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Horst Meyer and Quantum Evaporation

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Abstract With their 1963 article in *Cryogenics* Horst Meyer and his collaborators triggered intense research activity on the evaporation of superfluid helium. Discussing this subject with him in 1975 was enlightening. Fifty years later, the analogy between the photoelectric effect and the evaporation of superfluid helium in the low temperature limit is not yet clear, although remarkable progress has been made in its observation and its understanding. This special issue of the Journal of Low Temperature Physics is an opportunity to recall the history of quantum evaporation, and to express my gratitude to Horst Meyer. It describes quickly most of the experimental and theoretical works which have been published on quantum evaporation during the last 50 years, but it is not a comprehensive review of this fascinating subject.

Keywords Superfluidity · Helium · Evaporation

1 Meeting Horst Meyer

The first time I met Horst Meyer must have been in 1975. I was a graduate student at ENS in Paris and Horst was visiting my supervisor, Albert Libchaber.

Twelve years earlier, together with Meyer, Halliday and Kellers, Horst had published an article [1] with the title “Beams of helium atoms at temperatures below 0.5 K.” When I described my observations on similar beams, we started a long discussion during which I discovered that, in 1969, A. Widom [2] and Anderson [3] had made remarkable

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predictions about the evaporation of He atoms in the low temperature limit. At that time they did not describe it as a close analog of Einstein's photoelectric effect [4], probably because they considered evaporation from a liquid bath at thermal equilibrium, not the extreme case where ballistic rotons¹ could evaporate ballistic atoms one by one. But, as noticed by Maris [7], it is really what the evaporation of superfluid ⁴He should be in the low-temperature quantum limit where heat is decomposed into well-defined elementary excitations, whose energy may exceed the binding energy of atoms in the liquid, that is the latent heat per atom. This situation is a consequence of the large value of the de Boer parameter, which is the ratio of the kinetic energy due to quantum fluctuations to the strength of the attractive potential between atoms. In reality, Widom and Anderson had predicted that the quantization of heat shows up in the evaporation of superfluid helium as the quantization of light shows up in the photoelectric effect.

In the photoelectric effect, photons incident on a metal surface eject electrons in a one-to-one elastic process so that their kinetic energy is the difference $E_q = E_{ph} - E_b$ between the photon energy E_{ph} and the binding energy E_b of the electron in the metal. Widom and Anderson proposed that single rotons incident on the liquid–gas interface could evaporate single atoms in a similar one-to-one elastic process so that the evaporated atoms should have a minimum kinetic energy $E_q = 1.5$ K since the roton minimum energy Δ equals 8.65 K while the binding energy of atoms E_b equals 7.15 K. A few months later, Hyman, Scully, and Widom published a more elaborate article [8] where they treated evaporation as a quasiparticle tunneling process and they gave some arguments for rotons to be transmitted as atoms through the liquid–gas interface in such a one-to-one elastic process. Further theoretical developments were soon published. Griffin [9] claimed that a study of evaporation “*should give information about the condensate fraction*” of superfluid He, while Kaplan and Glasser [10] argued that “*the roton branch contributes most strongly [to the evaporation] as suggested by Anderson*”. On the contrary, Cole [11] wrote that “*the evaporation spectrum does not manifest the quasiparticle density of state*”.

I was not aware of these somewhat controversial ideas. It is really by accident and partly thanks to my discussion with Horst that I realized the interest of my results, the first experimental evidence of the nonclassical character of evaporation from superfluid He.

