20. *Study on the Elasticity near the Melting Point. Part I. Nature of Dilatational Wave.*

By Daisuke SHIMOZURU,

Earthquake Research Institute.
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Introduction.

Recent discussions on the constitution of the earth's interior, especially those on the state of the earth's core, have brought out the scantiness of relevant experiments. Particularly, the elastic behavior of rocks in the neighborhood of the melting point is one of the most urgent questions. But, our knowledge concerning this is very poor because of the lack of definite theory of fusion and experimental facts.

Theory of fusion has been advocated by many authorities such as Brillouin[1], Born[2], Fürth[3], Lindemann[4], Lennard-Jones and Devonshire[5], Kirkwood[6]. But there exists no exact theory based upon the concept of the statistical mechanics.

On the other hand, there are a few experimental data in the literature on the behavior of the elastic moduli of solids in the neighborhood of their melting point.

Birch and Bancroft[7] reported the temperature variations of the rigidity modulus of polycrystalline Al up to 650°C i.e. 13° below the melting point.

Siegel and Cummerov[8] compared the above data with other data[9],[10],[11] such as Young's modulus of a single crystal of Pb at temperatures up to 325°C, 2° below the melting point. He stated that these data do

1) L. BRILLOUIN, *Phys. Rev.*, **54** (1938).
2) M. BORN, *Jour. Chem. Phys.*, **7** (1939).
3) R. FÜRTH, *Proc. Cambr. Phil. Soc.*, **37** (1941).
4) F. A. LINDEMANN, *Phys. Zeits.*, **11** (1910).
5) LENNARD-JONES and DEVONSHIRE, *Proc. Roy. Soc. A*, **169**, **170** (1939).
6) J. G. KIRKWOOD, *Jour. Chem. Phys.*, **9** (1941).
7) BIRCH and BANCROFT, *Jour. Chem. Phys.*, **8** (1940).
8) SIEGEL and CUMMEROV, *Jour. Chem. Phys.*, **8** (1940).
9) QUIMBY and SIEGEL, *Phys. Rev.*, **54** (1938).
10) F. C. ROSE, *Phys. Rev.*, **49** (1936).
11) M. DURAND, *Phys. Rev.*, **50** (1936).

not agree with Sutherland's curve[12] quoted by Brillouin, in which the melting temperature was assumed to be that at which the rigidity vanishes.

Hunter and Siegel[13] measured the principal elastic moduli of single crystal rods of NaCl over the temperature range from 20°C to 804°C, that is, the melting point. According to them, the shear constants C_{44} and C_{11}-C_{12} decrease almost linearly with temperature, but they do not become zero at the melting point.

Kornfeld[14] has investigated the propagation of transverse vibrations in rosin for different temperatures between 20°C and 70°C. Using Maxwell's theory, Kornfeld obtained for rigidity modulus a value of the order of 10^{10} dynes/cm^2 at 20°C, which decreased by a factor of 10 as the temperature was raised to 70°C. In the case of stearin the approach to the melting point is accompanied by a catastrophic drop in rigidity modulus.

Bordoni and Nuovo[15] measured the frequency of fundamental resonance of a small cylinder by an electro-acoustic method for five metals at elevated temperatures up to the vicinity of the fusion point, and the logarithmic variation with temperature of the velocity of propagation was compared with the behavior of cubic dilatation.

Pochapsky[16],[17] measured the variation of the velocity of propagation of a short train of ultra high frequency compressional acoustic waves in liquid sodium with temperature between 371°K and 545°K. He calculated the temperature coefficient of velocity and adiabatic compressibility between the aforementioned temperatures. He stated that no change in the temperature coefficient of velocity was detected in the neighborhood of the freezing point.

These experimental investigations give, however, no definite figure of the variation of elasticity in the neighborhood of the melting point and its relation to the variation of density, specific heat or other physical quantities. As for the field of seismology or the research on the constitution of the earth's interior, it is very interesting to know how the elasticity or the velocity and amplitude of elastic waves change when

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12) SUTHERLAND, *Phil. Mag.*, **32** (1891).
13) HUNTER and SIEGEL, *Phys. Rev.*, **61** (1942).
14) M. O. KORNFELD, *C. R. Ac. Sci. U.R.S.S.*, **36** (1942).
15) BORDONI and NUOVO, *Ric. Sci.*, **23**, **4** (1953).
16) POCHAPSKY and QUIMBY, *Phys. Rev.*, **79** (1950).
17) T. E. POCHAPSKY, *Phys. Rev.*, **84** (1951).

the first phase transition from solid to liquid occurs.

