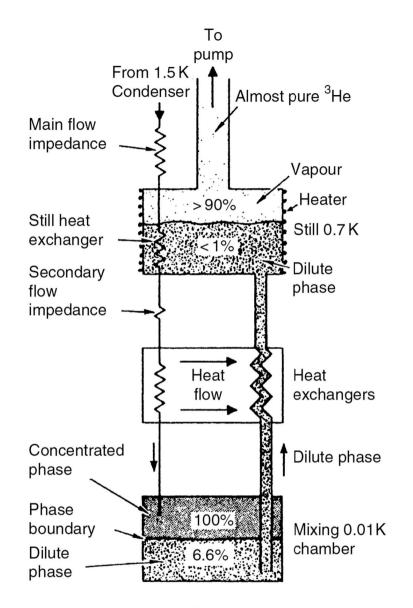


Fig. 7.9. Schematic ${}^{3}\text{He}{-}^{4}\text{He}$ dilution refrigerator. This part will sit in a vacuum chamber that is immersed in a ${}^{4}\text{He}$ bath at 4.2 K. The incoming ${}^{3}\text{He}$ gas is condensed on a continuously operating ${}^{4}\text{He}$ pot at 1.5 K (Sect. 5.2.4)

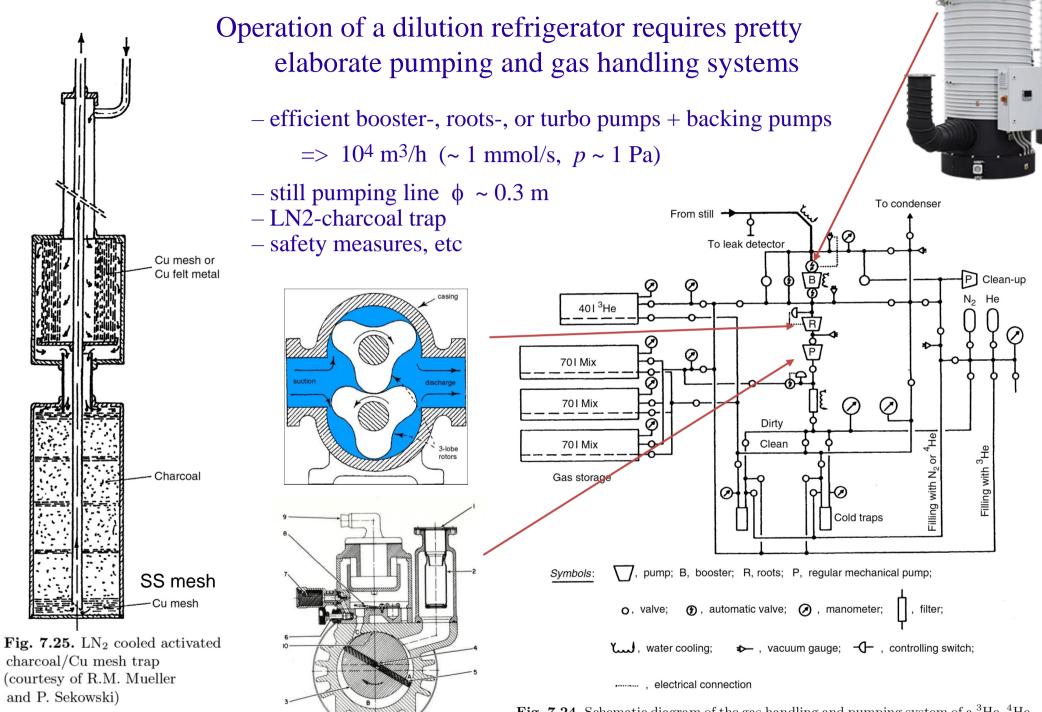


Fig. 7.24. Schematic diagram of the gas handling and pumping system of a ³He–⁴He dilution refrigerator (courtesy of P. Sekowski, Universität Bayreuth)

Dilution units are commercially available (price around 200-300 k€...700k€)

- Oxford Instruments, UK
- Leiden Cryogenics, The Netherlands
- Bluefors Cryogenics, Finland
- etc.

