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SOUND SPEED MEASUREMENTS IN LIQUID LEAD AT HIGH TEMPERATURE AND PRESSURE*

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Abstract

Recently sound speed measurements in high temperature liquid lead have been made over a wide density range. Such measurements may be combined with other thermophysical properties measured with the isobaric expansion experiment (IEX) to yield several additional thermodynamic quantities. Results of calculations based on the sound speed measurements are presented, and their impact on liquid metal phenomenology is discussed.

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1. Introduction

The measurement of thermophysical properties of materials at very high temperature and high pressure under laboratory conditions can be difficult. Such high temperatures and pressures are required for the study of liquid metals because of the location of the thermodynamic critical point.[1] The critical point is at low enough temperature and pressure to be studied with conventional static techniques for only a very few metals (Hg, Cs, Rb and K). Of these metals, Hg has been studied most extensively. The estimated location of the critical point of Pb is well above that of the above metals, making it impossible to study fluid Pb in the critical region statically. Other fluid metals such as W must be studied at even higher values of temperature and pressure.

Our technique for reaching high temperature, expanded liquid states in metals is resistive pulse heating and can be used both above and below the critical point. The metallic sample is resistively heated in an inert high pressure gas atmosphere over a time scale of about 10^{-4} s. Expansion takes place along an isobaric path, so our technique is called the isobaric expansion experiment (IEX). Details are given elsewhere.[2] During the course of an experiment, enthalpy, temperature, and specific volume are measured. From these quantities the specific heat at constant pressure and thermal expansion coefficient may be calculated. Such properties of liquid metals are needed for several applications, such as design and modeling of exploding wires, foils, and fuses, and for the development of fluid metal models.

Recently we have begun to measure sound speeds in liquid metal samples. The first step in such a measurement is to generate the sound wave. This is done by focusing a low energy laser pulse onto a spot on one side of the sample.[3] After the resulting stress wave (which is spherically diverging and degrading into a sound pulse) traverses the sample, it emerges on the side opposite the source and produces a small amount of surface motion. The detection of this surface motion is the most difficult part of the technique. We have discussed various attempts to detect this motion elsewhere. [3,4,5] Recently we have been able to make measurements of the sound speed of liquid lead over a wide density range ($5\ Mg/m^3 < \rho < 10.0\ Mg/m^3$).[6]

2. Experimental Details

The technique mentioned above for the measurement of sound speeds in highly expanded liquid metals has the virtue of being simple to perform. Details of the application of the method to collecting data in liquid lead, as well as limitations, are given here.

The source of the sound wave that propagates through our sample is a laser pulse focused onto a spot of about 10^{-4} m diameter on the sample surface. This not only results in a stress wave being driven into the sample, but also causes a disturbance in the surrounding gas which propagates outward from the source. This wave can clearly be seen in framing camera sequences made during a sound speed measurement. The gas disturbance moves around the sample and will eventually obscure the line-of-sight used for the sound speed measurement. The sound speed in the surrounding gas increases with increasing pressure, and the sound speed in expanded lead is decreasing with decreasing density. Higher confinement pressures are needed to reach stable states at low density, and so eventually the gas disturbance appears to arrive at the opposite side of the sample before the sound wave traveling through the sample. This occurs at an expansion of about $V/V_o = 2.2$, or $\rho = 5.15 \times 10^3$ kg/m^3. Techniques to overcome this problem are being studied, as we are attempting reach even lower density values.

3. Discussion

The sound speed C, in liquid lead at high pressures (0.1, 0.2, and 0.3 GPa) and high temperatures (up to ~5200 K) has been measured.[6] The results indicate that C decreases linearly with decreasing density, and is independent of temperature within our experimental uncertainty (± 7%). The best fit to our present data has been found to be

$$C = A\rho - B \quad , \qquad (1)$$

with 5×10^3 kg/m^3 $< \rho < 10 \times 10^3$ kg/m^3 A = 0.2084, and B = 354.0. The data along with the above fit are shown in Fig. 1.

