
**Optics and photonics — Lasers and
laser-related equipment — Measurement
and evaluation of absorption-induced
effects in laser optical components**

*Optique et instruments d'optique — Lasers et équipement associé aux
lasers — Mesurage et évaluation de la déformation et de la distorsion
des composants optiques dans un faisceau laser*

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Foreword

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Introduction

This Technical Report has been promulgated in order to highlight the problem and to specify meaningful, standard measurement techniques in order to provide useful information to reduce conflict between users and suppliers of optical components.

When a laser beam impinges upon an optical component (lenses, windows and mirrors) some of the energy is absorbed. Depending on the intensity of the laser beam and the absorption properties of the component material the component temperature will rise. Even if a uniform intensity laser beam fills the whole area of the component, temperature gradients will be created across the aperture. Unless the material has a negligible expansion coefficient, temperature gradients lead to differential expansion, and this will lead to distortion, strain and a change in the birefringence properties of transmissive components. The refractive index of most optical materials is also temperature dependent. If the optical figure of the component changes, the transmitted and/or reflected beam will tend to change shape and/or change its divergence. If the beam path involves a polariser, a beam splitter or a beam deflector the power/energy output and/or the beam propagation of the laser system may change. These effects may be amplified if the component is rigidly restrained. If the strain is high enough the component may crack.

The absorption coefficients of most materials are usually only slightly non-linear with increase in temperature. However, transmissive components made from most semi-conducting materials exhibit a highly non-linear absorption coefficient with a sharp threshold, significantly below the melting point of the material. This phenomenon is termed "thermal runaway" and effectively limits the optical power loading at which these materials can be used. This thermal runaway threshold is accompanied by a sharp increase in the absorption and an increase in the accompanying distortion and thermal lensing. The refractive indices and linear expansion coefficients are both temperature sensitive but do not necessarily have the same sign.

Distortion will occur when a component is irradiated nonuniformly and especially when it is held rigidly and constrained from expanding. The material will expand because of the thermal loading and if this is not uniform the component will bow or lens, thus changing the optical figure. In addition, in the case of transmissive components, it is possible that the temperature dependence of the refractive index of the material will cause a thermal lensing effect. In general, non-uniform expansion changes the focussing properties of the component. In the case of non-linear absorbing windows and mirrors (e.g. Ge, ZnS and ZnSe used with infrared beams) the effects have been observed to severely affect the transmitted beam divergence. In the case of planar reflective interferometric components these have been observed to become convex. In practice, even minor distortion of *in-situ* laser components leads to changes in the divergence and beam propagation ratio of the laser beam and to loss of laser output.

Strain in crystalline components leads to induced birefringence and thus to changes in transmission/reflection and this leads to fluctuations in the system output, to the necessity for raising the input power and to non-linearity in the input/output characteristics of the system. Even homogeneous materials can exhibit birefringence if the thermal loading is non-uniform and the component mounting constrains the material from expanding. The laser output of optically thin laser rods in a flash-tube pumped close coupled or elliptical pumping chamber is governed by the circularly symmetric induced birefringence. Changes in birefringence commonly lead to a change of the transmission of the laser beam through the system, particularly if polarisation sensitive elements are in the beam train.

Thermally induced strain, due to non-uniform irradiation, can occur even in a freely mounted component. Most optical components are, however, mounted in a holder, which is used to control the angular position of the component. If this mount does not allow differential expansion to take place, then the component will become increasingly strained as the component is irradiated. When this strain reaches the elastic limit the component will crack. This is perhaps one of the main causes of the unusually low laser induced damage thresholds encountered in practice. The effect is mainly encountered in high prf (pulse repetition frequency), long pulse and cw (continuous waves) laser systems. Thermally induced strain, due to rigidity in the component mounting and/or lack of expansion gaps in the component/mount combination, is perhaps the greatest cause of laser

components failing in the active system context. The induced strain either cracks the component or lowers the thermal loading at which melting occurs.

The thermally induced effects are minimised if the component is held freely and maximised if clamped hard. The stress involved can be positive or negative depending on the relative differences in the coefficients of thermal expansion between the holder and the component. As this is the decisive factor it is necessary to make the measurement with the component in its holder under, as near as possible, the system working conditions and environment. In practice, this may make it hard/impossible to perform some of the measurements suggested. Therefore, although the measurement of distortion is the most basic and relevant, it may be necessary to monitor either the changes in birefringence or the changes in beam propagation ratio of the transmitted beam. Measurement of the change in the laser induced damage threshold between free and clamped components is not expected to be routine as it is catastrophic.