Special types may be adapted to specific conditions:

• Dry cryostats

- pulse tube cooler provides the ~ 2 (3.5) K base
- have become the mainstream of mK-refrigerators
- just one vacuum space for the pulse tube and the dilution fridge

• Miniature cryostats

- can be dipped into a storage dewar (often max diam ~ 5 cm)

– quick operation, $T_{\min} \sim 10 \dots 20$ mK in couple hours, limited power

• Fully plastic dilution fridges

- can be operated in very high magnetic fields
- Kapitza resistance between helium and plastics is smaller than He-metal

• Monster machines

 $-dn/dt \sim 10 \text{ mmol/s}$ with $A_{\text{hex}} > 2000 \text{ m}^2$



Mixing chamber diameter 490 mm

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CF-CS110-1500 Maglev-2PT Tmin < 7 mK

Q > 1500 microW @ 100 mK

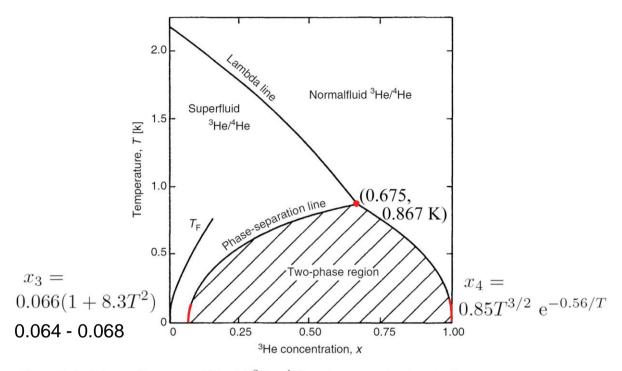


Fig. 7.1. Phase diagram of liquid ³He–⁴He mixtures at saturated vapour pressure. The diagram shows the lambda line for the superfluid transition of ⁴He, the phase separation line of the mixtures below which they separate into a ⁴He-rich and a ³He-rich phase, and the line of the Fermi temperatures $T_{\rm F}$ of the ³He component

Helium isotopes T < 0.5 K ⁴He - Bose condensate - no thermal excitations $-\eta = 0, S \sim 0, C \sim 0$

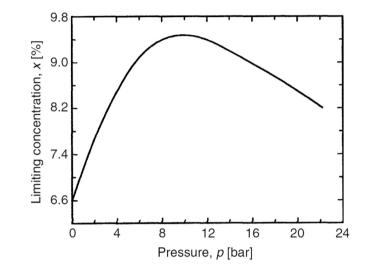
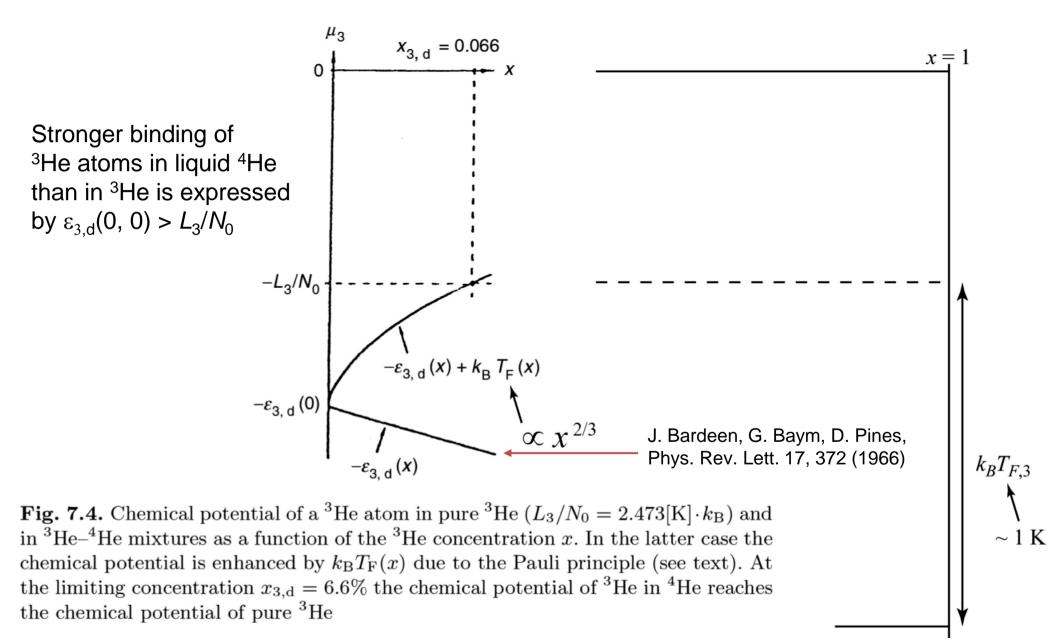
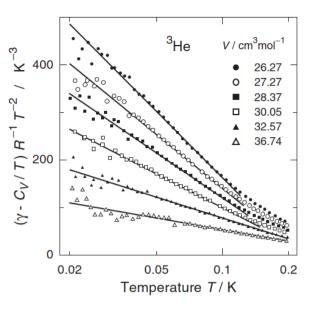


