Ultrasonic monitoring of float zones in germanium bars.

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RESUME

L'article fournit des résultats préliminaires caractérisant la mesure ultrasonique de zones de flottaison. Le prototype ci-décrit fournit en ligne des estimations de la position, de l'épaisseur, et de la plénitude de la zone de flottaison. Les résultats obtenus à ce jour indiquent que l'erreur de position est caractérisée par un écart-type inférieur au millimètre, et qu'une bonne indication de la plénitude de la zone de flottaison peut être obtenue.

SUMMARY

This paper gives preliminary results obtained with a prototype float zone ultrasonic measurement system. This prototype produces on-line estimates for the float zone position thickness, and completeness. Results obtained to date indicate a standard deviation of less than 1 mm for the float zone position accuracy and an accurate indication of the float zone completeness.

INTRODUCTION.

Future ultrapure monocrystalline germanium may well be produced in the microgravity environment offered by space.

Reasons for this include the fact that, in order to build the increasingly sensitive detectors that are required by scientific, military, and industrial application, germanium crystals of a diameter larger and purity better than what is now produced on earth will be required. While the necessary purity appears to be achievable by a containerless zone-refining process called float zoning, in which a liquid zone moves through a vertical bar of the material to be refined, without any contact with a crucible, the application of the process to germanium bars of sufficient diameter appears difficult in earth gravity, in which the hydrostatic pressure of the liquid germanium exceeds the retaining power of the germanium surface tension.

Automatic process control, which is particularly important for space applications, includes in this case the control of the float zone geometry. The latter, which is a critical parameter of the float zoning process [1], comprises the float zone position, thickness, completeness, and liquid solid interface shapes.

While the physics of the propagation of ultrasound along anisotropic germanium rods, as well as the interaction of ultrasound with the liquid phase of germanium have been discussed in a recent paper [2], the present paper gives preliminary results on signal processing aspects of the work. A more in-depth examination of signal processing for float zone detection will be published elsewhere.

Specifically, the following aspects are examined:

- Choice of operating mode and frequency;
- Dispersive propagation and matched filtering;
- Self-calibrating algorithms for float-zone position and thickness extraction;
- Float zone completeness estimation.

EXPERIMENTAL SET-UP.

In the float-zone processing, a narrow zone of liquid germanium (melting point: approx. 937.4°C) is slowly moved along a vertical bar of the single crystal germanium to be purified. The float zone is created and moved by the effect of the heat supplied to the bar from a high power radio-frequency coil (figure 1).

Ultrasonic float-zone sensing is accomplished by means of two transducers, which are connected via commercial and custom-made ultrasonic hardware to the data analysis subsystem. The latter consists of a Data 6000A Data Precision signal analysis computer controlled by a Compaq 286 computer, and provides on-line information on the float zone position and completeness. The germanium bar extremities are cooled in order to stabilize the float zone and to protect the ultrasonic transducers against excessive temperatures. In addition, to avoid oxidation of the germanium, the bar is placed in a nitrogen gas flow.

The first results obtained with this set-up were discussed in some detail in [2], together with the propagation of longitudinal and transverse waves in <100> germanium bars, as well as their

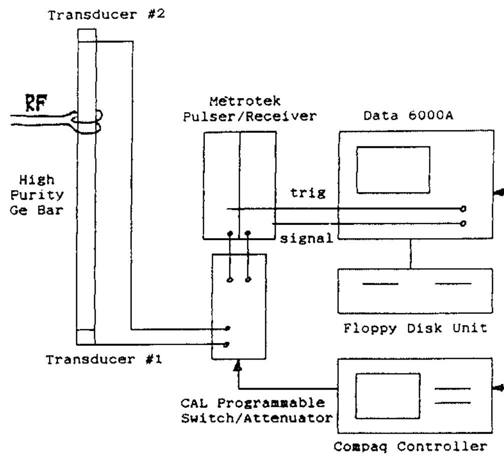

Figure 1. Flow chart of the experimental set up.

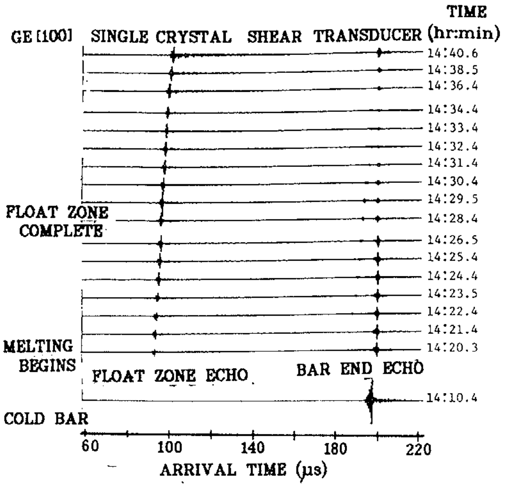

Figure 2. The reflected signals from the solid-liquid interface and the end of a 35cm long, 1cm diameter, <100> orientation, germanium rod with a 5 MHz, 0.5" diameter shear wave transducer.

reflection at solid-liquid interfaces. Additional information is contained in [3-6].

