Interfacial melting of ice in contact with SiO_2

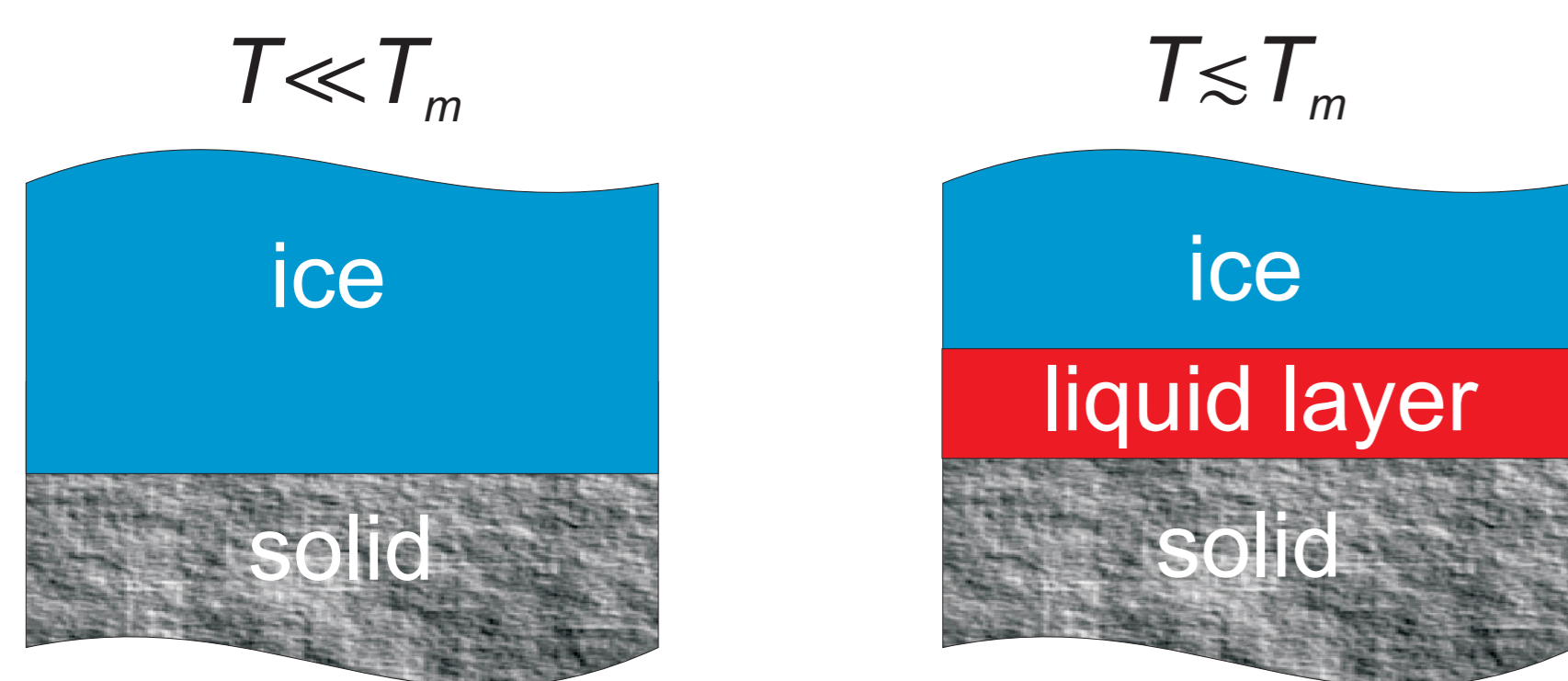S. Engemann*, H. Reichert*, H. Dosch*, J. Bilgram[‡], V. Honkimäki[§], A. Snigirev[§]

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Introduction

$T \ll T_m$
$T \lesssim T_m$

At the interface between ice and another solid, a premelting layer can form at temperatures below the bulk melting point. The liquid layer may serve as a nucleation site for the bulk melting. The phenomenon has also consequences for many important processes in nature and technology, as the motion of glaciers, permafrost and ice films on airplane wings. We have studied well defined ice-SiO_2 interfaces which might serve as a model for ice-mineral interfaces as they occur in nature.

