LASER-ULTRASONICS: A NON-CONTACT METHOD TO CHARACTERIZE ELASTIC PROPERTIES OF METAL UP TO THE MELTING POINT

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Abstract

The aim of this work is to determine the elastic behavior of metals up to their melting temperature by measuring the elastic bulk velocities. Because of the high temperature domain and the solid to liquid transition, an innovating non-contact (Laser-Ultrasonics) has been optimized. This paper is especially applied to the characterization of the Tin (Sn), from 300K to 550 K (T_m = 505 K). We analyze the experimental results concerning the elastic wave velocity and the modeling of the shear elastic modulus falling down to zero at T_m. For tin, elastic velocities in the solid and the liquid phase are determined. The experimental results show a linear decrease followed around T_m by a jump of the longitudinal wave velocity and a drastic fall to zero of the shear velocity. This approach is quite attractive because measurements in both phases (solid and liquid) with the same experimental device is performed.

INTRODUCTION

The elastic moduli (Young modulus E, shear modulus G and adiabatic bulk modulus B) are the parameters needed to implement the constitutive laws. These mechanical properties can be measured for a solid or a liquid with different ultrasonic methods [1].

Because the temperature dependence of the shear modulus G(T) must be accounted for in any elastic-plastic constitutive model [2], this paper deals with the determination of G(T) especially during the solid-liquid phase transition. Elastic wave velocity determination is performed by an original laser-ultrasonics device [3, 4, 5]. Indeed, owing its non-contact characteristic, this technique has demonstrated its potential for high temperature measurement [6, 7, 8]. Nakano *et al* [9] determined the longitudinal velocity in solid and liquid Tin. The present work focuses on Tin too and proposes the simultaneous determination of the longitudinal and the shear velocities from the room temperature T_0 up to the melting point T_m. Indeed, a special effort was made to measure shear-wave velocity despite a strong attenuation [10] function of the temperature.

A continuous function G(T) from T_0 to T_m is proposed, taking into account the drastic jump of the shear modulus at T_m, validated by experimental measurements.

MATERIAL AND LASER-ULTRASONICS SETUP

This study is devoted to the characterization of white Tin samples obtained by cold laminating. Tin has a tetragonal crystallographic structure and its melting point is 505 K (the present work takes place at ambient pressure). White Tin exhibits a solid-solid phase transition at 286 K out of the temperature range of this study. The grain size is smaller than 200 µm. The solid is considered as a polycrystal and therefore is supposed to be isotropic. The samples have a parallelepiped shape with a square surface (15×15 mm^2) and their thickness range from 1 mm and 5 mm.

The experimental setup (figure 1) is composed of three main components: a Q-switched Nd:YAG laser operating at the wavelength of 1.064 µm to generate ultrasounds; a Mach-Zehnder heterodyne interferometer [5] to measure the normal component of the mechanical displacement; a furnace with infrared lamps to control the temperature of the sample (1-10 K/min - temperature level better than 1/10 K).

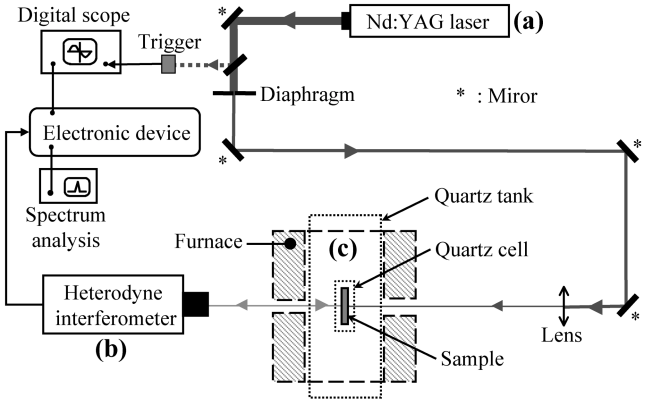

Figure 1: Experimental set-up

A sample is introduced inside a quartz cell located in a greater quartz tank filled by inert gas (Ar) to avoid oxidation effects, especially around the melting point. All the signals are recorded and digitized by using an oscilloscope. The sampling rate is 500 MHz. Average function is used (256 shots). The full details of this setup are described in a previous paper [4].

DETERMINATION OF ELASTIC VELOCITIES

The experiment principle is to determine the times of flight respectively of the longitudinal and shear waves in the solid, and of the longitudinal waves in the liquid. The principle of elastic velocity determination is discussed in references [4, 11] keeping in mind that optical power density is adjusted in order to ensure a thermoelastic generation mechanism [3]. Measurements are performed in a transmission configuration at the epicenter. Correcting the thickness for thermal expansion [12], velocity is then calculated.

First of all, elastic velocities must be measured versus temperature (V_{SL} longitudinal wave velocity in the solid; V_{LL} longitudinal wave velocity in the liquid; V_{SS} the shear waves velocity in the solid).