The ultimate object of the present study is the measurement of the variation of the elastic moduli of rocks at elevated temperatures. As is well known, rocks are in general composed of glass and minerals, which in turn are solid solutions. Accordingly, phenomena of fusion of rocks is exceedingly complex. Therefore, it will be the proper course to take at first the chemical elements or binary or ternary eutectic alloy as the test materials.

As a preliminarly experiment, the present paper describes a series of experiments on the temperature variation of the velocity of ultrasonic compressional wave in Wood's metal for the range between 10°C and 90°C.

Apparatus.

The ultrasonic apparatus has been constructed for the measurement of the velocity of elastic wave in rocks. The apparatus is similar to that which has been described by Hughes *et al.*[18] The basic instrument is composed of an electronic apparatus for generating pulses and that for measuring the transmission time (Fig. 1).

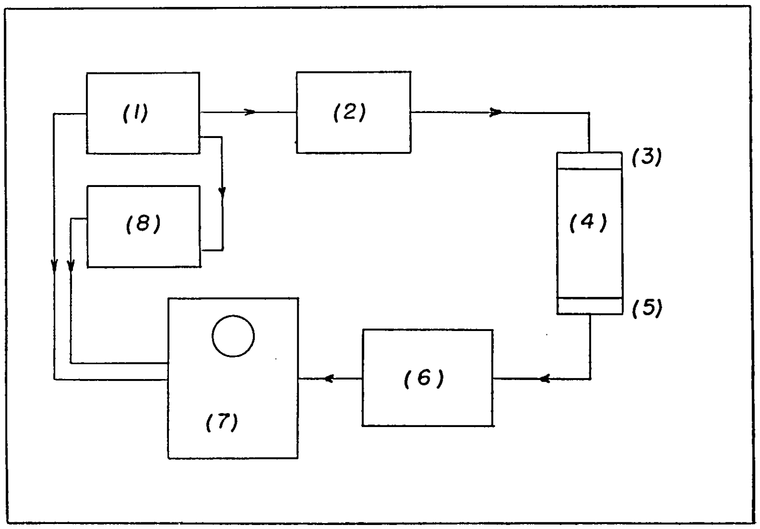

Fig. 1. Functional Diagram of the Apparatus. (1) Main trigger generator. (2) Pulse generator. (3) Driving crystal. (4) Sample. (5) Receiving crystal. (6) Video amplifier. (7) Cathode ray oscilloscope. (8) Time mark circuit.

18) Hughes *et al.*, *Phys. Rev.*, **75** (1949), 1552–1556.

This pulse generator provides pulses with a rise time of about 0.2 microsecond. These sharp pulses are impressed on a 1 MC crystal which generates very sharp elastic pulse waves in the specimen. These waves are received at the opposite end of the specimen by a similar crystal. Although, X-cut quartz crystals are common as a driver and a receiver, 1 MC barium-titanate of 25 mm diameter was used in the present experiment. The output of the receiving crystal goes through a video-amplifier to a cathode ray oscilloscope. It is equipped with sweeps of two durations 100 and 10 microseconds. Time marks are indicated on each sweep at the interval of 1/10 sweep. Two kinds of sweeps are ultilized as follows. When the transmitted pulse appears on the screen of the oscilloscope, say, between 20 and 30 microseconds, units and fractions of the transmission time in microsecond are measured by switching the interval on the 10 microsecond sweep which has the time mark of each 1 microsecond.

Recently, Hughes and others found in their study of the velocity measurements of elastic waves that a plane dilatational pulse travelling parallel to the axis of a circular cylinder gives rise at the cylindrical boundary to rotational pulses which leave the boundary at the critical angle given by

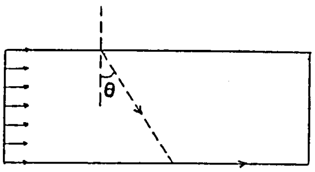

Fig. 2.

$$\sin\theta = \frac{V_R}{V_D},$$

where V_R is the velocity of rotational waves and V_D the velocity of dilatational waves. This rotational pulse reaches the opposite boundary of the cylinder and gives rise to a dilatational pulse travelling parallel to the axis. Thus, if the cylinder has sufficient length, the original dilatational pulse may be followed by many later phases. Transmission time of these pulses is easily shown to be

$$t_{mn} = m\frac{L}{V_D} + nR\frac{[V_D^2 - V_R^2]^{1/2}}{V_D V_R}, \qquad mL \geqslant nR\tan\theta,$$

where m : odd integers,
n : integers including zero,
L : length of the cylinder,
R : diameter of the cylinder.

