LBL-10640 c.2
Preprint

Lawrence Berkeley Laboratory
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Submitted to Metallurgical Transactions

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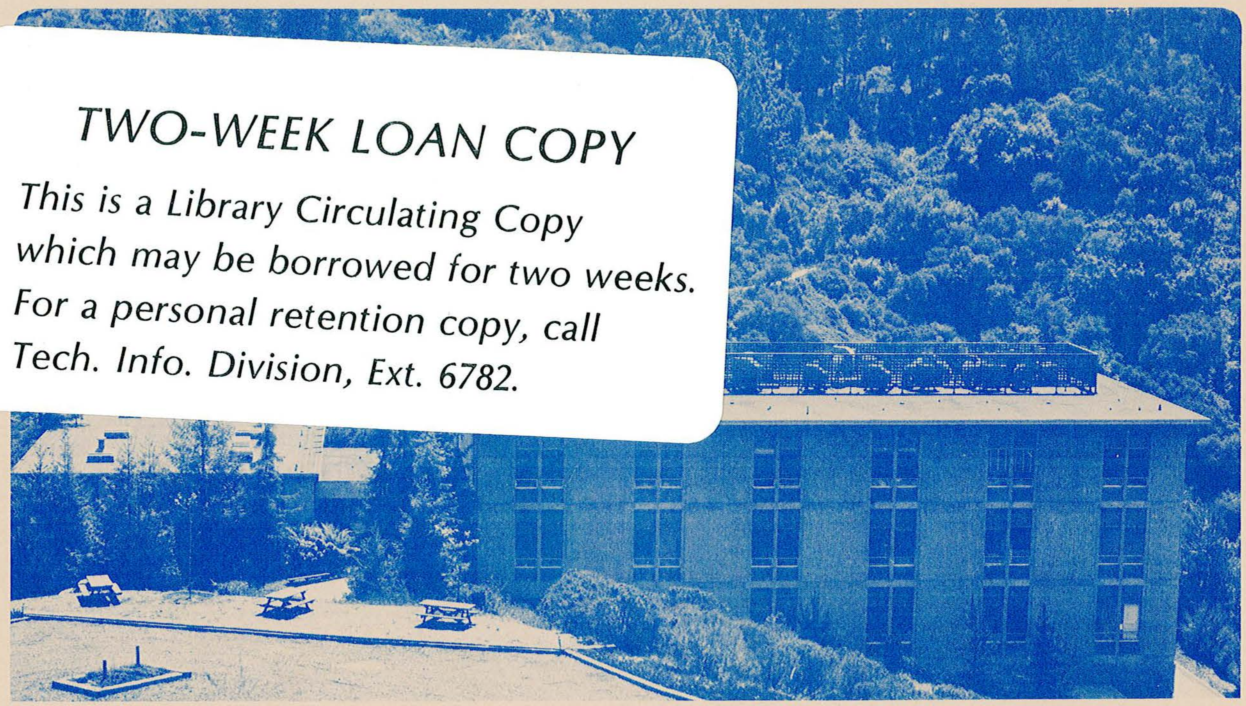
March 1980



Prepared for the U.S. Department of Energy under Contract W-7405-ENG-48

DISCLAIMER

THE THERMAL EXPANSION OF THE DIRECTIONALLY SOLIDIFIED Al-$CuAl_2$ EUTECTIC

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ABSTRACT

Alloys of Al-$CuAl_2$ eutectic composition were prepared from 99.999% pure materials and directionally solidified in a temperature gradient of about 45°C/cm at different growth rates R. The λ^2R = constant relation was verified and lamellar spacings of 7.5, 3.5, 2.6, 1.8 and 1.4 μm were obtained. Dilatometer specimens were machined with axes aligned in the principal lamellae coordinate directions. Thermal expansion was measured by standard dilatometry (Cu standard) using a set point program cycling between room temperature and 500°C. Thermal expansion of the directionally solidified Al-$CuAl_2$ eutectic is greatest in the growth direction (in the plane of the lamellae), least in the transverse direction (orthogonal to the growth direction in the plane of the lamellae) and intermediate in the vertical direction (normal to the lamellae). The most significant finding of the study is that the thermal expansion increases with decreasing lamellar spacing between limits defined approximately by the thermal expansion of the $CuAl_2$ phase alone and the predicted thermal expansion of an isotropic elastic model of the composite.

INTRODUCTION

The Al-CuAl$_2$ eutectic, when directionally solidified under conditions of high purity and high thermal gradients[1] has a regular lamellar microstructure of period λ. This period is controlled by the rate R at which the melt is withdrawn from the furnace according to the relation: $\lambda^2 R$ = constant as shown in Fig. 1. The constant is dependent on material parameters which are held constant in this work.

A universal law of composites proposed by Z. Hashin[2] is

$$\left[\frac{V_A}{K_A} + \frac{V_B}{K_B}\right]^{-1} \leqslant K_C \leqslant V_A K_A + V_B K_B \tag{1}$$

where V is the volume fraction, K is a physical property, the subscripts A and B refer to their respective phases, and C refers to the composite. This expression is independent of all microstructural parameters except volume fraction. Analogous formulae for electrical conductivity, dielectric constants, permeability, and heat conduction are known to be valid. B. Paul[3] and R. Hill[4] have shown that the right-hand side holds for elasticity. The objective of this work was to measure the thermal expansion of the Al-CuAl$_2$ directionally solidified eutectic and determine whether the thermal expansion of the in-situ composite conforms to Hashin's universal law of composites and also to determine any anisotropy or lamellar spacing dependence of the thermal expansion.

If the rule of mixtures is applied to an elastic model of the ideal microstructure (Fig. 2) with the constraint that no slip at the interface occurs,[5,6,7] the following equations for the thermal strains result:

$$(2) \qquad \varepsilon_{11_C} = \alpha_{\mathrm{long}_C}\Delta T = \left[\frac{\left(\alpha_{\mathrm{long}_\theta} - \alpha_{\mathrm{Al}}\right) E_\theta V_\theta}{V_\theta E_\theta + V_{\mathrm{Al}} E_{\mathrm{Al}}} + \alpha_{\mathrm{Al}}\right]\Delta T$$

$$(3) \qquad \varepsilon_{22_C} = \alpha_{\mathrm{trans}_C}\Delta T = \left[\frac{\left(\alpha_{\mathrm{trans}_\theta} - \alpha_{\mathrm{Al}}\right) E_\theta V_\theta}{V_\theta E_\theta + V_{\mathrm{Al}} E_{\mathrm{Al}}} + \alpha_{\mathrm{Al}}\right]\Delta T$$

$$(4) \qquad \varepsilon_{33_C} = \alpha_{\mathrm{vertical}_C}\Delta T =$$

$$= \left[\alpha_{\mathrm{Al}} V_{\mathrm{Al}} + \alpha_{\mathrm{vertical}_\theta} V_\theta + \frac{V_\theta V_{\mathrm{Al}} (E_{\mathrm{Al}} - E_\theta)\left(2\alpha_{\mathrm{Al}} - \alpha_{\mathrm{long}_\theta} - \alpha_{\mathrm{trans}_\theta}\right)}{(1 - \nu)\left(E_{\mathrm{Al}} V_{\mathrm{Al}} + E_\theta V_\theta\right)}\right]\Delta T$$

where E is Young's modulus, ν is Poisson's ratio, V is volume fraction, ΔT is the change in temperature, and α is the coefficient of thermal expansion. The longitudinal direction ε_{11} is the direction in the plane of the lamellae closest to the growth direction, the vertical direction ε_{33} is the direction normal to the lamellae, and the transverse direction ε_{22} is the remaining orthogonal direction. The equations fall nearly into the universal law of composites formulation, except for terms that can be categorized as accounting for Poisson effects.

