Effects of Detector Nonlinearity and Specimen Size on the Apparent Thermal Diffusivity of NIST 8425 Graphite

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With the use of a graphite thermal conductivity standard it is demonstrated that optical detector non-linearity, coupled with excessive laser pulse energies, is primarily responsible for the anomalous specimen size dependence of the thermal diffusivity measured by the laser-pulse technique. High laser pulse energies also result in an anomalous positive temperature dependence for thin specimens near room temperature, in contrast to the expected negative temperature dependence. Using moderately thick specimens and attenuated laser pulses yields excellent agreement with thermal diffusivity calculated from standard thermal conductivity data.

1. INTRODUCTION

The flash technique, pioneered by Parker *et al.* [1], has become a widely accepted method for measuring the thermal diffusivity of a wide variety of materials over a wide temperature range. It requires samples of small size and simple geometry and allows rapid data acquisition, which is highly advantageous when compared to the time-consuming steady-state methods for measuring thermal conductivity. Further refinements in data analysis, which account for the effect of pulse-width and heat losses [2, 3], yield accurate data for thermal diffusivity with an experimental error of only a few percent.

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In general, any method used to determine physical property data should yield results which are independent of operating conditions and which agree with the data obtained with other techniques. In this respect, the flash technique has yielded results which appear to be anomalous. Taylor [4–6] reported that the calculated values (without correcting for detector nonlinearity) for the thermal diffusivity of thin samples decreases with increasing pulse energy above a threshold energy but that this problem could be overcome by decreasing laser energy or by correcting for detector nonlinearity.

Similar results were reported by Groot [7]. Also, a pronounced specimen size effect, characterized by a decreasing thermal diffusivity with decreasing specimen thickness, was observed for stainless steel, iron, molybdenum, copper, and aluminum nitride [4–11]. This effect of specimen size has been attributed to laser-beam nonuniformity [7], temperature-dependent thermal diffusivity [3, 11], nonlinearity of the IR detector commonly used to measure the transient specimen temperature [4–7, 11], and a contribution of radiative heat transfer between the front and the rear face of the specimen [12]. The latter effect could be operative in dielectric materials such as AlN but is not expected to play a role in metallic materials, such as copper.

The effect of specimen size on thermal diffusivity is inherent in the characteristics of the solid-state lasers most commonly used as the flash source for thermal diffusivity measurements. For reliable data, the radiation intensity across the width of the laser pulse should be as uniform as possible. Generally at low laser pulse energies, i.e., low discharge voltages for the flash-lamp, the intensity profile across the pulse can be highly nonuniform. The discharge voltage, therefore, should exceed some minimum value in order to assure beam uniformity. This implies that the flash energy is not adjusted to the specimen thickness. As a result, thin specimens will experience a higher temperature rise than thicker specimens. This can lead to two effects.

First, for crystalline dielectric materials such as AlN, with a thermal diffusivity/conductivity which exhibits a strong negative temperature dependence, especially near room temperature, a higher mean temperature level for thinner specimens will lead to a lower value for the measured thermal diffusivity than for the thicker specimens which will have a lower mean temperature rise. This effect, however, probably does not explain the factor of two to three variation in thermal diffusivity observed over the practical range of AlN specimen thickness, nor does it explain a similar range for Cu [10] with a thermal diffusivity with a modest temperature dependence when compared to AlN.