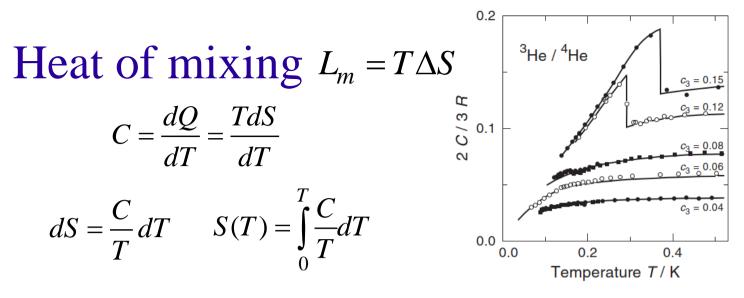
Fig. 7.2. Limiting low-temperature concentration of ³He in ⁴He at T = 50 mK as a function of pressure

³He - Fermi fluid - $T_F \sim 1 \text{ K}$ - $C = \pi^2 R/2 T/T_F$ - $m^* \sim 2.8 (3.0) \text{ m}_3$ $T_F = \frac{\hbar^2}{2m_3^* k_B} (3\pi^2 n_3)^{2/3}$

Finite solubility of ³He in ⁴He at T = 0







$$C_3 = 2.7 RT = 22 T J/(\text{mol K}^2)$$

 $S_3 = 22 T J/(\text{mol K}^2)$

– empirical fact– no first-principles theory

$$C_V = \gamma T + \Gamma T^3 \ln \left(\frac{T}{\Theta_{\rm c}}\right)$$

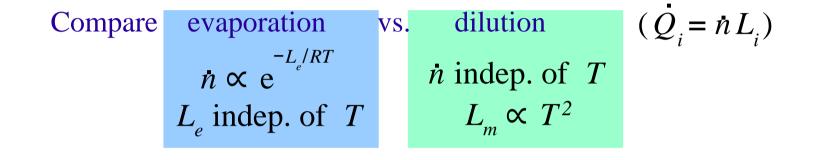
Fermi-liquid theory works fine:
$$C_d = N_A k_B \frac{\pi^2}{2} \frac{T}{T_F} = \frac{0.745 \text{ J}}{\text{mol K}} \frac{T}{\text{K}} \frac{m^*}{m_3} \left(\frac{V_m \text{ mol}}{x \text{ cm}^3}\right)^{2/3}$$

 $x = 0.066, \quad m^* = 2.5 m_3$
 $x \to 0, \qquad m^* = 2.34 m_3$
 $=> C_d (0.066) = 106 T \text{ J/(mol_3 K^2)}$
 $=> S_d (0.066) = 106 T \text{ J/(mol_3 K^2)}$

THUS: $L_{\rm m} = T\Delta S = T^2 (106 - 22) \text{ J/(mol K}^2) = 84 (T/\text{K})^2 \text{ J/mol}$