## Method: High-energy x-ray reflectivity
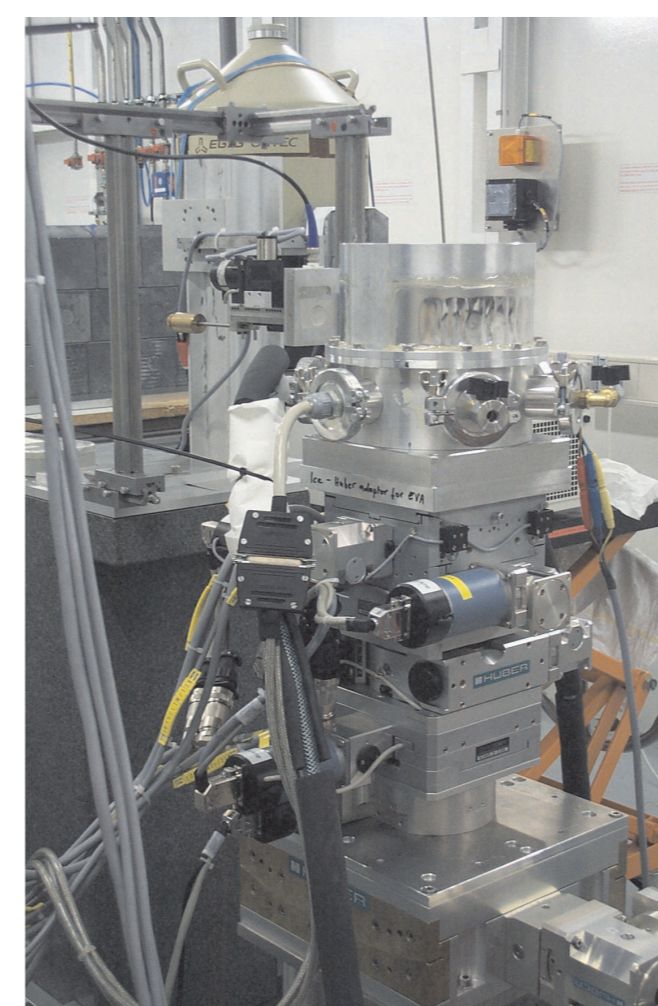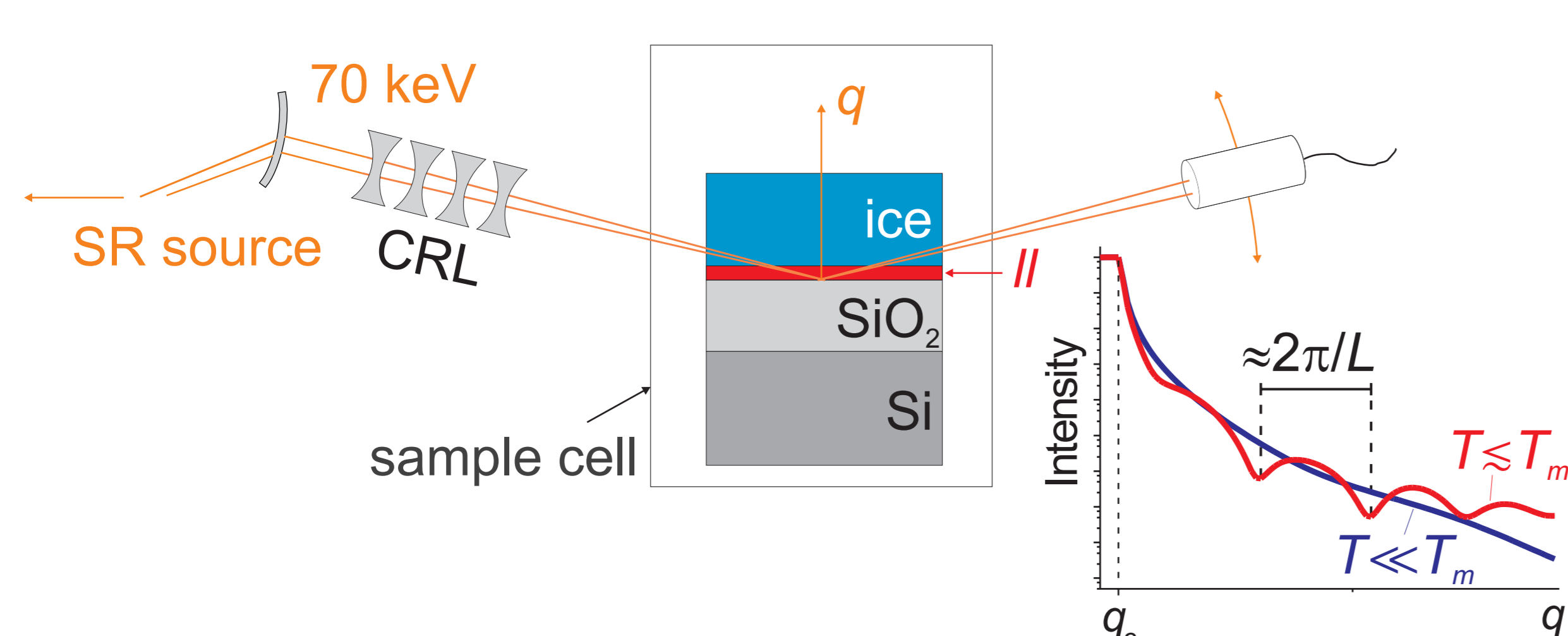
For studying these deeply buried interfaces, a novel X-ray transmission-reflection scheme has been developed. It is based on the use of high-energy (>70 keV) X-rays from a Synchrotron Radiation (SR) source. Compound refractive lenses (CRL) allow to produce micro-beams. The angular dependence of the reflected intensity allows to deduce the density profile across the interface.