Figure 2 shows the evolution of the elastic velocities versus temperature, up to the melting point. The uncertainties of T, V_{SL} and V_{SS} are respectively estimated to be lower than 0.6%, 0.7% and 3%.

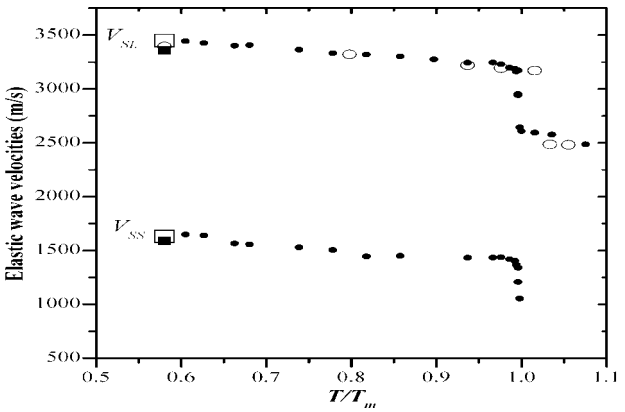

Figure 2: Elastic velocities of Tin versus temperature up to the melting point, (●) this study by laser-ultrasonics, (❍) Nakano et al [9] by a similar method, (☐) this study at room temperature by a contact ultrasonic method (■) and calculated from literature [13] for isotropic Tin.

The values of longitudinal and shear velocities both decrease with temperature. Assuming a linear behavior of the elastic velocities, the following equations are obtained for Tin:

$$V_{SL}(T) = 3810 - 604.7\,(T/T_m) \qquad (1)$$

$$V_{SS}(T) = 1941 - 542.7\,(T/T_m) \qquad (2)$$

These values of V_{SL} are in good agreement with the results of Nakano et al [9], even if a difference exits around the melting point because of a shift in the measurement of the temperature (in the quartz cell and not in the sample). Measurements of velocities using an ultrasonic contact method [11] (with piezoelectric transducers) and values from literature for isotropic Tin [13] are also indicated at room temperature.

The shear-wave attenuation, due to the approach of the melting, leads to a crucial difficulty of the shear velocity measurement. Indeed, the shear attenuation is much greater than the longitudinal one [10].

The present work is particularly relevant because of the simultaneous determination of $V_{SL}(T)$ and $V_{SS}(T)$ in the solid phase.

As a scientific anecdote, we precise that the Lindemann constant (critical fraction of the lattice spacing reached at the melting) can be considered linked to the melting mechanism and determined by the drastic fall of the shear modes [14, 15, 16, 17].

BEHAVIOR OF THE ELASTIC MODULI VERSUS TEMPERATURE

The elastic moduli are deduced from the previous results by using the well known following equations given for an isotropic solid.

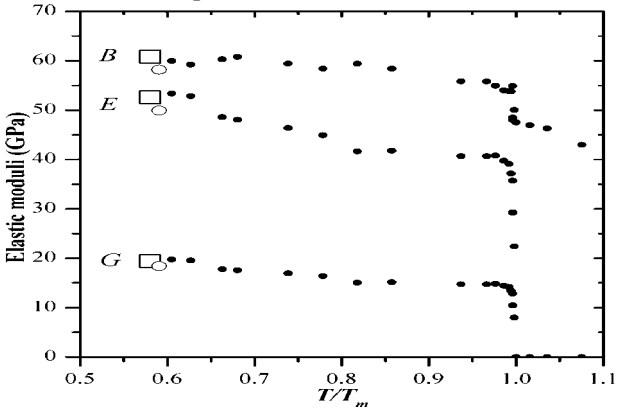

Figure 3: Elastic moduli (E, G, B) versus T/T_m for the Tin sample, (●) this study by laser-ultrasonics, (☐) this study at room temperature by a contact ultrasonic method and (❍) literature data [13] at room temperature.

The density is considered as an input datum, taking into account the thermal expansion coefficient of the material for the solid state. The density at room temperature $T_0 = 293\,K$ is $\rho_0 = 7282\ kg.m^{-3}$. Furthermore, the density of the liquid Tin is given by a linear function of the temperature [18].

First approach: linear dependence of the elastic moduli versus temperature

Figure 4 shows the elastic moduli according to the temperature. The total uncertainty of moduli is found by combining the uncertainty of each source. Estimating the uncertainty of ρ to 0.13%, the uncertainties of B, E and G are calculated to 4.8%, 4.9% and 6.1%.

At room temperature our results using an ultrasonic contact method (with piezoelectric transducers) and values of literature data [13] are also indicated. In a first approach, the linear dependence (over the temperature range from 305 K to 501 K) of the elastic moduli, leads to:

$$B(T) = 70.5 - 15.6(T/T_m) \quad (3)$$

$$E(T) = 71.0 - 32.4(T/T_m) \quad (4)$$

$$G(T) = 26.5 - 12.6(T/T_m) \quad (5)$$

Linear behavior of G(T) is validated until 501 K. Acoustic signals exhibit an attenuation rise a few Kelvin before T_m. G(T) (or V_{SS}) falls to zero in a few Kelvin around T_m. In order to take into account this phenomenon, a more accurate description of $G(T)$ up to the melting point is proposed in the next section.