Knowledge of C over this range of densities allows the calculation of several thermophysical properties in addition to those normally measured. Lead has been previously studied with the IEX, and in addition to P, V, and T data, it was found that [7]:

$$C_p = \left(\frac{\partial H}{\partial T}\right)_p = 157 \text{ J/kgK} \quad .$$

In this expression, T is temperature, P pressure, H the enthalpy, and C_p the specific heat at constant pressure. We found that C_p was approximately constant over the entire measurement range. We found that α, the thermal expansion coefficient, varied with temperature and pressure. The thermal expansion coefficient may be calculated from the normal IEX data from

$$\alpha = \frac{1}{V}\left(\frac{\partial V}{\partial T}\right)_p \quad .$$

With a knowledge of C we can then calculate Gruneisen's gamma,

$$\gamma_G = V\left(\frac{\partial P}{\partial E}\right)_V = \frac{\alpha C^2}{C_p} \quad . \qquad (2)$$

This calculation has been performed over the relevant density range, and the results are shown in Fig. 2.

The adiabatic bulk modulus is defined by

$$B_s = \rho\left(\frac{\partial P}{\partial \rho}\right)_s \; ; \qquad (3)$$

but

$$C^2 = \left(\frac{\partial P}{\partial \rho}\right)_s \quad ,$$

and so $B_s = \rho C^2$. Next, the fit to C has been substituted into this equation and we find

$$B_s = C_1\rho^3 - C_2\rho^2 + \rho C_3 \quad ,$$

where $C_1 = A^2$, $C_2 = AB$, and $C_3 = B^2$. The adiabatic compressibility is calculated from

$$K_s = \frac{1}{B_s} \quad . \qquad (4)$$

The specific heat at constant volume is given by

$$C_v = \left(\frac{\partial E}{\partial T}\right)_V \quad .$$

From straightforward manipulation,

$$C_v = \frac{C_p^{\ 2}}{C_p + \alpha^2 T C^2} \quad . \tag{5}$$

This expression, as the preceeding ones, contains C^2 and α^2 and so the experimental uncertainties in the calculated quantities are higher than those in the measured quantities. The values of C_v over our range of data have been calculated, and once C_v is known, we may find

$$\gamma = \frac{C_p}{C_v} \quad . \tag{6}$$

The calculated values of γ and C_v are shown in Fig. 3. Since C_p is constant, γ is proportional to the inverse of C_v as is clearly seen.

The isothermal bulk modulus B_t and compressibility K_t can be found from

$$B_t = \rho \left(\frac{\partial P}{\partial \rho} \right)_t = \frac{B_s}{\gamma} \tag{7}$$

and

$$K_t = \frac{1}{B_t} \quad .$$

The variations of these quantities with density are shown in Fig. 4.

From the bulk moduli we can find equations for isentropes and isotherms. Since

$$B_s = \rho\left(\frac{\partial P}{\partial \rho}\right)_s = C_1\rho^3 - C_2\rho^2 + \rho C_3 \quad ,$$

$$\left(\frac{\partial P}{\partial \rho}\right)_s = C_1\rho^2 - C_2\rho + C_3 \quad . \qquad (8)$$

The form of the isentropes may be found from Eq. (8) to within an integration constant. Similarly from B_T the form of the isotherms may be found

$$\left(\frac{\partial P}{\partial \rho}\right)_T = \frac{1}{\gamma}(C_1\rho^2 - C_2\rho + C_3) \quad ,$$

but in this case the integration is not analytic, since for liquid lead γ is a function of ρ.

From the above calculations, we can see the value of a sound speed measurement for finding thermophysical properties. Such measurements have further value for addressing basic physics issues.

The behavior of C_v over an extended density range can be estimated from

$$C_v = C_v^l + C_v^e \quad ,$$

where C_v^l is a fluid term, and C_v^e is the electronic contribution. Models have been used to approximately calculate both of these terms [8], but until now no real data existed to compare with. Predicted sound speeds in liquid lead based on these calculations agree only roughly with our data. We are able to find values of C_v with moderate precision based on our sound speed measurements, and this should provide a more accurate estimation of the contributions of each term to the overall value of C_v.

The thermodynamic critical point of a material has a number of unusual properties. For example, when the critical point is approached along some thermodynamic path, the sound speed reaches a local minimum value, and attenuation becomes very high. This has been observed to occur for mercury [9], and is related to the critical fluctuations. Work is ongoing to use our technique to locate the critical point of several metals, beginning with lead.