Sample
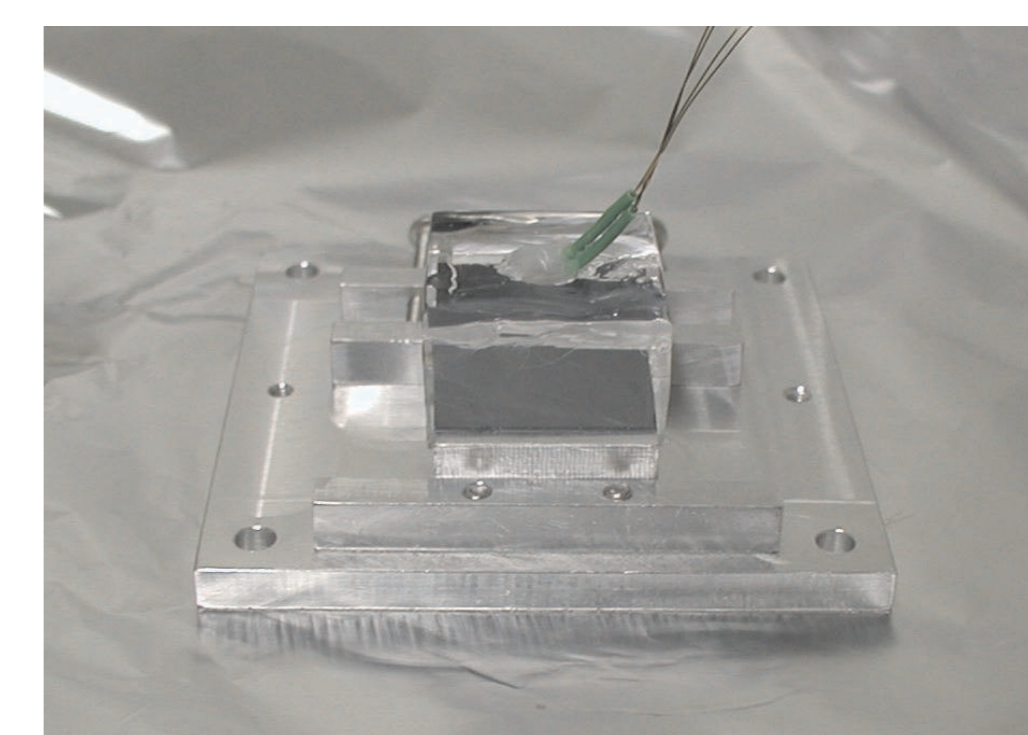
The samples are prepared in a walk-in cold room. Single crystals of ice with basal orientation are frozen onto carefully cleaned Si surfaces terminated with a native oxide layer.