Some of the conclusions pertinent to the present situation are summarized thus:

- **Mode.** The parameters of the elastic anisotropy of <100> germanium together with mode conversion effects result in the propagation of the longitudinal waves being far more dispersive than that of shear waves. This fact is illustrated by the traces marked "cold bar" in figures 2 and 3, which represent the bar and echoes obtained by a shear wave and a longitudinal wave transducer respectively, operating at comparable wavelengths. Most of the subsequent work was thus performed using shear wave transducers.
- **Frequency.** A large operating bandwidth is desirable from the resolution viewpoint. Very large bandwidth ($Q \leq 1$) transducers were selected with a top frequency of the order of 5 MHz, limited by what was commercially available rather than by the attenuation in <100> germanium, which is quite low at these frequencies.
- **Transmission and reflection at solid-liquid interfaces.** When an ideal shear wave strikes a solid-liquid boundary ar normal incidence, it is totally reflected as a shear wave. However, as the angle of incidence increases, mode conversion takes place, and a portion of the incident energy is transmitted into the liquid as a longitudinal wave, while some of that energy is also reflected as a longitudinal wave. Similarly, liquid-borne longitudinal ultrasound can be partially converted into a shear wave in the solid for angles of incidence different from normal. The effects of this phenomenon are illustrated in figure 2, where it is seen that, as the float zone develops, its echo increases relative to the bar end echo. However, even for a complete float zone (that is, when the bar has melted through), a residual bar end echo can be observed, due to the mode conversion described above. For reference, selected acoustic properties of germanium have been collected in Table 1.

TEMP (K°)	DENSITY (Kg/m³)	V_L (m/s)	V_S (m/s)
298	5323	4826	3582
573	5297	4761	3527
973	5257	4617	3436
1155	5240	4552	3372
1209	5235	4529	3351
LIQUID	5510	2710	

Table 1. Acoustic properties of <100> germanium at various temperatures.

SHEAR WAVE DISPERSION.

Shear wave dispersion, even though much smaller than longitudinal wave dispersion, nonetheless adversely affects signal-to-background ratio and time-of-arrival estimation. This is illustrated in figure 4, which compares shear wave impulses received in the "pitch-catch" mode (i.e. after one way propagation) in various lengths of 1 cm diameter germanium <100> bars.

The dispersion is seen to increase with the distance, from a near perfect replica of the transmitted 5 MHz bandwidth pulse in the shortest (3.52 cm) distance to an impulse elongated by several periods for the longest (35.0 cm) distance travelled.

As an analytical description for this dispersion does not appear to be available [6], or easy to arrive at, this difficulty is solved pragmatically by matched-filtering the received signals with observed (rather than computed) replicas. The replica can be obtained in the pitch-catch mode, and is approximately appropriate for echo returns originating in the central portion of the bar, as would be the case for float zone echoes.

The pulse compression effect obtained by the autocorrelation of arrival (d) figure 4 is illustrated in figure 5. All the results described in the sequel are obtained with a matched filter as the first stage of processing. In this manner, a time-of-arrival resolution of the order of - or better than - the bandwidth of the received impulse can be obtained even though the raw signal received may be dispersed over many times this duration. Likewise, the amplitude of the original (undispersed) pulse tends to be restored.

FLOAT ZONE POSITION AND THICKNESS ESTIMATION.

The estimation of the float zone position and thickness is made difficult by two circumstances:

(i) The non-planarity of the liquid-solid interface, which makes the position definition ambiguous. In what follows, we are mainly concerned with the position of the melt closest to the bar ends, which generally corresponds to the melt position on the outside of the bar.

(ii) The shear wave speed in the <100> germanium varies with the temperature, especially in the vicinity of the melting point: for example, Table 1 shows a speed variation of some -20cm/s°K close to room temperature, reaching some -40/s°K in the vicinity of the melting point. Significant errors in distance estimates obtained on the basis of echo time delay would result from relatively small errors in the mean temperature estimation.