In liquid mercury Susuki, et.al., have also observed that at a density of 9 x 10^3 kg/m^3 there is a change in slope of the sound speed-density curve.[9] At this same point it is known that certain other properties of mercury (such as Knight shift) also undergo a change associated with electron localization.[1] We are attempting to determine if such a change occurs in liquid lead as we reach lower densities. If this behavior is seen in liquid lead it will provide a means of finding the density at which electron localization occurs in metals for which the temperatures are much too high for static measurements, such as NMR studies, to be performed.

Acknowledgement

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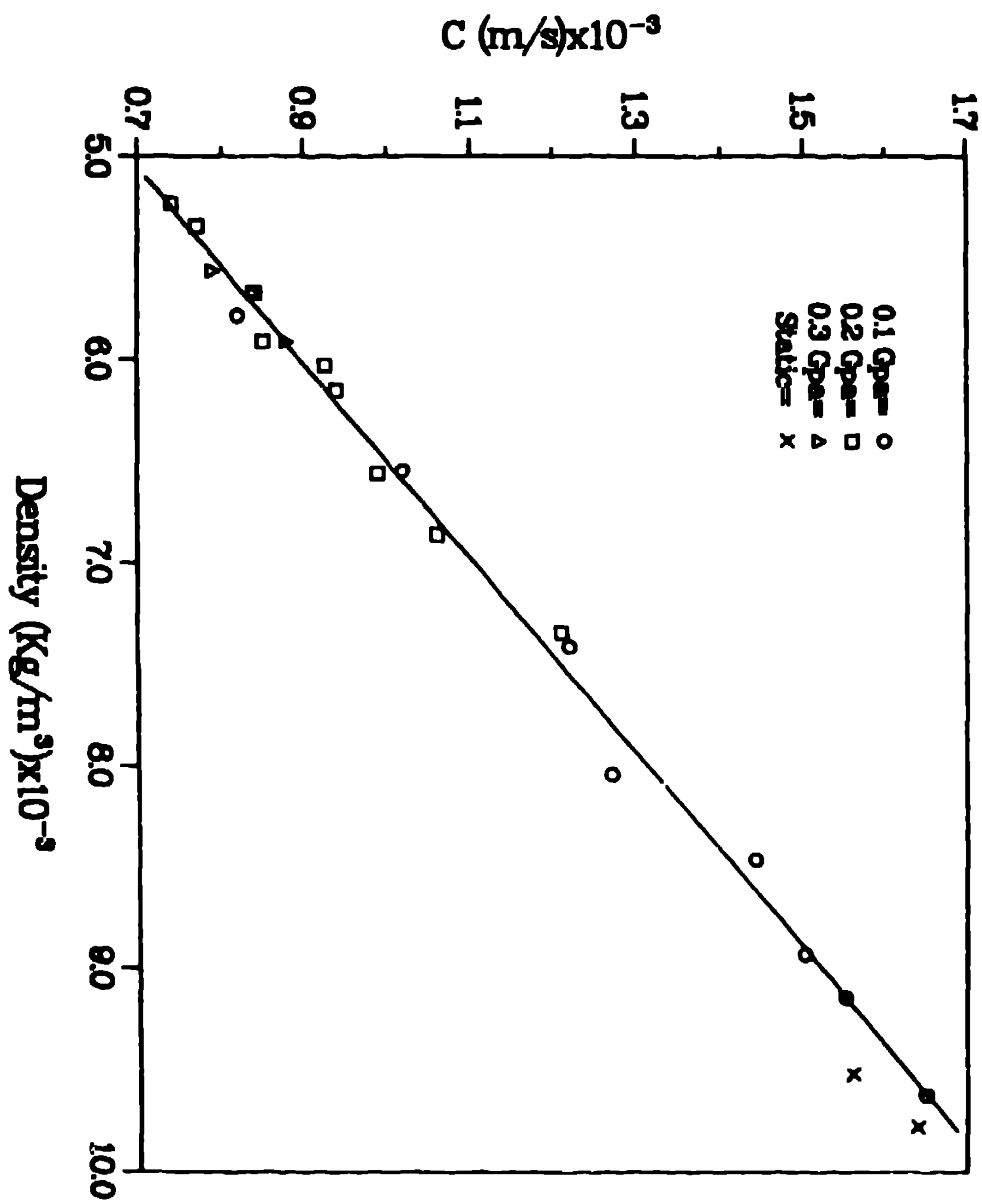
Figure Captions

FIG. 1. Experimentally measured sound speeds for liquid lead vs density.