All the effects mentioned lead to a shortening of the component life and/or a change in the output characteristics of the laser [1]. They also form the main source of friction between the component suppliers and the system manufacturers/users. The effects are most commonly observed in the case of high prf, long pulse and cw laser systems (e.g. welding lasers). However they have also been seen to influence the energy/power output of single-shot Q-switched Nd:YAG lasers operating at 1 064 μm and the transmission and divergence of planar Ge windows under short pulse CO_2 , 10,6 μm laser irradiation.

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Optics and photonics — Lasers and laser-related equipment — Measurement and evaluation of absorption-induced effects in laser optical components

1 Scope

This Technical Report specifies standard measurement and evaluation techniques for determining the absorption-induced effects caused by lasers in laser optical components in order to provide useful information to reduce conflict between users and suppliers of optical components.

2 Absorption

2.1 General

Absorption is a fundamental property of a material. It is directly related to the electronic structure of the material and the wavelength of the probe radiation. For transmitting materials the absorption is directly related to the band gap. A schematic of the spectral transmission of a material is shown in Figure 1. The exact placing and spacing of the different absorption edges are defined by the material structure, including impurities. In the case of materials used for optical windows, which transmit in the visible, the absorption coefficient is small and hardly varies with increasing temperature. In the case of both ultra-violet and infra-red transmitting materials the absorption coefficient is non-negligible and has to be taken into account. In addition, most, but not all, infra-red transmitting materials are semi-conducting and exhibit non-linear optical absorption. These materials have low, usable, absorption characteristics at low/room temperatures but exhibit a thermal runaway threshold at elevated temperatures. Above this thermal runaway temperature the absorption coefficient increases sharply and the transmission of the window drops. In addition any non-linearity in the incident beam is reflected in a lensing effect which changes the quality of the transmitted beam drastically.

Most optically transmitting window materials are, nowadays, homogeneous. However the absorption of a material may be vitally affected by impurities, both localised and diffused through the lattice.

The reasons for absorption may be broken down into a variety of effects.

a) Bulk absorption

$$I = I_0 e^{-\alpha x}$$

This absorption may be permanent or induced. Absorption in the visible, for example, may be induced by absorption of ultra-violet radiation, forming colour centres (trapping of electrons at negative ion vacancies). This commonly occurs in the halides (NaCl and KCl) and in Nd:YAG laser crystal [2]. The latter example is the reason why many Nd:YAG lasers gradually lose output with time and why it is sensible to fit a Nd:YAG pump cavity with an ultra-violet filter. Most instances of colour centres can be nullified by suitable heat treatment.

Absorption is a function both of the electronic structure of a material and the wavelength at which it is irradiated. Single-photon absorption will occur if the photon energy is great enough to bridge the energy gap between the valence band and the conduction band. This is independent of the energy density (Beer's law) but is crucially dependent on the wavelength of the probe radiation. Two-photon absorption can occur if the two photons arrive simultaneously and the sum of the energies exceeds the band gap. At

constant pulse length this process is linearly dependent on energy density. Multi photon absorption can occur as long as, again, the photons arrive simultaneously and the sum of the photon energies exceed the band gap. This process becomes more likely as the pulse length decreases but the total energy density remains constant. There is also an intermediate absorption path where electrons elevated to energy levels within the band gap can then absorb a second photon and thus populate the conduction band [3]. Figure 2 shows a comparison of the absorption behaviour of CaF_2 at 248 nm, 193 nm and 157 nm. The lowest trace (measured using 248 nm radiation) indicates a constant absorption indicating an absence of 2- and 3-photon absorption. At 193 nm there is a strong energy dependent absorption indicating two-photon absorption. At 157 nm only weak two-photon absorption was shown as the combined energy of two photons exceeds the vacuum level, however the linear absorption at 157 nm is about three times that at 193 nm. It will be noticed from Figure 2, where data from two different samples are shown, that the absorptions are strongly sample dependent. This is confirmed by the measurements shown in Figure 3 where the β_{eff} data from 19 different samples are plotted versus their corresponding A_0 values. It will be noticed that there is a linear relationship. This points to a dominant two-step absorption process as opposed to an intrinsic two-photon absorption. Ratification of this analysis is that the comparison of β_{eff} versus pulse length, τ , also shows a linear relationship (see Figure 4).

