X-ray Standing Wave Study of the Low-Temperature Phase Transition for 1/3 Monolayer of Pb on the Ge(111) Surface

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Introduction

Pb adsorption on a Ge surface presents a model two-dimensional system for the study of a metal on a semiconductor due to their mutual, near insolubilities.[1] The focus of this study is the low-temperature, structural transition for 1/3 monolayer (ML) of Pb on the Ge(111) surface. At room temperature, the Pb forms a (√3×√3)R30° reconstruction and is typically modeled with a single atom on the T_4 adsorption site.[2] When the temperature is lowered to ca. 100K, the reconstruction undergoes a gradual and reversible transition to a 3×3 phase.[3] A surface x-ray scattering analysis concluded that the 3×3 phase has three Pb atoms per unit cell with a 0.4 Å vertical corrugation in their heights, where one atom is higher than the other two atoms in the unit cell.[4] Also reported were severe displacements of the Ge atoms near the surface that extend down to the third bilayer. X-ray standing wave (XSW) results are model independent and provide the bulk-extrapolated positions of the Pb adatoms without assuming positions for Ge atoms near the surface. By measuring at both room and low temperatures, the average position and distribution for Pb can be compared to determine if the time-averaged structure changes.

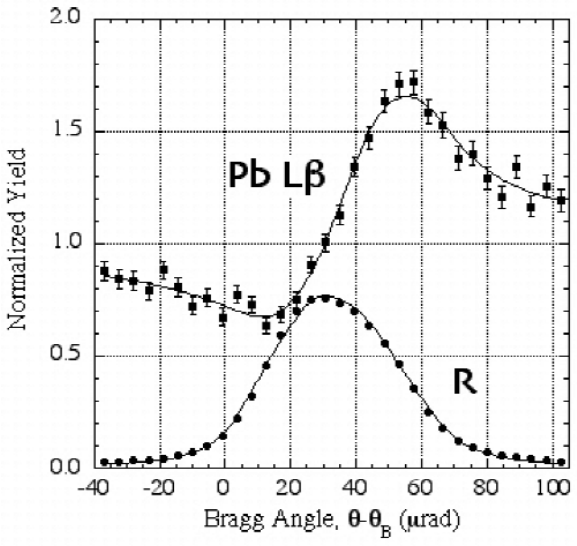

FIG. 1. XSW measurement at low temperature for the (111) normal reflection for sample with 0.27 ML of Pb. The lower curve is the normalized diffracted intensity for the (111) peak. The upper curve is the normal yield for Pb Lβ fluorescence collected during the scan.

Methods and Materials

The sample preparation and measurements were performed in a UHV chamber coupled to a six-circle diffractometer located at the DND-CAT 5-ID-C station. The Ge(111) surface was sputter cleaned and annealed until low-energy electron diffraction (LEED) showed a clean, well-ordered c(2×8) reconstruction. Pb was evaporated onto the clean surface, and the sample was annealed to obtain the (√3×√3)R30° reconstruction. Upon cooling the sample to 115K, the surface exhibited a 3×3 structure.

During our XSW measurements, the H^{th} Bragg peak for the Ge single-crystal substrate is scanned and the induced modulation in the Pb Lβ fluorescence intensity is measured. Dynamical diffraction theory was used to fit the rocking curve and fluorescence spectrum to obtain the coherent position (P_H) and coherent fraction (f_H) for the Pb atoms. To verify that the data collection and analysis procedure, the Ge Kα fluorescence from the substrate was analyzed and found to agree with the bulk crystal structure Ge positions. A representative XSW scan is shown in Fig. 1, the lower curve is the rocking curve for the (111) reflection normal to the surface and the upper curve is the modulation in Pb Lβ fluorescence. For this analysis the origin is located at the center of the Ge bilayer (see Fig. 2).

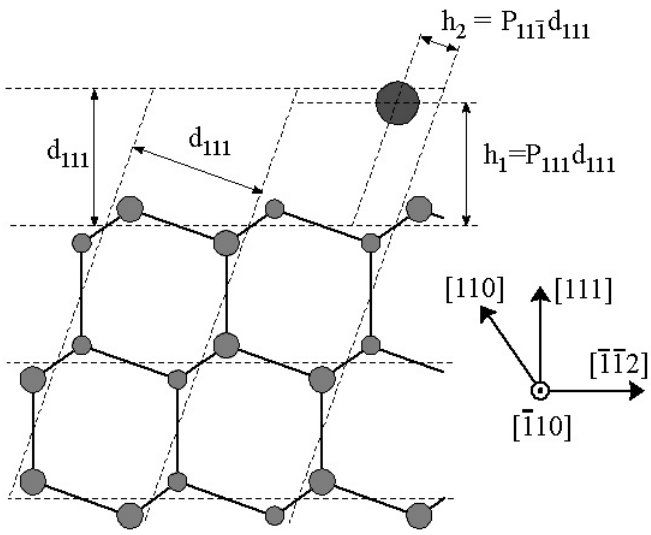

FIG. 2. A side-view representation of Pb on the Ge(111) surface. The coherent position of Pb is triangulated by combining the results for the normal (111) and off-normal (11$\bar{1}$) measurements.

XSW measurements were made with a 16.35 keV x-ray beam on two sample surfaces with different Pb coverages, 0.32 ML and 0.27 ML. The coverages were determined by comparing the Pb Lβ fluorescence yield with an ion-implanted standard (calibrated with Rutherford backscattering). The (111) and (333) reflections normal to the surface were measured at room and low temperatures. Additionally, an off-normal (11$\bar{1}$) reflection was measured at room temperature.

Results and Discussion

The results for five measurements are compiled in Table 1. By using the normal (111) and off-normal ($11\bar{1}$) data, the position of the Pb can be triangulated, as shown in Fig. 2. The first point to note is that this XSW data does not agree with a strictly T_4-site model. It appears that the location of the Pb atom is more consistent with an off-centered T_1 site or a Pb atom population split between T_1 and T_4, with a higher fraction on T_1 sites. A similar conclusion was reached in an XSW study of 1/3 ML Pb ($\sqrt{3}\times\sqrt{3}$)R30° phase on Si(111).[5]

Table 1: XSW experimental results for 1/3 ML Pb on Ge(111)

	Θ Pb		Experimental values		Calculated from T^4-site model	
(hkl)	Temp.	(ML)	P_H	f_H	P_H	f_H
(111)	Room	0.32	0.93(3)	0.70(2)	0.71	0.88
(111)	Room	0.32	0.28(2)	0.47(2)	0.40	0.94
($11\bar{1}$)	Low	0.27	0.91(2)	0.71(2)	0.71	0.88
(111)	Room	0.27	0.89(2)	0.67(2)	0.71	0.88
(333)	Room	0.27	0.95 (3)	0.43(6)	0.09	0.29

A second result can be determined by comparing the (111) measurements at room and low temperatures. At both temperatures, the XSW results are the same, within error. While the LEED images showed that the long-range order of the surface changed, the time-averaged local atomic structure remained the same. This supports a dynamical fluctuations model, where the time-averaged structure does not change, but the atomic motions are locked into place at lower temperature.

Acknowledgments

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