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MAGNETOSTRICTION OF RARE EARTH-IRON COMPOUNDS RFE3 AND R6FE23.(U)
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MAGNETOSTRICTION OF RARE EARTH-IRON COMPOUNDS; RFe_3 AND R_6Fe_{23}

BY R. ABBUNDI A. E. CLARK

RESEARCH AND TECHNOLOGY DEPARTMENT

1 APRIL 1980

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20. ABSTRACT (Continue on reverse side if necessary and identify by block number)

The magnetostriction of the RFe$_3$ and R$_6$Fe$_{23}$ compounds was measured from room temperature to 80 K (R = Sm, Tb, Dy, Ho, Er and Tm). In the rhombohedral RFe$_3$ series, TbFe$_3$ possesses the largest magnetostrictive strain with $\lambda_{\parallel} - \lambda_{\perp} = 990 \times 10^{-6}$ at room temperature and 1725×10^{-6} at 80 K. SmFe$_3$, although potentially a highly magnetostrictive compound, possesses a strain of only -316 ppm at room temperature due to the fact that the easy axis of magnetization is along the c-axis. The temperature

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dependence of the magnetostriction of $TmFe_3$ is complex. In the cubic R_6Fe_{23} series the magneto-strains of Er_6Fe_{23} and Tm_6Fe_{23} increase substantially with decreasing temperature, due to the rapid ordering of the rare earth sublattice. The strain in Er_6Fe_{23} increases nearly 7-fold from -57 ppm at 300 K to -392 ppm at 80 K. Tm_6Fe_{23} shows a nearly 10-fold increase from -37×10^{-6} to -362×10^{-6}. The signs of the magnetostriction for both the RFe_3 and R_6Fe_{23} series are consistent with those predicted by the Stevens' factor.

FOREWORD

The magnetostriction study reported here is part of a research program undertaken to determine the nature of the magnetostriction in the rare earth-iron intermetallic compounds. In this report is detailed the temperature dependence of the magnetostriction for a series of RFe_3 and R_6Fe_{23} compounds. Studies were made as a function of applied field at various temperatures from room temperature to 80 K.

In the RFe_3 series $TbFe_3$ possesses the largest magnetostrictive strain with $\lambda_{\parallel} - \lambda_{\perp} = 990 \times 10^{-6}$ at room temperature and 1725×10^{-6} at 80 K. $SmFe_3$, although potentially a highly magnetostrictive compound possesses a strain of only -316 ppm at room temperature due to the fact that the easy axis of magnetization is along the c-axis.

In the R_6Fe_{23} series the magneto-strains of Er_6Fe_{23} and Tm_6Fe_{23} increase substantially with decreasing temperature, due to the rapid ordering of the rare earth sublattice. The strain in Er_6Fe_{23} increases nearly 7-fold from -57 ppm at 300 K to -392 ppm at 80 K. Tm_6Fe_{23} shows a nearly 10-fold increase from -37×10^{-6} to -362×10^{-6}.

The study was carried out in the Solid State Branch of the Radiation Division as part of the research program on magnetostrictive materials. The research was sponsored by the Office of Naval Research (PO-4-0081, NR 039-110) and the NSWC Independent Research Program (IR-011).

B F Desavage

B. F. DESAVAGE
By direction

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INTRODUCTION

Previous work on the cubic Laves phase rare earth-Fe_2 compounds has shown that these materials possess enormous magnetostriction constants. In some of these compounds this huge strain is even present at room temperature. Both $TbFe_2$ and $SmFe_2$ possess a room temperature magnetostriction $|\lambda| > 2000$ ppm.[1]

Although the magnetostriction of the RFe_2 compounds (R = Sm, Tb, Dy, Ho, Er and Tm) has been extensively studied,[1-4] little attention has been paid to the magnetostriction of the remaining rare earth-iron intermetallic compounds. Here we report measurements of the magnetostriction of the RFe_3 and R_6Fe_{23} compounds from room temperature to 80K. The room temperature magnetostrictions of the R_2Fe_{17} compounds were also investigated.

In the rare earth-iron series the Curie temperatures are the highest for the RFe_2 compounds (560-710K)[5,6] and decrease with increasing iron concentration. Just the opposite is true for the rare earth-cobalt and rare earth-nickel series.[7,8]

Mössbauer effect measurements[9-13] have been reported on the RFe_3 series at various temperatures. These results indicate the direction of the magnetization in these compounds. An attempt will be made to correlate the direction of the magnetization with the behavior of the magnetostriction as a function of temperature.

EXPERIMENTAL RESULTS

For all R, the magnitude of the magnetostriction at room temperature in the RFe_3 and R_6Fe_{23} compounds is smaller than that in the RFe_2 compounds.[1] However, as shown in Table I, the trend reverses with an increase in the magnitude of the magnetostriction as the concentration of rare earth decreases from R_6Fe_{23} to R_2Fe_{17}.

TABLE I. Room temperature magnetostriction
$(\lambda_{\parallel} - \lambda_{\perp}) \times 10^{-6}$ at 25 kOe.

R	RFe_2[a]	RFe_3	R_6Fe_{23}	R_2Fe_{17}
Sm	-2340	-316	††	-95
Tb	2630	991	**	-21
Dy	650	514	**	-90
Ho	120	85	88	-159
Er	-449	-103	-57	-83
Tm	-185	-66	-37	-45

a) Compiled by A. Clark. Handbook on the Physics and Chemistry of Rare Earths, Ed. K. Gschneider and L. Eyring, North Holland Publishing Co. (1979), Volume II.

†† The compound Sm_6Fe_{23} does not form.

** Attempts to prepare single phase Tb_6Fe_{23} and Dy_6Fe_{23} were unsuccessful.

Figures 1-3 show the room temperature magnetostriction as a function of applied field for the RFe_3, R_6Fe_{23} and R_2Fe_{17} series. Unexpected results were obtained for the R_2Fe_{17} compounds, where the sign of the magnetostriction for all the compounds was negative. The only exception was Tb_2Fe_{17} in the as-cast (unannealed) state, where $\lambda_{\parallel} - \lambda_{\perp} = 197 \times 10^{-6}$ at H = 25 kOe. This is in contrast to the results on a Tb_2Fe_{17} sample after annealing which yields $\lambda_{\parallel} - \lambda_{\perp} = -21 \times 10^{-6}$ at 25 kOe. However examination of the field dependence in Figure 3 of the annealed specimen does give some indication that there could possibly be a change in sign for this sample at larger fields. The R_2 Fe_{17} compounds crystallize into a hexagonal Th_2Ni_{17}-type structure for R = Dy, Ho, Er and Tm. However, Strnat et al.[14] have reported that the x-ray pattern of Tb_2Fe_{17} displays lines belonging to this hexagonal structure as well as a rhombohedral Th_2Zn_{17}-type structure, with the hexagonal form being the high temperature modification. This possibly accounts for the difference in the magnetostriction observed for Tb_2Fe_{17} in the annealed and unannealed state.

A. RFe_3 COMPOUNDS

The RFe_3 compounds crystallize into a rhombohedral $PuNi_3$-type structure. Figure 4 shows the field dependence of the magnetostriction at various temperatures for $TbFe_3$. This compound possesses the largest magnetostrictive strain in the RFe_3 series with a room temperature $\lambda_{\parallel} - \lambda_{\perp} = 990 \times 10^{-6}$ at H = 25 kOe. Although the basal plane is easy throughout the entire temperature range with the magnetization along a b-axis,[9,10,12] the lack of saturation in the field dependence is indicative of the large magnetocrystalline anisotropy found in the Tb-Fe intermetallics.[15] Figure 8 shows the temperature dependence of the magnetostriction at H = 15 and 25 kOe. The strain in $TbFe_3$ increases monotonically with decreasing temperature reaching 1725×10^{-6} at 80 K.

The magnetostriction as a function of applied field for $SmFe_3$ at various temperatures is shown in Figure 5. While highly magnetostrictive at the lower temperatures, $\lambda_{\parallel} - \lambda_{\perp} = -1192 \times 10^{-6}$ at 80 K, $SmFe_3$ possesses only a moderate strain of -316 ppm at 300 K, resulting from the hard basal plane (c-axis easy) in this compound.[9] Although there has not been any determination of the easy axis in $SmFe_3$ below room temperature, the curves in Figure 5 do show a substantially larger strain at low fields below T = 150 K. The low temperature data also displays a "knee" in the curves which is totally absent near room temperature. The temperature dependence of the strain of $SmFe_3$ at H = 15 and 25 kOe is shown in Figure 8. $SmFe_3$ possesses the largest negative magnetostriction in the RFe_3 series.

Mössbauer effect measurements on $DyFe_3$ show that the magnetization rotates out of the basal plane toward the c-axis with decreasing temperature, making an angle of ≈ 26° with the basal plane at T = 77 K.[9-11] This accounts for the behavior of the magnetostriction displayed in Figure 8. The room temperature strain at H = 25 kOe was found to be 540×10^{-6} with a peak in the magnetostriction of 565 ppm occurring at T = 270 K. The rotation of the moment out of the basal plane results in a decrease in λ below this temperature. Near 100 K the magnetostriction reaches a minimum of 265 ppm and then begins to increase in value with further temperature reduction as rotation of the moment ceases. The strain vs. field curves in Figure 6 for $DyFe_3$ saturate fairly well at room temperature but as the temperature is reduced the curves become "harder" as the moment pulls up out of the basal plane. At the lowest temperature measured T = 65 K the magnetostriction once again saturates, giving further evidence that the moment has ceased its rotation.