If the ratio of Young's modulus for the Al and θ phases is independent of temperature, the Poisson's ratios are taken as equal, and the temperature dependence of α_θ is not a function of crystallographic orientation; then, using literature data[5-10] the model predicts that the thermal expansion of the composite in the longitudinal direction will be greatest, followed by the thermal expansion in the vertical and transverse directions. At 500°C, the mean coefficients of thermal expansion have been calculated as $24.7 \times 10^{-6}{}^\circ C^{-1}$, $22.6 \times 10^{-6}{}^\circ C^{-1}$, and $21.3 \times 10^{-6}{}^\circ C^{-1}$ respectively.

The shortcomings of the model, aside from microstructural defects, are that other temperature effects and mechanical phenomena are not fully appreciated.

EXPERIMENTAL

Alloys of 33.25 w/o 99.999% Cu-99.999% Al were directionally solidified in a horizontal furnace in an alumina boat under an argon atmosphere and a thermal gradient of about 45°C/cm at different rates R. After solidification, the alloys were milled square, polished and etched, revealing the grain structure. The largest grains were chosen to be sectioned out of the ingots. Specimens were machined by milling parallelopipids of square cross section, turning them down and facing them off in a square collet. Dilatometer specimens were nominally 0.5 cm in diameter and 1 cm long, the exact dimensions being controlled primarily by the finite size of the single crystals from which the specimens were taken. Dilatometer specimens for the polycrystalline ingot with $\lambda = 1.4\ \mu m$ were machined along ingot coordinates.

Thermal expansion measurements were made using a Theta IIIR dilatometer which consists of a fused silica pushrod driving a linear voltage differential transformer (LVDT) with a sensitivity of 1×10^{-5} cm and an accuracy of 5×10^{-5} cm as the sensing device. The temperature was measured and controlled by a 10% Rh-Pt, Pt thermocouple inserted in a blind hole drilled in each specimen and the edges of the hole peened lightly to hold the thermocouple in place. The thermal expansion was measured at set points approximately 20°C apart from room temperature to 500°C. 99.999% Cu was used to calibrate the dilatometer.

RESULTS

Figures 3A, 3B and 3C are optical micrographs of a typical well ordered specimen as viewed along the dilatometer axes. In the transverse axis specimen, an ideal microstructure is evident, at least on a local scale. The longitudinal axis specimen shows that the lamellae are not ideal, but bend and terminate at dislocations. The lamellar spacings in these two figures are the same since the view is edge-on. The lamellar spacing for the vertical axis specimen appears extremely large because of the shallow projection angle. The micrograph magnifies any departure from the ideal microstructure.

Figures 4A and 4B show polycrystalline material of $\lambda = 1.4$ μm, in which the growth direction lies in the plane of the lamellae but the lamellae are disordered or degenerate. The transverse and vertical directions for macroscopic specimens cannot be defined.

To check the lamellae crystallographic orientation, transmission Laue photographs were taken using nickel filtered CuKα radiation (Fig. 5A) and stereographic projections were made (Fig. 5B). The Laue photographs for all ingots were consistent with the crystallographic relationship $(121)_\theta \parallel (111)_{Al}$, $\langle 1\bar{1}0 \rangle_\theta \parallel \langle 10\bar{1} \rangle_{Al}$ as found by R. W. Kraft and A. L. Albright,[11,12] L. Valero[13] and others.[5,6,8,14]

Figures 6A, 6B, 6C show results of the thermal expansion measurements.

It should first be noticed that the greatest thermal expansion occurred in the longitudinal direction, that is, near the growth direction; the least thermal expansion was in the transverse direction; and the thermal expansion in the vertical direction was intermediate between these. These results are in qualitative agreement with the trends predicted using Eqs. (2-4). In the polycrystalline specimen with $\lambda = 1.4$ µm, the vertical and transverse directions are not well defined but clearly there is a difference in the thermal expansion in these two directions normal to the growth direction.

The second trend apparent in Figs. 6A,6B,6C is that the thermal expansion is a function of the lamellar spacing, being least for large lamellar spacings and increasing as the lamellar spacing decreases. This is the most significant finding of this study. For the slowest growth rate specimens, the thermal expansion of the composites were approximately equal to those calculated for the θ phase alone. For intermediate growth rates, the thermal expansion lay between values for the θ phase and those predicted by the elastic rule of mixtures model.

DISCUSSION

An empirical approach to the interpretation of these findings that does not involve a more detailed and/or other continuum mechanics model is to consider the effects of lamellar spacing and temperature on the mechanical properties of the composite.

As a function of the lamellar spacing, yield stress,[15,16,17] as well as hardness[14,16] and elastic modulus[18] increase as λ decreases. This suggests that the Al phase bears more of the load as λ decreases if the effective yield stress of the θ phase remains the same. The constraint in the composite is that at the interface no slip occurs.[5,6,7] Therefore thermal stress builds up at the lamellar interface and across both lamellae. In the elastic model thermal stress is proportional to the change in temperature. The yield stress of the Al phase is a function of both temperature and lamellar spacing. This establishes that the thermal expansion of the Al phase could almost immediately (small ΔT) conform to that of the θ phase for large λ, but that the elastic model should be more correct as λ decreases.

The minimum and maximum of the thermal expansion of the composite are approximately equal to those of the θ phase and values predicted by the elastic rule of mixtures, respectively. However, since thermal stress is a function of the difference in thermal expansion between the two phases rather than the sum, it is unclear what function of λ should be used to represent the thermal expansion of the composite. No mechanical property of the composite has yet been shown to be consistently predicted by a specific function of λ.[19,20]

There are several mechanical phenomena neglected by the elastic rule of mixtures model. Plastic deformation is most likely to occur in the Al phase as a function of the thermal stresses at the lamellae interface which in turn is a function of both ΔT and T. However, the aluminum solid solution is known to work harden as it is deformed. This effect tends to be offset at higher temperatures where the aluminum anneals. Creep or relaxation also occurs.

Damage to the microstructure of the composite can occur because of repeated cycling of the thermal stresses induced by raising and lowering its temperature. Thermal cycling can be ignored in this work, however, because of the large number of cycles ($\sim 10^4$)[21] needed for thermal cycling damage to become evident.

G. Garmong[22] has attempted to model this system to account for plastic deformation, creep, and thermal cycling; however, he could not supply direct evidence for his standard linear solid model which works well for the Al-$NiAl_3$ directionally solidified eutectic (fibrous) composite. One could use his approach and take advantage of the fact that the microstructure allows one to assume plane stress conditions.

There are several thermal effects. At high temperatures, the directionally solidified Al-$CuAl_2$ eutectic will coarsen, i.e., the θ lamellae will become thicker and shorter, and begin to spheroidize. Significant changes in stable lamellar material will not be evident for many hours at temperatures near the melting point of the eutectic[23] and hence coarsening can be ignored in this work because of the relatively short times involved at high temperatures. For precipitation to occur in the Al phase of the Al-$CuAl_2$ directionally solidified composite, a supersaturation of Cu in the Al solid solution must be obtained by quenching the material. If it is not quenched drastically enough, the excess Cu solute will diffuse directly to the θ phase lamellae. None of the specimens in this work were quenched; all measurements were made in the as-grown condition and relatively slow cooling rates were employed in vacuum. It is concluded that precipitation is not a major consideration. A related consideration is that the solubility of Cu in the Al solid solution varies with temperature. Thus the change in thickness of the θ lamellae from solidification to room temperature is ~10%.[24] The shrinkage and expansion of the θ phase as the temperature increases and decreases has been ignored in this work.

