

## Two melts phase separation in the liquid Sb-Sb<sub>2</sub>S<sub>3</sub> system: critical sound wave propagation and metal-non-metal transition

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**Abstract.** The sound velocity and magnetic susceptibility as a function of temperature and composition were measured to investigate critical sound wave propagation and metal-non-metal transition in the liquid Sb-Sb<sub>2</sub>S<sub>3</sub> system. The sound velocity in a homogeneous alloy around 60 at.% of Sb decreases very rapidly and the rate of decrease increases as the two melts phase is approached, which is the typical temperature dependence of the sound velocity in a liquid with a miscibility gap. Below the critical point, the sound velocity was measured along the phase boundary. Using those data, the phase boundary was precisely determined. The critical point is located at  $901 \pm 2$  °C and  $41.5 \pm 0.5$  at.% S, and the critical exponent of the phase boundary is about 1/3. On the other hand, the magnetic susceptibility as a function of temperature and composition indicates that the electronic state is metallic in liquid Sb and non-metallic in molten Sb<sub>2</sub>Se<sub>3</sub>, and crossover from the metallic to non-metallic state occurs around the critical composition.

### 1 Introduction

There reported many binary elemental systems which have miscibility gaps in the liquid state. Most of their phase boundaries are, however, qualitative especially for systems having high critical temperature [1]. The critical indices of the concentration differences between the coexisting two liquids for most of those systems have been undetermined and it is not known whether or not other thermodynamic quantities exhibit critical behavior along the phase boundary as well.

In the previous investigations, we studied, for example, the sound velocity in the Ag-Se system by measuring sound velocity [2]. This system has two immiscible regions between Ag-Ag<sub>2</sub>Se and Ag<sub>2</sub>Se-Se. For both immiscible regions, the critical sound propagation in alloy close to a critical composition has been observed as the temperature is approached to the two-melt critical point from above. The sound velocity along the phase boundary has not been determined, however, because such measurements are time-consuming and it is not known the characteristics of the sound velocity along the phase boundary so far.

Recently, we have developed a new sound velocity measuring system which enables to determine the sound velocity almost continuously with changing temperature once the initial setup of a specimen has been made [3]. In this paper, we report the results of Sb-Sb<sub>2</sub>S<sub>3</sub> system. In addition, very detailed magnetic susceptibility measurements have been made. From the latter we have obtained information on changes in the electronic structure with increasing S.

### 2 Experimental procedure

The present system for the sound velocity measurements consists of LeCroy LT262 (350MHz) oscilloscope, Panametric 5077 pulsar-receiver, and Okura EC5500 or Yamatake-Honeywell SDC40 digital temperature controller, which are controlled by PC. The program is a home-made program on Visual Basic.

Ultra-sonic transducer was PZT operating at about 9 MHz. A cell was made of fused quartz, of which the thickness of sample reservoir was determined by measuring the sound velocity in distilled water as a function of temperature in the temperature interval from room temperature to 96°C. The sound velocity could be fitted to the reference data [4] within to less than 0.1% over the above mentioned temperature interval by taking the thickness of sample reservoir as a single disposable parameter. In the actual measurements, the thermal expansion of a cell was neglected because it is very small as compared with changes in the sound velocity in a specimen. The time interval that the sound propagates in a sample was measured using the time function of the LeCroy oscilloscope, the resolution of which is approximately 0.32 ns. The sound velocity in a quartz buffer rod has a temperature dependence, however the change does not affect the results because the reflection method was used, in which the time required for the sound pulse to propagate in the rod is automatically cancelled. In the measurements we changed temperature by 1 K step, and checked whether the temperature was equal to the setting temperature or not. The check was made 3 times at every 0.5 min, and then measurements were made after few minutes waiting time