b) Surface absorption

Scratches, digs and adsorbed contaminants all offer the possibility of increased absorption. It is not always possible to clean this absorption off the surface without damaging it. Figure 5 shows the absorption measured as a function of the resistivity, for n-doped single-crystal germanium [2]. The upper trace shows the absorption measured in air while the lower trace shows the absorption measured under vacuum. Analysis indicated that the germanium surface in air had a 100 μm layer of water adsorbed on its surface. Further experimentation indicated that this water-absorbing layer could be eliminated temporarily by drying or cleaning but that the water layer grew back fairly fast if the component was left in a humid atmosphere.

c) Sub-surface absorption

Machining and cutting the surface of a material involve straining the surface and, it has been proved, leaving a damaged layer under the visible surface. A number of measurement programmes have come up with a figure for the depth of the damaged layer, under tightly controlled machining conditions, of 100. Polishing, etching and annealing can do a lot to remove this disordered, polycrystalline layer but there is usually a remnant of the effect left. Some polishing materials, if chosen wrongly, may also leave absorbing material lodged just underneath the surface.

d) Localised absorption

Some materials contain particulate material dispersed throughout the bulk. When this is sub-micron the absorption related to this material simply raises the bulk absorption. When the particles are larger they may absorb radiation without heating the lattice. In these cases differential strain will occur and catastrophic damage result. Platinum inclusions were a common occurrence in the early Nd-doped laser glass. Dust particles and misoriented crystallites are the chief problem in solution grown materials and in natural crystals.

e) Transient absorption

Colour centres may give rise to intensity dependant absorption. This has been observed in fused silica [4], Ruby [5, 6], Nd:YAG laser crystal [2] and MgF_2 [3]. The colour centre absorption may only be low at the wavelength of operation but it may give rise to absorption, which is only effective while the radiation is high. Figure 6 shows the transient absorption (lower trace) measured, at 0,63 μm , when a Nd:YAG laser rod was flash-pumped (upper trace). The extra absorption could only be measured during the pulse duration.

The induced absorption in MgF_2 [3] measured at 193 nm as a function of probe intensity is shown in Figure 7. This Figure indicates both the energy dependent two-photon absorption and the increase in this absorption with irradiation level.

f) Non-linear absorption

Both stimulated Brillouin and Raman scattering or multi-photon absorption may occur at high optical intensity levels. These effects occur in many optical fibres and effectively limit the transmission of these materials at the wavelength of incoming laser light.

g) Free-electron absorption

This, non-linear absorption, occurs in all semi-conducting materials and results in a thermally non-linear absorption and thermal runaway. The thermal runaway temperature is usually well below the melting point of the semi-conducting material but effectively acts as a limit to the temperature at which the material can be used. Figure 8 shows traces of the absorption, measured at 10,6 μm , as a function of the ambient temperature for both germanium and zinc selenide.

h) Conduction electron absorption

The absorption in electronically conducting materials, e.g. metals, is a function of the plasma frequency and is therefore affected by the temperature of the material. The skin-depth and the thermal conductivities dominate the absorption coefficients of metal mirrors although it has also been shown that the surface roughness has a marked effect [2] (see Figure 9). This indicates that as the surface is heated the absorption rises. This gives rise to extra heating, distortion and a lowering of the laser-induced damage threshold (LIDT). However if the substrate is cooled sufficiently these effects do not occur.

2.2 Measurement of absorption

It has been shown that, as long as the measurement rules are followed, it is possible to make corroborative measurements of the absorptance of optical components at different laboratory locations. ISO/TC 172/SC 9 has produced a standard procedure document for these measurements (ISO 11551 [2¹]) and also, in the process of doing so, made a range of comparative measurements.

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The main problems with obtaining agreement on the absorption of specified components have been:

- Attempting to make measurements with too large or too small a probe spot size. Radiation scattered around the edge of a component will add to the measured absorption. If there is any particulate matter or surface scratches present they may cause an increase in the absorption, particularly if the probe spot is comparable with the discontinuity. Unless the optical component under measurement is uniformly homogeneous the probe spot size and placement may be critical.
- Differences in the calibration. It is important that the “standard” sample, where used, is well characterised and that it is tested at all laboratories in the investigation.
- Measurements may be made using either pulsed or cw probe radiation. It must be realised that the conductivity of the sample may affect the temperature measured. If the more accurate pulsed measurement procedure is used it must be noted that it is sensible to make measurements at a range of different probe pulse lengths and/or pulse energy levels to ensure that the differences between the thermal diffusivity of the sample and the standard do not lead to wrong measurements.
- The absorption measurement must be made under identical ambient atmospheric conditions and at the same wavelengths. Measurements made under vacuum and in air may give radically different results because most surfaces adsorb water. Water vapour absorbs strongly at specific wavelengths in the infrared. In practice, it is recommended that the measurement should be made under the identical conditions that pertain in the system in which the components are to be used.