For a metallic cylinder, many later phases have been observed. With rocks, however, number of later phases is rather few. In this case, the first and the second phases must be used to measure the velocity of dilatational and rotational waves. Fig. 3 shows the example of the screen pattern of the pulse transmitted respectively through an aluminum rod and a sandstone rod.

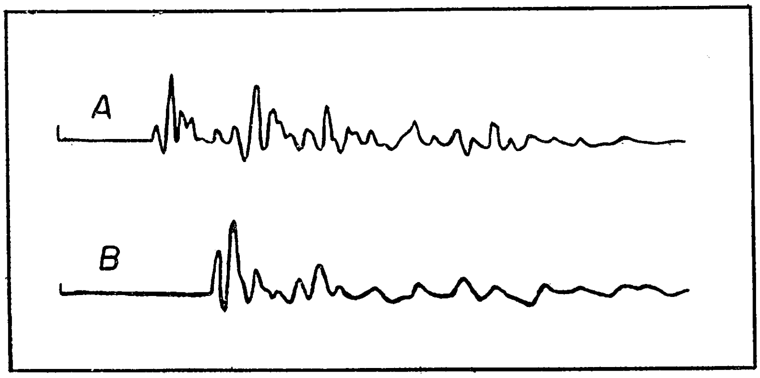

Fig. 3. Examples of the screen pattern of received pulse.
(A) Aluminum. (B) Sandstone.

Specimen Material.

As a preliminary experiment, a kind of low melting point alloy, generally called "Wood's Metal", was used. It is composed of cadmium, tin, lead and bismuth and has a eutectic point at 73°C. At this temperature, the first phase transition occurs between solid phase and liquid phase. The density is about 9.81 gr./cm^3 at 15°C. Wood's metal was casted in a thin brass pipe of 5 cm in length and 2.5 cm in diameter. Generally, a substance which melts at low temperature has a large coefficient of thermal expansion. Moreover, sudden volume expansion occurs at this fusion point. From these reasons the pipe has a cut along the generating line so as not to compress the containing sample when the latter is melted. To keep the specimen at the desired temperature, a thermostat has been used.

Experimental Results.

As the barium-titanate has a Curie point at 120°C, the measuring temperature was restricted below 90°C. As the velocity of sound in brass is much larger than that in Wood's alloy, the first arriving pulse ought to have the direct path along the environing brass pipe.

However, the amplitude of such pulse is very small compared with that of pulse propagated through the specimen material in question.

Therefore, below 60°C when the gain of amplifier is not high, the phase through brass is not distinguishable. Fig. 4 shows one of wave patterns obtained in the present experiment. Ordinate indicates the temperature at which the velocity was measured. In this figure, A-phase is the arrival of pulse propagated along the container pipe. B-phase is the