The second effect arises from the general nonlinearity of the infrared

(IR) detectors used for the remote sensing of the transient temperature of the rear surface of the specimen [4–7]. It is common practice to determine the thermal diffusivity from the time period $(t_{1/2})$ required for the specimen rear face temperature to reach one-half of its final value. This value readily permits calculation of the thermal diffusivity, taking into account the effect of the finite-pulse width and/or heat losses [2, 3], if required. Large increases in specimen temperature cause the detector output to rise proportionally higher than the relative change in specimen temperature. If this effect is not accounted for in the analysis of the data, the resulting values for the thermal diffusivity are expected to deviate from the true value. Using excessively high pulse energies to result in large temperature increases, Hasselman and Merkel [11] found a variation in the thermal diffusivity of AlN at room temperature of about a factor of three for specimen thicknesses ranging from about 1 to 5 mm, when the thermal diffusivity was evaluated assuming that $t_{1/2}$ for the transient detector output represented $t_{1/2}$ for the transient temperature of the specimen rear surface. Clearly, this indicates the need for a more detailed data acquisition or analysis, which takes the detector nonlinearity into account. Hasselman and Merkel [11] found that detector nonlinearity could affect the thermal diffusivity values for specimen temperature increases as low as 0.5°C. Experimentally, specimen temperature increases can be reduced significantly, thereby simplifying the data analysis, by attenuating the pulses from a ruby or Nd-glass laser by passing them through an aqueous solution of $CuSO_4$ of the proper concentration [4–7]. The effectiveness of this approach was demonstrated by Hasselman and Merkel [11], who found the specimen size effect for AlN greatly reduced, and a residual size effect attributed to the negative temperature dependence of the thermal diffusivity. The nonlinearity of the detector, however, was the major contributing factor for the observed size effect. The highest thermal diffusivity values obtained with strongly attenuated beams occurred at a value of specimen thickness near 3.5 to 4.0 mm. Unfortunately, for the particular AlN studied by Hasselman and Merkel, no data for the thermal diffusivity/conductivity obtained by methods other than the laser-flash technique were available. Therefore, no independent check on the true value of the thermal diffusivity for their material was possible.

The purpose of the present study was to conduct an investigation of the effects of pulse energies, pulse attenuation, and specimen thickness on thermal diffusivity for a standard graphite material for which the thermal conductivity was obtained by the National Institute of Standards and Technology (NIST).

2. EXPERIMENTAL

2.1. Material

The thermal conductivity standard selected for this study consisted of graphite RM 8425 obtained from the National Institute of Standards and Technology (NIST) in the form of a circular rod with a diameter of 1.27 cm and a length of 5.0 cm. The density and electrical resistivity of this particular piece were $1730 \, kg \cdot m^{-3}$ and $14.5 \times 10^{-6} \, \Omega \cdot m$. Using a slow-speed diamond saw, a number of disk specimens ranging in thickness from 0.075 to 5.15 mm were cut from the rod.

2.2. Measurement of the Thermal Diffusivity

The thermal diffusivity of the disk samples was measured by the flash technique using a Nd–glass laser (Model 936G4L-1, Laser Applications, Inc., Winter Park, FL) as the flash source. The beam diameter was 1.25 cm. The maximum pulse energy was approximately 55 J. The pulse duration was approximately 800 μs. The pulse energy as a function of time could be most closely approximated by a triangular shape with the constant b in the theory of Heckman [3] near zero. As judged by the flash imprint on photographic paper a minimum value for the lamp discharge voltage of 5 kV, with corresponding pulse energy of approximately 20 J, was required to yield a pulse with reasonably uniform intensity distribution across its width. Attenuation of the pulse energy was achieved by passing the pulse through an aqueous solution of $CuSO_4$ contained within spectroscopic cells. Based on data established by Groot [7], $CuSO_4$ concentrations were prepared to result in beam attenuations of 60, 78, and 90%. The transient temperature of the rear surface of the specimens was measured with an InSb infrared detector (Model IS-030, Electro-Optical Systems, Inc., Malvern, PA). An optical filter (Part T-LPS-2.0, Infrared Industries, Orlando, FL), which transmits radiation with a wavelength greater than 2 μm, was placed directly against the sapphire window of the detector, in order to protect the sensing element from possible damage as the result of direct laser impact due to pulse flash-by around the specimen or other reasons. For measurement at room temperature (21°C), the specimens were mounted directly in front of the detector window at a distance of approximately 6 mm and separated from the window by a 5 cm diameter thermally insulating rubber disk, which covered the front face of the detector. The specimen was viewed by the detector through a 5 × 5-mm square hole cut into the center of the rubber disk. The specimens were supported by two thin pins located below the hole in the disk. Any possible effects of laser flash-by and associated heating of the rubber disk or direct viewing

by the detector were eliminated by shielding the outer edges of the graphite specimens by about 0.1 mm, using a circular diaphragm.

For measurements above room temperature up to about 300°C, the specimens were placed in a graphite holder held within a 5-cm-diameter band heater. Using a controller with a thermocouple in direct contact with the side of the specimen, the temperature was held constant to within 1°C. The distance from the specimen to the detector was approximately 20 cm. The radiation from the sample was focussed on the detector with a sapphire lens. The detector viewed a circular area of the specimen approximately 8 mm in diameter.