2 Evaporation: Early Experiments at ENS and in Horst Meyer's Group

In the early 70's, I was hesitating on various subjects for my Ph.D. My memory of that time is a little fuzzy but here is what I remember. Libchaber's group had invited Professor Yasuji Sawada from Tohoku University. He liked being called “Koji” in the lab and he initiated me to elementary heat pulse techniques to study the propagation of heat at low temperature. At the same time in Libchaber's group, my neighbors Hulin, Perrin, and Laroche were searching for the Josephson effect in superfluid helium. When

¹ It has been predicted by L.D. Landau [5] and demonstrated by Henshaw and Woods [6] followed by many other neutron scattering experiments that, in superfluid helium at low temperature and pressure, the phonon excitation spectrum has a peculiar branch called the “roton branch” with an energy gap $\Delta = 8.65$ K

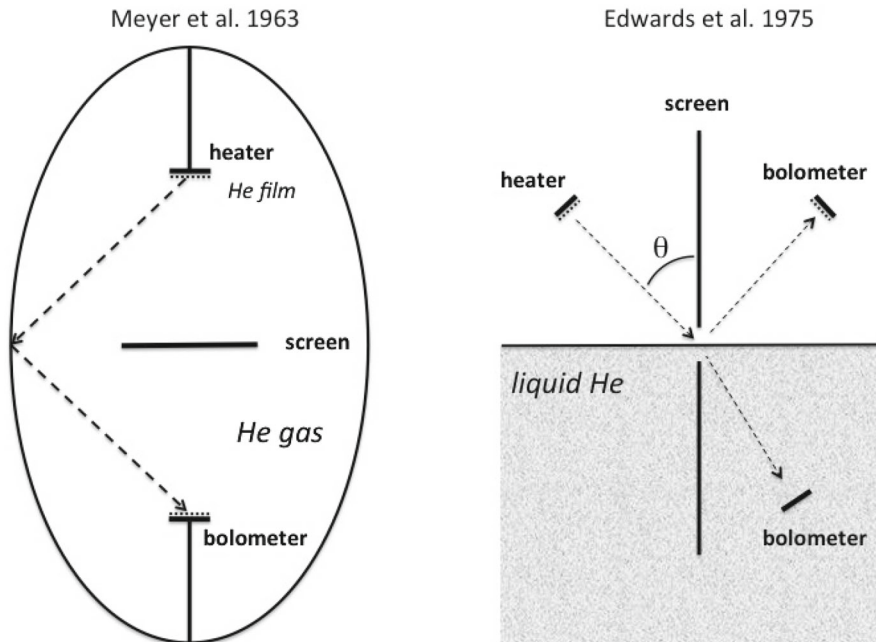


Fig. 1 The experimental setups used in 1963 by Meyer [1], and in 1975 by Edwards [15]

trying to detect vortices produced by a superfluid flow through a small orifice, we built a little setup to study the propagation of heat pulses through the flow, and we realized that, during the filling of the cell, heat pulses could propagate not only in superfluid liquid helium but also in the vapor. I started studying this phenomenon and found that heat pulses could even be transmitted through the liquid–gas interface. When cooling down to 0.45 K in a ^3He fridge, I observed the splitting of a heat pulse into a phonon pulse and a roton one, which propagated at different velocities so that they reached the detector at different times. At that temperature, the phonon–roton collision time was long enough for the heat pulses to split into these two separate pulses [12, 13]. With some bulk liquid between the heater and the detector, a large signal was visible which I associated to the evaporation of atoms by rotons. Evaporation by phonons looked much more difficult to detect [14]. This is basically what I described to Horst Meyer in 1975.

Horst Meyer was one of the coauthors of the clever experiment they published in 1963 [1]. Together with Meyer, Halliday, and Kellers, they had built a metallic ellipsoid with a heater and a bolometer at its focal points (see Fig. 1). With one necessary reflection on the walls all the paths from heater to detector had the same length and it was a clever method for precise measurements of the speed of second sound. In a way somehow similar to my accidental discovery, Meyer observed that heat pulse propagation occurred not only when the ellipsoid was full of superfluid helium but also with helium gas at the vapor pressure so that both the heater and the detector were covered by a thin liquid film only. In the latter case, the heat propagation had to result from the propagation of a beam of He atoms, which originated in the