There are different cases of misorientation to be considered. The first is that of lamellar misorientation, which is due partly to faulty specimen preparation, but primarily because the lamellar directions within the ingot are not planar. The second case is that of crystallographic interphase misorientation. This is impossible to avoid since the relation $(121)_\theta \parallel (111)_{Al}$, $[12\bar{3}]_\theta \parallel [1\bar{2}1]_{Al}$ is not exact but only a low indice expression for the true relationship. The interface relationship between the phases has been found to vary within the same specimen[25,26] by as much as 12°.[27] The thermal expansion of a volume element would have to be calculated by summing these components of the thermal expansion for the local volume element and performing an appropriate integration over a macroscopic volume.

CONCLUSION

The thermal expansion of the directionally solidified Al-$CuAl_2$ eutectic decreases with increasing lamellar spacing for the range 1.4 μm to 7.5 μm. This lamellar dependence is not accounted for by the rule of mixtures based on volume fractions. The thermal expansion is greatest in the growth direction, least in the transverse direction, and the thermal expansion in the vertical normal to the lamellae is only slightly greater than it is in the transverse direction. These results confirm that the $CuAl_2$ phase is anisotropic with regards to thermal expansion.

The most important implication of the results of this work is that the designer not only must take into account the fact that for high temperature applications the yield strength of the directionally solidified eutectic declines, but that the dimensional tolerances will change due to the change in thermal expansion as coarsening proceeds.

ACKNOWLEDGMENTS

This work was supported by the Division of Materials Science, Office of Basic Energy Sciences, U.S. Department of Energy under Contract No. W-7405-ENG-48.

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FIGURE CAPTIONS

Fig. 1. $\lambda^2 R$ = constant growth relation for the directionally solidified Al-CuAl$_2$ eutectic.

Fig. 2. Idealized microstructure with thermal expansion specimen axes.

Fig. 3. Optical micrographs of a set of directionally solidified Al-CuAl$_2$ regular microstructure thermal expansion specimens; viewed along

A: transverse axis,

B: longitudinal axis,

C: vertical axis.

Fig. 4. Al-CuAl$_2$ directionally solidified eutectic degenerate microstructure thermal expansion specimens optical micrographs viewed along

A: longitudinal axis,

B: "vertical" axis.

Fig. 5A. Transmission Laue photograph of the Al-CuAl$_2$ directionally solidified eutectic, λ = 7.5 μm.

Fig. 5B. Stereographic projection corresponding to Fig. 5A.

Fig. 6. Al-CuAl$_2$ directionally solidified eutectic thermal expansion in

A: longitudinal direction,

B: transverse direction,

C: vertical direction.