FIG. 2. Values of the Gruneisen gamma calculated from experimental data plotted against density.

FIG. 3. Calculated values of C_V and γ plotted against density.

FIG. 4. Calculated values of B_T and K_T plotted against density.



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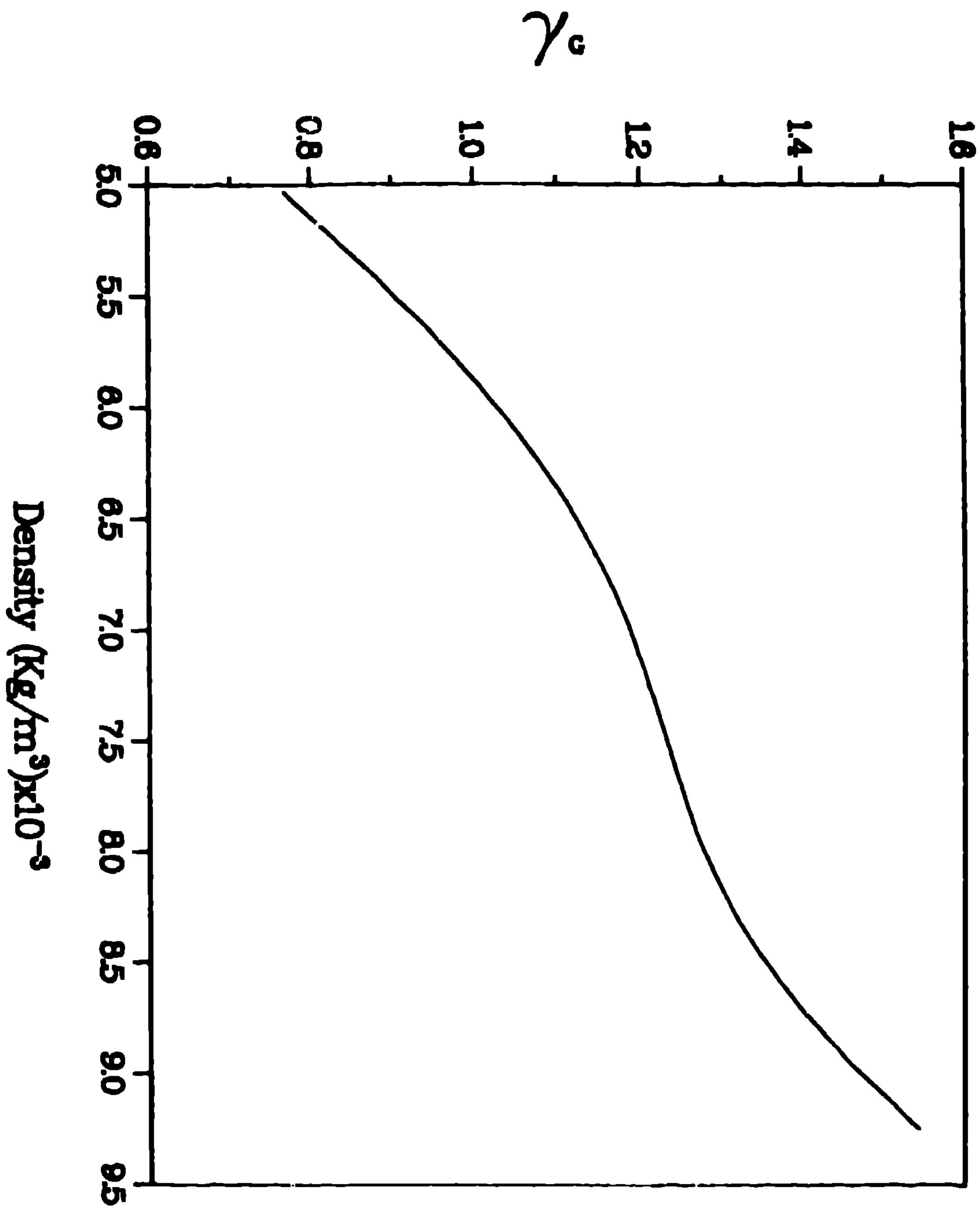

FIGURE 2. Values of the Gruneisen gamma calculated from experimental data plotted against density.

