ESRF	**Experiment title:** Liquid structure of the Sn/Ge(111) interface	**Experiment number:** Si 477
Beamline:	**Date of experiment:** from: 12-6-1999 to: 19-6-1999	**Date of report:**
Shifts:	**Local contact(s):** Kevin Peters	*Received at ESRF:* 1 - MAR. 2000

Names and affiliations of applicants (* indicates experimentalists):

M.F. Reedijk*[1], J. Arsic*[1], F.K. de Theije*[1], E. Vlieg*[1], M. McBride*[2], K. Peters*[3]

[1] *RIM dept. of Solid State Chemistry, University of Nijmegen, 6525 ED Nijmegen, The Netherlands*

[2] *Lawrence Livermore National Laboratory, California*

[3] *Surface diffraction beamline ID03, ESRF, Grenoble*

Report:

The aim of the experiment was to find the atomic structure of an ultrathin liquid Sn layer in contact with a Germanium (111) crystal.

Two ways of characterising this structure were used. The first consists of measuring the normal crystal truncation rods of the substrate. The liquid at the interface will contain small density modulations with the substrate periodicity, and will thus contribute to the diffraction signal. The second characterisation method was to directly measure the diffuse scattering from the liquid.

A Ge (111) crystal was used, on which we deposited between 1.2 and 1.45 monolayers of Sn, the exact amount was being determined with specular reflectivity. This system has a solid-liquid transition near 170°C. One data set was obtained below and two above this transition. Each data set consisted of crystal truncation rods, specular data, in-plane data and diffuse scattering.

Below the solid- liquid transition, the Sn structure appeared to be reconstructed, in agreement with earlier RHEED experiments [1]. Unfortunately there was not sufficient time to collect fractional order data, so this structure could not be determined exactly. Nevertheless, from the integer-order data we derived important information about the structure projected in a 1x1 unit cell. An exact description of the reconstructed surface will require more data.

The Sn layer measured at the two higher temperatures shows obvious liquid behaviour, as is proved by the appearance of a ring of diffuse scattering. The position of this ring shows a small shift to higher momentum transfer at higher temperatures (figure 1) indicating that the average Sn-Sn distances become slightly smaller at increasing temperature. The influence of the germanium surface on the liquid Sn should appear in the modulation of the liquid ring, but this effect was not clearly measurable.

The Sn layer shows a dual character. Some of the Sn atoms are 'solid-like'. They reside on the three highly symmetric lattice sites of the germanium crystal (the T1, T4 and H3 sites). The other Sn atoms are 'liquid-like': they have a high diffusibility and contribute to the liquid ring intensity. The 'solid-like' atoms are visible in the crystal truncation rods, whereas the 'liquid-like' atoms contribute to the liquid ring intensity. Both types of Sn atoms are visible in the specular reflectivity, because this specular curve is only sensitive to the vertical density and insensitive to in-plane diffusion and disorder. This gives us a tool to determine the total number of Sn atoms present and the numbers of 'solid-like' and 'liquid-like' atoms. As a function of time, 'solid-like' atoms will convert into 'liquid-like' ones and visa versa.

Several changes can be observed in the liquid structure as the temperature is increased. First, at higher temperatures more Sn atoms become liquid-like, going from 14% at the lowest liquid temperature to 21% at the highest temperature, as determined from the crystallographic data. This implies that the intensity of the liquid ring should increase (more atoms contribute to this liquid ring). Qualitatively speaking, this is the case, but a quantitative measurement is difficult. The intensity appears to be increased by approximately a factor of 2.

Another important parameter is the Debye Waller parameter (an indication of the thermal vibration of the atoms). At temperatures below the solid - liquid transition the Debye Waller parameter was 4.5, above this transition this had increased to 10. Experiments performed by de Vries et al. [2] on a solid - liquid transition of lead on Ge (111) showed an increase from 6.3 to 35. The difference might be due to the difference in size between Sn and Pb.

Some crystallographic structural changes can also be observed, affecting mainly the position of the Sn atoms above the germanium surface. In the solid structure the Sn atoms are ordered approximately like an α-Sn crystal. As the temperature rises above the critical temperature, the z - coordinates of the Sn atoms remain the same. However, upon further heating of the liquid, the Sn layer becomes compressed and the height of the Sn atoms above the surface changes. This could be an indication that the liquid becomes less influenced by the germanium surface.

Thus, all data suggest an increase in liquid-like and a decrease in solid-like behaviour as a function of temperature. This is the first time that such continuous behaviour has been measured.

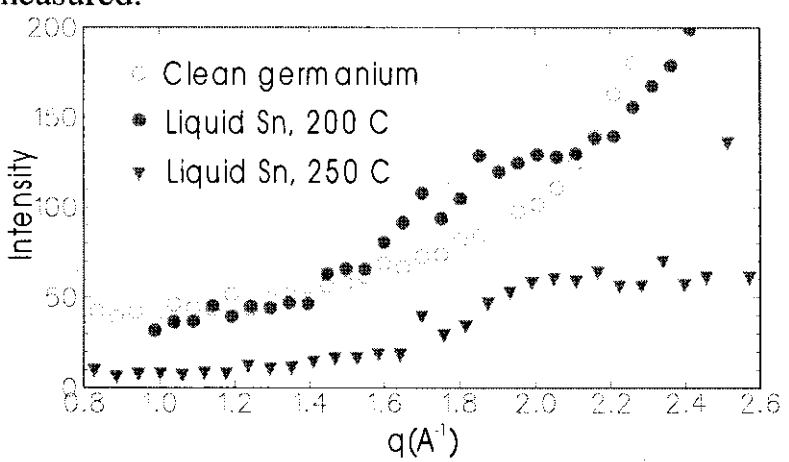

Figure 1, radial scans through the liquid ring at different temperatures (dark symbols) and on clean germanium.

[1] R. Ichikawa, Surface science **140** (1984) 37,

[2] S.A. de Vries, P. Goedtkindt, P. Steadman and E. Vlieg, Phys. Rev. B. **59** (1999) 13301.