partial evaporation of the film covering the heater. By cooling the cell below 0.5 K, they observed a transition in the propagation of the beam in the vapor from a hydrodynamic regime to a ballistic one. In the hydrodynamic regime, the propagation speed was that of sound in the vapor, proportional to the square root of T , and independent of the pulse amplitude. On the contrary, in the ballistic regime, the speed was independent of T but increasing with the pulse amplitude. Meyer observed that the repetition rate of their heat pulses had to be low enough—typically less than 0.5 Hz—for the liquid film on the heater to recover after each partial evaporation. From the time of arrival of the maximum of their signal, they obtained evidence that the typical kinetic energy of the evaporated atoms was well related to the heating of the liquid film covering the heater. However, the velocity distribution was sharper than expected from a Maxwell–Boltzmann distribution, and this suggested that the reflection of atoms on the liquid film covering the walls of the ellipsoid was not purely specular. The article by Meyer triggered research on the microscopic processes of evaporation/condensation at the interface between a superfluid and its vapor phase.

Strangely enough, Widom and Anderson had been attracted to this subject by an experiment which was later found not reproducible. In 1966, that is 3 years after Meyer’s experiment, Johnston and King [16] had obtained astonishing results in an experiment at MIT, which had some similarities but also some differences with the one by Meyer [1]. Johnston and King measured the velocity distribution of a beam of atoms evaporating from a small heater covered by a superfluid film by measuring flight times through a long (98 cm) narrow (3 mm) tube thanks to a rotating chopper and a detector at room temperature. The source temperature (0.6 K) was obtained from vapor pressure measurements, and they found a beam temperature at 1.6 K, that is 1 K hotter than the source. This was a quite astonishing result, which they tried to attribute “*to the well-known quantum properties of superfluid He.*” They did not mention the existence of rotons, as Widom and Anderson did in 1969, but the formation of eight-atom microcrystallites predicted by Toda [17]. Contrary to Meyer who imposed reflexions of the atom beam on cold walls covered by liquid films, Johnston and King assumed negligible effects from possible collisions inside the beam or with warm walls along their long narrow tube.

The article by Meyer was published in *Cryogenics* and did not catch the attention of theorists contrary to the astonishing results of Johnston and King published in *Phys. Rev. Letters*. My motivation in recalling all these details is that it illustrates how physics progresses, sometimes. Indeed, J.G. King published a short abstract [18] 3 years later in the *Bulletin of the American Physical Society*, where he explained basically that his previous results were not reproducible, probably because collisions with walls were modifying the beam temperature, but, in the mean time, the question had been asked: should the peculiar existence of rotons lead to anomalies in the velocity distribution of atoms evaporated from superfluid He? It is not that surprising to find artifacts in experiments. More surprising is to realize that, sometimes, wrong experiments trigger theoretical predictions that may be correct.²

² In this conference abstract, King et al. consider their new results as “*contradicting theories predicting significant warming of the evaporating atoms due to the effects of rotons.*”

In 1973, Andres, Dynes, and Narayanamurti published a new series of measurements of “the velocity spectrum of atoms evaporating from a liquid He surface” [13]. For this, they applied a heat pulse to a small heater again covered by a thin He film, as Meyer had done. They detected the propagation of pulses of He atoms reaching a bolometer 2.35 mm away. The atoms were supposed to condense in the film that was covering this bolometer. From the signal shape as a function of arrival time, they obtained a velocity distribution. At low enough temperature for the propagation of atoms to be ballistic in the very low pressure vapor, they found good agreement with a Maxwell–Boltzman distribution at a temperature larger than the ambient temperature but no anomaly that could be attributed to the existence of rotons, no singularity around a kinetic energy of 1.5 K. There was no visible difference with a classical source of atoms at a variable temperature depending on the heat pulse magnitude.