Cooling power

$$\dot{Q} = n L_m = 84 n (T/K)^2 J/mol$$



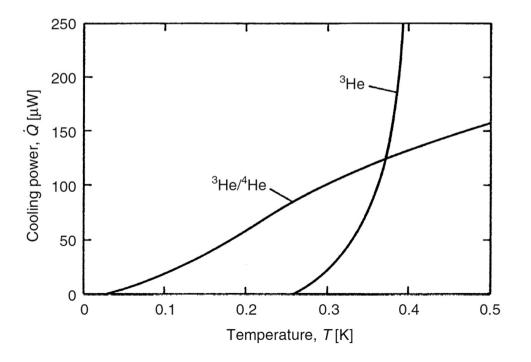


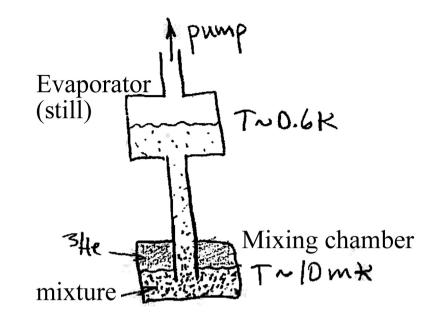
Fig. 7.3. Cooling power of a ³He evaporation cryostat and of a ³He–⁴He dilution refrigerator, assuming that the same pump with a helium gas circulation rate of $5 l s^{-1}$ is used [7.9]

Osmotic pressure

Large thermal gradient in DR causes concentration gradients, x(T)

Ideal mixture model:

 $T > T_{\rm F} \qquad \pi V_{\rm m,4} = x R T$ $T < T_{\rm F} \qquad \pi V_{\rm m,4} = 0.4 R T_{\rm F}$ $\pi \text{ is osmotic pressure}$



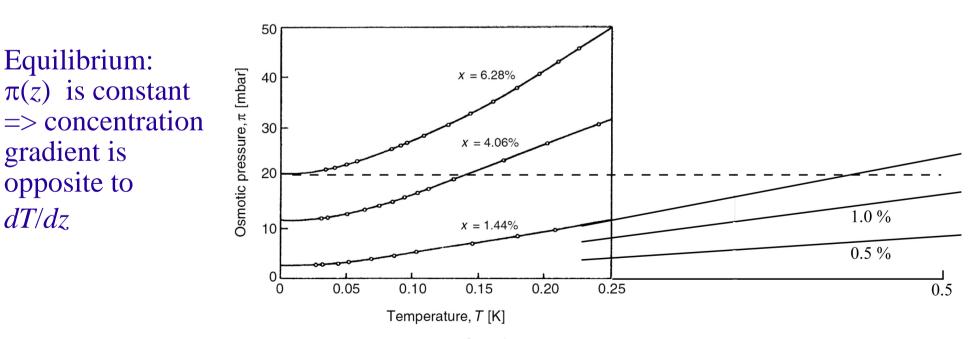
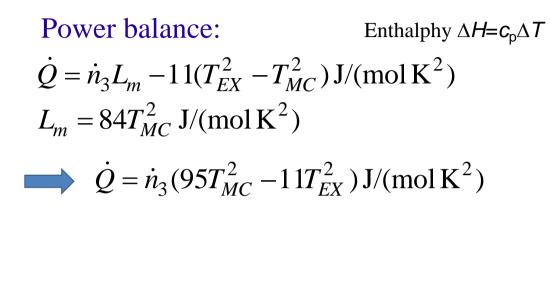
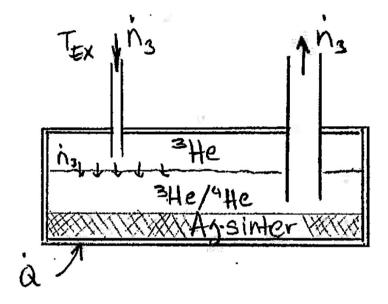


Fig. 7.7. Osmotic pressures of some dilute ${}^{3}\text{He}{-}^{4}\text{He}$ mixtures at a pressure of 0.26 bar (from [7.11] who used the data of [7.31])