This difficulty can be circumvented as follows under the condition that the **average** sound speeds in the top and bottom solid (i.e. non-melted) sections of the germanium bar are equal. First, the propagation time of the longitudinal waves though the liquid germanium, t_f is estimated as:

$$t_F = t_{PC} - (t_T + t_B) / 2$$

where t_{PC}, t_T, and t_B respectively denote the pitch-catch, top and bottom transducer float zone echo time delays. Next, as the temperature of the melt is kept very close to the melting point, the melt thickness l_F is found with a good approximation as:

$$l_F = V_F t_F,$$

where V_F denotes the longitudinal wave speed in liquid germanium at its melting point (~ 2710 m/s, see Table 1). Finally, the top l_T and bottom length l_B of solid germanium are found by a proportionality rule, for example:

$$l_T = (L - l_F)\ t_T / (t_T + t_B),$$

where L is the bar length. Note that no numerical value is needed for the shear wave velocity in the solid part of the bar.

Figure 6, based on the on-line output of the prototype float zone sensing system, illustrates the performance of the above algorithms. The control measurements (represented by + and squares) are derived from the approximate visual estimates of the float zone boundaries, subject to an estimated error of the order of 1 mm or more. The figure demonstrates a standard error of a fraction of a mm in the ultrasonic measurement.

FLOAT ZONE COMPLETENESS.

When germanium is refined by float-zoning, it is particularly important that the float zone be complete, i.e. that the float zone extend over the whole bar thickness.

This condition can be sensed ultrasonically in at least two manners, as explained below.

- **Completeness index.** As a partial float zone develops, its echo received at each bar end increases with the radial extent of the float zone, whereas the pitch-catch signal decreases. It is thus natural to take a function of the ratio of the echo energy over the pitch-catch signal energy as an indicator of the melt completeness. Further, interfaces closer to a plane normal to the bar axis will increase this ratio, as reflection is more efficient and mode conversion is less efficient for smaller incidence angles, as discussed above. An example of the application of such an index is given in Figure 7. The correlation between the completeness index, and the control measurement

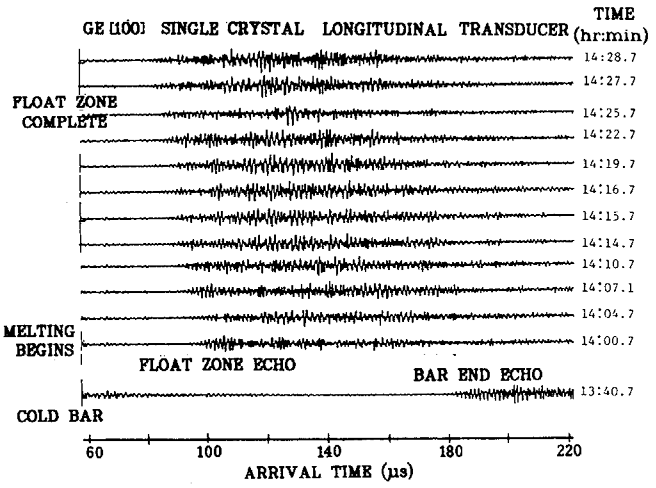

Figure 3. The reflected signals from the solid-liquid interface and the end of a 35cm long, 1cm diameter, <100> orientation, germanium rod with a 2.25 MHz, 0.25" longitudinal wave transducer.

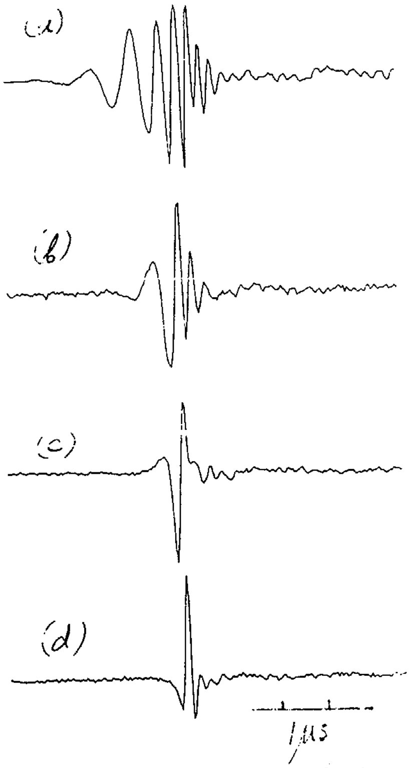

Figure 4. Pitch-catch impulses, transmitted and received by a shear wave 5MHz, 0.5" diameter transducer, through a 1cm diameter <100> orientation germanium bar of length (a) 35.0cm, (b) 16.02cm, (c) 6.86cm and (d) 3.52cm.