A change in the easy axis of magnetization has also been observed in $HoFe_3$. In this compound the Mössbauer effect reveals that near 100 K a spin reorientation occurs, with the magnetization moving from a b-axis to an a-axis as the temperature is lowered.[9,10] Figure 7 shows the magnetostriction as a function of applied field for $HoFe_3$ at various temperatures. The temperature dependence of the magnetostriction at H = 15 and 25 kOe is shown in Figure 8. The magnetostriction of $HoFe_3$ remains small over the entire temperature range in spite of the fact that the magnetization remains in the basal plane. At room temperature we find $\lambda_{\parallel} - \lambda_{\perp} = 85 \times 10^{-6}$ at 25 kOe. Below T = 225 K the strain vs. field curves become magnetically "harder", while in the temperature region 175 K > T > 125 K the high field magnetostriction is temperature independent. This behavior is presumably a result of the intermediate direction of the magnetization at these temperatures. Saturation, as well as an increase in λ, again occurs at temperatures below 100 K, with $\lambda_{\parallel} - \lambda_{\perp} = 183 \times 10^{-6}$ at T = 80 K.

The magnitude of the strain in $ErFe_3$ was found to undergo the largest change of any of the RFe_3 compounds. Figure 11 shows that the magnetostriction at H = 25 kOe increases nearly 6-fold from -103 ppm at 300 K to -595 ppm at 80 K. $ErFe_3$ possesses a compensation point in its magnetic moment at T = 239 K. This is reflected in the sharp "dip" in the magnetostriction at this temperature with an accompanying change in sign. The RFe_3 compounds previously discussed (with the exception of $SmFe_3$) also possess compensation points but they all occur above room temperature. The magnetostriction as a function of applied field for $ErFe_3$ at various temperatures is shown in Figure 9. Below the compensation temperature the curves do not saturate with the available fields. Neutron diffraction measurements[16] have determined that for T > 50 K a complex noncollinear magnetic structure occurs in $ErFe_3$. The moments on the two inequivalent Er sites, rather than being parallel, are oriented toward the c-axis and the basal plane respectively. All Fe moments are considered to be equal and parallel and are located ≈ 56° from the c-axis.

The temperature dependence of the magnetostriction of $TmFe_3$ is complex, as seen in Figure 11. The room temperature strain of -65 ppm is the smallest of the RFe_3 series. Below 300 K the magnitude of the magnetostriction begins increasing as the temperature is lowered, reaching a maximum negative strain of -325×10^{-6} at 175 K. At this point the magnetostriction decreases in magnitude with further temperature reduction until T = 100 K, where λ was found to remain constant with temperature until a compensation point at 78 K causes a sign reversal. Following this, the magnitude of the strain increases rapidly with temperature reaching -179×10^{-6} at 59 K, the lowest temperature measured. Figure 10 shows the field dependence of the magnetostriction at various temperatures for $TmFe_3$. Below 175 K the strain vs. field curves become increasingly harder to saturate with $|\lambda|$ decreasing with temperature for all field values. At the lowest temperature measured T = 59 K, Figure 10 shows that a large spontaneous magneto-strain of -100 ppm appears at 1 kOe, increasing to -179 ppm at 25 kOe as the magnetization rotates against a large anisotropy. In an effort to further investigate this behavior magnetic moment measurements from 4 - 300 K were performed on $TmFe_3$. The findings from this experiment closely parallel the results from the magnetostriction. Figure 12 shows the magnetization as a function of applied field at T = 300, 188 and 4.2 K. The moment curves depart from saturation at approximately 190 K and are extremely "hard" by 4.2 K. The temperature dependence of the magnetization at H = 3, 10 and 16 kOe is shown in Figure 13. The moment was found to decrease with temperature, even at 16 kOe, with the largest slope occurring in the region 200 K > T > 125 K. Coupling these results with the magnetostriction data, we infer that the easy axis of $TmFe_3$ is in the basal plane at high temperature with a rotation occurring between 100 K and 175 K, leaving the magnetization either along or close to the c-axis.

B. R_6Fe_{23} COMPOUNDS

The R_6Fe_{23} compounds crystallize into a cubic Th_6Mn_{23}-type structure. The magnetostriction of these compounds was found to be well behaved.

The strain in Ho_6Fe_{23} is rather small as in the RFe_3 and RFe_2 compounds. Figure 14 shows the field dependence of the magnetostriction at various temperatures while Figure 17 displays the temperature dependence at H = 25 kOe. At room temperature $\lambda_{\parallel} - \lambda_{\perp} = 88 \times 10^{-6}$ and increases only to 269×10^{-6} at 80 K. A compensation point is observed at 193 K.

The magnetostriction of Er_6Fe_{23} and Tm_6Fe_{23} increases substantially with decreasing temperature due to the rapid ordering of the rare earth sublattice in these two compounds. Figure 17 shows that the magnetostriction of Er_6Fe_{23} at 25 kOe increases nearly 7-fold from -57×10^{-6} at room temperature to -392×10^{-6} at 80 K. A compensation point was found at T = 100 K. Figure 15 shows the magnetostriction of this compound as a function of applied field for various temperatures. The magnetostriction is well behaved with an increasing anisotropy occurring at the lower temperatures.

Tm_6Fe_{23} undergoes the largest change of any of the R_6Fe_{23} compounds (see Figure 17) increasing nearly 10-fold from -37 ppm at 300 K to -362 ppm at 80 K and reaching -388 ppm at T = 55 K, the lowest temperature measured. Tm_6Fe_{23} is expected to have a compensation point below 40 K. Figure 16 shows that the field dependence of the magnetostriction saturates fairly well at all temperatures.

The magnetostrictions of Tb_6Fe_{23} and Dy_6Fe_{23} are not reported on since attempts to prepare single phase compounds were unsuccessful.

The signs of the magnetostriction for the R_6Fe_{23} compounds, as well as all compounds of the RFe_3 series, are consistent with those predicted by the Stevens' factor, α.[17]

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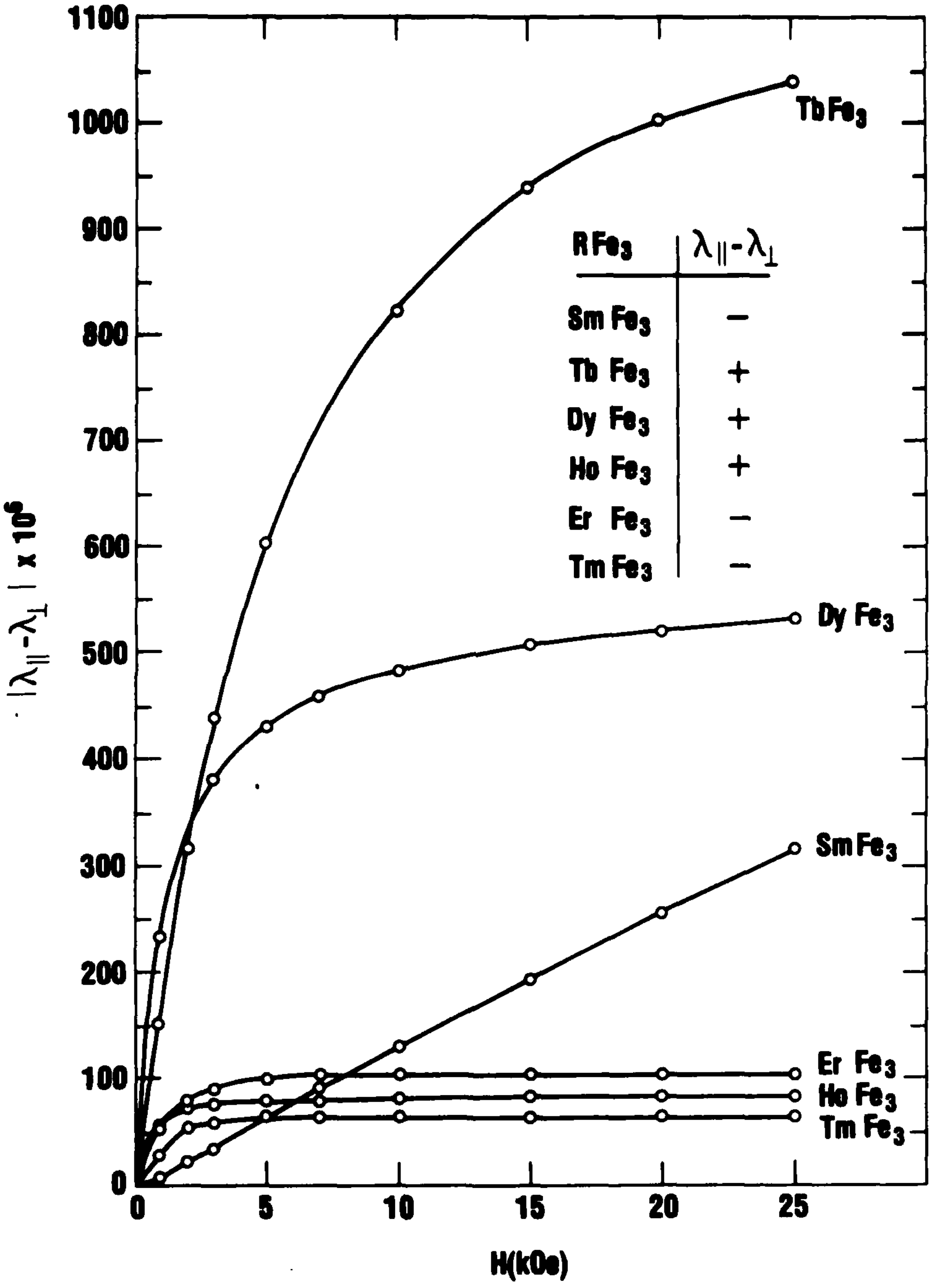

