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Kapitza resistance at the H\textsubscript{\downarrow}/liquid He interface

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Résumé. — Dans l'hydrogène atomique polarisé, l'accès aux basses températures et fortes densités est bloqué par la résistance thermique de Kapitza. Cette résistance est calculée, ainsi que le temps caractéristique associé, pour un canal significatif : du gaz hydrogène aux modes de surface de l'hélium liquide.

Abstract. — In spin-polarized atomic hydrogen, the way to low T and high density is restricted by the Kapitza resistance. This is calculated, together with the corresponding characteristic time, for one channel : from H gas to liquid helium ripplons.

1. In the past three years a set of important results, both experimental and theoretical, have been gathered on the physics of spin-polarized atomic hydrogen [1] : recombination and relaxation rates, including the B\textsuperscript{-2} magnetic field dependence, adsorption energies on liquid 4He and 3He, sticking probabilities [2], etc. The relaxation bottleneck [3] in the population time-evolution has been observed [4], as well as the anisotropy in the surface dipolar relaxation rate [5]. As a result, higher and higher H\textsubscript{\downarrow} densities have been stabilized on long time scales, at lower temperatures — but these achievements seem to be levelling off at about 3 × 10\textsuperscript{17} at/cm\textsuperscript{3} and 200 mK, which is still way out of the Bose-condensation border (16 mK for 10\textsuperscript{18} cm\textsuperscript{-3}, 75 mK for 10\textsuperscript{19} cm\textsuperscript{-3}). A fundamental limitation to achieving lower temperatures, which we discuss in this letter, is the Kapitza thermal resistance \( R_K \) between H\textsubscript{\downarrow} gas and walls, which fastly increases at lower \( T \), thus building up a larger and larger temperature difference \( \Delta T \) between cold walls and a hot gas — due to the continued release of recombination heat (even though it is relaxationally bottlenecked) : about 5 eV per elementary reaction.

One is led, therefore, to think of compressing the gas — but this brings in a related problem. Even with a small \( \Delta T \), it takes a finite time, \( \tau_K \), to thermally equilibrate the system. \( \tau_K \) is given by the product of \( R_K \) and the gas heat capacity :

\[ \tau_K = R_K C_H. \] (1)

And for the compression to be roughly isothermal, it should proceed on a time scale \( \tau_{\text{comp}} \) long compared with \( \tau_K \). The compression time, which of course must remain short on the scale of relaxation time \( T_1 \), must therefore obey the double inequality

\[ \tau_K < \tau_{\text{comp}} < T_1. \] (2)
2. One can think of various channels for the heat transfer from gas to helium film ($^3$He/$^4$He liquid mixture; see Ref. 5). One is the gas-phonon channel (phonon in liquid helium). We write:

$$\dot{Q} = R_K^{-1} \Delta T.$$  

(3)

Here $\dot{Q}$ is the heat flux, and $\Delta T$ is assumed to be much smaller than $T$ (thereby allowing a linearization procedure).

$R_K^{-1}$ is proportional to $\bar{v}_{th} = \sqrt{\frac{2k_B T}{\pi m_H}}$ an average impact thermal velocity (1) and to a statistical integral proportional to $T^3$ (3-dimensional integration over phonon wave-vector). So, one expects: $R_K \sim \tau_K \sim T^{-7/2}$. This result was obtained long ago by Landau [6] (his « accommodation coefficient » is defined without $\bar{v}_{th}$ and is therefore proportional to $T^3$). It would yield rather long thermal times below 100 mK.

A more efficient channel, which we now investigate is the gas-to-ripplon transfer. The principle is the same as for phonons, but the integration is now two-dimensional and the dispersion law is different. Let $T_G$ and $T_L$ be the gas and liquid helium temperatures, $\Delta T = T_G - T_L < T$, $n_H$ the atomic density of the hydrogen gas. Then $\frac{1}{2} n_H \bar{v}_{th}$ is the mean number of atoms which hit the helium surface per unit time and unit area. The energy transfer rate $\dot{Q}$ between gas and liquid is given by

$$\dot{Q} = \frac{1}{2} n_H \bar{v}_{th} A \int_{q_{\min}}^{q_{\max}} A q \frac{dq}{2 \pi} h \omega_q \left[ (n_q + 1) \exp \left( -\frac{h \omega_q}{k_B T_G} \right) - n_q \right] K_q,$$

