Melting of Indium at High Pressure Determined by X-ray Diffraction

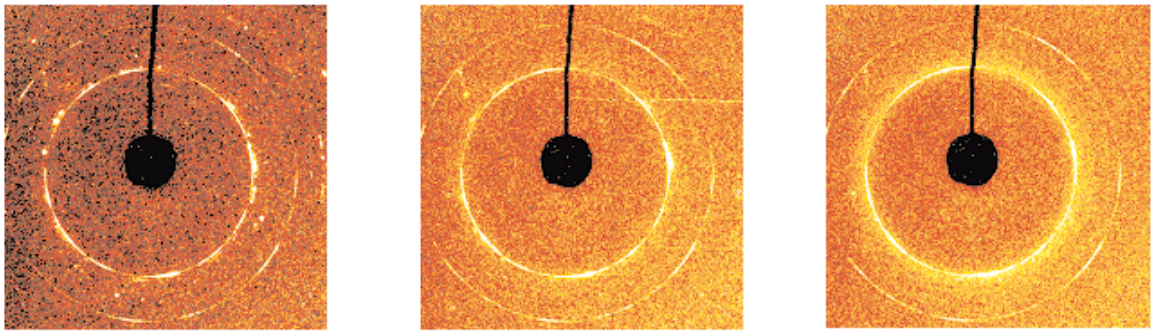
G. Shen,[1] N. Sata,[1] M. L. Rivers,[1,2] S. R. Sutton[1,2]

[1] *Consortium for Advanced Radiation Sources, University of Chicago, Chicago, IL, U.S.A.*

[2] *Department of Geophysical Sciences, University of Chicago, Chicago, IL, U.S.A.*

Introduction

In recent years, the generation and measurement of simultaneous high pressures and high temperatures has undergone rapid development with the diamond anvil cell (DAC) technique.[1-3] The amount of available data on melting behavior at high pressure is increasing. However, at high pressures, detection of the onset of melting is challenging. The difficulties lie in the characterization of small samples (e.g., unambiguous melting criteria) and in the measurement of pressure and temperature under extreme conditions. In this study, x-ray diffraction/scattering data were used to identify melting unambiguously by the observation of diffuse scattering from the melt with simultaneous disappearance of all crystalline diffraction peaks.



(a) T=519±1 K (b) T=526±1 K (c) T=531±0.7 K

FIG. 1. X-ray scattering images at the onset of melting of indium at about 2 GPa. Two sharp rings are the diffraction of NaCl. (a) Crystalline indium shows a spotty pattern. (b) The indium diffraction spots fade out and diffuse scattering appears. (c) A clear homogenous diffuse ring of the melt can be seen.

Experimental Procedures and Results

An externally heated DAC (DXR-7, Diacell) was used in this study. The main feature of the DAC design is the use of four cartridge heaters inserted in the cell, providing heat to the whole cell and resulting in a uniform temperature distribution inside the cell. Indium powder (99.999%, Alfa Aesar) was loaded into the rhenium gasket aperture (100 mm in diameter and 30 mm thick). NaCl was used as the pressure transmitting medium and the internal calibrant for pressure measurement.

The x-ray diffraction experiment was performed at GeoSoilEnviroCARS, beamline 13-BM-D. A CCD detector (Bruker-2K) was used to collect diffraction patterns. The x-ray beam size was controlled by a slit system to 150x150 mm and then focused to a beam size of 7(vertical) x 10(horizontal) mm full width at half maximum by Kirkpatrick-Baez mirrors.[4]

Figure 1 shows recorded diffraction patterns at ~2 GPa near the onset of melting. When the temperature was raised to 519K, the crystalline nature of indium was still clearly identified (spotty pattern in Fig. 1a). As the temperature was increased to 526K, we observed that the indium diffraction spots faded out and diffuse scattering appeared (Fig. 1b). When the temperature was raised to 531K, the scattered diffuse area (Fig. 1b) became a homogenous broad ring (Fig. 1c) accompanied by the disappearance of all diffraction spots from indium, indicating a completely molten state. Upon slight cooling, to 523K, the homogeneous broad ring disappeared, and the diffuse scattering was now distributed only in some localized areas. The CCD images were integrated using the software Fit2D as a function of diffraction angle 2θ. The broad diffuse ring is clearly seen in the integrated pattern as a broad band centered near 9 degrees 2θ. Figure 2 shows the area of the diffuse band as a function of temperature near the melting point. There is a sharp change in intensity of the diffuse scattering upon melting over a temperature interval of about 6K. The solid-liquid transition is reversible with negligible hysteresis within the temperature precision in this study. As the temperature was further decreased, we found that indium did not show crystalline diffrac-

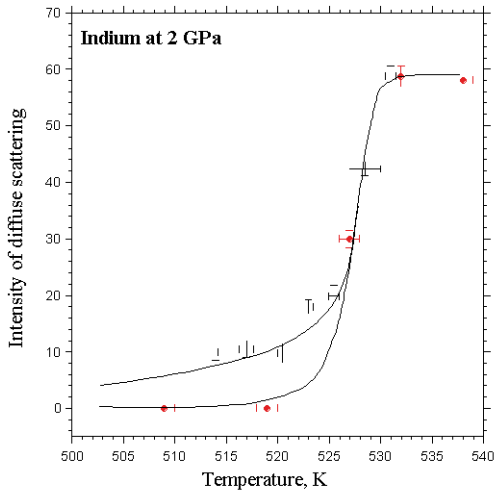


FIG. 2. Intensity change of the diffuse scattering band at the onset of melting. Solid circles are the data upon heating; while the crosses are those on cooling. Lines are freehand fits to the heating and cooling paths. The hysteresis of complete solidification appears at the low temperature side of the sharp transition. Error bars for temperature are from the multiple measurements during the exposure of 5 min. Errors for the intensity of diffuse scattering are obtained from the standard errors in fitting the band with >95% confidence limits.

tion immediately below melting point. Some hysteresis in the intensity occurred at the bottom of the sharp change as can be seen from Fig. 2.

The observed melting curve is in good agreement with previous determinations based on resistivity measurements in piston cylinder apparatus.[5,6] These results demonstrate the successful melting experiments in a diamond anvil cell using the x-ray diffuse scattering as the melting criterion. The current method provides an objective way of identifying melting, an important extension of the visual observation methods generally used in previous DAC studies. This technique, using an area detector and micro-x-ray beam, should be applicable to many other high pressure melting studies, such as laser-heated DAC experiments on melting at ultrahigh pressures.

Acknowledgments

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