FIGURE 1 ROOM TEMPERATURE MAGNETOSTRICTION AS A FUNCTION OF APPLIED FIELD FOR VARIOUS RFe_3 COMPOUNDS

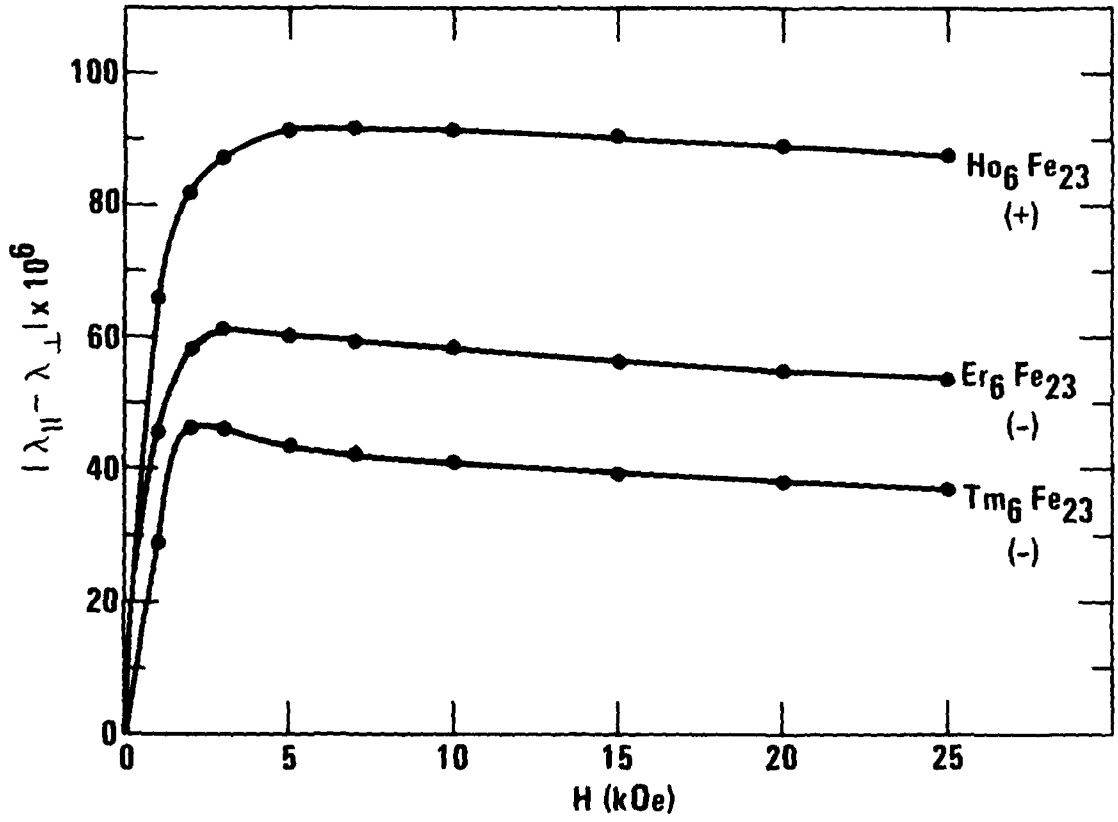

FIGURE 2 ROOM TEMPERATURE MAGNETOSTRICTION AS A FUNCTION OF APPLIED FIELD FOR VARIOUS R_6Fe_{23} COMPOUNDS

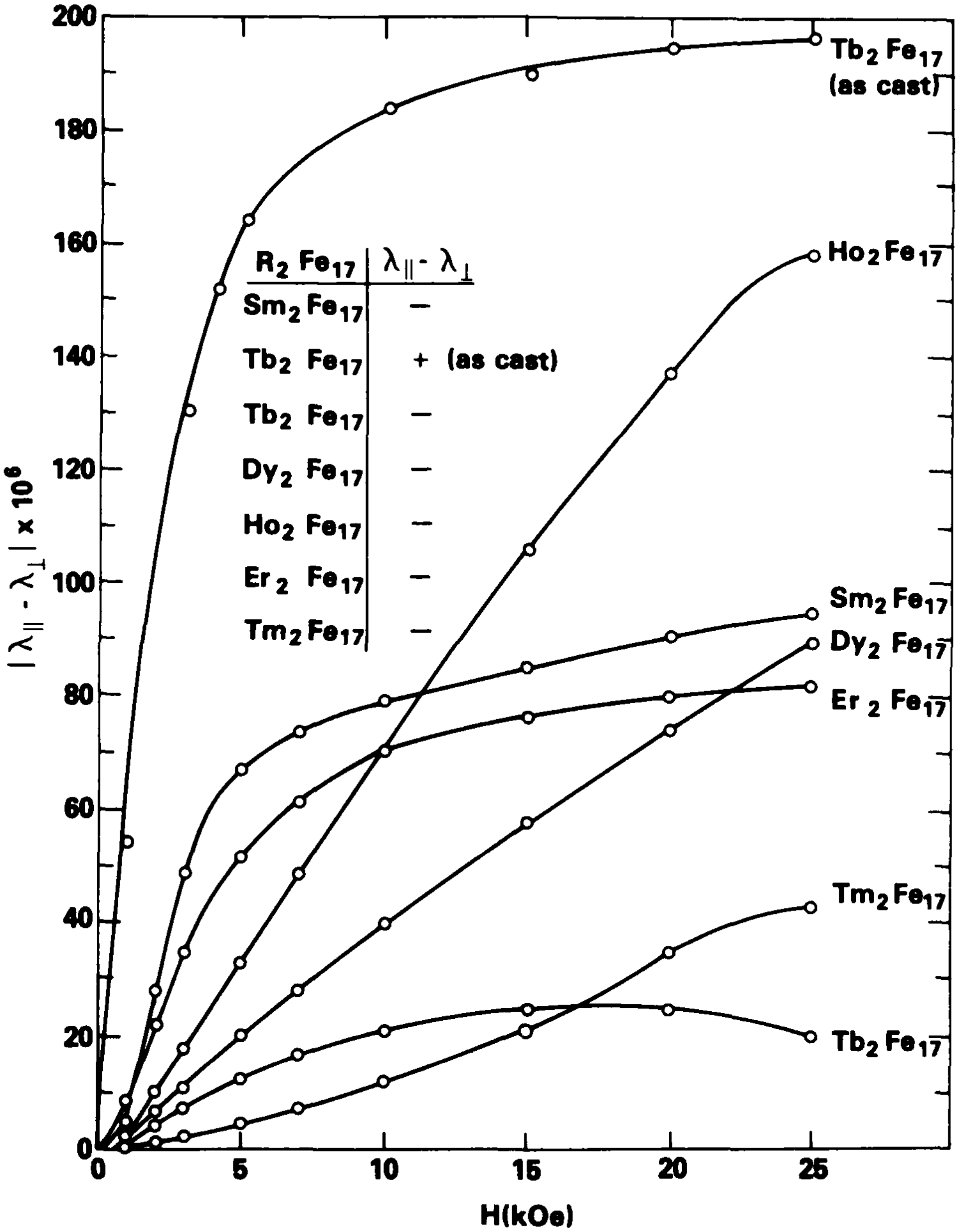

FIGURE 3 ROOM TEMPERATURE MAGNETOSTRICTION AS A FUNCTION OF APPLIED FIELD FOR VARIOUS R_2Fe_{17} COMPOUNDS

