

Microscopic Surface Structure of Liquid Potassium

O.G. Shpyrko,¹ P. Huber,¹ H. Tostmann,² P. S. Pershan,¹
B.M. Ocko,³ A. Grigoriev,¹ M. Deutsch⁴

¹Department of Physics, Harvard University, Cambridge, MA, U.S.A.

²Department of Chemistry, University of Florida, Gainesville, FL, U.S.A.

³Department of Physics, Brookhaven National Laboratory, Upton, NY, U.S.A.

⁴Department of Physics, Bar-Ilan University, Israel

Introduction

We present x-ray reflectivity (XRR) and diffuse scattering measurements of the liquid surface of pure potassium. They confirm the existence of atomic layering at the free surface of a pure liquid metal with low surface tension. Before this study, layering was observed only for metals like Ga, In, and Hg, the surface tension of which is five- to sevenfold higher than that of K and hence comes closer to the ideal “hard wall” condition. The measurements required a quantitative analysis of the contribution from thermally excited capillary waves to surface scattering. Our measurements confirm the predicted form for the differential cross section for diffuse scattering over a larger range of incident angles than any previous measurement. The partial measure of the surface structure factor that we obtained agrees with computer simulations and theoretical predictions.

Results

We report x-ray scattering results of the microscopic structure of the surface of a pure liquid alkali metal. In contrast to measurements of dielectric liquids, which show a monotonically varying density profile at the liquid-vapor interface, liquid metals have been shown to exhibit a phenomenon known as surface-induced layering. This was predicted to occur because the Coulomb interactions between the two constituents of any liquid metal—the free electron Fermi gas and the classical gas of positively charged ions—suppress local surface fluctuations that would otherwise conceal the layered atomic ordering that occurs at hard walls [1]. Most metallic systems studied so far [2-4] exhibit relatively high values for surface tension, leaving unanswered the fundamental question of whether surface atoms just order against the effective “hard wall” generated by the high surface tension, which suppresses the long-wavelength capillary waves, or whether the layering is caused by the intrinsic metallic properties of the studied systems.

Surface-induced layering is manifested in the surface structure factor (SF) [2], which is a Fourier transform of the derivative of the surface-normal density profile averaged parallel to the surface. The rise of the SF above unity is strong evidence for the presence of surface-induced layering. However, in order to obtain SF from the

measurements, one needs to deconvolve thermally excited capillary wave contributions from a measured reflectivity signal. These contributions become especially important when the reflectivity measurements are extended to high values of q_z [5]. Since thermally excited capillary wave fluctuations increase with decreasing surface tension, the deconvolution is more difficult to do for low-surface-tension liquids like K than for high-surface-tension liquids like Ga and Hg [6].

Figure 1 shows the x-ray diffuse scattering from the liquid K surface taken with a position-sensitive detector (PSD) at the Complex Materials Consortium Collaborative Access Team (CMC-CAT) sector for a wide range of q_z . As q_z increases, it becomes increasingly more difficult to separate the specular reflectivity “peak” from the “wings” because of the capillary wave diffuse scattering. When combined with the fast decay of the reflectivity with increasing q_z , this effect puts a limit on the range of q_z for which meaningful XRR can be measured for a liquid K surface.

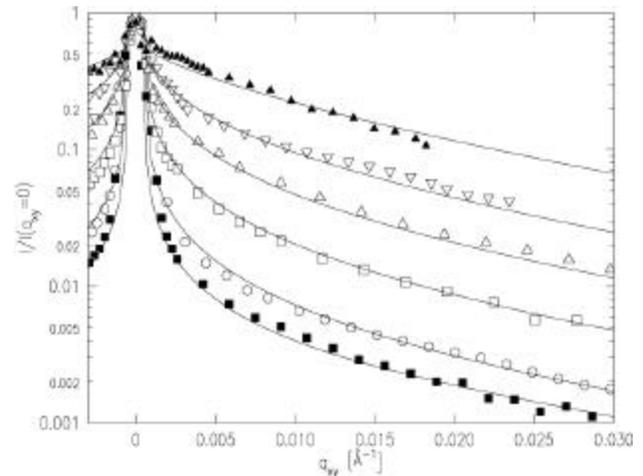


FIG. X-ray diffuse scattering from the surface of liquid K. The specular condition corresponds to (bottom to top): $q_z = 0.3, 0.4, 0.6, 0.8, 1.0,$ and 1.1 \AA^{-1} . The scans are normalized to unity at $q_{xy} = 0$ to allow comparison. Lines represent theoretically simulated scans for the given values of q_z .

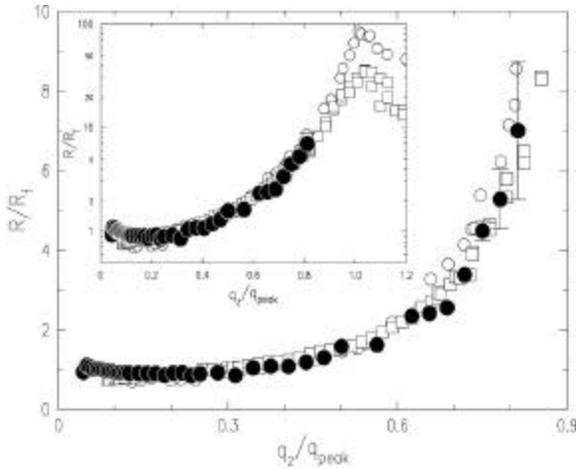


FIG. 2. The surface structure factor for liquid Ga (open circles), liquid In (open squares), and liquid K (filled circles) obtained from XRR data by deconvolving resolution, Fresnel reflectivity, and capillary wave contributions. Here q_{peak} is the value of q_z at which the layering peak is observed or expected to be observed. The inset shows the same three sets of data over an extended range.

The good agreement between the experimental data and the theoretical modeling of the capillary wave effects allows deconvolution of the surface SF from the raw XRR measurements. Fig. 2 shows the liquid K reflectivity data normalized to the Fresnel reflectivity and corrected for the capillary waves' scattering. This result is the surface SF of liquid K.

The surface SF rises clearly above unity, which is evidence for the occurrence of surface-induced layering in liquid K. The comparison in the figure with surface SFs measured for liquid Ga [3] and In [4] shows that over the measured range of q_z , the surface layering in the present low-surface-tension K appears to be comparable to that of the high-surface-tension liquids.

Acknowledgments

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