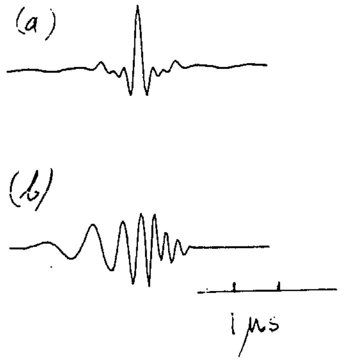

Figure 5. Example of the pulse compression obtained by matched filtering the received signal with its replica. (a) Matched filter (autocorrelation), (b) Replica.

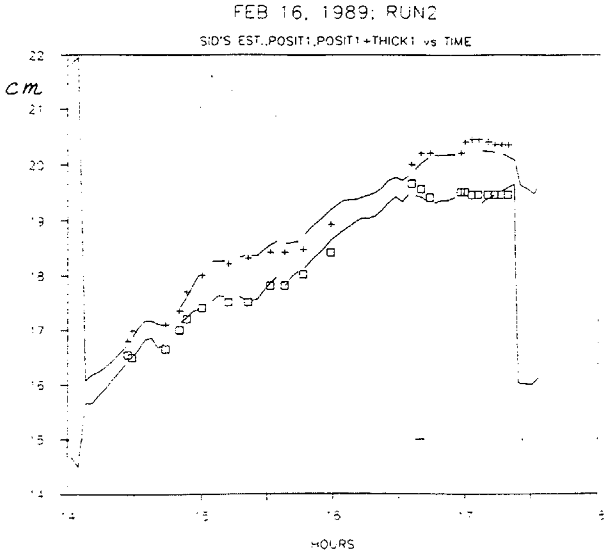

Figure 6. Ultrasonic estimates (-----) of float zone position with approximate control measurements (+ square).

(obtained by lightly twisting one bar end during melting) is seen to be good.

Time of arrival. Another sensitive indicator of the presence of a fully developed float zone is the time of arrival of the pitch-catch signal. Figure 8 shows this parameter for the same run as Figure 7 together with the control measurement. Here again, the correlation is seen to be good.

CONCLUSION.

The paper has given preliminary results of on-line float zone measurements in monocrystalline germanium rods. The examples show the feasibility of ultrasonic float zone position measurements with a standard deviation of 1 mm or less, and of reliable float zone completeness estimation by means of ultrasonic measurements.

ACKNOWLEDGEMENTS.

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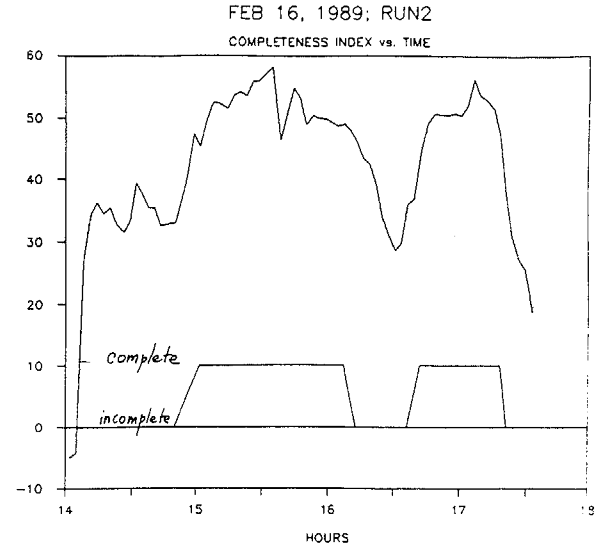

Figure 7. Ultrasonic estimate of float zone completeness index (top curve) with a control measurement (bottom curve).

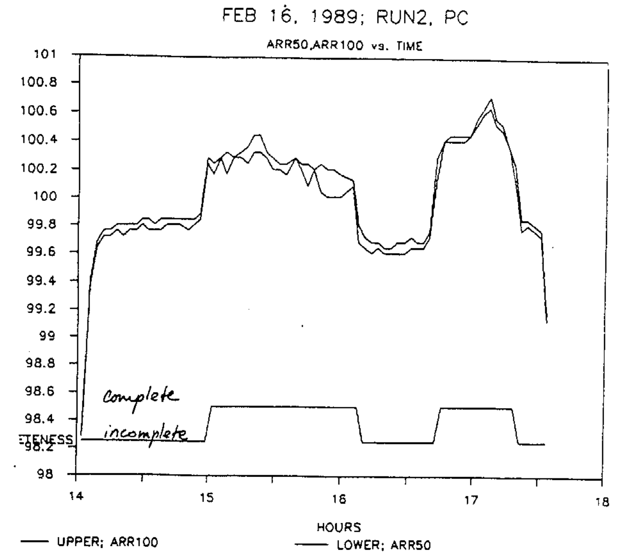

Figure 8. Two pitch catch time of arrival measurements as a completeness estimate.

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