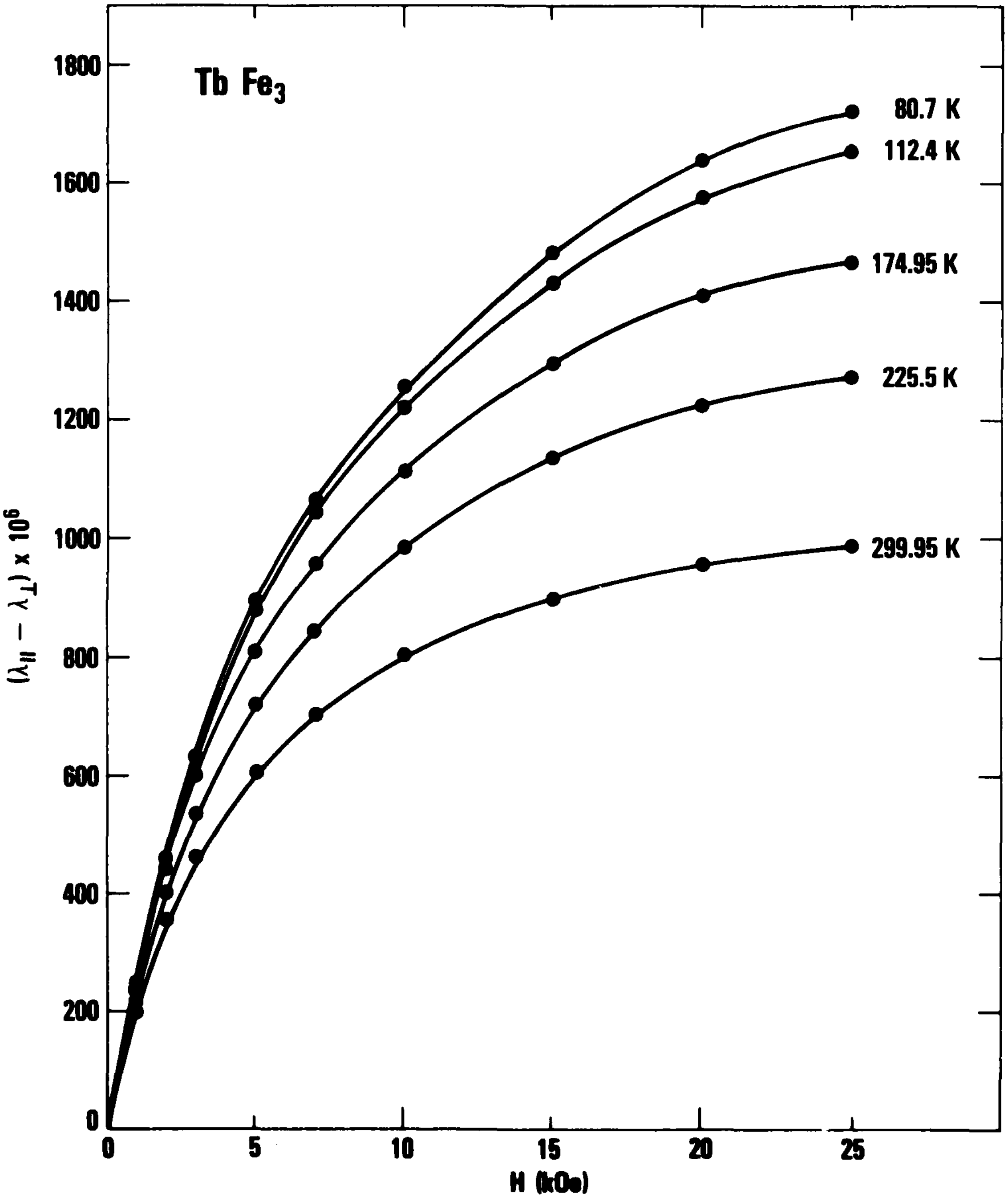

FIGURE 4 MAGNETOSTRICTION AS A FUNCTION OF APPLIED FIELD FOR $TbFe_3$ AT VARIOUS TEMPERATURES

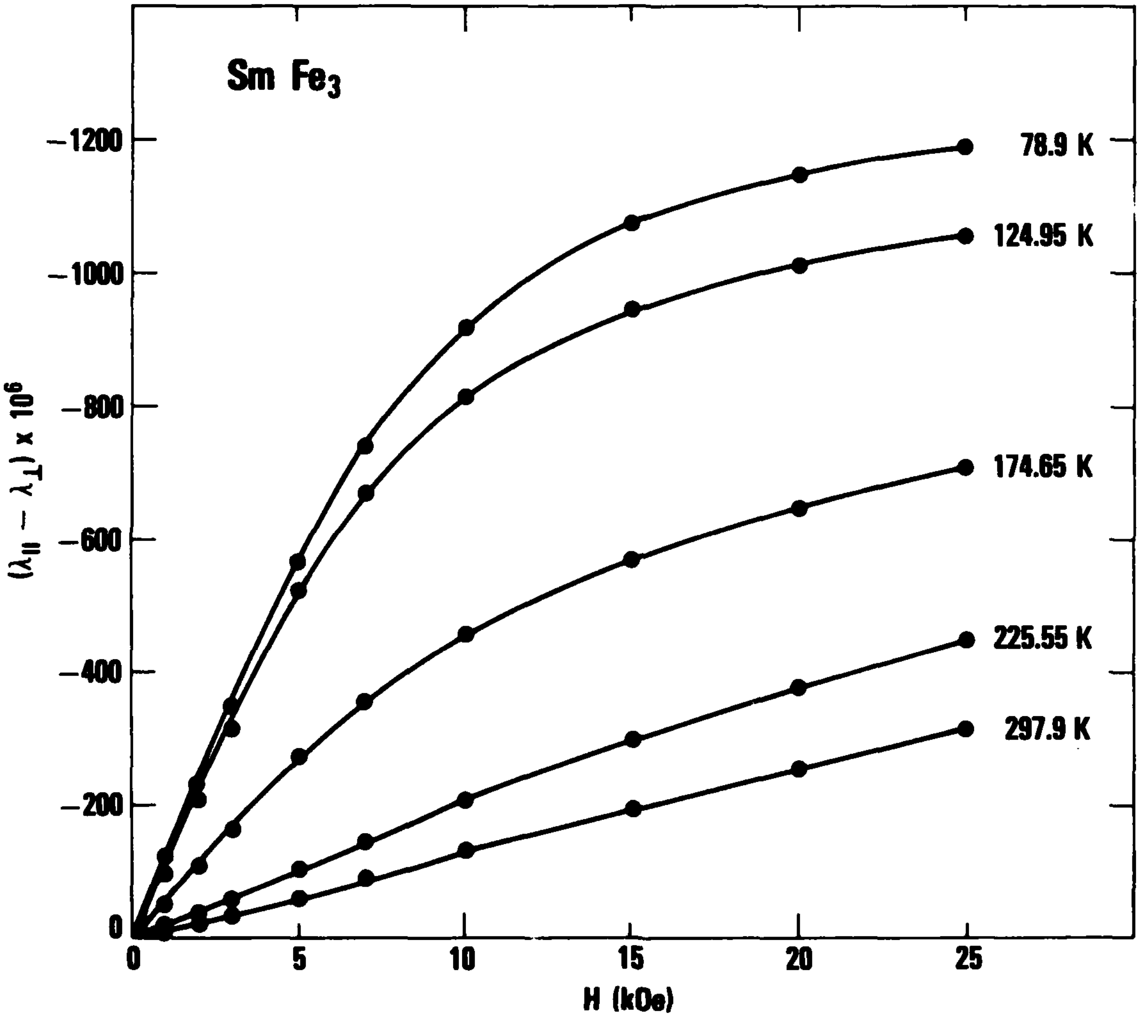

FIGURE 5 MAGNETOSTRICTION AS A FUNCTION OF APPLIED FIELD FOR $SmFe_3$ AT VARIOUS TEMPERATURES

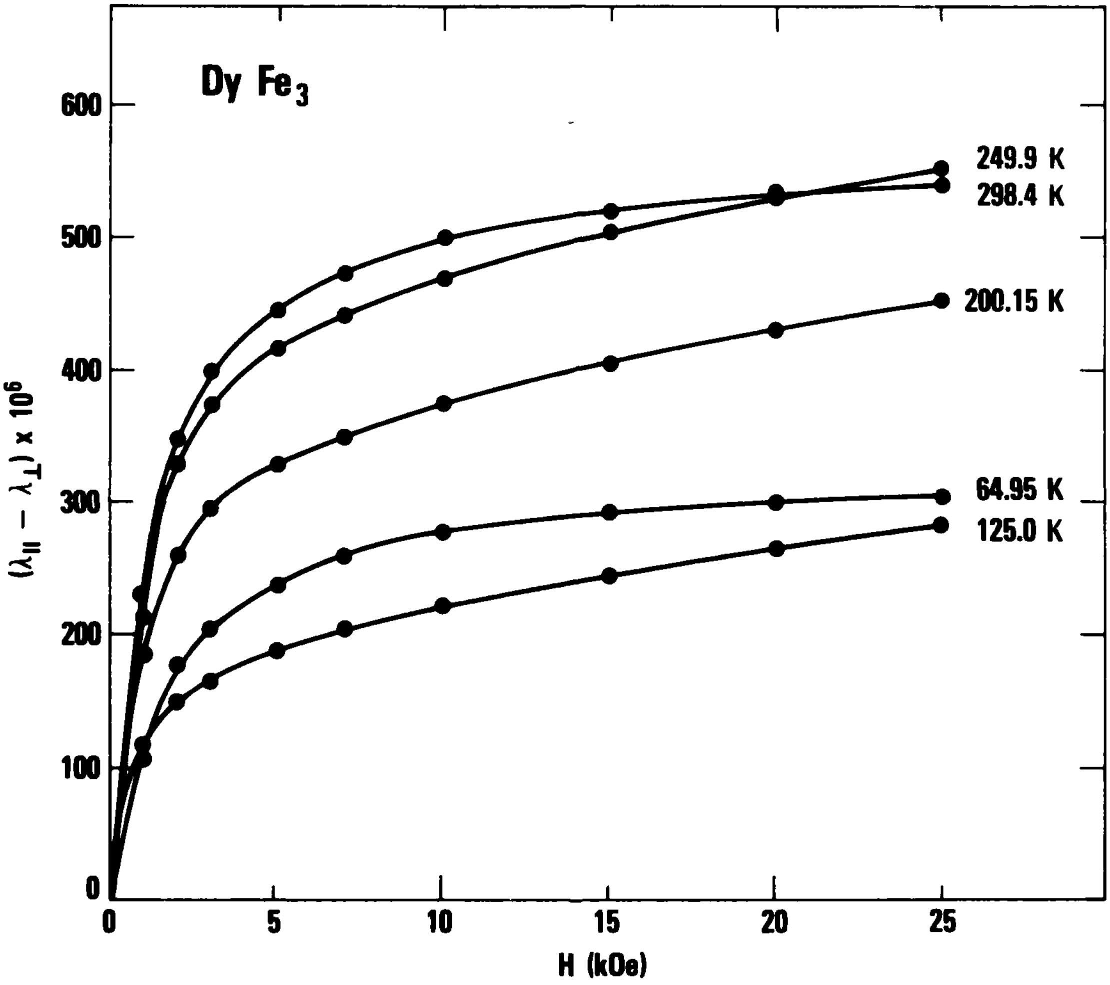

