X-ray Standing Wave Measurement of 1/3 ML of Pb on the Ge(111) Surface

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Introduction

The focus of this study is to measure the atomic-scale structure of the 2-D, temperature-induced, reversible phase transition for 1/3 monolayer (ML) of Pb on the Ge(111) surface. At room temperature, Pb forms a $(\sqrt{3}\times\sqrt{3})R30°$ reconstruction and is typically modeled with a single atom on the T_4 adsorption site [1]. As the temperature is lowered to near 100K, the reconstruction undergoes a gradual and reversible transition to a (3×3) phase [2]. A surface x-ray diffraction (SXRD) analysis concluded that the low-temperature (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. The authors also reported severe displacements of the Ge atoms near the surface that extend down to the third bilayer [3]. X-ray standing wave (XSW) results are element-specific and model-independent and provide the bulk-extrapolated positions of the Pb adatoms. With measurements at both room and low temperatures, the average position and distribution of Pb can be compared to determine if the time-averaged structure changes. This investigation improves upon our previous measurements by achieving higher resolution in the data and including additional off-normal Bragg reflections.

Methods and Materials

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 is 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 procedures are responding correctly, 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 curves represent the modulation in Pb Lβ fluorescence measured with XSW and calculated for the SXRD model [3]. For our analysis, the origin is located at the center of the Ge bilayer, as shown in Fig. 2.

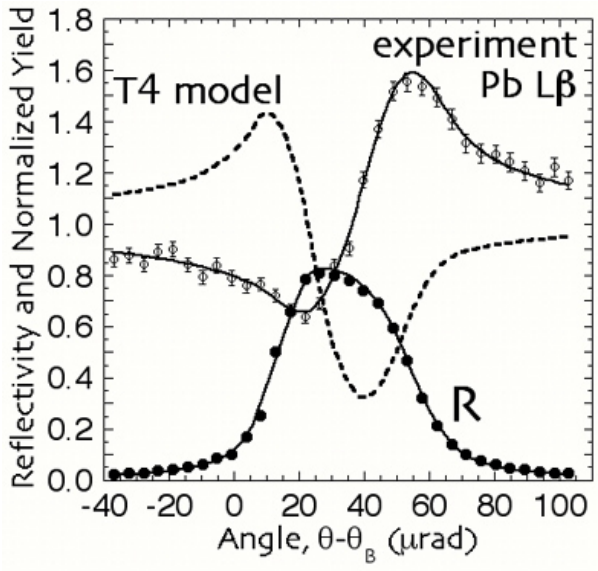

FIG. 1. Comparison of normalized fluorescence yield for XSW experimental data at room temperature and the expected result on the basis of an SXRD model [3]. The lower curve is the normalized diffracted intensity for the (111) Bragg reflection.

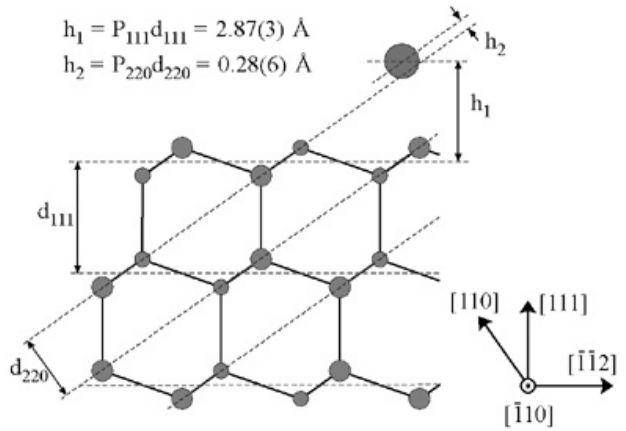

FIG. 2. A side-view representation of Pb on the Ge(111) surface. The coherent position of Pb is triangulated by combining the results for the normal (111) and off-normal (220) measurements.

The sample preparation and measurements were performed in an ultrahigh vacuum (UHV) chamber coupled to a six-circle diffractometer located at the DuPont-Northwestern-Dow Collaborative Access Team (DND-CAT) sector 5-ID-C. 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 a sharp $(\sqrt{3} \times \sqrt{3})$R30° reconstruction. Upon cooling the sample to 110K, the surface displayed a (3×3) reconstruction.

XSW measurements were made with a 16.35-keV x-ray beam on a sample with 0.4 ML Pb. The coverages were determined by comparing the Pb Lβ fluorescence yield with an ion-implanted standard (calibrated with Rutherford backscattering). The normal (111) and (220) off-normal reflections (to the surface) were measured at room and low temperatures.

Results and Discussion

The results for these four measurements are compiled in Table 1. For both the (111) and (220) measurements, the XSW results are the same, within error, at room and low temperatures. While the low-energy electron diffraction (LEED) images show that the long-range order of the surface changed, the time-averaged local atomic structure remained the same. This is consistent with an order-disorder phase transition and supports a dynamical fluctuations model, where the time-averaged structure does not change but the atomic motions are locked into place at low temperature [4].

Table 1. XSW experimental results for 0.4 ML Pb on Ge(111).

		Results from XSW experiment		Calculated from SXRD model [3]	
(hkl)	Temp (K)	P_H	f_H	P_H	F_H
(111)	298	0.87(1)	0.70(2)	0.71	0.88
(220)	298	0.14(3)	0.51(9)	0.11	0.76
(111)	110	0.88(1)	0.71(2)	0.71	0.92
(220)	110	0.14(2)	0.50(7)	0.11	0.85

By using the normal (111) and off-normal (220) data, the location of Pb can be triangulated, as shown in Fig. 2. The measured position for Pb above the Ge surface (2.87 Å) is significantly higher than the result for the SXRD model (2.34 Å). Also, these XSW results do not agree with a strictly T_4-site model. It appears that the location of the Pb atom is inconsistent with a single high-symmetry adsorption site. The measured coherent positions and fractions for the normal (111) reflections are similar to our prior XSW measurements.

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