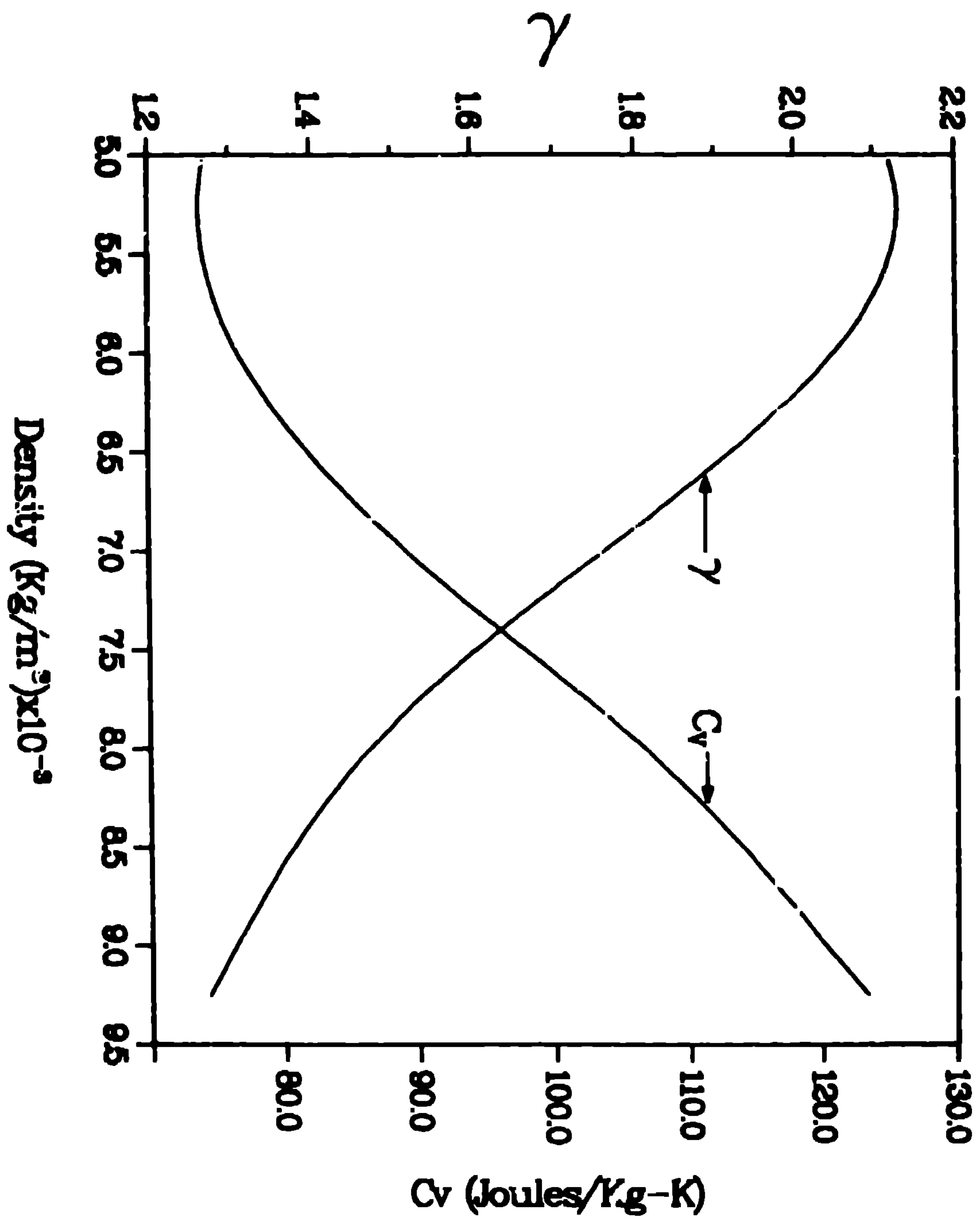

FIGURE 3. Calculated values of C_v and γ plotted against density.