FIGURE 6 MAGNETOSTRICTION AS A FUNCTION OF APPLIED FIELD FOR DyFe$_3$ AT VARIOUS TEMPERATURES

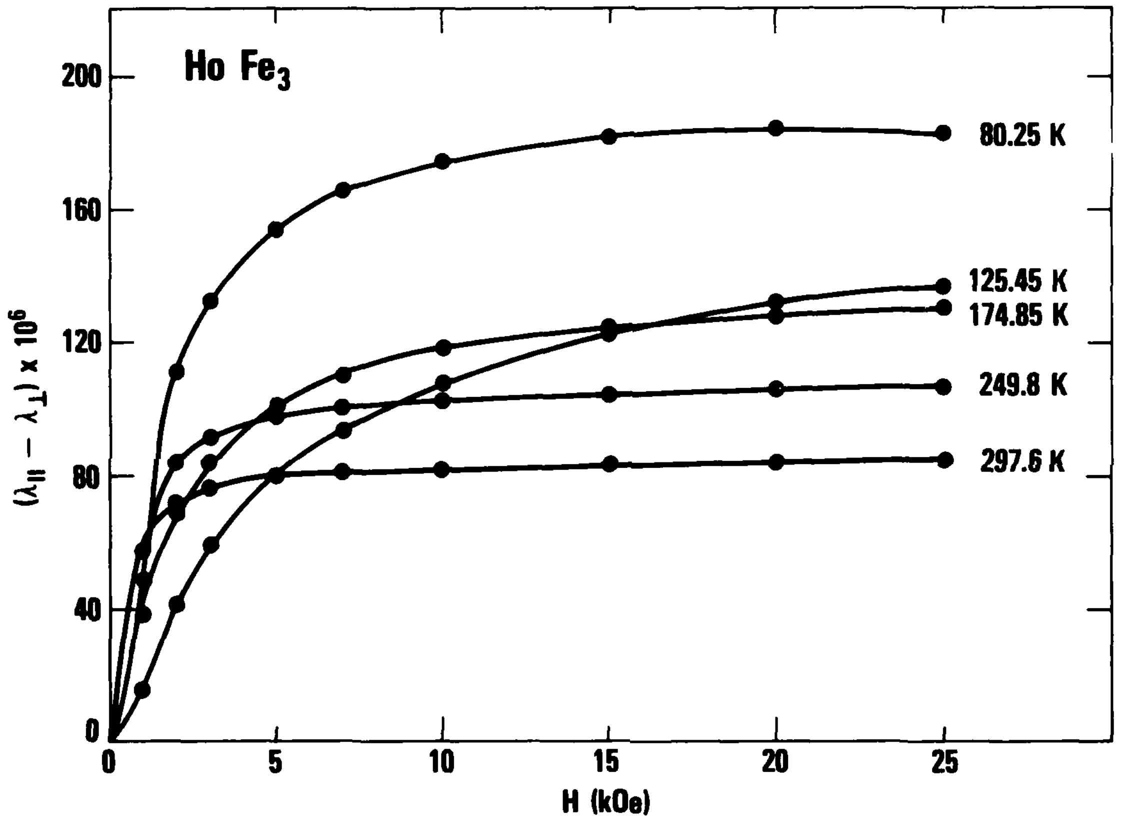

FIGURE 7 MAGNETOSTRICTION AS A FUNCTION OF APPLIED FIELD FOR $HoFe_3$ AT VARIOUS TEMPERATURES

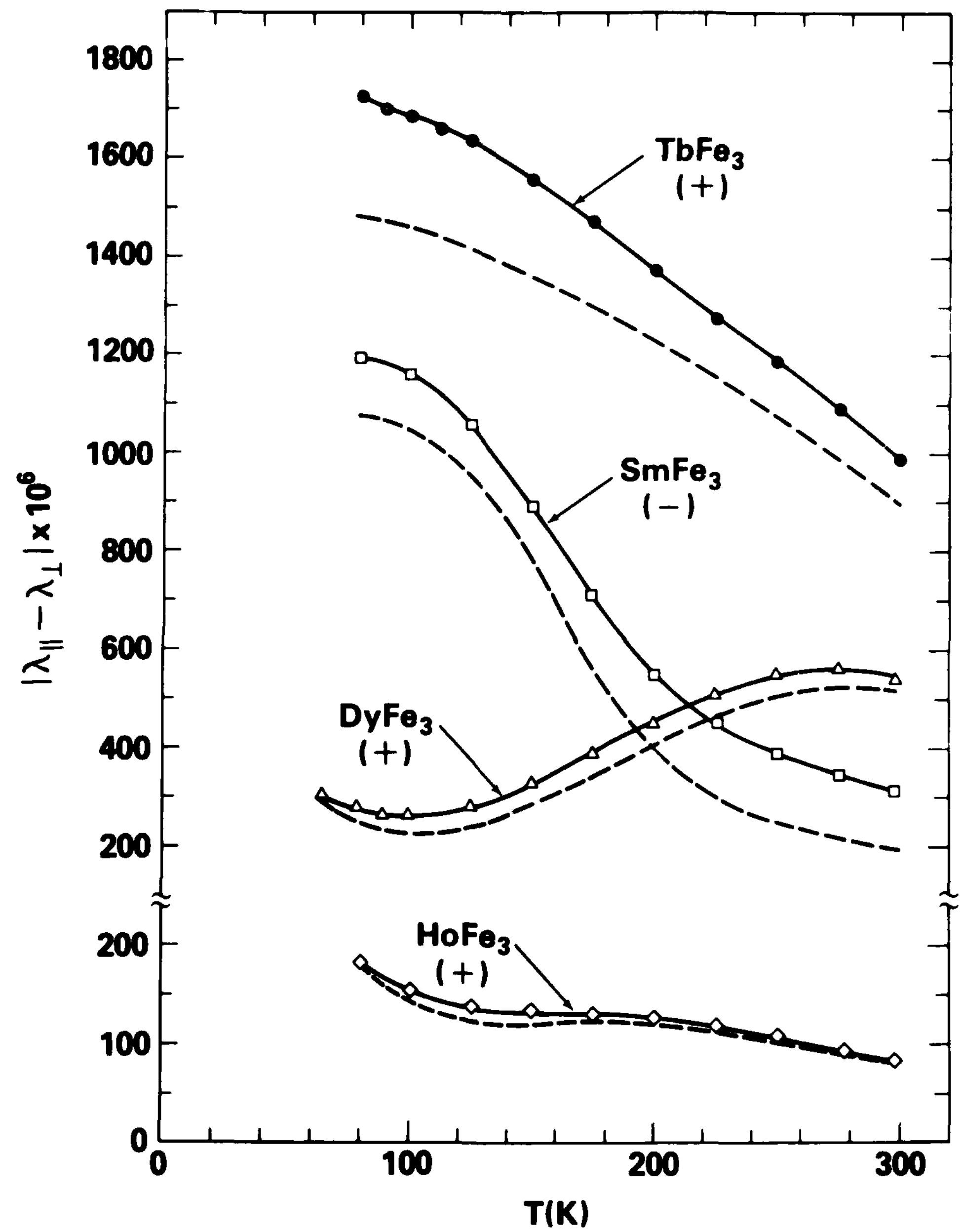

FIGURE 8 TEMPERATURE DEPENDENCE OF THE MAGNETOSTRICTION FOR TbFe$_3$, SmFe$_3$, DyFe$_3$, AND HoFe$_3$ (—— 25kOe, – – –15kOe)