For measurements at all temperatures, the transient output from the detector was recorded on the screen of a storage oscilloscope. The value for $t_{1/2}$ was measured directly from the oscilloscope screen. A total of five measurements of $t_{1/2}$ was made at any given value of specimen thickness, temperature, and laser pulse energy. The specimen was allowed to return to thermal equilibrium between successive measurements.

For the calculation of thermal diffusivity it was assumed that $t_{1/2}$ for the transient detector output equaled $t_{1/2}$ for the transient temperature rise of the rear surface of the specimens. The effect of the finite pulse-width and heat losses were taken into account with the theory of Heckman [3]. The effect of heat loss over the duration of $t_{1/2}$ for all values of specimen thickness and pulse energies was found to be insignificant. For the thinnest specimen subjected to a pulse energy of 20 J and attenuated by 90%, the finite-pulse correction factor amounted to 1.264 and was lower for higher values of specimen thicknesses and pulse energies. The change in specimen thickness with increasing temperature was taken into account using a value for the coefficient of thermal expansion of $7.4 \times 10^{-6} °C^{-1}$, ascertained from the data of Hust [13]. For the thicker specimens, thermal diffusivity could not be determined at the lower values of pulse energy and higher values of attenuation, as the temperature rise of the specimens was too low to yield a detector signal of sufficient strength for reliable measurement.

The output of the detector as a function of specimen temperature was measured to establish its degree of nonlinearity. For the specimen at room temperature the detector output was measured using a 5-mm-thick graphite disk with a thermocouple inserted in a hole near the midplane of the disk, drilled to a depth near the outer edge of the viewing area. The detector viewed the graphite disk through the identical hole in the thermally insulating disk. The graphite disk was heated uniformly by a remote radiation source. The detector output and the temperature of the graphite disk were recorded simultaneously.

For temperatures in excess of 100°C, the detector output was measured with the identical equipment used for measurement of the

thermal diffusivity. The specimen temperature was obtained from the temperature controller. For measurement of the detector output at room temperature and above 100°C, the optical filter was kept in place, thereby permitting a quantitative assessment of the effect of the detector non-linearity on the experimental thermal diffusivity results.

The specific heat of the graphite was measured using a differential scanning calorimeter (Model DSC-4, Perkin–Elmer, Norwalk, CT).

From the values of thermal conductivity (K) listed by Hust [13], the thermal diffusivity (κ) was calculated from

$$\kappa = K/\rho c$$

where ρ is the density and c is the specific heat. The change in density with temperature was accounted for using the coefficient of thermal expansion of $7.4 \times 10^{-6}\,°\text{C}^{-1}$, inferred from the data of Hust [13]. As this report lists only data for the specific heat above 400 K (127°C), the thermal diffusivity was calculated from the NIST data using specific heat data obtained for the sample of this study from room temperature to 300°C.

3. RESULTS, DISCUSSION, AND CONCLUSIONS

Figure 1 shows the dependence of measured thermal diffusivity on specimen thickness for four values of pulse energy at 21°C, without beam attenuation. These data increase with decreasing pulse energy and cover a range of almost a factor of three. At the higher values of pulse energy the thickness dependence agrees with the corresponding data for AlN and Cu [10, 11]. The maximum values were obtained at a thickness near 4 mm. The decrease in thermal diffusivity at the highest value of specimen thickness is attributed to an unfavorable ratio of specimen thickness to diameter.

Included in Fig. 1 is the value for thermal diffusivity calculated from the thermal conductivity reported by NIST and the density and the specific heat listed in Table I. All data in Fig. 1 lie below the calculated value.

Figure 2 shows the data for thermal diffusivity measured with a pulse energy of 20 J attenuated by 0, 60, 78, and 90%. In general, the thermal diffusivity increases with increasing degree of attenuation. Within the scatter of the data, the values of thermal diffusivity measured at the highest degree of attenuation show excellent agreement with the values calculated from the data provided by NIST [13] and show no effect of specimen thickness.