3 More Experiments in the Seventies

Shortly after my Ph.D. [19], I summarized my results in an article on the evaporation of helium, which I published with several collaborators in 1978 [20]. I had first demonstrated that, by working at low enough temperature with heat pulses propagating in *bulk* liquid helium before reaching the free liquid surface, it was possible to measure evaporation by ballistic rotons. In the case of a thin liquid film, the heat pulse generated excitations which thermalized rapidly with each other. On the contrary, in the case of a bulk liquid at sufficiently low temperature, phonons and rotons are sufficiently dilute to propagate ballistically so that one could study separately how phonons and rotons evaporate atoms. Furthermore, this experiment gave direct evidence that Landau’s rotons [5] can be free quasiparticles. Then, we had measured the velocity of the evaporated atoms. To do that we had measured the time of flight on a 6.85 mm path with a variable height of the liquid–vapor interface in between (see Fig. 2). From this, we found evidence for a minimum kinetic energy of 1.5 K for the atoms evaporated by rotons, as had been predicted by Widom [2, 8] and by Anderson [3]. This was a quite encouraging preliminary evidence for quantum evaporation. But we were not able to fit our data with a precise calculation of the time-dependent signal which would need to include two unknown quantities, namely a spectrum for the emitted rotons and a momentum-dependent probability of evaporation. More precise work was obviously necessary, for which I had proposed a collaboration to Adrian Wyatt who was moving to Exeter in 1977. During my short postdoc stay in his laboratory, I started building an experiment with a rotating emitter and a rotating detector. Our main goal was to look at the angle of refraction which we predicted to be anomalous if the elastic one-to-one transmission of rotons into atoms was confirmed. Of particular interest was the case of so-called “ R^- ” rotons whose momentum is opposite to their group velocity. Indeed, since the conserved quantity should be the momentum k_x parallel to the surface, an R^- roton propagating to the right should evaporate an atom in the left direction (see Fig. 3). I was not able to achieve these measurements in a few months, but Wyborn, Tucker, and Wyatt did it in a beautiful series of experiments from 1990 to 1999 [21, 22] (see below).

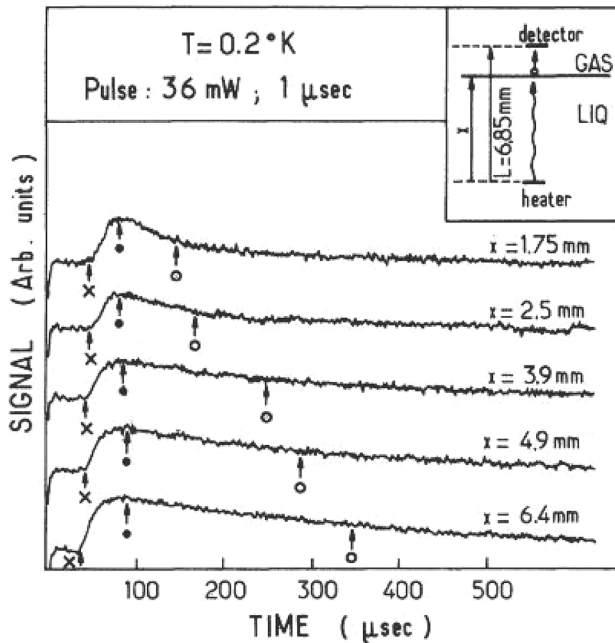


Fig. 2 Some of the results by Balibar et al. [20]. Crosses indicate the starting time of the pulse arriving on the detector, which was a bolometer covered by a liquid film. Black circles indicate the signal maximum and open circles to its half height. A quick analysis of these times as a function of liquid height x indicated that the kinetic energy of evaporated atoms had a minimum kinetic energy $\Delta - E_b = 1.5$ K