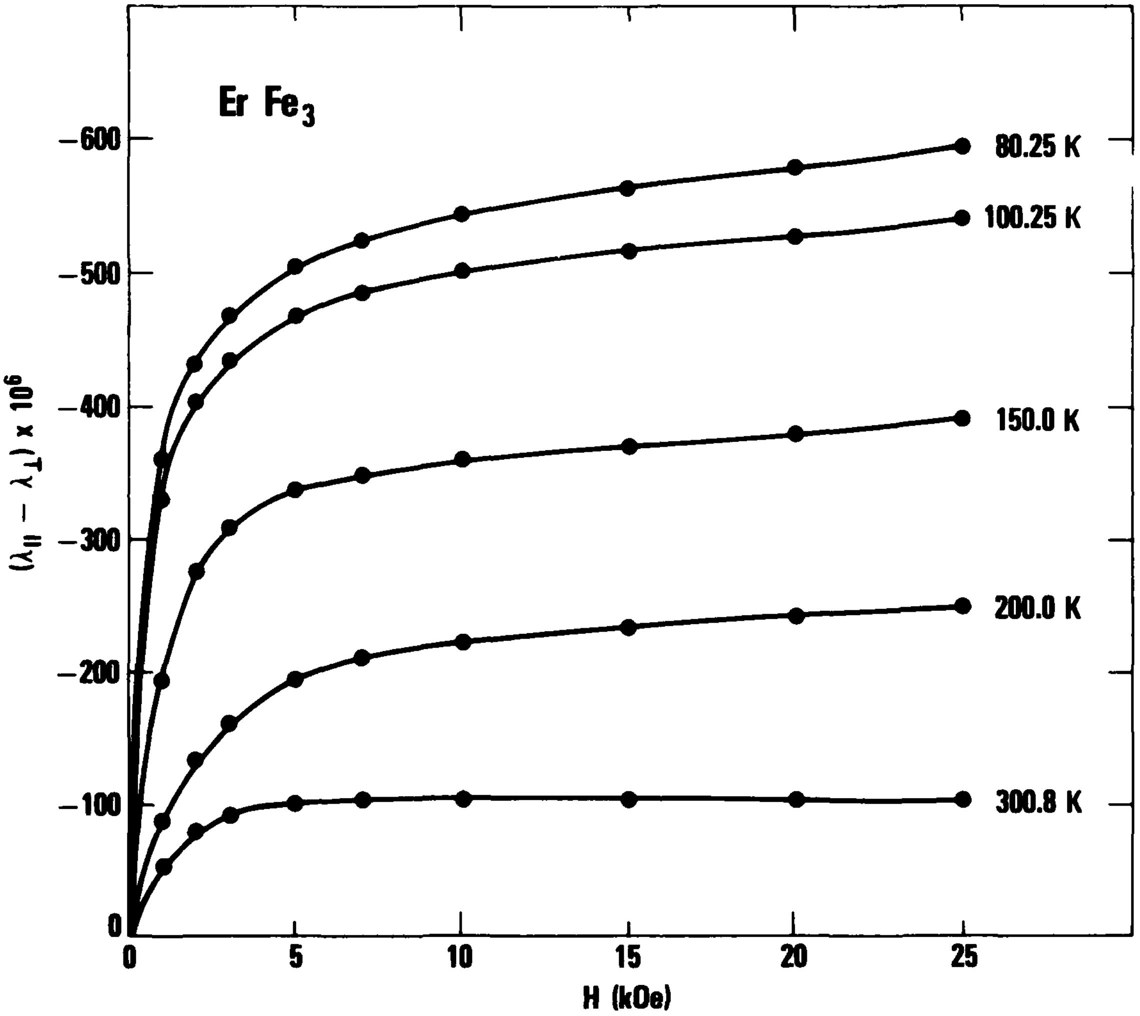

FIGURE 9 MAGNETOSTRICTION AS A FUNCTION OF APPLIED FIELD FOR ErFe$_3$ AT VARIOUS TEMPERATURES

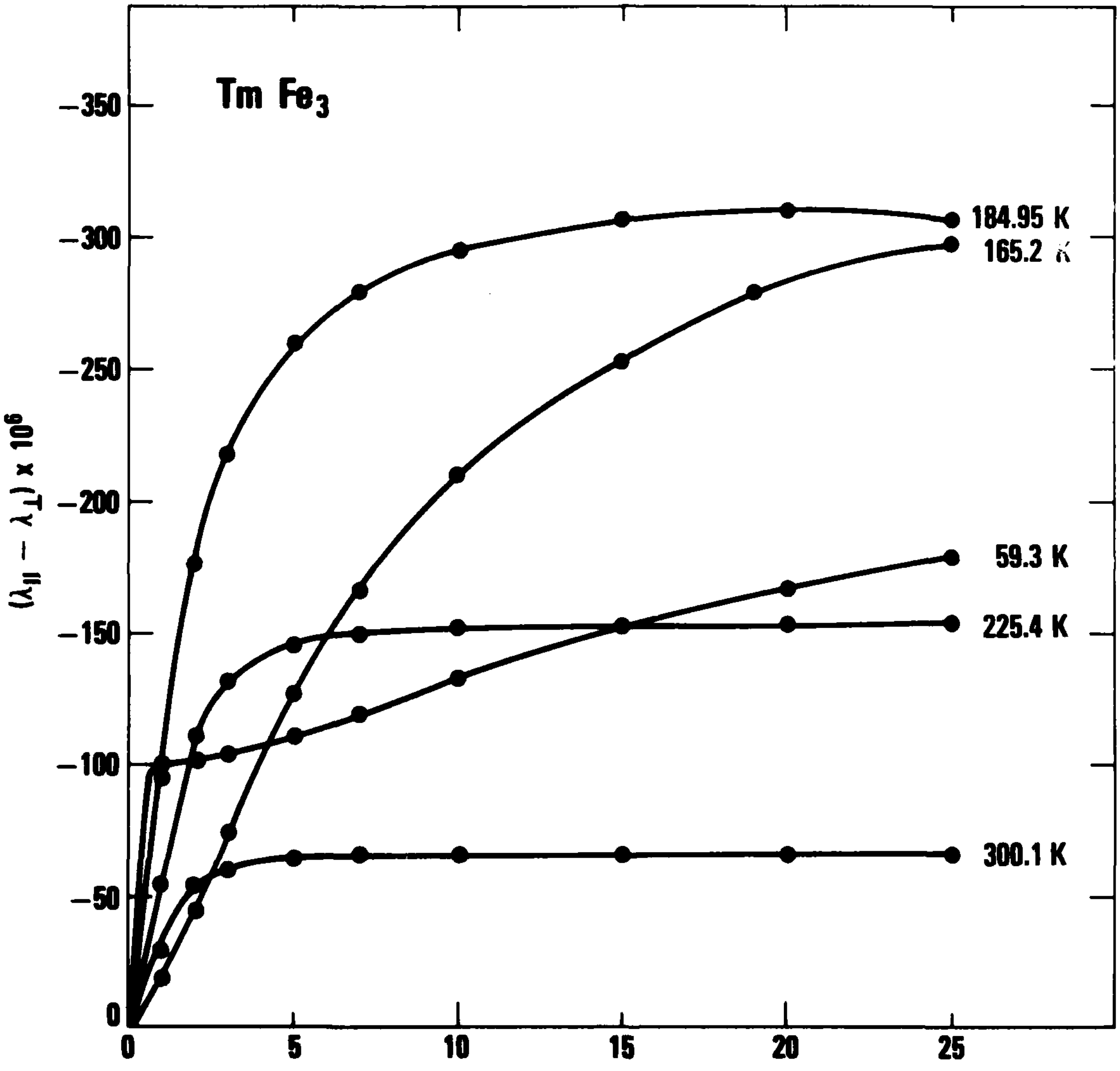

FIGURE 10 MAGNETOSTRICTION AS A FUNCTION OF APPLIED FIELD FOR $TmFe_3$ AT VARIOUS TEMPERATURES

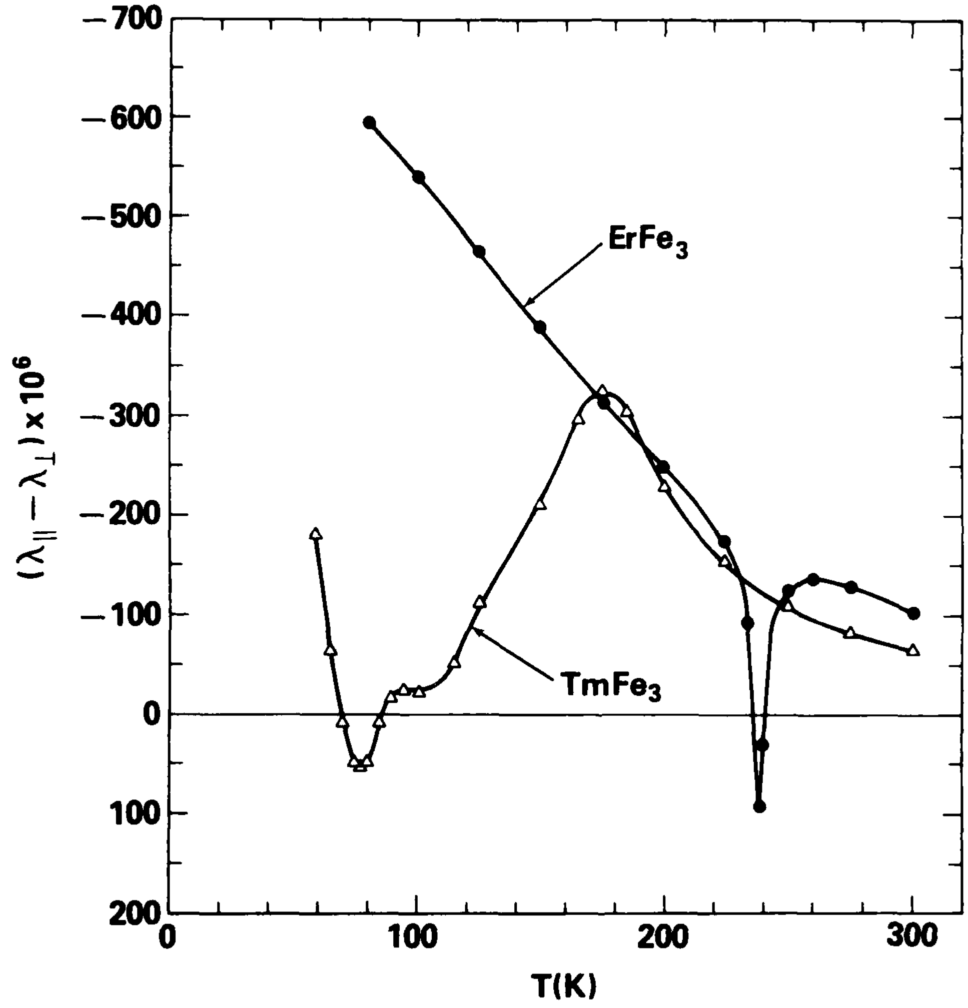

FIGURE 11 TEMPERATURE DEPENDENCE OF THE MAGNETOSTRICTION AT H = 25kOe FOR $ErFe_3$ AND $TmFe_3$

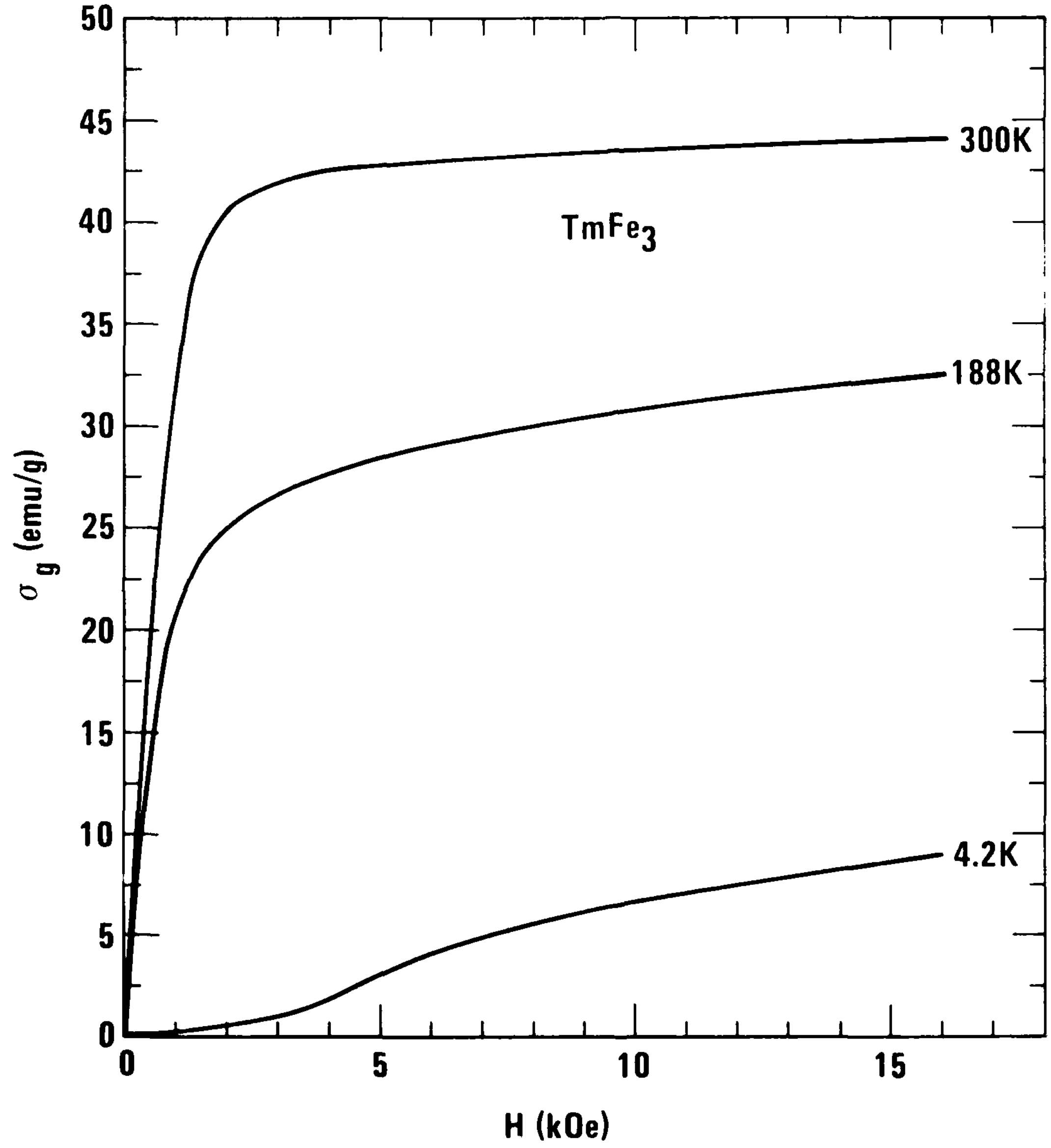