Figure 3a, b, and c show the temperature dependence of the measured thermal diffusivity for specimen thickness of 0.075, 0.217, and 0.404 cm, respectively, for a range of values of pulse energies and attenuation. These
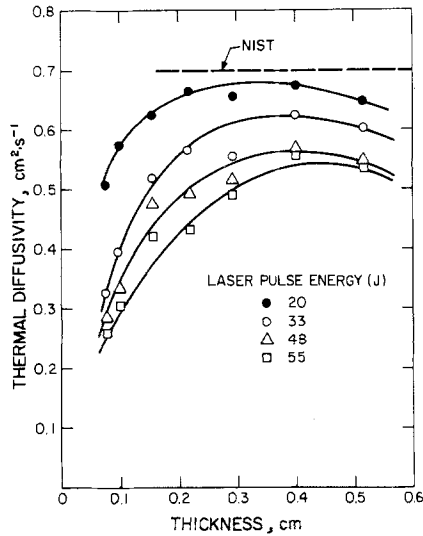
Fig. 1. Effect of specimen thickness on measured values for the thermal diffusivity of NIST 8425 graphite at 21°C for a range of laser pulse energies without laser beam attenuation.

data indicate that the effect of pulse energy and specimen thickness decreases with increasing temperature. For the specimen with a thickness of 0.404 cm, no significant effect of pulse energy on the measured value of thermal diffusivity exists at 300°C.

Noteworthy is the effect of pulse energy on the relative temperature dependence at the lower temperatures. For the thinnest specimen, the ther-

Table 1. Specific Heat of NIST 8425 Graphite

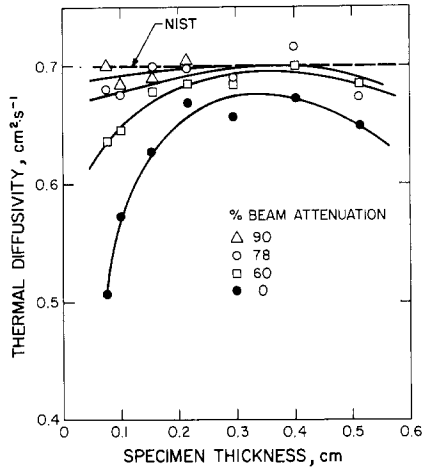| Temperature (°C)	Specific heat ($J \cdot g^{-1} \cdot K^{-1}$)
25	0.751
50	0.787
100	0.904
150	1.018
200	1.128
250	1.232
300	1.338
350	1.470
400	1.556
450	1.601

Fig. 2. Effect of specimen thickness on measured values for the thermal diffusivity of NIST 8425 graphite at 21°C for a pulse energy of 20 J and range of values of beam attenuation.

mal diffusivity exhibits a positive temperature dependence at the highest value of pulse energy, in direct contrast to the expected strongly negative temperature dependence. This effect is less pronounced for the specimen with a thickness of 0.217 cm. The expected strongly negative temperature dependence is observed, however, for the specimen with thickness of 0.404 cm. Critical to note is that for all three specimens, at the lowest values of pulse energies and highest values of pulse attenuations at which data could be obtained, the data for the thermal diffusivity as well as its temperature dependence show excellent quantitative agreement with the data ascertained from the values for the thermal conductivity data supplied by NIST [13].