A special sample cell has been designed which allows to control the temperature via peltier elements. Several Pt100 sensors monitor the temperature. Temperature stability is in the mK regime.
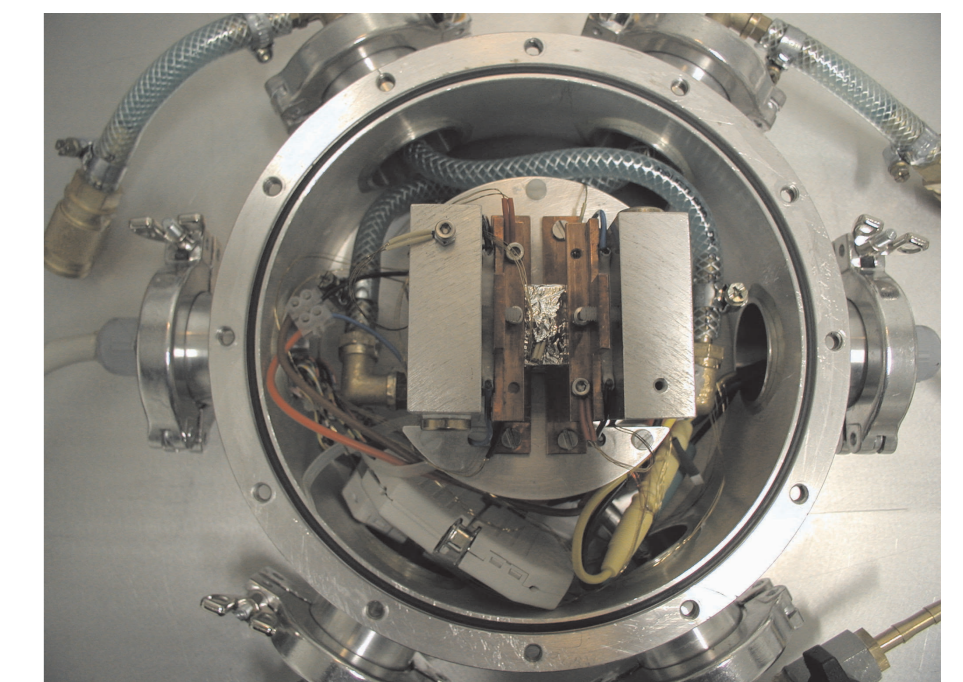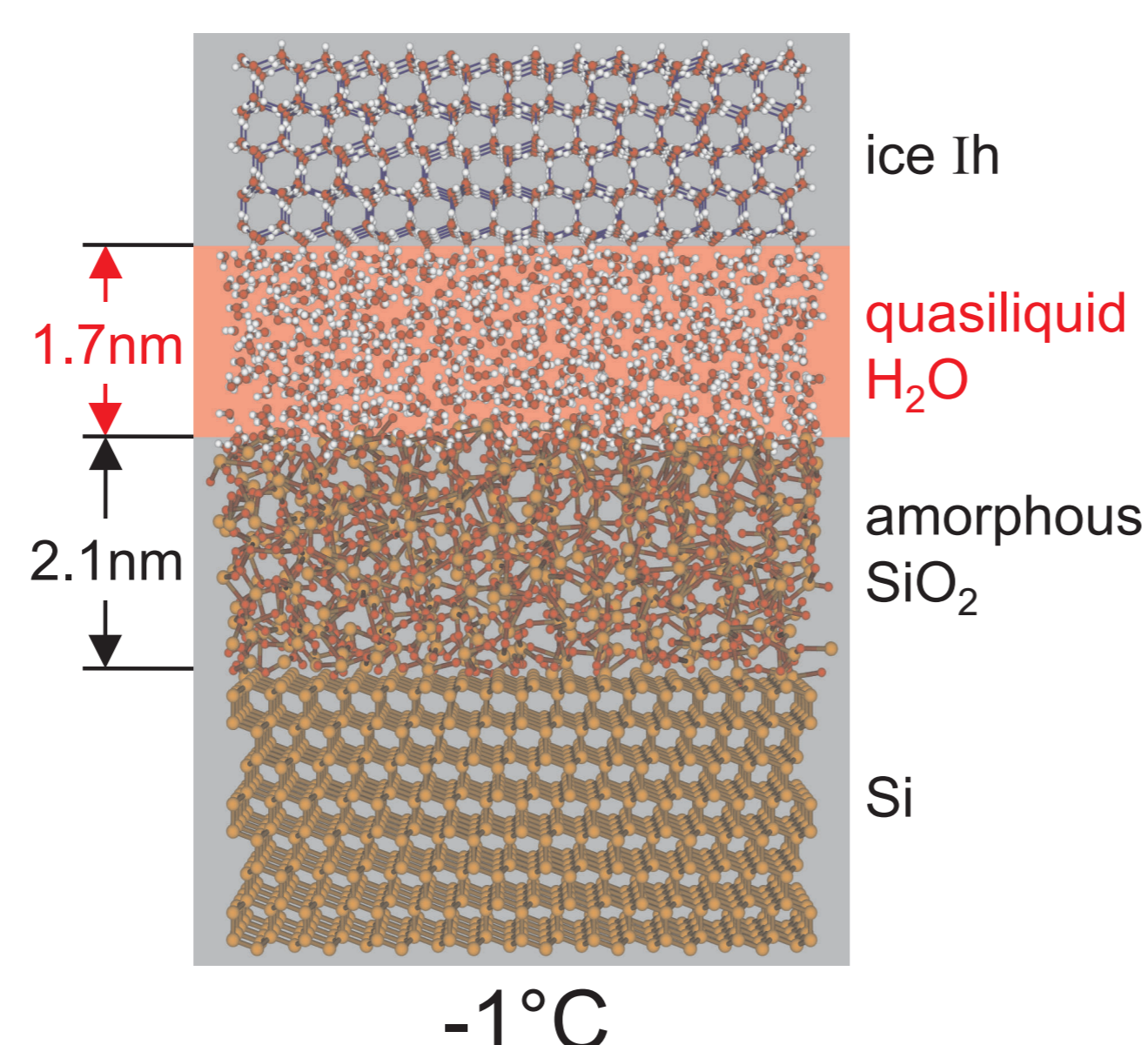
Results 1
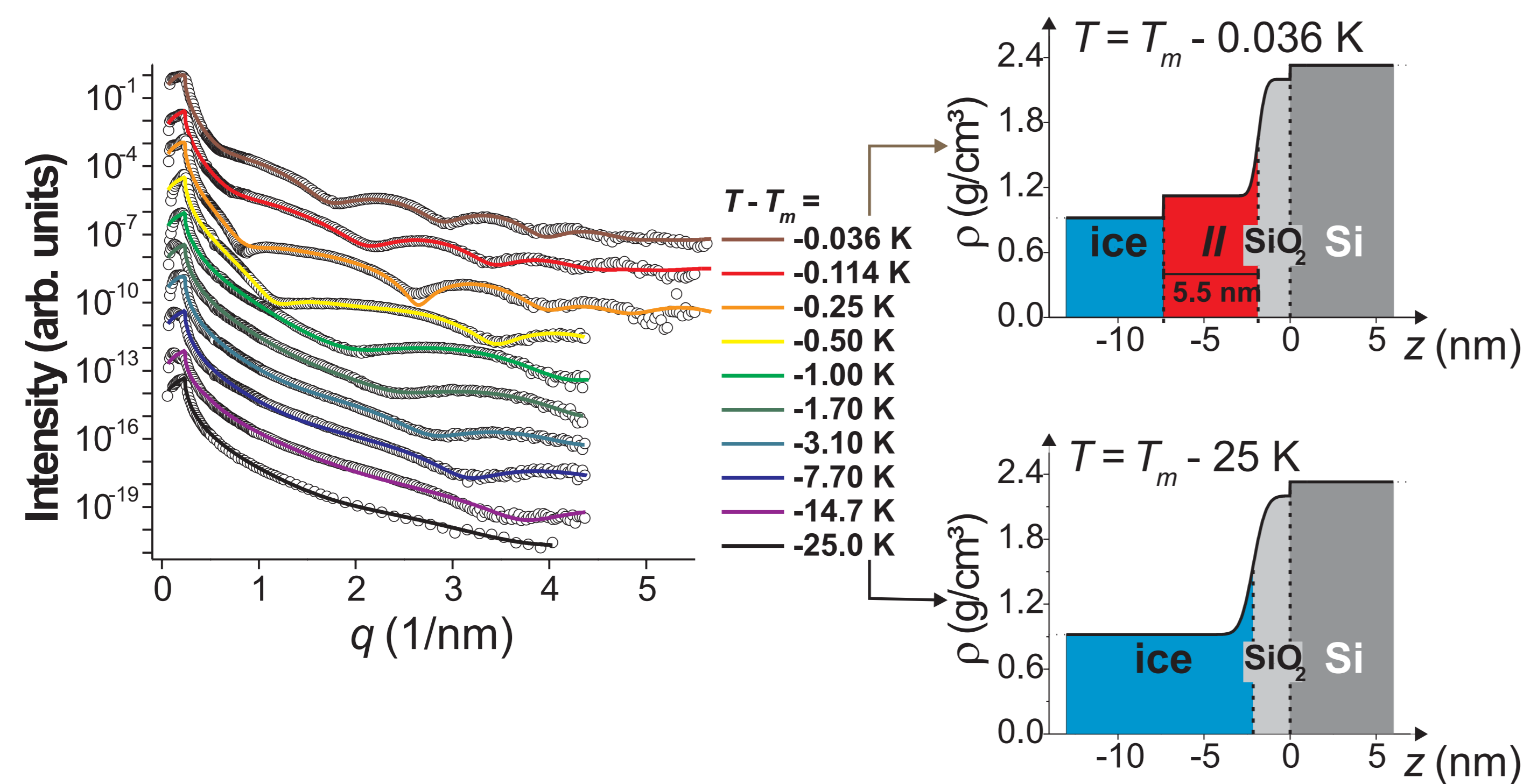
Upon heating, a drastic and reversible change in the measured X-ray reflectivity profiles can be seen (top left). The increasing number of interference fringes shows that a new layer with different density and growing thickness is formed.