FIGURE 12 MAGNETIZATION AS A FUNCTION OF APPLIED FIELD FOR $TmFe_3$ AT T = 4.2, 188 AND 300K

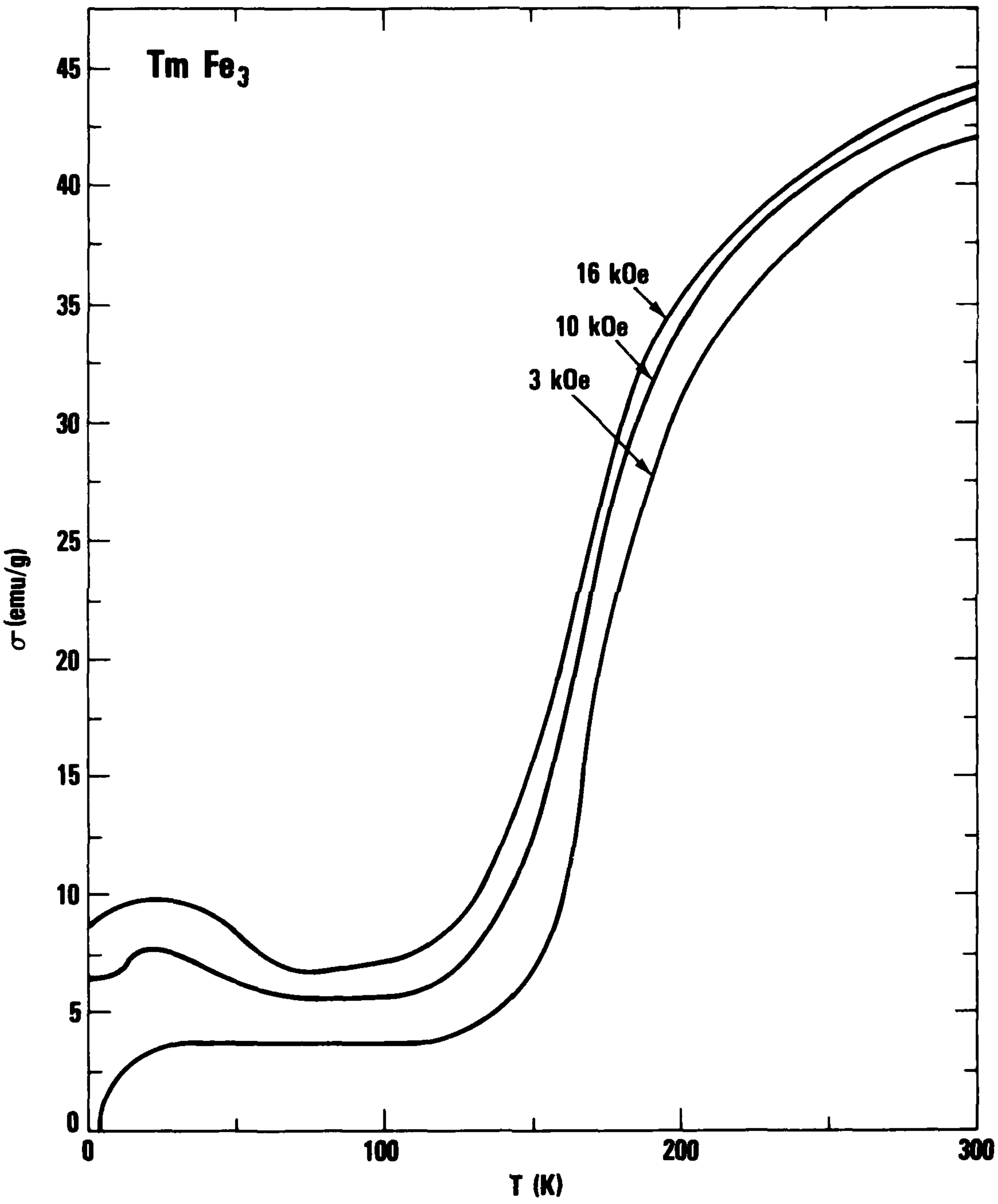

FIGURE 13 TEMPERATURE DEPENDENCE OF THE MAGNETIZATION FOR TmFe$_3$ AT H = 3, 10 AND 16kOe

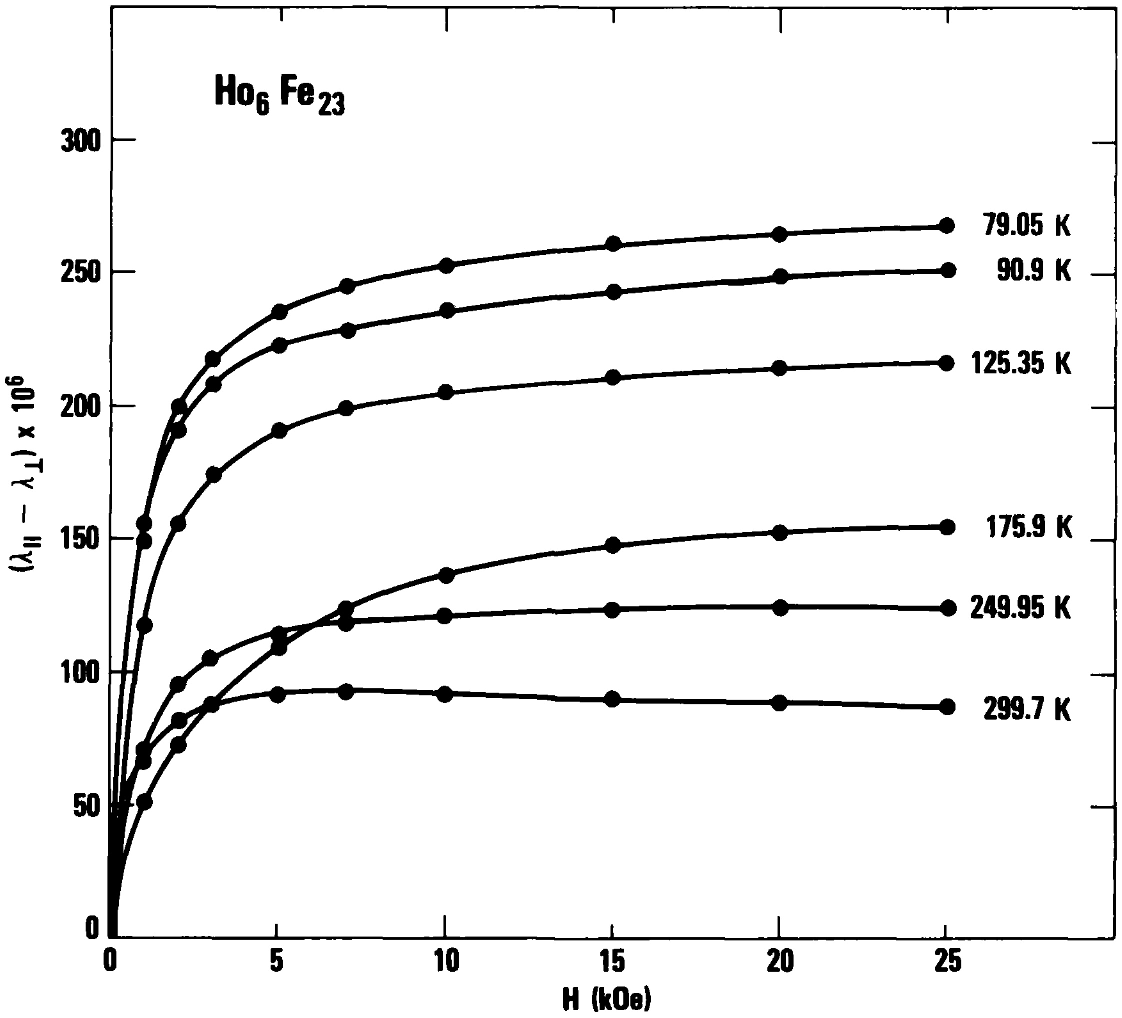

FIGURE 14 MAGNETOSTRICTION AS A FUNCTION OF APPLIED FIELD FOR Ho_6Fe_{23} AT VARIOUS TEMPERATURES

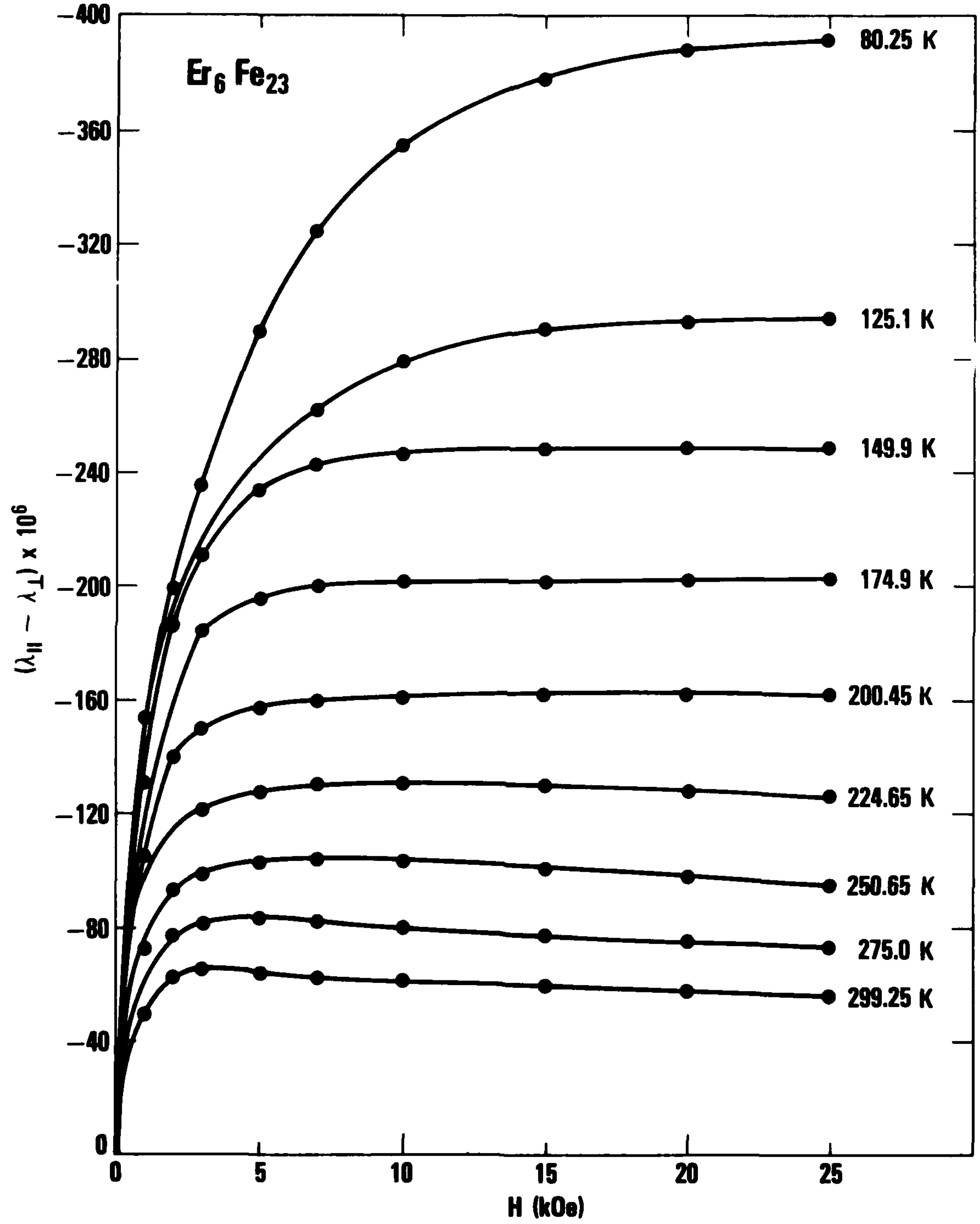