At the same time, David Edwards and his group [15,23,24] used a method inspired by Meyer's experiment to measure the reflectivity of He atoms at the liquid surface. I was not aware of the work by David's group, but their studies had an obvious link to ours. Indeed, consider a system at equilibrium. It should be invariant by time reversal if one neglects inelastic processes, and the detailed balance principle should apply. As a consequence, the probability that a roton with momentum $\hbar \vec{k}$ and energy $E(k) = \hbar \omega(k)$ emits an atom with a momentum $\hbar \vec{q}$ and energy $\hbar^2 q^2 / (2m)$ should be equal to the probability that an atom hitting the surface with a momentum $\hbar \vec{q}$ condenses as a roton with momentum $\hbar \vec{k}$. (once more if one neglects inelastic processes). As a consequence, the sum of certain probabilities of evaporation or condensation should be equal to one (unitarity). One expected that, if the probability of evaporation was large, the probability of condensation should also be large and that of reflexion should be small. But, as we shall see, there are many channels, all of them may depend on incidence angle, momentum and energy, and inelastic processes may well exist, so that the whole issue is far from simple.

P.W. Anderson had written that the problem could be "treated in terms either of particles impinging on the surface from the vapor, or of elementary excitations coming from the liquid side, since there is a detailed balance relationship between these two points of view" [2]. At equilibrium, the evaporated flux of atoms with momentum $\hbar \vec{q}$ and angle θ has to be balanced by the flux of atoms condensing with the same momentum

and direction [15]. But, if the existence of rotons showed up in the velocity distribution of evaporated atoms out of equilibrium, the reverse phenomenon should also show up, namely the emission of single rotons by atoms condensing from the vapor.

Anyhow, Edwards used a heater in the vapor, which was covered by a liquid film and consequently able to emit a pulsed beam of atoms as Horst Meyer did before, but rotating, and collimated (see Fig. 1). Their bolometer was also rotating and consequently able to be in the vapor or in the liquid. Moreover, their setup was such that they could align the heater and the bolometer in the vapor in order to calibrate the atom beam as a function of the pulse energy. They could thus measure the probabilities of either reflexion or condensation. Let us call P_{ij} the probability that an atom $i = a$, a phonon $i = p$, an ordinary roton $i = +$, or an anomalous roton $i = -$ is either reflected or transmitted into $j = a, p, +$, or $-$, as introduced by Dalfovo et al. [25]. Edwards found that a He atom striking a liquid He surface had a probability close to one to condense into it. The probability P_{aa} for specular reflection of atoms was smaller than 5×10^{-2} , and decreasing as a function of the perpendicular component $q_z = q \cos(\theta)$ of the atom momentum only. However, they could not investigate the reflexion of atoms at normal incidence. By varying the angle of incidence, they found that the reflexion probability was independent of the parallel momentum $q_x = q \sin(\theta)$. The probability for inelastic scattering of atoms by the surface was found to be less than 2×10^{-3} , so that the condensation of atoms creating excitations in the liquid had to be close to 1. Moreover, there was no evidence for any singularity in the reflectivity near $0.5 \text{ \AA}^{-1}$, the momentum corresponding to atoms with a 1.5 K kinetic energy, the threshold for roton emission.

A year later, Edwards [24] detected the emission of ballistic rotons by an atom beam hitting the free surface. They analyzed the angular distribution and flight time

