Oscillations in the ultrasonic velocity during heating of liquid gallium

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Highly sensitive measurements have revealed oscillations in the temperature dependence of the ultrasonic velocity in liquid gallium. The oscillations are interpreted as a compressibility-oscillation effect and attributed to the particular nature of the interaction between the medium and microscopic regions in which the crystal structure has been partially retained.

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The temperature dependence of the ultrasonic propagation velocity in liquid gallium has been measured by the method of Ref. 1. The measurements were carried out in an atmosphere of spectrally pure argon. Gallium samples with a purity no worse than 99.999% Ga were selected and placed in a cell; measures were taken to prevent their oxidation. The cell and the acoustic media were made of fused quartz. In the course of the measurements we observed regular oscillatory anomalies in the temperature dependence of the ultrasonic velocity.

Since the object of this study was a rather subtle manifestation of the properties of the sample, in this case its compressibility and thermal expansion, we made a major effort to improve the sensitivity of the experimental procedure. Refinements in the thermal part of the apparatus to maximize the rigidity of the structure, a special procedure for detecting the phase of the ultrasonic wave, and the use of "brightening" coatings of boron anhydride to arrange stable acoustic contact improved the sensitivity of the measurements to at worst 1 m/s. The error due to diffraction was held at a negligible level by choosing an appropriate acoustic baseline (about 20 mm). The average value of the absolute random error of 20 measurements at a fixed temperature was $\pm$ 1.5 m/s.

Experiments were carried out with various samples, subjected to various types of heat treatment: cyclic heating and cooling. The results reveal clearly defined oscilla-

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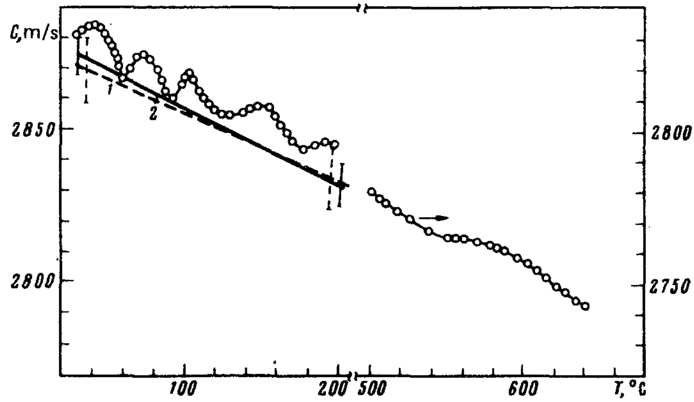

FIG. 1. Temperature dependence of the ultrasonic velocity in liquid gallium. Circles—data of the present experiments (the diameter of a circle shows the absolute error); lines 1, 2—data from Refs. 2 and 3, respectively.

tions in the temperature dependence of the ultrasonic velocity. The oscillation amplitude reaches 8 m/s at the beginning of the temperature interval studied and 3 m/s at the end. Figure 1 shows the final temperature dependence of the ultrasonic velocity in liquid gallium, constructed from the data of 20 experiments. Shown for comparison by the dashed and solid lines are the data of Refs. 2 and 3. The vertical error bars correspond to the indicated studies. We see from this figure that the oscillations observed in the present experiments might have been attributed to experimental error by other investigators because of less sensitive experimental procedures.

In the hydrodynamic approach the propagation velocity of elastic waves in a liquid medium depends on the compressibility and density of the medium. Since the temperature dependence of the density of liquid gallium has no oscillatory anomalies, and since the decrease in the ultrasonic velocity upon heating predicted by the hydrodynamic expression, $C = (\beta\rho)^{-0.5}$, where ρ is the density and β and compressibility, contradicts the changes observed in the density, it may be suggested that the oscillations of the ultrasonic velocity result from oscillations of the compressibility. The effect observed here may thus be called a compressibility-oscillation effect. The reason for the effect evidently lies in the structure of liquid gallium and the particular nature of the interaction of the constituent structures. Semimetal and semiconductor melts have microscopic regions in which the crystal structure is partially retained. The existence of solidlike microscopic regions in samples heated comparatively slightly beyond melting has been confirmed independently by experiments on internal friction, the Hall effect, the acoustics of ultrashort elastic waves, and thermal expansion.[4] Direct diffraction studies of liquid gallium itself have confirmed that there are solidlike microscopic regions in the melt even when heated well above the melting point.[5,6] When a gallium melt is heated, the solidlike microscopic regions dissolve. They apparently do not disappear completely, however, and the dissolution is accompanied by simultaneous conversions which lead to thermally more stable forms. In all probability, it is the dissolution and conversion into progressively higher-temperature forms of the solidlike microscopic regions that are being seen in the compressibility-oscillation effect.

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