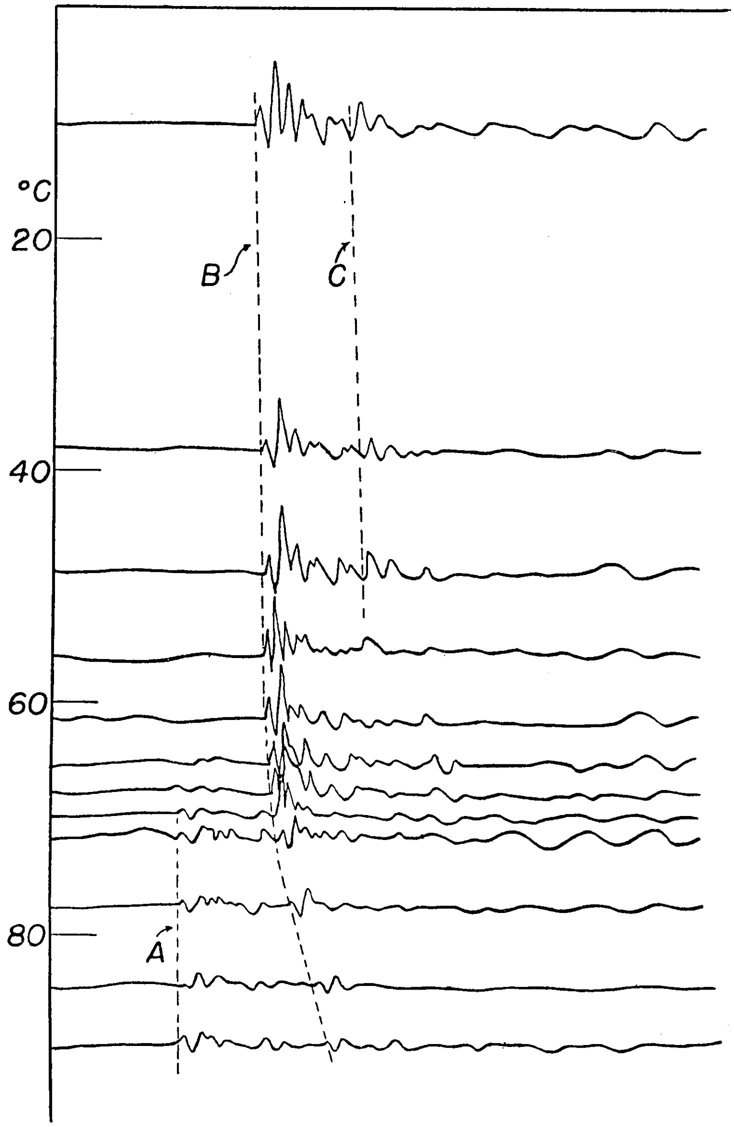

Fig. 4. Wave patterns as the function of temperature.

arrival of dilatational wave transmitted directly through the specimen material. C-phase is the second arrival of dilatational wave which was partly transmitted as rotational wave. Table I shows the tabulated values of the velocity of dilatational wave for various temperatures as well as the amplitudes of B-phase in arbitrary scale. At room temperature, dilatational pulse gives rise easily to rotational pulse at the boundary. But as the temperature is raised, in consequence of the increase of solid viscosity of the specimen, the dilatational pulse shows difficulty in giving rise to rotational pulse. Therefore, C-phase could not be detected distinctly in this experiment. This is the reason why we confine our study only to the nature of dilatational wave. Velocity of dilatational wave or amplitude versus temperature are plotted in Fig. 5 and Fig. 6 respectively.

Table I.

Temp. °C	Velocity m/sec.	Ampl.
10.0	2785	21.0
38.0	2665	21.0
48.5	2650	20.8
55.7	2620	19.0
61.5	2585	17.8
65.9	2556	16.2
67.9	2520	15.5
69.8	2510	14.8
73.5	2345	13.7
77.7	2320	9.8
84.9	2190	7.0
90.1	1953	3.5

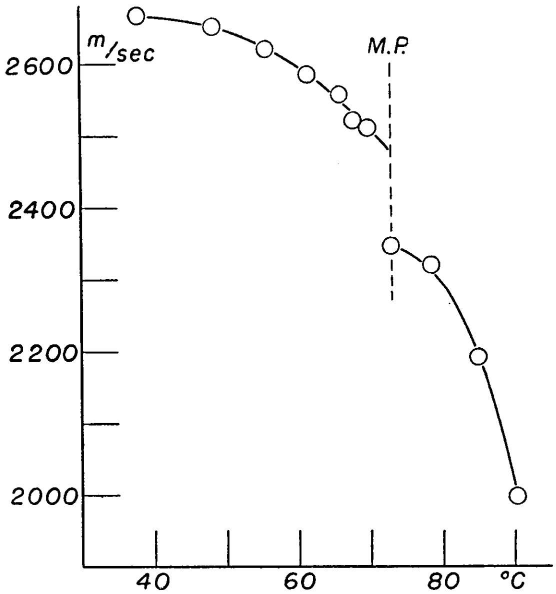

Fig. 5. Velocity of dilatational wave vs. temperature. M.P. stands for melting point.

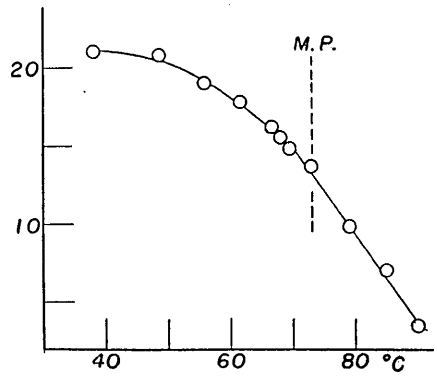

Fig. 6. Amplitude of dilatational wave vs. temperature.