Mixing chamber

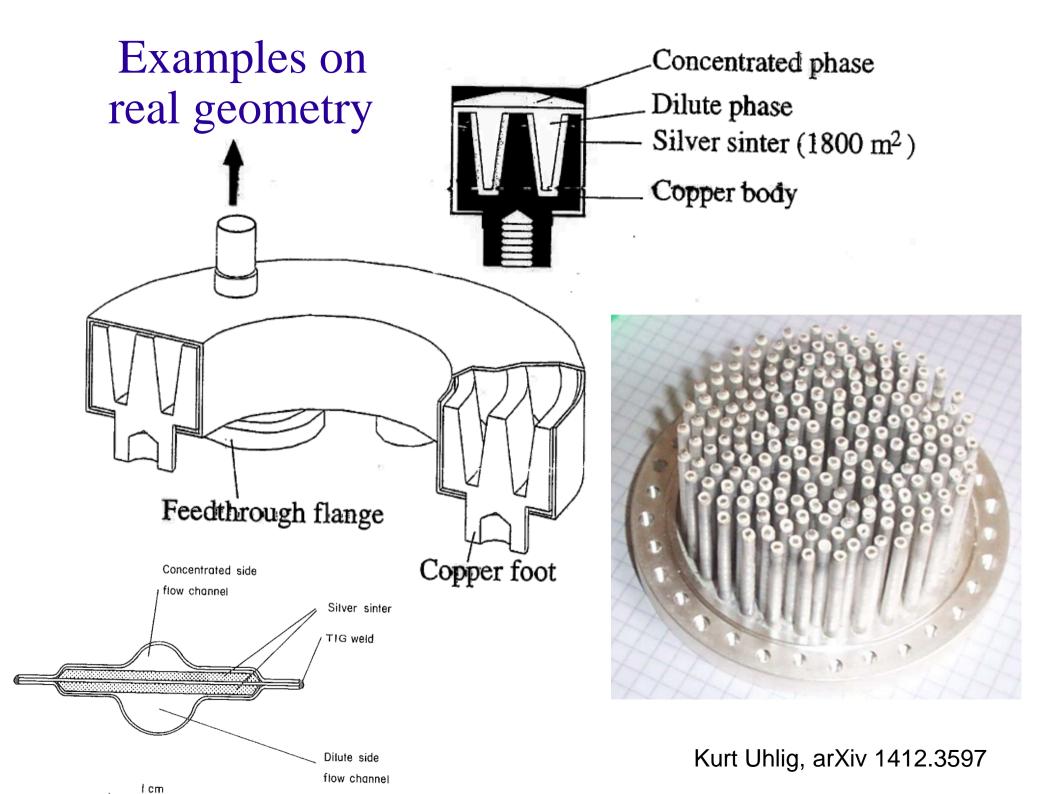


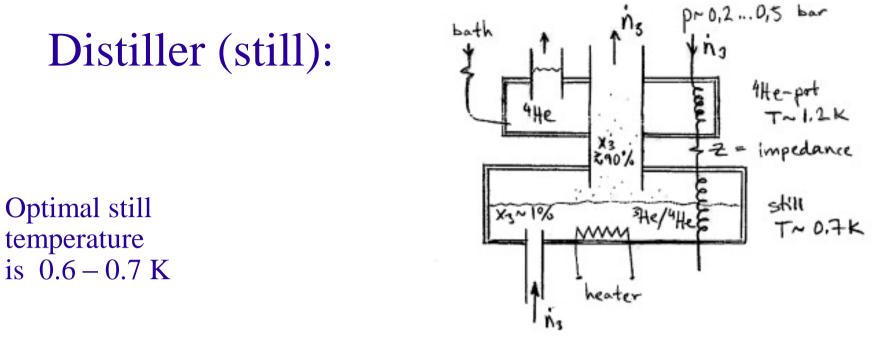


If $T_{\text{EX}} = T_{\text{MC}}$ (or operation in single cycle) $\dot{Q} = \dot{n}_3 84 T_{MC}^2 \text{ J/(mol K}^2)$

if $\dot{Q} \rightarrow 0 T_{MC} = T_{EX} / 2.9$

=> best possible heat exchangers to obtain minimum $T_{\rm EX}$





| <i>T</i> /K | <i>x</i> ₃ /% | $p_3 + p_4 / Pa$ | $p_3/(p_3 + p_4) /\%$ |
|-------------|--------------------------|------------------|-----------------------|
| 0.6 | 1.2 | 4.6 | 99 |
| 0.7 | 1.0 | 8.8 | 97 |