Density profiles across the interface have been reconstructed using the dynamical Parrat formalism (top right). An atomic scale illustration of the interface can be seen on the right.

Results 2 and discussion
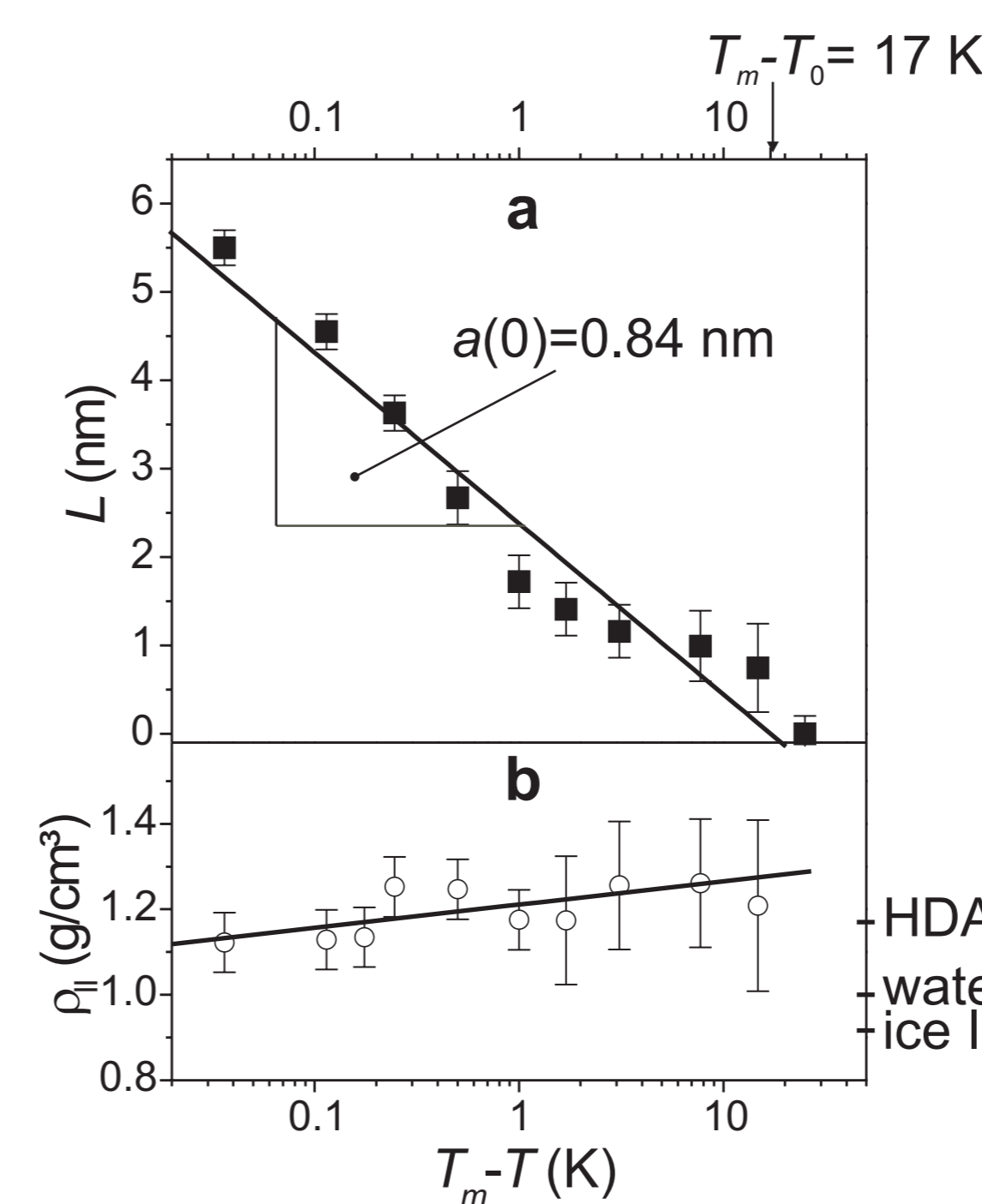
Interfacial premelting starts to emerge at an onset temperature of $T_o = T_m - (17\pm3)$ K and follows in a good approximation a logarithmic growth law which is well-known from wetting phenomena (a). The decay length of the average density in the liquid layer $a(0) = (0.84\pm0.02)$ nm can be compared to the correlation length in bulk water with literature values from 0.45 to 0.8 nm.

The density of the liquid layer (b) is significantly higher than in bulk water and close to the value for high-density amorphous (HDA) ice ($\rho_{HDA} = 1.17$ g/cm^3), suggesting a close structural relationship.

Since interface melting is an equilibrium phenomenon, one must assume that the liquid layer is not governed by the metastable HDA phase, but rather by (equilibrium) fluctuations of the postulated high-density liquid (HDL) phase.

Current water theories trying to explain the (often anomalous) properties of water are based on a liquid-liquid transition in water terminating at a second critical point (see right). Interfaces might offer a way to stabilize and study the two liquid phases of water which could not be isolated so far.

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