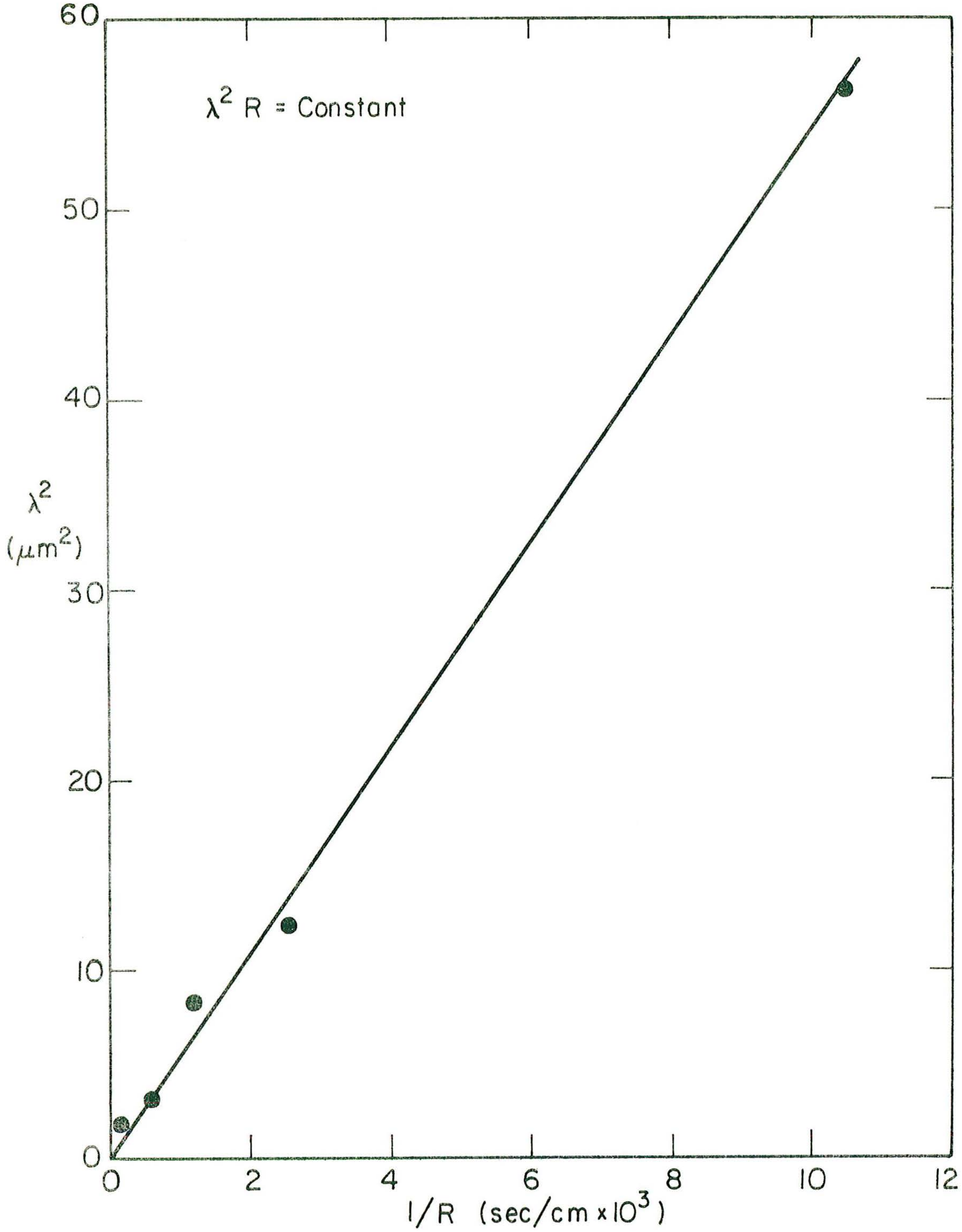

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Fig. 1

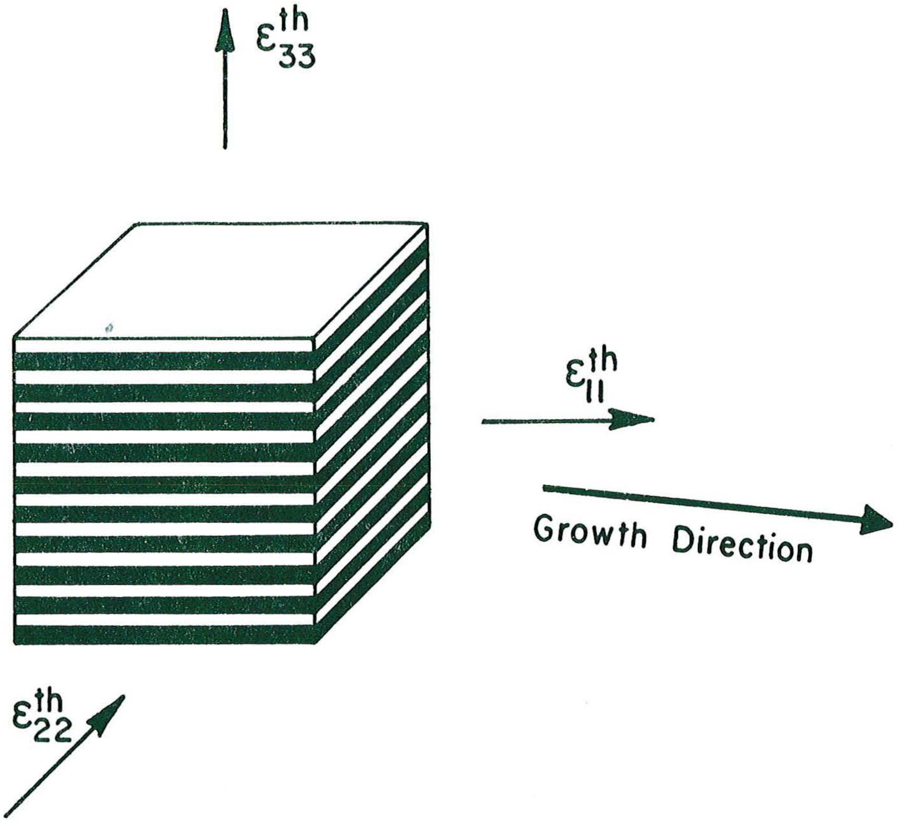

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Fig. 2

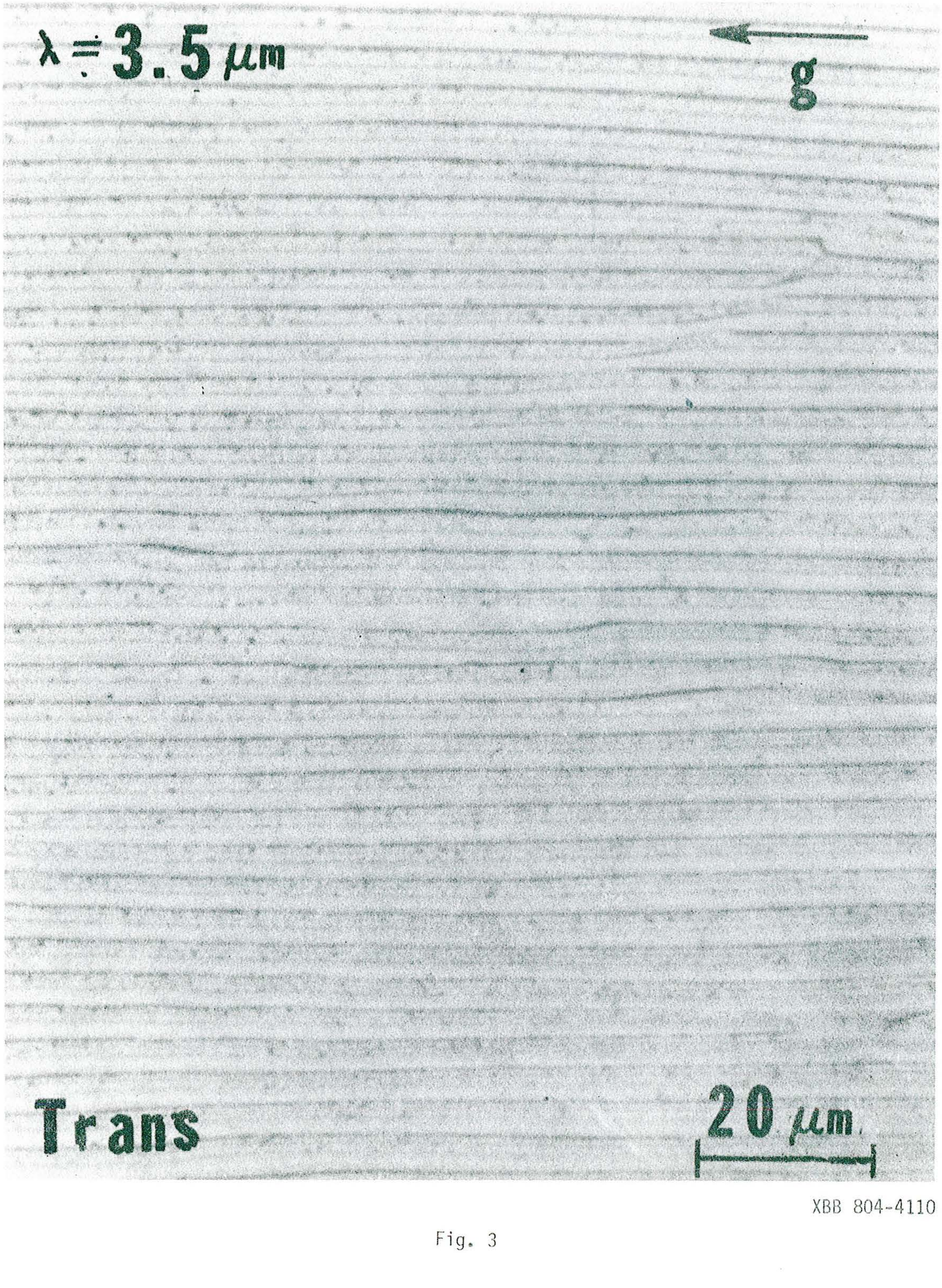