Nonlinear description of the shear modulus

With the aim of describing a more realistic evolution law G(T) (especially near the absolute temperature), a Varshni function is adapted [19, 20]. Nevertheless, if this approach corresponds to a suitable fit of the material behavior, this kind of modeling is entirely dependent of some mathematical parameters. Depending on the assumption of linear or nonlinear behavior of $G(T)$ at low temperature range, approaching the absolute temperature, the $G(T)$ modeling combines some Varshni functions [21].

Another work [22] extends these previous studies by introducing a condition compelling the shear modulus to zero at the melting point. In these cases, the $G(T)$ modeling uses fit parameters and is not matched to the behavior of metals at low temperatures.

For pure polycrystal metals, a $G(T)$ modeling is proposed made of a continuous analytical expression over a large temperature range, from the absolute temperature to the melting point. In order to describe the drastic fall of $G(T)$ at T_m a Varshni function [21] is divided by the function $\Im(T/T_m)$ defined for $T/T_m \in [0;1+\varepsilon]$ leading to:

$$G(T) = \left(G_0 - \frac{s}{e^{T_{cal}/T} - 1} \right) \Big/ \Im(T/T_m) \quad (6)$$

where s (GPa) and T_{cal} (K) are constants [21] and G_0 is the shear modulus at 0 K. In the temperature range of our study, $T_{cal} << T$, then a linear function substitutes the Varshni function leading to:

$$T/T_m \in [0;1+\varepsilon] \quad G(T) = \frac{G_0 [1 - a\ (T/T_m)]}{\Im(T/T_m)} \quad (7)$$

with the following definitions:

- $\Im(T/T_m) = 1 + \exp\left[\frac{T/T_m - 1}{\varepsilon \left\{ 1 - \frac{T}{T_m(1+\varepsilon)} \right\}} \right]$
- $a = sT_m / G_0 T_{cal}$. The slope of the linear behavior of $G(T)$ is aG_0/T_m far from the melting point (a is a positive factor).
- ε is a shift value around $T/T_m = 1$ yielding to: $G(T) = 0$ for $T = T_m(1+\varepsilon)$

This model is entirely characterized by the slope parameter a and the parameter ε.

According to the intrinsic characteristics of the material, the model [Eq. (7)] becomes a full consistent model to describe the $G(T)$ evolution versus the complete temperature range of our study.

Experimental validation: shear modulus of Tin between 300 K and T_m (505 K)

The model [Eq. (7)] can be applied to the present study of Tin. Nevertheless, the temperature range of this model is reduced this way: $T/T_m \in [286/T_m;1+\varepsilon]$. Indeed, white Tin exhibits a solid-solid phase transition at 286 K, which is out of the temperature range of this study. The modeling predicts a linear behavior of $G(T)$ at low temperature. It is therefore impossible to predict the behavior of G below 286 K because the corresponding phase is not studied in this work.

Figure 4 shows the comparison between some experimental data of the shear modulus function of temperature and the modeling.

The value G_0 cannot be exploited because the phase transition of Tin at 286 K is not taken into account in the modeling. More generally speaking, the model is given for $T/T_m \in [286/T_m;1+\varepsilon]$.

The fall of $G(T)$ around $T/T_m = 1$ is described rather well by the modeling. The results show $\varepsilon = 0.05$. It can be noted that the drastic fall of $G(T)$ around the melting point can be described by a function falling down to zero in a weak temperature range.

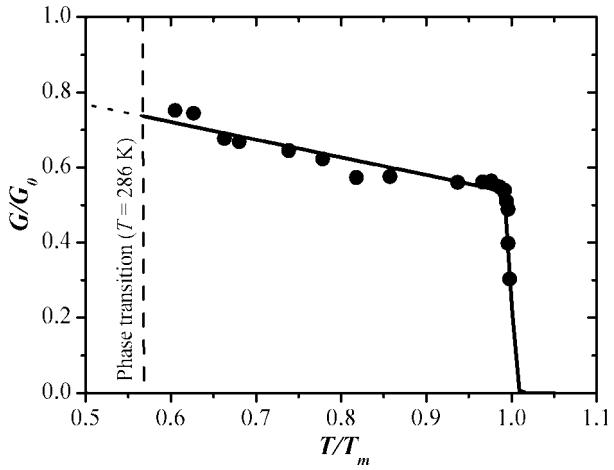

Figure 4: Evolution of the shear modulus of Tin versus T/T_m up to the melting point: (●) the present experimental study and (—) the model.

Conclusion

This paper shows the high potential of Laser-Ultrasonics to study the solid to liquid transition. Applied to Tin, this work succeeded in the determination of both longitudinal and shear wave velocities and the elastic moduli from ambient to the liquid. An analytical and continuous description of G(T) is proposed and validated by the experimental data. The drastic fall of G(T) at T_m is then described.

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