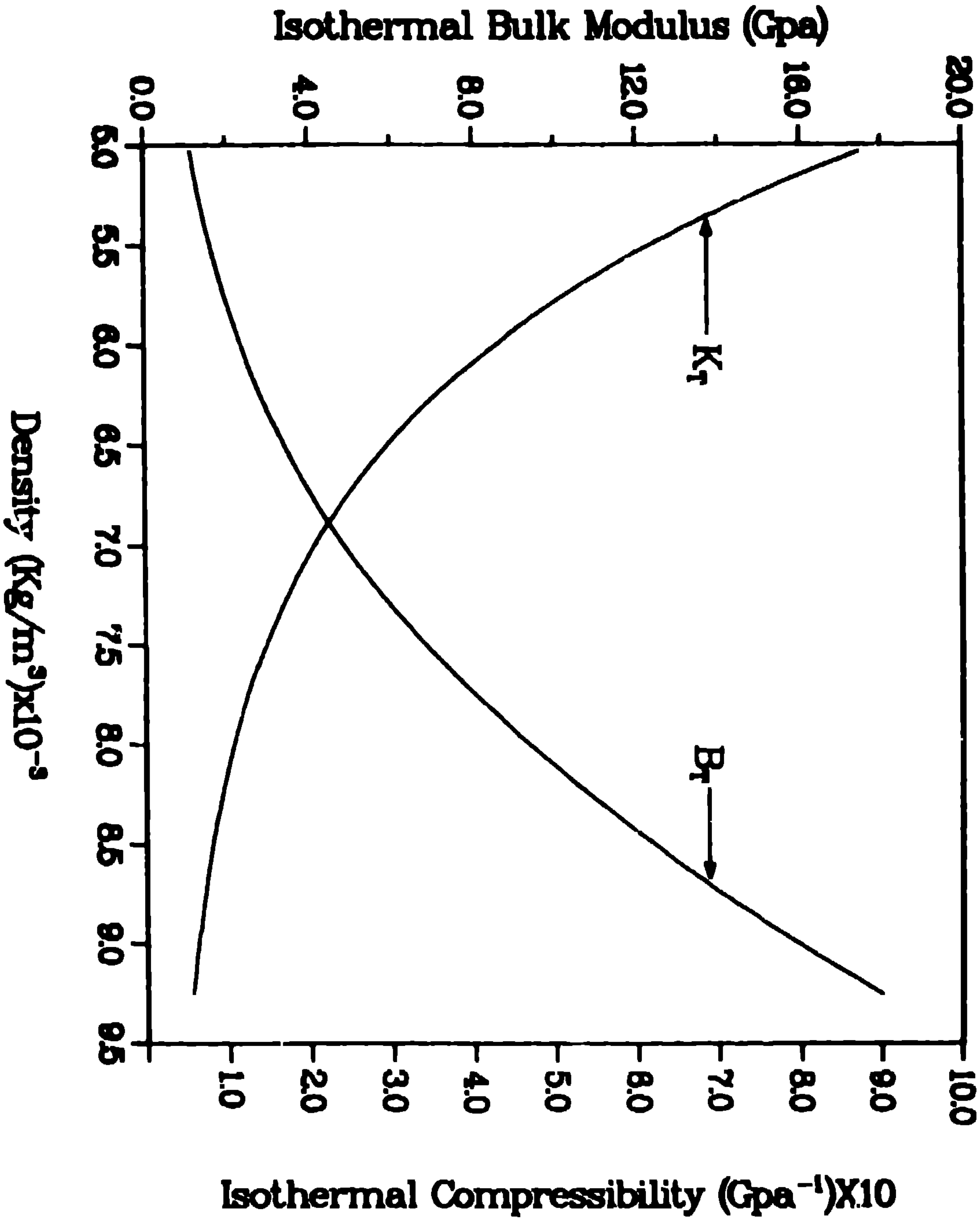

FIGURE 4. Calculated values of B_T and K_T plotted against density.