Summary

This thesis deals with the transfer of heat in pure fluids in the neighbourhood of the liquid-vapour critical point in the absence of gravity. In particular, an experiment is described which took place in the low gravity environment of a spaceshuttle in orbit.

The study of critical phenomena in fluids is troubled seriously by the dynamics of pure fluids near their liquid-vapour critical point. In the earth's gravitational field the divergence of several quantities leads to a strong stratification of the density and the advanced appearance of convective motion. Together with the phenomenon of critical opalescence, the precise measurement of thermal quantities close to the critical point becomes unusually difficult and good agreement between critical constants measured in different ways is hard to achieve. Consequently, the study is facilitated substantially by turning to a low gravity environment. In the described space experiment a measuring technique is used, similar to that introduced by Becker and Grigull, in which the propagation of a plane thermal disturbance into an otherwise homogeneous sample is observed by interferometry and by temperature measurements. SF_6 was chosen as the fluid under investigation mainly because of its convenient critical values for the pressure and the temperature.

As Onuki already pointed out, on the thermal response of a critical fluid to heating one needs to account for what has come to be known as the piston effect. Fundamentally, this piston effect is not a mechanism of true heat transport; rather, it is a temperature change associated with isentropic compression of the fluid. In chapter 2 the respons of the temperature-density field in the absence of gravity to the applied way of heating is considered analytically. It confirms the profound influence of the piston effect. In particular, it confirms the conclusion by Ferrell and Hao that, because of the piston effect, the thermal properties of all materials with which the contained critical fluid comes in contact with, play an important role in the thermal behaviour of the critical fluid. The as found quantitative description of the piston effect enables to predict in any geometry the piston effect, with which the contribution of the piston effect can be eliminated from the measurements of temperature changes before they are interpreted in terms of a simple conduction equation. This is necessary because, as in this investigation, from these kind of measurements often the thermal diffusivity is determined. An important additional result of the analytic exercise in this chapter is the proposal for a new, intrinsically accurate method to determine the isochoric specific heat, which is difficult to measure for fluids in general but even more so close to the critical point. This method is based on simultaneous measurements of temperature and density changes.

The space experiment was performed using a custom-made test cell mounted in ESA's 'Critical Point Facility' which was part of 'Spacelab' in NASA's 'Spaceshuttle Columbia' during the IML-2 mission. The experimental equipment is described in chapter 3. The complete set up offers, besides the described experiment, the possibility for light scattering experiments. Unfortunately, the functionality of this part appeared inadequate for a thorough investigation. As a consequence the light scattering results are not discussed in this thesis. In a project like this one, because of its uniqueness, partial failure is quite defendable and this project shows once more the limitations of experimenting in space. Therefore, this thesis may be regarded, besides a scientific dissertation, as a report on a space-related project.

Density changes in the fluid were monitored by interferometry because of its essentially non-invasive nature. In the applied Twyman-Green interferometry the recombination of two parts of a beam of laser light results in an interferogram. The density changes can be followed because they

affect the optical path of the part passing through the fluid, thereby affecting the interference pattern in the interferogram. A difficulty arises because light rays are deviated in an inhomogeneous density field resulting in a non-trivial relation between a light ray and his optical path. To a large extent, this effect may be dealt with conveniently by proper focusing of the optical system. Unfortunately, the CPF optical system, also utilized by other experiments, was not in an optimum configuration for our experiment and the development of an alternative procedure to determine the thermal diffusivity was required. This procedure returns knowledge of the density distribution of principally one dimension less than the intended procedure which led to less accurate results than hoped for. The conversion of an interferogram to a density distribution and the alternative procedure are discussed in chapter 4.

The applied interferometry is based upon the relation between the density and the refractive index of the fluid. It is generally assumed that this relation is most accurately described by the Lorentz-Lorenz relation. However, this relation is only approximate and it was far from evident that this approximation is sufficiently accurate for our purpose. For this reason we have conducted precision measurements in our laboratory of the refractive index and the density of SF_6 in a region around the critical density, presented in chapter 5, utilizing a set up which resembles CPF. Our results show that, for SF_6, the Lorentz-Lorenz relation is not always adequate when, as in our case, the slope of this relation plays an important role. Of the determined critical values for the density and the refractive index, we find that the latter differs significantly from the up to now assumed value.

The results of the space experiment, discussed in chapter 6, confirm our understanding of the piston effect both qualitatively and quantitatively. Furthermore, we find that the thermal properties of all materials with which the contained critical fluid comes in contact with can be described by a single set of phenomenological parameters, with which even in a container of complex geometry the piston effect can be separated from true heat transport effects. This allowed the determination of the thermal diffusivity as close as 5 mK to the critical point. The resullts closer to the critical point than 10 mK differ significantly from the only other measurements this close by Wilkinson et al., which was also a space experiment. A possible cause lies in the difference in the process which is observed by the two methods; we have deduced values from the early, rapid and local response to a thermal disturbance whereas Wilkinson et al. inferred values from slow and non-local behaviour in the late stage of thermal equilibration.

Finally, the space experiment has demonstrated that the newly developed method to determine the isochoric specific heat provides a necessary, complementary method for an accurate determination in the critical region. Although the space experiment was not optimized for this method, the results are in good agreement with both an existing equation of state and results from the microgravity experiment by Straub et al., but differ from earth-based measurements. With improvements, suggested in this thesis, in future experiments of this kind, this method offers an excellent tool for assessing the quality of existing equations of state in the near critical region.