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Fig. 3

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Fig. 3B

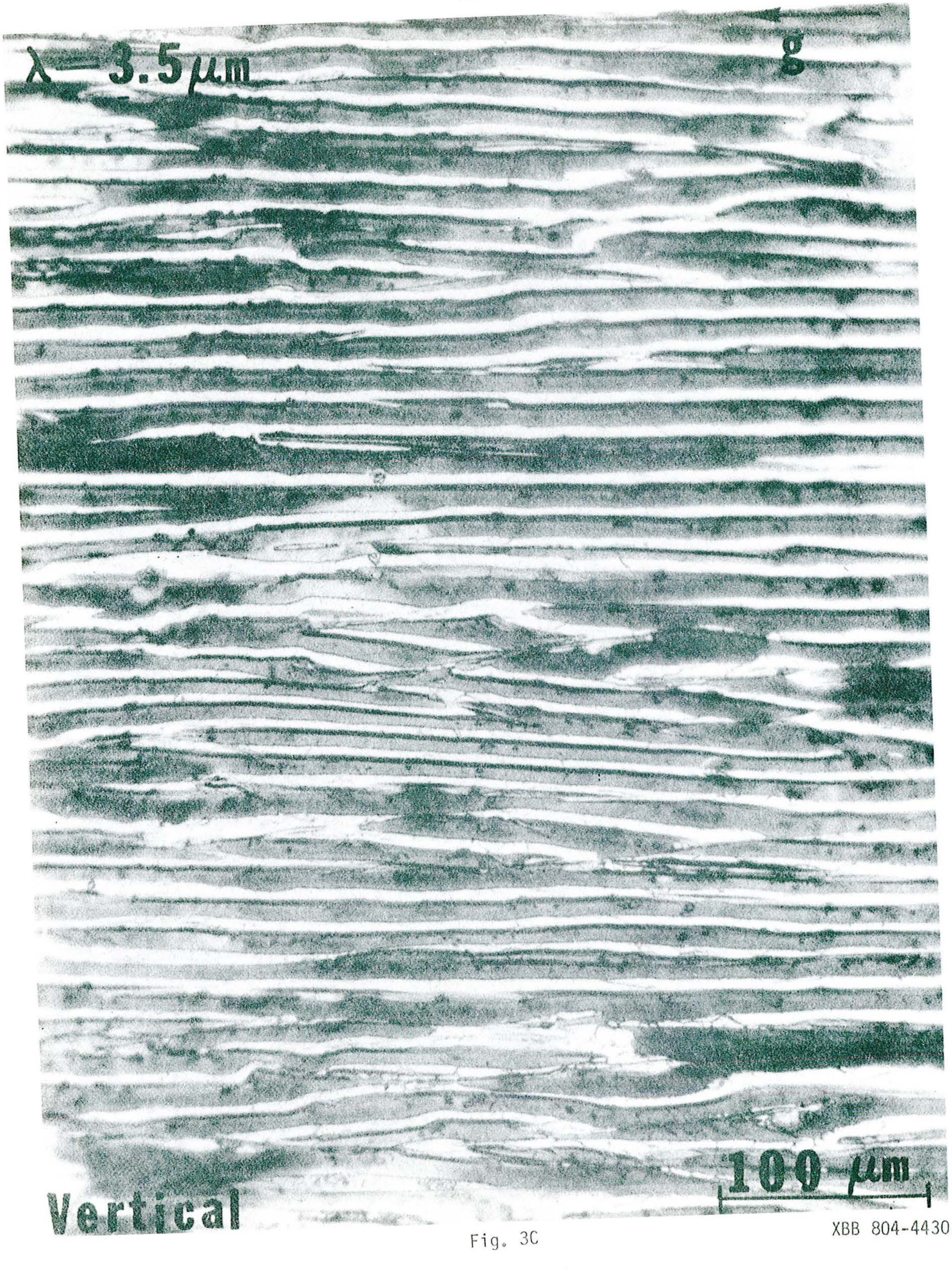

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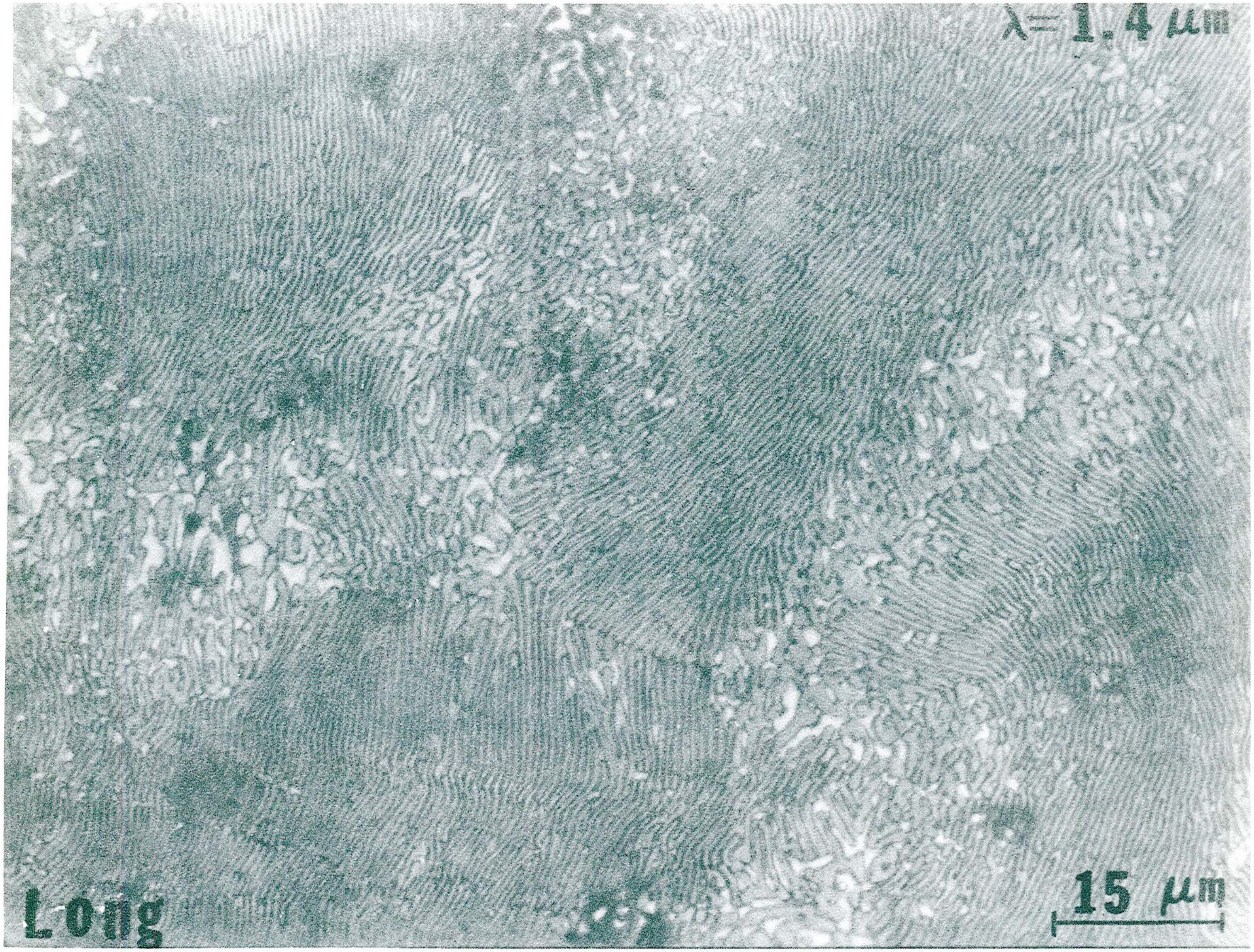
Fig. 3C