-³He coming from RT is condensed in ⁴He evaporator (pot), $T \sim 1.2$ K

– further cooling of returning ³He occurs in still, $T \sim 0.7$ K

- simple spiral tube heat exchangers

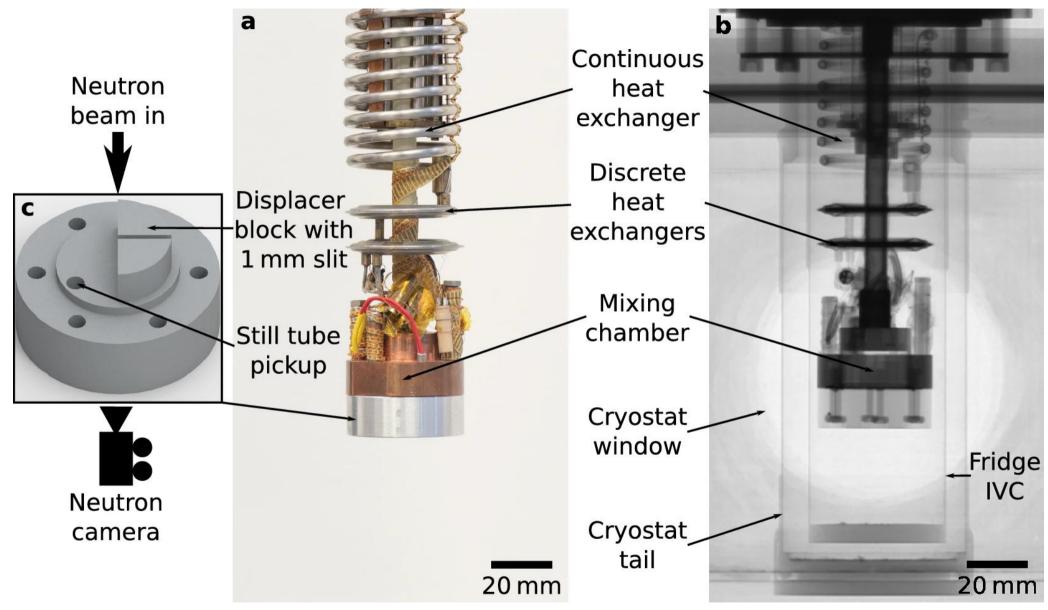
- impedance $Z \sim 10^{12} \text{ cm}^{-3}$

Still must be heated up:

- latent heat of ³He evaporation is $L_3 \sim 20$ J/mol
- incoming ³He flow gives a load $\Delta H_3 \sim 2$ J/mol
- additional heating $dQ/dt \sim 18 dn/dt$ J/mol (~ few mW typically) must be provided to keep up circulation
- still is a good thermal-anchor point (additional load up to $\sim 1 \text{ mW}$ is OK)

Important to prevent ⁴He from evaporating in the still. **Superfluid film may creep** up to a point where $T \sim 2$ K => higher vapor pressure

- may become as much as 10 ... 20 % unless attention is paid
- pumps are loaded by additional mass flow
- heat exchangers are loaded as **mixture has higher heat capacity than pure ³He**
- before MC 3 He separates from 4 He and produces heating (reverse to mixing)
- -⁴He may accumulate to heat exchangers if geometry is not right (gravity)
- down at the mixing chamber extra ⁴He does no harm, except that some ³He is away from taking part in the mixing process
- check ³He/⁴He ratio in circulation by a leak detector (mass spectrometer)



https://www.nature.com/articles/s41598-022-05025-0#Sec13

Lawson, C.R., Jones, A.T., Kockelmann, W. *et al.* Neutron imaging of an operational dilution refrigerator. *Sci Rep* **12**, 1130 (2022). https://doi.org/10.1038/s41598-022-05025-0