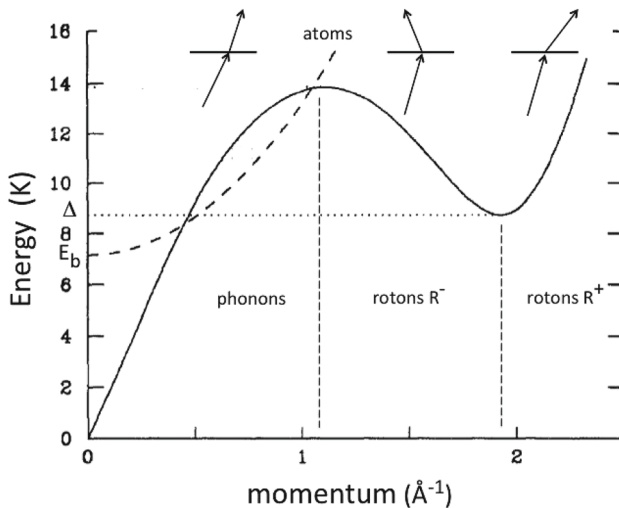


Fig. 3 The phonon–roton dispersion relation in superfluid helium (*solid line*), the kinetic energy of free He atoms (*broken line*), and typical angles of evaporation by single phonons, normal rotons R^+ , and anomalous rotons R^- . $\Delta = 8.65 \text{ K}$ is the minimum energy of rotons and $E_b = 7.15 \text{ K}$ is the binding energy of atoms to the liquid phase, which is equal to the latent heat per atom in the low temperature limit

of these rotons, and concluded that some of the atom energy had to be lost from the emission of surface excitations (ripples). However, several parameters like the amplitude of diffuse scattering from walls and the dependence of the detector efficiency on the momentum and incidence angle of the rotons made a fully quantitative analysis difficult. For example, they could not determine the precise energy loss in the transmission of atoms into rotons. They considered their results as consistent with those of Balibar [14,19] at least in the sense that their observation of roton emission by atom absorption confirms the existence of the reverse process. However, in their calculations of condensation probabilities, Echenique and Pendry [26], Edwards and Fatouros [27], Goodman and Garcia [28], Usagawa [29], Swanson and Edwards [30], and Campbell [31] included the generation of ripples, which means that, according to them, condensation and evaporation are *not* purely elastic one-to-one processes.

4 Later Experiments and Theoretical Developments

A.F.G. Wyatt and his group made a long series of new experiments, some of which are described in a review article published in 1992 [32]. In 1983, they discovered the evaporation by phonons [33]. A year later, they showed that the parallel momentum is conserved in the evaporation by both ballistic phonons and ballistic rotons [34]. In 1990, Brown and Wyatt repeated some of the condensation experiments by Edwards and found no evidence for inelastic effects [35,36]. However, they published a detailed study of “quantum condensation” in 2003, where they explained the apparent contradiction with Edwards [37]. Their careful study of the condensation of atoms into phonons, rotons, or surface ripples demonstrated that incident atoms emit *either* phonons, rotons, *or* ripples, depending on energy, momentum, and incidence angle. According to them, the condensation processes are one to one as previously predicted, there is no energy loss in the condensation into phonons or rotons but the emission probability of surface ripples is comparable to that of bulk excitations, and the production of ripples by some atoms create a surface roughness which enlarges the angular distribution of the bulk excitations emitted by other atoms.

In the years 1990–1999, Wyatt and his group discovered [21] and analyzed [22,38] the evaporation by anomalous rotons R^- , which once more occurs with conservation of the parallel component of the momentum so that the refraction angle is opposite to what is usually observed with light (see again Fig. 3). Among their most recent results are the measurements of the evaporation probability for R^+ and R^- rotons. This is particularly interesting since the calculation of this probability has been a challenge for several groups of theorists [7,25,31,39–43].

The calculation of transmission and reflexion probabilities is obviously difficult and the purpose of this article is not to make a comprehensive review of all the works that have been published, only to cite some of them. Both Widom and Anderson predicted a sharp enhancement in the spectrum of evaporated atoms at the roton threshold, which was a consequence of the divergence of the density of states at the roton minimum. In 1976, Caroli [39,40] explained that it was incorrect to treat the evaporation problem

with an effective transfer Hamiltonian coupling the liquid to the gas phase. They found a square root singularity at the roton threshold, whose exact shape could be a cusp or a rounded wedge.