XBB 804-4112

Fig. 4

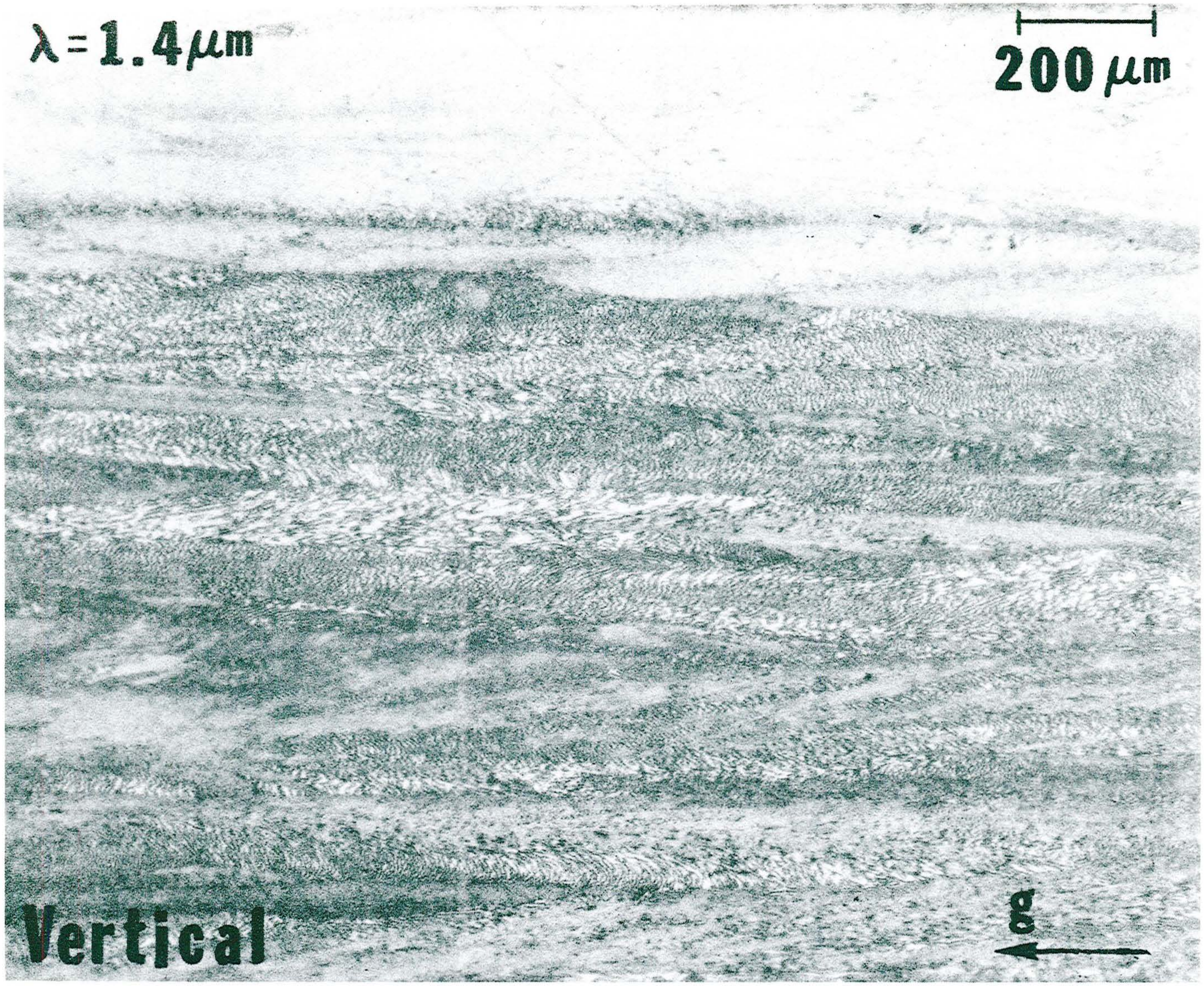


Fig. 4B

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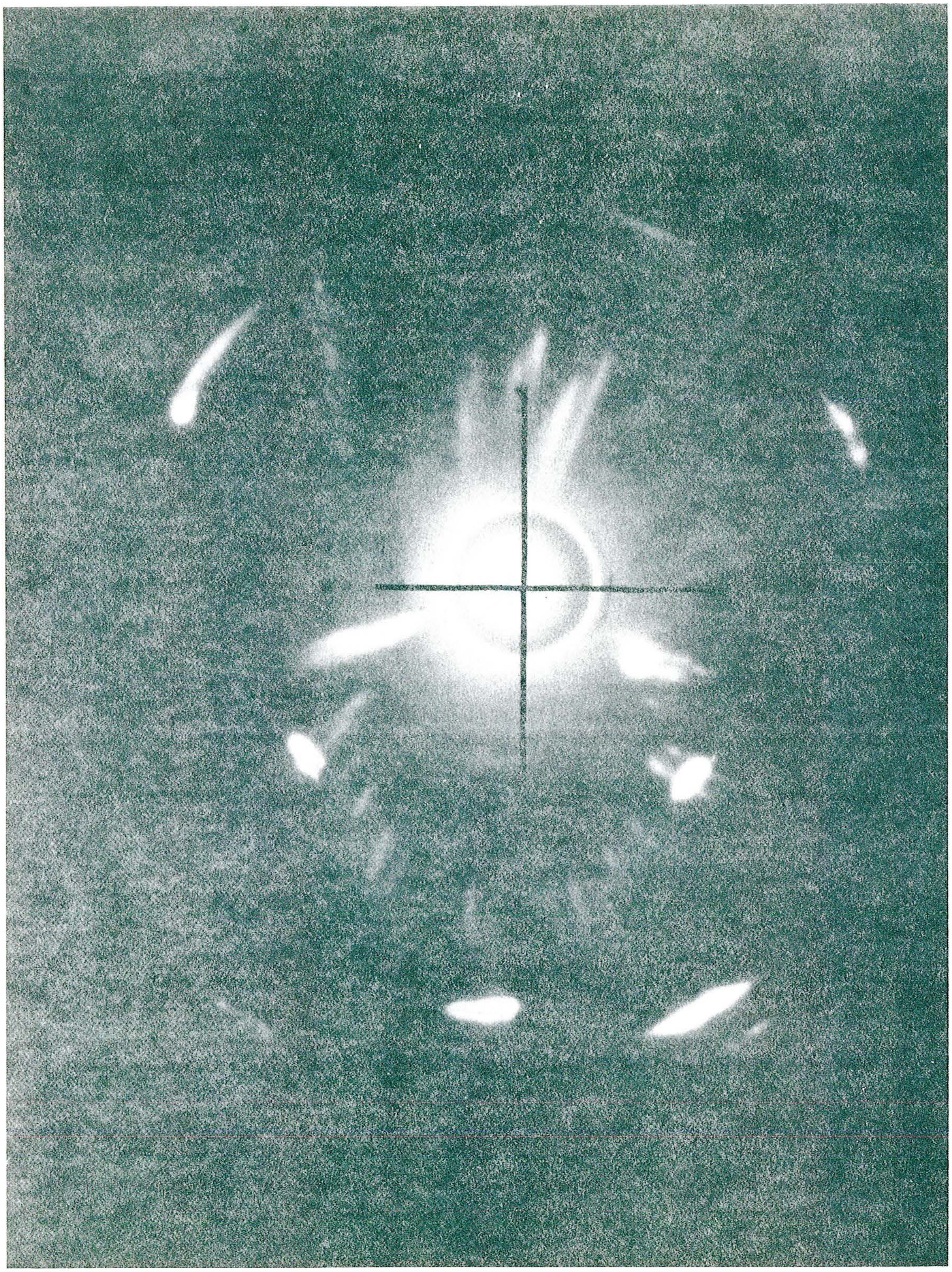