In general, the data presented in Figs. 1, 2, and 3 indicate that the use of excessive pulse energies, especially for thin specimens near room temperature, can result in measured values of thermal diffusivity which can be a great deal lower than the true values. In order to offer a plausible explanation of this effect, Figs. 4a and b show the voltage output of the detector as a function of specimen surface temperature, for temperature rises of about 20 K above a laboratory ambient temperature of 293.75 K and specimen temperatures above 373 K (100°C) to as high as 600 K (323°C), respectively. Figure 4a clearly indicates that for specimens initially at room temperature, the effect of detector nonlinearity should already influence the results obtained for increases in temperature of only 2°C. In
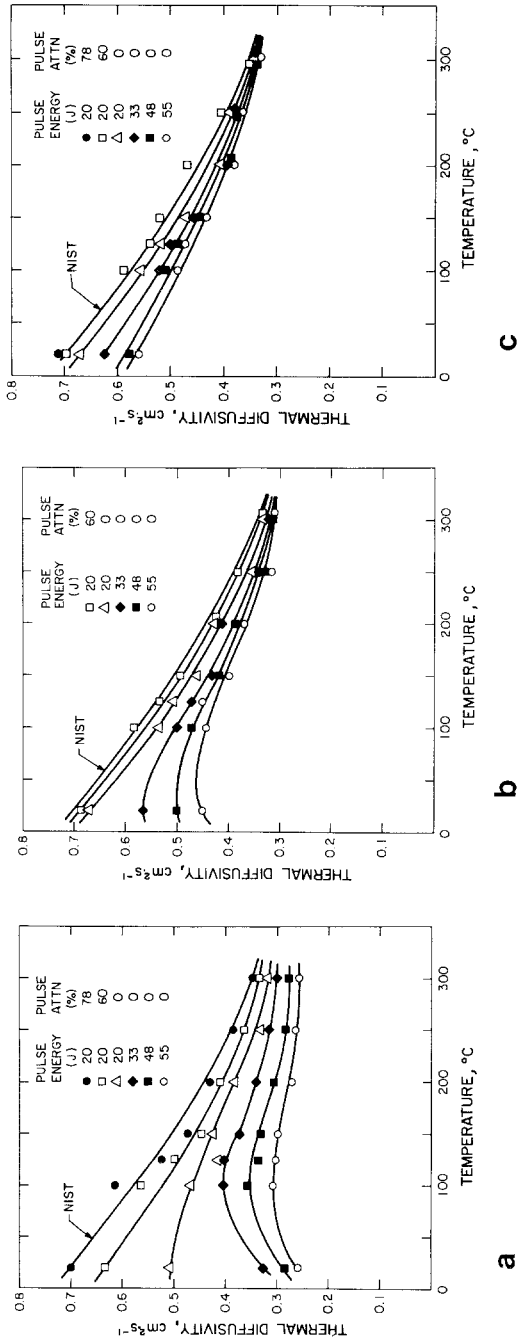
Fig. 3. Temperature dependence of the thermal diffusivity of NIST 8425 graphite for range of values of pulse energies and attenuation for values of specimen thickness: (a) 0.075; (b) 0.217; (c) 0.404 cm.
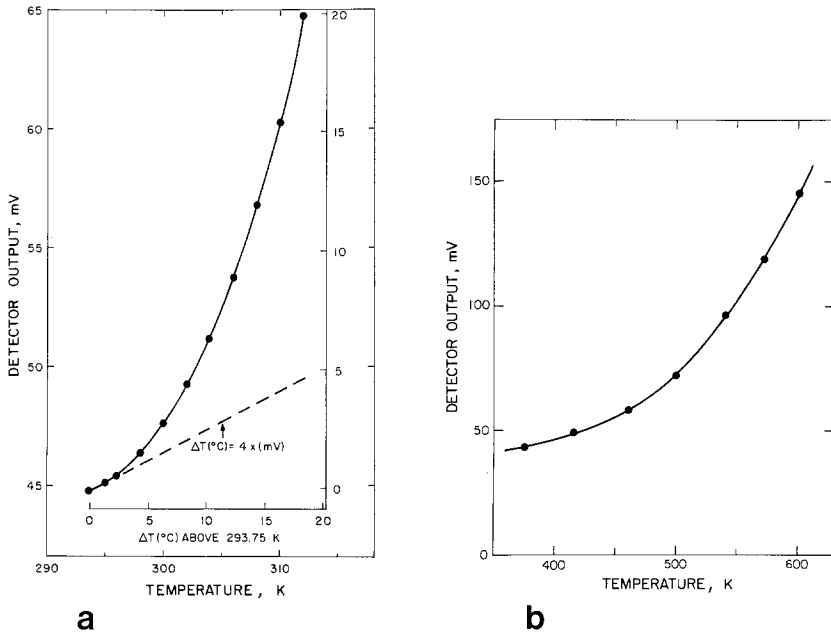
Fig. 4. Dependence of output of InSb detector at 21°C on surface temperature of graphite sample viewed by the detector (a) near room temperature and (b) above 373 K.

contrast, as indicated by Fig. 4b, for the same temperature rise of 2°C and initial temperature above 100°C, the detector output deviates little from the tangent drawn to the curve at the initial temperature in question. For instance, for the specimen with thickness of 0.075 cm subjected to a pulse of 20 J attenuated by 60%, the change in detector output was approximately 0.54 mV. As indicated by Fig. 4b, the detector nonlinearity should have a negligible effect. The effect of detector nonlinearity on experimental results is expected to be much more pronounced for initial specimen temperatures near room temperature rather than at elevated temperatures. At the latter temperatures, specimen temperature increases approaching 50 K are required for the detector nonlinearity to become effective.