In his 1992 article [7], Humphrey Maris neglected inelastic processes and considered that the liquid–gas interface could be wide enough for excitations coming from the liquid to be adiabatically transformed into atoms emerging in the gas. A simple interpolation between the phonon–roton dispersion relation and the quadratic one for atoms in the gas led him to a surprising result: the transmission probability at normal incidence could be zero in a very large momentum range, from 1.13 to $2.31 \text{ \AA}^{-1}$, where R^+ rotons should be anomalously reflected into R^- rotons and vice versa. Since the roton momentum is $1.92 \text{ \AA}^{-1}$, well inside the range of zero transmission, and since experiments had found a definite transmission, Maris suggested that tunneling effects could restore a nonzero transmission probability in this range. He also suggested that excitations could be attenuated as they pass through the interface.

With the opposite assumption of a sharp interface, Tanatarov [43] recently found that the probability of rotons to evaporate atoms vanishes at the roton minimum as the square root of the departure from the roton minimum energy Δ , consequently in a very narrow region. In their two articles, Dalfovo [25,41] had used a density functional theory to describe the dynamics of excitations in an interface of definite thickness. They had already found that the transmission probabilities $P_{+,a}$ and $P_{-,a}$ tend to zero when the roton energy E_r tends to its minimum [$\Delta = 8.65 \text{ K}$; $k = 1.92 \text{ \AA}^{-1}$], but in a much larger momentum domain than Tanatarov. They had also found that the ratio $P_{+,a}/P_{-,a}$ is always larger than 1 but tends to 1 in the $E_r = \Delta$ limit.

Unfortunately, the measurements by Forbes and Wyatt [44] followed by Tucker and Wyatt [45] are not sufficiently precise in the region of the roton minimum to discriminate between all calculations and to fully clarify the situation. To investigate the vicinity of the roton minimum Δ , experiments are difficult because the roton group velocity vanishes so that the flight time diverges. Tucker's data for R^+ rotons show a decrease of the evaporation probability as E_r tends to Δ but they do not extend below 9.5 K . For R^- rotons, they found a much smaller probability than for R^+ but it *increases* below 9.5 K . In summary, there is a qualitative agreement about R^+ rotons being more efficient than R^- rotons to evaporate atoms, but no quantitative agreement on the values of evaporation probabilities, no clear experimental evidence that the probabilities $P_{\pm a}$ tend to zero at the roton minimum.

Wyatt found no measurable energy loss in the evaporation by rotons [32–34]. However, Edwards [15,46] found evidence of inelastic processes due to the emission of ripplons during the approach of the surface by incoming atoms, as found also in a calculation by Echenique and Pendry [26]. In the end, there seems to be no consensus either on unitarity, that is on the exact relation between evaporation, reflexion, and condensation probabilities. Most calculations neglect inelastic processes and consider only one-to-one elastic processes.

5 Conclusion

When coming back to the issue of quantum evaporation 40 years after my PhD, I had the somewhat painful feeling that a full understanding of this effect remains to be achieved. A number of remarkable experiments have brought evidence for the existence of several interesting properties of evaporation, condensation, and atom or excitation reflexion at the liquid–gas interface in the low temperature limit where heat is quantized into long lifetime elementary excitations. The theory has obviously progressed a lot using various approaches based on assumptions which may appear contradictory to one another. Even the weight of possible inelastic processes is not really known so that the analogy with the photoelectric remains questionable. One would like to see experiments discriminate between all the theoretical results but, as far as I can see, it is unfortunately not really possible yet.

In 1963, Horst Meyer contributed launching a research subject, the evaporation of superfluid helium. More than 50 years later, it is a little surprising that it “deserves further studies,” as one often says. I am very grateful to him not only for having oriented my research on this evaporation a long time ago, but also for the deep friendship we kept strengthening all the time since then. By the way, the list of references at the end of the present article shows that the Journal of Low Temperature Physics has published a large fraction of the work devoted to quantum evaporation. I am convinced that the quality of this journal is largely due to all the time Horst generously gave to its management. For the numerous discussions we had together on quantum evaporation, I am also grateful to Christiane Caroli, Bernard Roulet, Daniel Saint-James, Albert Libchaber, Philippe Nozières, David Edwards, Humphrey Maris, and Franco Dalfovo.

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