FIGURE 15 MAGNETOSTRICTION AS A FUNCTION OF APPLIED FIELD FOR Er_6Fe_{23} AT VARIOUS TEMPERATURES

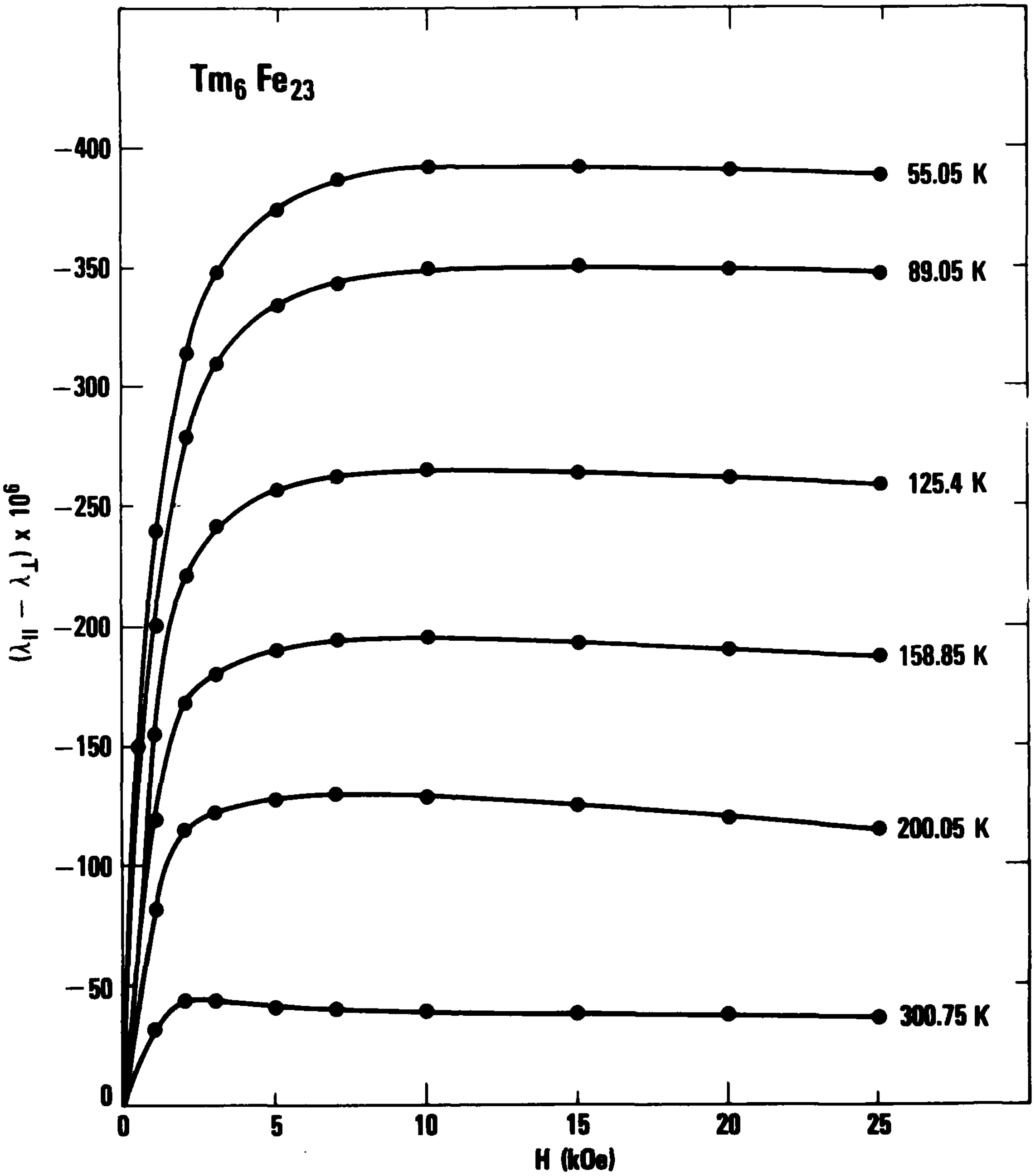

FIGURE 16 MAGNETOSTRICTION AS A FUNCTION OF APPLIED FIELD FOR Tm_6Fe_{23} AT VARIOUS TEMPERATURES

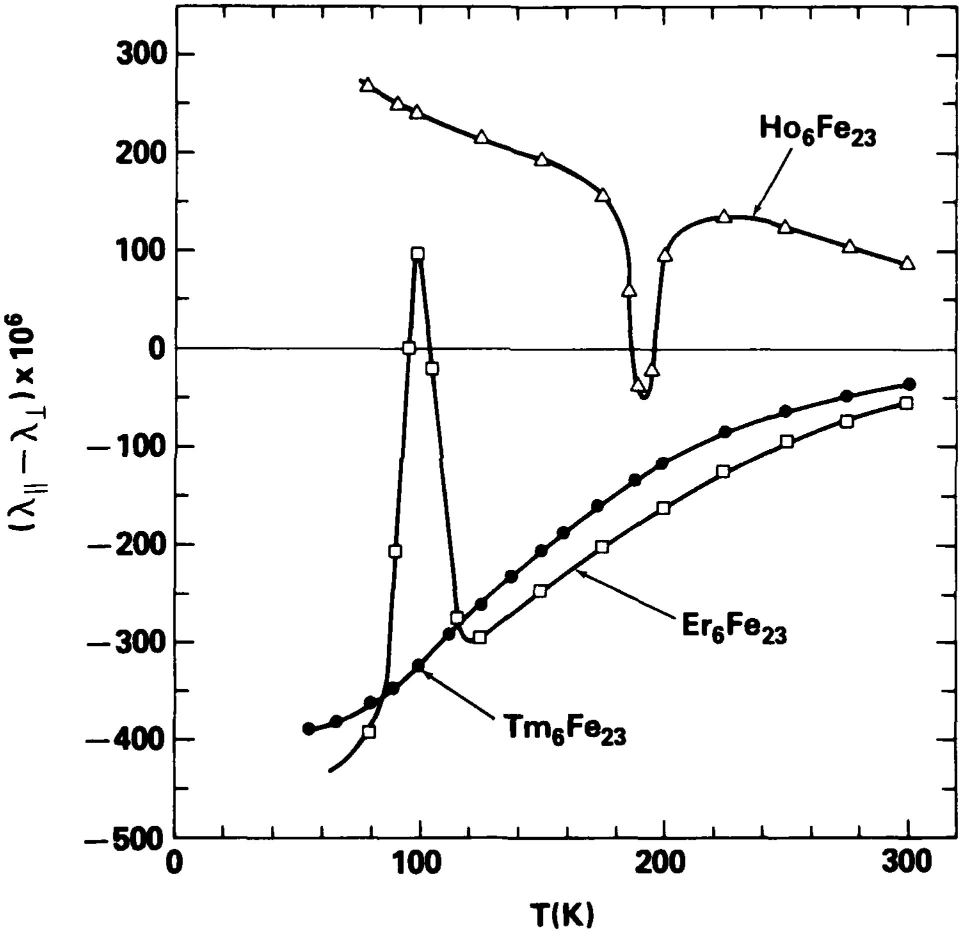

FIGURE 17 TEMPERATURE DEPENDENCE OF THE MAGNETOSTRICTION AT H = 25kOe FOR Ho_6Fe_{23}, Er_6Fe_{23} AND Tm_6Fe_{23}

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