Figure 5 compares the theoretical temperature response for an initial temperature of 293.75 K and specimen temperature rise of 10°K calculated from the data for the dependence of detector output on temperature rise shown in Fig. 4a. The value of $t_{1/2}$ for the detector response is about 30% higher than for the corresponding temperature response. For this reason, if $t_{1/2}$ of the detector output is assumed to be equal to $t_{1/2}$ of the transient temperature, the resulting calculated value for the thermal diffusivity will
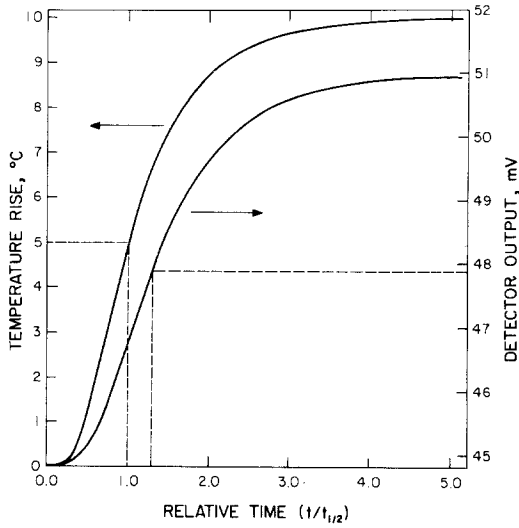
Fig. 5. Comparison of relative surface temperature rise and detector output and associated effect on value of $t_{1/2}$.

be 30% too low. If $t_{1/2}$ for the detector output is to equal $t_{1/2}$ for the transient temperature, the specimen temperature rise should remain small enough so that the detector response can be considered reasonably linear.

In order to ascertain the maximum rise in specimen temperature for which the non-linearity of the detector does not play a role, Fig. 6 shows the
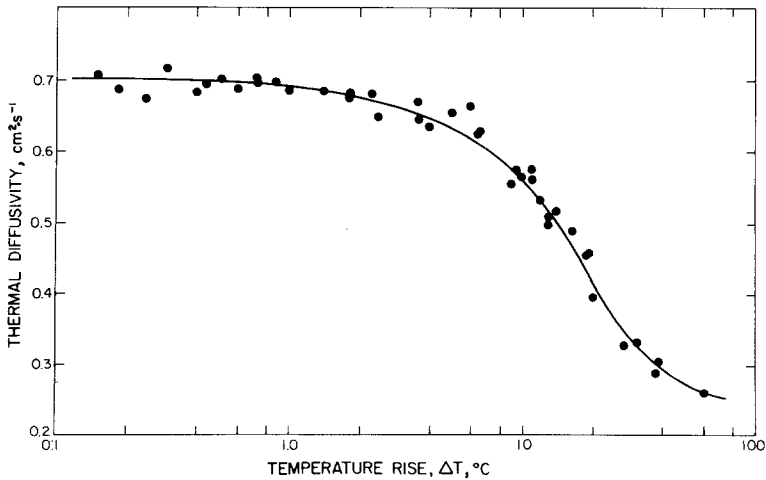

Fig. 6. Dependence of measured values for the thermal diffusivity of NIST 8425 graphite at 21 °C on rear-surface temperature rise for all values of specimen thickness, pulse energies, and attenuations.

measured value of the thermal diffusivity as a function of specimen tem-
perature rise, determined from the detector output, for an initial specimen
temperature of 21°C and for all values of specimen thickness, pulse
energies, and attenuations. The maximum temperature rise was near 60°C,
obtained by extrapolation of the data in Fig. 4a using a fourth-order poly-
nomial fit to the data. The very low data for the highest values of tem-
perature increases can be attributed in part to the effect of the nonlinearity
of the detector. For the specific material of this study, the negative
temperature dependence of its thermal diffusivity/conductivity is a further
contributing factor. A numerical analysis can determine the relative
contributions of these two effects. The detector nonlinearity is expected
to be the major contributor.

The data in Fig. 6 indicate that invariant values for the thermal dif-
fusivity can be obtained if the temperature rise of the specimens does not
exceed about 1°C. It appears reasonable to conclude that literature reports
of an anomalous size effect for thermal diffusivity obtained by the flash
method, certainly at least in part, can be attributed to the use of excessive
flash energies. It has always been the practice of the present authors and
their co-workers to use the minimum flash energy which will yield a
measurable detector output, if only for the purpose of extending the
longevity of the laser flash-lamp. If however, high values of flash energy are
required, for instance, in order to increase the signal-to-noise ratio or other
reasons, modifications will be required in the data acquisition and/or
analysis system which will correct for the detector nonlinearity. Avoiding
the use of very thin specimens, coupled with using minimum pulse energies,
appears to be a more simple alternative.

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