(4)

where $q$ and $\omega_q$ are the ripplon wave-vector and frequency, $n_q = \left[ \exp \frac{h \omega_q}{k_B T_L} - 1 \right]^{-1}$ is the ripplon occupation number, if assumed at the liquid He temperature $T_L$, and $K_q$ is the intrinsic probability for an H atom to be inelastically scattered off a ripplon mode $q$. $A$ is the total interfacial area. The term $\frac{1}{2} n_H \bar{v}_{th} K_q h \omega_q n_q$ thus represents the energy transfer rate from mode $q$ toward to gas. In the limit where the H atom is treated as a classical particle, this rate can be written

$$\frac{1}{2} n_H \bar{v}_{th} \left( \frac{1}{2} m_H \bar{u}^2 \right),$$

where $\bar{u}^2$ is the mean-squared normal velocity of the helium surface. It is then easy to obtain the expression of $K_q$ by considering the high-temperature case (where $n_q = k_B T/h\omega_q$):

$$K_q = \frac{2 m_H \bar{u}^2}{k_B T}.$$

On the other hand, in the same high-$T$ regime, the total energy of a ripplon mode is

$$A \frac{\sigma q^2}{\omega_q^2} \bar{u}^2 = A \frac{\rho}{q} \bar{u}^2 = k_B T,$$

where $\sigma$ and $\rho$ are the helium surface tension ($\sigma = 0.15$ erg cm$^{-2}$ for liquid $^3$He) and mass density leading to: $\omega_q^2 = \frac{\sigma q^3}{\rho}$, the capillary dispersion. Thus

$$K_q = \frac{2 m_H q}{\rho A}.$$

(1) See, for example, Landau and Lifshitz, Statistical Physics.
Injecting this classical expression into equations 4 and 3, we get in the low-temperature limit
\( x_{\text{Max}} = \frac{\hbar \omega_q}{k_B T} \text{Max} \gg 1 \)

\[ R_K^{-1} = \frac{\pi^3}{45} A \frac{k_B^3}{\hbar^2} \frac{m_H}{\sigma} n_H \bar{v}_{\text{th}} T^2. \]  

(6)

Note that the classical-trajectory approximation we made for H atoms, amounts to neglecting any horizontal momentum transfer. Put another way: to neglect the hydrogen de Broglie wavelength, \( \lambda_B \), as compared with that of the ripplon, \( \lambda_r \) — since the only efficient ripplons are those with \( \lambda_r \gg \lambda_B \). These two lengths turn out to be nearly equal at temperatures \( \sim 100 \text{ mK} \), and their ratio is slowly varying with \( T \). Therefore, although an approximate one, our calculation yields the right order of magnitude for \( R_K \).

For a perfect classical gas in volume \( V \), the heat capacity is \( C_H = V \frac{3}{2} k_B n_{He} \) and the characteristic thermal equilibration time, defined by equation 1, can be written

\[ \tau_K = 6.5 T^{-2} \left( \frac{2 V}{\bar{v}_{\text{th}} A} \right), \quad T \text{ in K}, \]

(7)

or, equivalently : \( \tau_K \approx 1.6 \times 10^{-3} T^{-5/2} \frac{V}{\bar{v}_{\text{th}} A} \), in seconds, if \( T \) in K and \( \frac{V}{A} \) in cm. So, compared with the gas-to-phonon channel, we gain one power in temperature, and \( \tau_K \) is reasonably short at moderately low \( T \), allowing the double inequality 2 to be easily fulfilled. For example, for a flat cell of width \( h = 2 V/A = 1 \text{ mm} \), \( \tau_K \approx 40 \text{ ms at } T = 80 \text{ mK} \).