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Fig. 5A

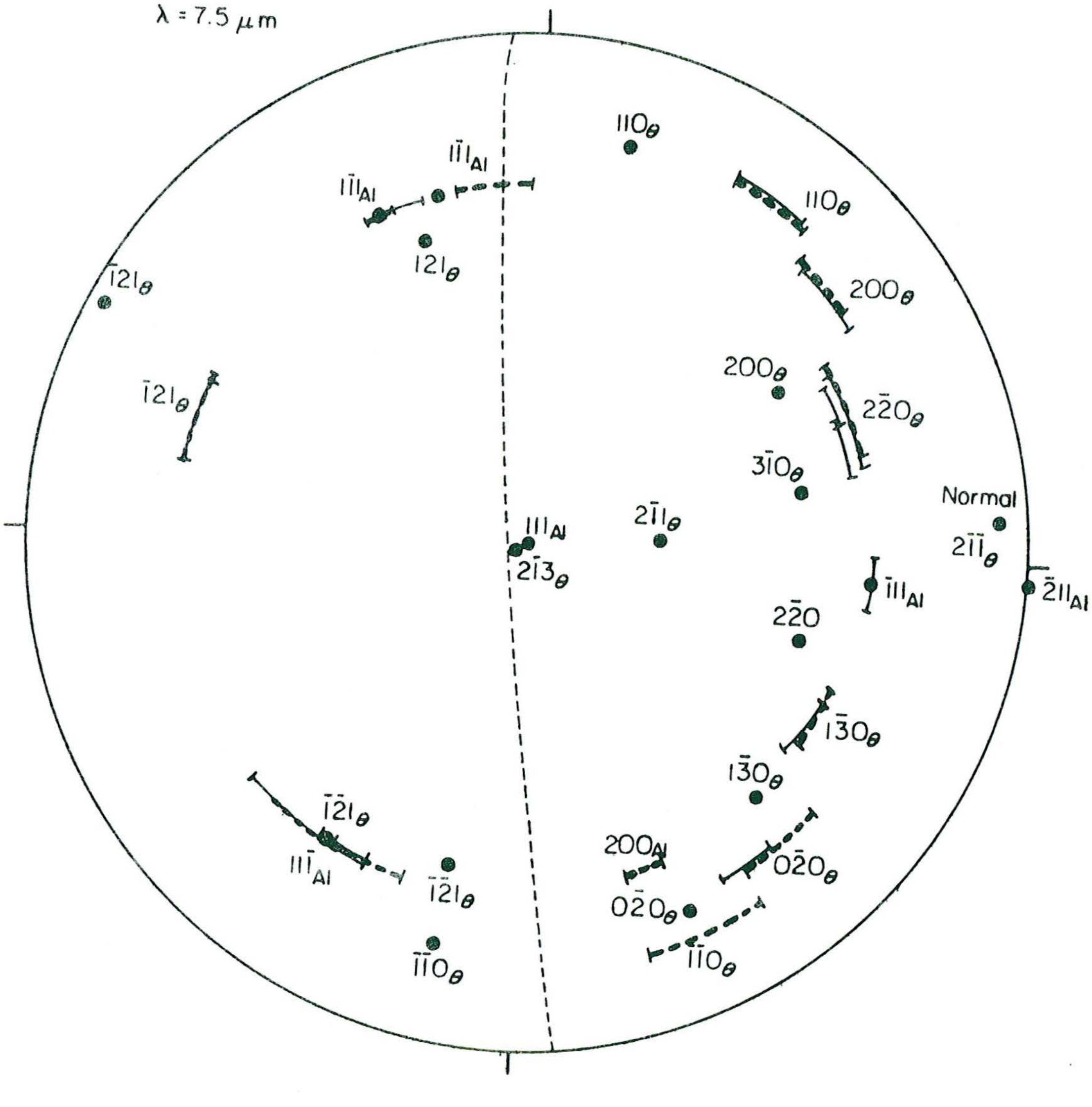

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Fig. 5B

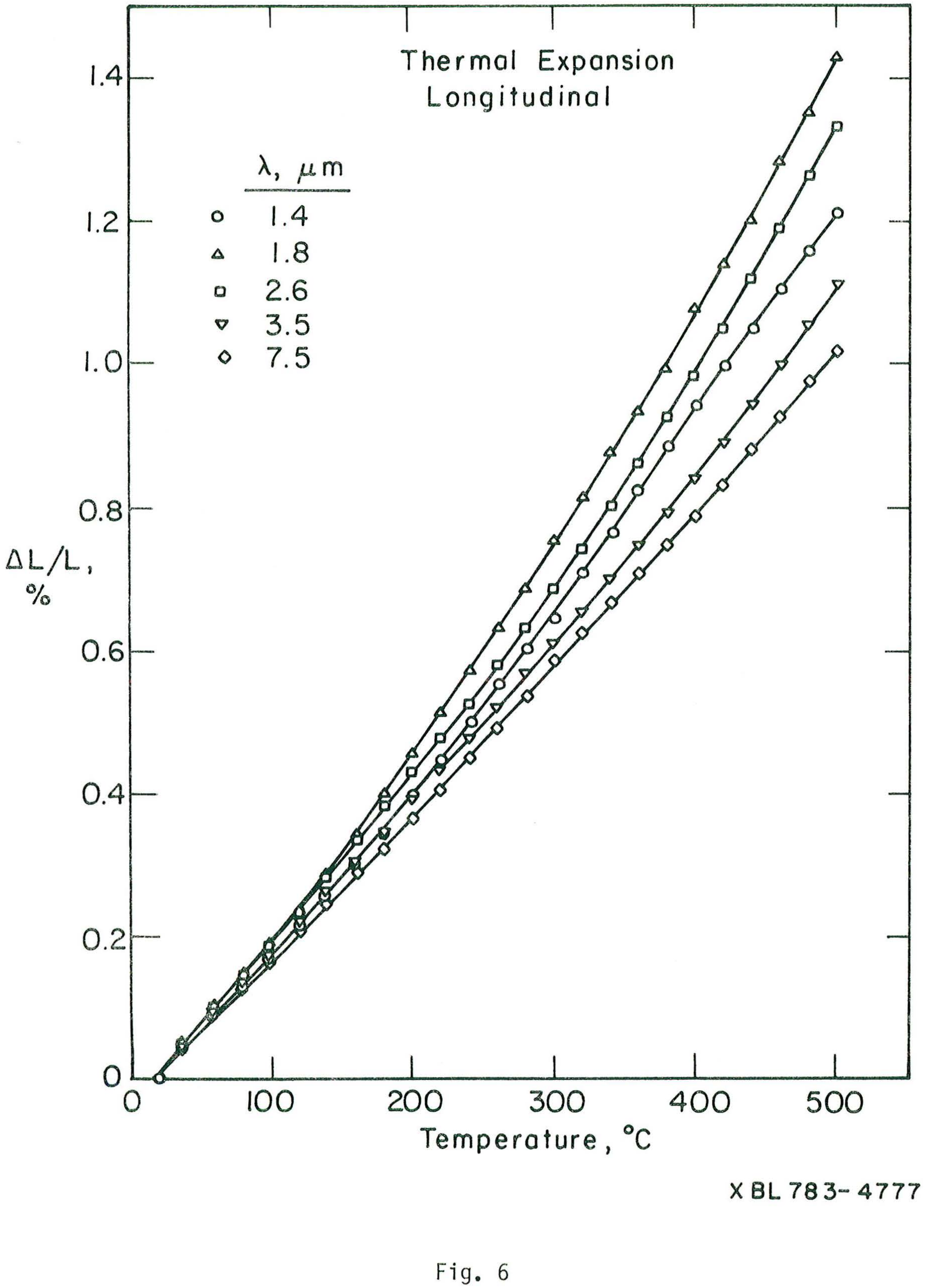
Thermal Expansion
Longitudinal
λ, μm
○ 1.4
△ 1.8
□ 2.6
▽ 3.5
◇ 7.5
ΔL/L, %
1.4
1.2
1.0
0.8
0.6
0.4
0.2
0
0
100
200
300
400
500
Temperature, °C
XBL 783-4777

