

# Chapter 6

## X-Ray Scattering Studies of Expanded Fluid Metals

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### ABSTRACT

*Fluid metals exhibit significant properties of thermodynamic-state dependence, since the inter-particle interaction among the constituents (electrons and ions) considerably changes depending on their thermodynamic conditions. The authors have thus far carried out X-ray scattering experiments of fluid metals in the expanded state, which have enabled them to gain insight into microscopic understanding of the structural and electronic properties of fluid metals. The purpose of this chapter is to provide intriguing aspects of fluid metals originated from the existence of conduction electrons, which distinguishes fluid metals from non-conducting fluids, through the results of fluid rubidium and mercury.*

### INTRODUCTION

#### Liquid Metals

Liquid metals can be regarded as a two-component system which consists of highly mobile conduction electrons and much heavier ions. In particular, the existence of conduction electrons is inherent characteristics of liquid metals, which clearly distinguishes liquid metals from a typical insulating liquid such as inert gas liquids, where the constituents at ambient conditions are basically neutral atoms which interact with each other via van der Waals forces. Liquid metals near the melting point exhibit a typical metallic character: the electrical conductivities of simple liquid metals such as alkali metals actually take metallic values ( $10^{-40} \mu\Omega \text{ cm}$  in resistivity) near the melting point (Faber, 1972; Shimoji, 1977). These values are within the same order of those in the solid state and also explain high thermal conductivity of liquid metals. With the success of the pseudopotential approach (Harrison, 1966), it was demonstrated

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that the electron-ion interaction near the melting point can be treated as a weak perturbation. As a result, conduction electrons are well described with the nearly free electron model even though the atomic arrangements become disordered upon melting. The Ziman formula (Ziman, 1961) obtained within the framework of the nearly free electron approximation has reasonably explained most of the conductivities of liquid metals (Faber, 1972).

Although a liquid metal is a mixture of ions and conduction electrons, a liquid metal near the melting point behaves as a simple monatomic liquid. This monatomic picture is based on the fact that ions in a liquid metal can be regarded as neutral pseudo atoms (Ziman, 1979), i.e., the Coulomb forces between ions are well-screened by conduction electrons.

## **Fluid Metals: Liquid Metals in the Expanded State**

When a liquid is heated across the liquid-vapor coexistence curve, the density sharply decreases accompanying the liquid-vapor first order phase transition. Vaporization of the metallic elements across the liquid-vapor coexistence curve accompanies the transformation from a metal to an insulator of the elements. Thus, the liquid-vapor phase transition involves the electronic transition. The electrical conductivity abruptly decreases on evaporation across the liquid-vapor coexistence curve, which indicates its discrete nature of this electronic transition, where the itinerant nature of valence electrons vanishes and valence electrons are bound by atoms or molecules, i.e., the constituents of a vapor phase of metals.

However, a line of the liquid-vapor phase transition is terminated at the critical point at high temperature and high pressure. A substantial and continuous variation in the density becomes possible on a pressure-temperature phase diagram by circumventing the critical point, and the fluid phase provides a great opportunity to investigate variation of the physical properties of materials as a function of density over a wide range of thermodynamic conditions from a liquid phase near the triple point to a vapor one.

A more unique and interesting situation is brought about when a liquid metal circumnavigates the liquid-vapor critical point without crossing the saturated-vapor-pressure curve by controlling temperature and pressure, and “intermediate” density between a liquid and a vapor can be realized as shown in the arrow in Figure 1. Correspondence relation between the liquid-vapor and the metal-nonmetal transitions is no longer straightforward (Landau & Zeldovich, 1943). Earlier investigation indicated that the electrical conductivity gradually decreases with volume expansion and more sharply decreases when the fluid reaches around the critical point (Franz et al., 1980). In this continuous expansion of the fluid, a dilute metallic state is realized where the conduction electrons and ions forced to rarefaction to a large extent. The physical properties of fluid metals are considerably dependent on their thermodynamic state (Hensel & Warren, 1999). The situation makes fluid metals intriguing in terms of basic scientific interest, stimulating the investigation for the nature of an electronic transition from a metal to an insulator, its relation with the liquid-vapor phase transition, and the phase behavior particular to the electronic system.

## **Alkali Metals**

Among the metallic elements, alkali metals are of special concern since they are classified as simple metals. Valence electrons in alkali metals originate from s-state and alkali metals have an almost spherical Fermi surface, thus the electronic states in alkali metals are well described with the nearly free electron approximation. This physical picture of alkali metals is basically preserved in the melts. Volume change of alkali metals upon melting is only about 2.5% increase, which means reduction in the valence electron

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