Discussions.

The velocity of dilatational wave decreases continuously as the temperature is raised up to the melting point, where the velocity shows a discontinuous decrease. From the melting point up to 90°C, the velocity decreases more rapidly than at the solid state. The discontinuous jump in the velocity from 2510 m/sec. to 2345 m/sec. at the fusion point is the most conspicuous feature. As we must treat the test material as a visco-elastic medium, the velocity of dilatational wave propagated through it is given by the complex velocity. In order to calculate the complex moduli from this velocity, the density must be known at different temperatures. We could unfortunately find no data about the thermal expansion of Wood's metal nor the expansion coefficient of it. It is a fact however that Wood's metal shows a sudden remarkable volumic expansion by melting, so the density must undergo a sudden decrease. This decrease in the density must result in the increase of the propagation velocity, if the elastic modulus is unchanged. In reality, we have a very remarkable decrease in the propagation velocity of elastic waves. We can therefore conclude that the complex modulus must make a very sudden, remarkable decrease at the melting point more than enough to compensate the density decrease. Owing to the lack of numerical data of density variation, however, we can not, for the present, give any quantitative result. As to the separation of the real elastic moduli from the complex expression, frequency of the pulse wave must be changed, and we must also measure the velocity of rotational wave which, as cited above, is very difficult to measure at or near the melting point.

In order to measure the velocity of rotational wave, Y-cut quartz crystals are suitable for a driver or a receiver instead of X-cut quartz crystals.

Amplitude decrease is monotonous as the temperature is raised and no sudden decrease was detected in the neighborhood of the fusion point. It is to be noticed that this decay of amplitude is not wholly due to the increase of solid viscosity, but partly due to the effect of decrease in the wave energy transmitted from the crystal to the specimen or from the specimen to the crystal.

The cause of the velocity discontinuity of the first order at the melting point, which was detected in the present experiment with Wood's metal, will be studied in near future using more simple material such as alkali metal, of which the density variation or other physical quantities are well known as the function of temperatures.

Acknowledgments.

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20. 融解点附近の弾性の研究
其の一　縦波の性質

地震研究所　下 鶴 大 輔

最近の地球内部構造論に関しては，実験事実の貧困さが，その理論的発展を阻害している．特に，物質が融けた時に，その弾性的性質が，どうなるかという問題は，未だ殆んど手をつけられていない．融解点近傍までは，固有振動法に依っていくつかの研究があるが，この方法では，更に融けて液体になった後までの事が実験出来ない．著者は，岩石物性研究の超音波装置を試作し，先づその第一の実験として，岩石の融解点における弾性の研究をとり上げた．此の論文では，その研究の第一段階として，ウッド金と称する低融点合金を試料とした．実験装置の概略は，電気的なパルスをチタン酸バリウムの振動子に加え，それより，弾性縦波として試料中を通し，他端で同じく，チタン酸バリウムで，通過パルスを受けとり増巾して，ブラウン管オッシロスコープにて，その伝播時間を測定し，試料中を通る弾性縦波の速度を算出した．試料部分を恒温槽に入れ，測定温度を次第に変えてその速度の変化をしらべた．測定温度は 10°C より 90°C までであって，ウッド金の場合，73°C に共晶点(融解点)がある．此の実験の結果，次の事がわかった．即ち温度が上ると縦波の速度は次第に減少し，融解点において，2510 m/sec から 2345 m/sec へ不連続的に減少し，更に温度が高くなると，速度の減少の割合は，固体のときよりも更に著しくなる．此の様に融解点に於いて，縦波の速度に第一次の不連続が見られる事は非常に興味深い．粘弾性体として取扱わなくてはならないから速度は複素数の形で表現され，複素弾性常数を計算する為めには温度による密度変化が判らなくてはならない．然るにウッド金の場合，密度変化が不明なので，複素弾性常数を計算する事が出来なかった．故に，此の実験で得られた融解点における縦波速度の不連続の機巧を説明する為めには，是非，簡単な化学元素について実験を行う必要がある．或る種のアルカリ金属に就いては実験中であって，その結果は次の機会に述べる事にする．