Fig. 6

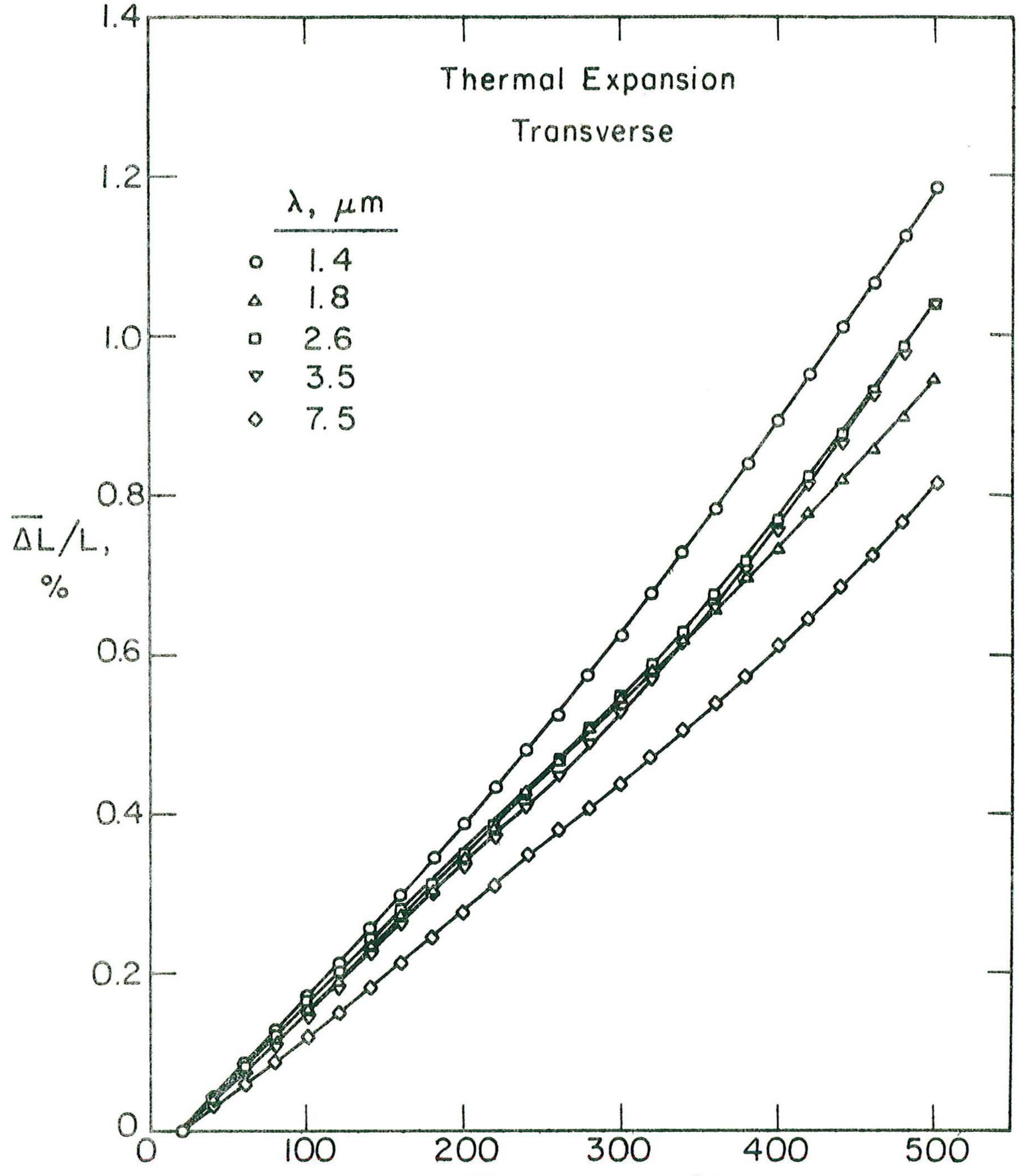

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Fig. 6B

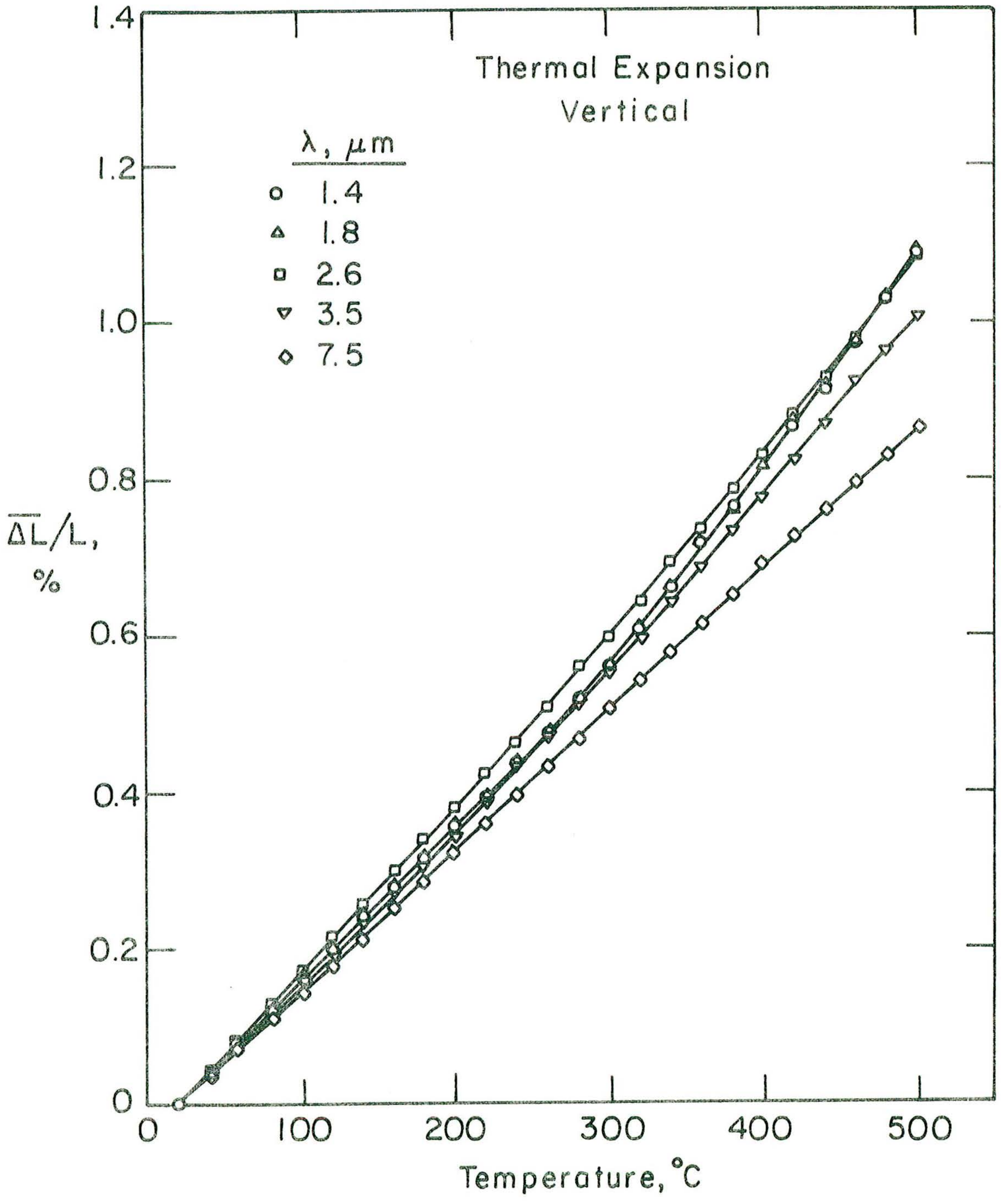

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Fig. 6C