3. Three remarks should be made at this point. First, from expression 6, the Kapitza resistance is lowered by a factor \( \frac{\sigma_4}{\sigma_3} = \frac{0.35}{0.15} \) (the ratio of free-surface tensions) for liquid \(^3\text{He} \), as compared with \(^4\text{He} \). Second, it is readily estimated from the capillary dispersion relation, that the penetration depth for the dominant ripplon \( (\hbar \omega_r \approx k_B T) \) will become comparable with the thickness of the helium film \( (\sim 300 \text{ Å}, \text{[1]}) \), only for temperatures below a few mK — the effect being, again, more favourable (slightly) for \(^3\text{He} \). Finally, the Fermi-liquid viscosity of \(^3\text{He} \) will ultimately damp off the ripplon modes, but again only for \( T \) lower than a few mK. Liquid \(^3\text{He} \)-%\(^4\text{He} \) mixtures [5] (i.e. \(^3\text{He} \) floating on top of the \( 6 \% \) solution) are to be favoured, therefore — the main reason, though, being that the \(^3\text{He} \) quasi-particles provide us with an efficient heat transfer channel from ripplons to bulk and walls, as we now discuss.

Were it is not for the quasi-particles (q.p.), the only channel on the liquid side would be the ripplon-to-phonon one. This can be shown to become very rapidly inefficient at low \( T : \text{as } T^7 \) (Ref. 7). The reason is that, due to momentum mismatch, two phonons are required to de-excite one ripplon mode (recall, for example, the analogous \( T_1^{-1} \sim T^7 \) argument for Raman spin-lattice relaxation in solids).

For the « gas » of q.p. the thermal resistance can be roughly estimated from equation 6 above, replacing \( m_H \) by \( m_3 \approx 6 m_{He} \), \( \bar{v}_{\text{th}} \) by \( v_F \) (same order of magnitude, at \( T \approx 50 \text{ mK} \), since \( T_F \approx 300 \text{ mK} \)), and — most important — \( n_H \) by \( n_{q.p.} \approx n_3 \frac{T}{T_F} \approx 10^{21} \text{ cm}^{-3} \). \( R_K \) is seen to be orders of magnitude lower for \(^3\text{He} \) quasi-particles. So, the total thermal resistance between \( \text{H-gas and bulk helium is essentially that given by equation 6. Similar arguments hold at the } \text{He/dilute solution interface (smaller number of q.p.'s, but smaller interfacial tension too). Therefore, the whole thermal transfer problem is largely governed by the results of section 2.} \)
There are other heat transfer channels which we have not discussed, however: for example via sticking to the liquid surface. Sticking coefficients $\alpha_{\text{st}}$ have been measured by NMR techniques [2]. At low coverage $n_\text{s}$, $\alpha_{\text{st}}$ was found to be $\approx 1.6 \times 10^{-2}$ for H against liquid $^3$He. Were this value independent of both $T$ and $n_\text{s}$, this channel would dominate at low $T$, with an incoming energy flux of order $(n_\text{H} v_\text{th} \varepsilon_\text{a}) \alpha_{\text{st}}$, where $\varepsilon_\text{a}$ is the adsorption energy. But the coverage-dependence of $\alpha_{\text{st}}$ is not an obvious problem; in a « site-adsorption » model, we expect $\alpha_{\text{st}}$ to be very small ($\sim 10^{-2} \exp -2013$ for a saturated layer. Although such a model is clearly not acceptable for the present problem, we think the question is open-especially with respect to volume Bose condensation which precisely requires completion of the adsorption hydrogen layer.

On the other hand, the H/q.p. transfer by direct H/$^3$He impact without the mediation of the ripplon bath, seems to be a rather poor channel too: due to the Van der Waals interaction, the incoming H atom interacts with ripplons before hitting the $^3$He quasi-particle.

Using equation 6 and measured values of the relaxation rate $G$, the Kapitza temperature difference $\Delta T$ can be estimated from equation 3 and the expression $Q_{\text{rec}} = V(5 \text{ eV}) G n_\text{H}^2$ for the steady-state recombination heat, at $n_\text{H} = 10^{17} \text{ at./cm}^3$ and $T = 80 \text{ mK}$ for example, we get: $\Delta T \approx 80 \text{ mK}$! this of course is an overestimate since expression 6 for $R_K$ assumes $\Delta T \ll T$, and it is expected that $R_K$ is reduced in the non-linear regime. Still, this figure shows that the Kapitza problem is a serious one at low temperature (more so for $\Delta T$ than for $\tau_K$, as we have seen). A very recent experiment, performed in Amsterdam [8], gives an upper bound of 15 mK upon $\Delta T$, at $T = 310 \text{ mK}$, $B = 5.7 \text{ tesla}$ and $n_\text{H} \sim 3 \times 10^{15} \text{ at./cm}^3$.

References