3 Distortion

3.1 General

When an optical component is irradiated by a laser beam, the material on the axis of the laser beam will expand, due to heating because of absorption of radiation. The periphery of the component is likely to remain cooler due to conduction of the heat away by the rest of the system.

A good example of the (reversible) thermal lensing effect in fused silica during 193 nm irradiation has been published [3]. This measurement was performed with a Shack-Hartmann wavefront sensor, using the transmission of a collimated 193 nm probe beam through the centre of the sample.

If the component is properly located but allowed to expand, the distortion may be minimised (the whole component expanding uniformly). If the component is held in a constricting holder distortion may well arise both from the non-uniform expansion and because of the restriction imposed by the holder. If there is a requirement to gauge the effect of irradiation on a component in a given system then it is necessary to measure the distortion under the same radiation geometry and power levels as will be expected in the final system. There are three different possibilities:

- a) Measurement of distortion under free standing conditions.

This is relatively easy, as long as the irradiation power levels and geometry can be duplicated. However, except as a means of measuring the absolute maximum power that could be placed on the component if the holder was perfect, this is not very useful in practice.

- b) Measurement of distortion in a sample holder of the correct dimensions, under the same irradiation levels that occur in practice.

- c) Measurement of distortion of the component inside the complete system.

This can be an extremely difficult measurement. It is however the final case on which the performance of the component depends.

The justification for undertaking this measurement lies in the possibility of helping the optics manufacturer and the user agreeing on a specification (including manufacturing tolerances) for the optical components for use in a given system. It is therefore in the interests of both the optics manufacturer and the systems manufacturer to agree on a measurement system which fulfills their requirements.

3.2 Measurement of distortion

The optics manufacturer and the systems buyer must identify an interferometer in which the distortion measurements can be made (see ISO 14999 series [25]). The component holder must be as close as possible to the system holder while allowing the measurement to be made. A simple schematic of a Michelson interferometer is shown in Figure 10 with the optical component in place of one of the standard reflecting components. The object of the measurement is to allow the measurement of the change of shape of the optical component when it is irradiated under the identical conditions as it would be in the final system. The procedure is to monitor the change of shape of the component under increasing irradiation levels, up to that expected in the system.

3.3 Discussion

Measurements of the distortion induced when a laser component is irradiated are not well documented. It is surmised that most manufacturers do not realise that this effect may be important or do not want to advertise the fact that their systems are not temperature insensitive. The best documented results are those from the manufacturers of He-Ne gas lasers in the 1980s where a lot of effort was put in to ensure that the output of the lasers did not degrade due to thermal expansion. Earlier than this He-Ne laser outputs were startlingly sensitive to the temperature of the components and most insisted that a warm up time of (e.g.) 30 min was

necessary to achieve a degree of output stability. The use of INVAR and ZERODURE in the construction of these lasers and coatings on Spectrosil for the window components were promoted for this specific reason.

Measurements were made in the 1980s at the GEC, Hirst Research Centre, Wembley on distortion induced at 10,6 μm using a cw CO_2 laser [2]. Free standing metal mirrors were shown to expand uniformly whilst free standing Ge substrates were shown to expand in a convex fashion and to ultimately crack catastrophically, see Figures 11 and 12. It was also found that the Ge blanks would shatter at lower power loading if the sample was constrained from expanding or if part of the blank was shielded from the radiation.

Measurement of the shape of a ruby laser rod, (and subsequent output beam divergence) under flash-pump irradiation in a large elliptical pumping chamber, showed that the laser rod (plane ended) expanded non-uniformly. The measurements indicated that by the time the Q-switch was opened the rod was both optically convex and continuing to expand.

Sparks [7] has considered the case where a spatially homogeneous incident laser intensity causes a temperature gradient to change both the thickness and the refractive index of a parallel-sided window. He came to the conclusion that optical distortion, rather than laser-induced damage, would be expected to be the factor that determined the usage of such a window in a long-length focal system. His analysis concluded that a large heat capacity and small values for absorption, stress coefficients, refractive indices and dn/dT would be more important than the melting point, tensile strength and Young's modulus. He also concluded that the net distortion was generally smaller for ionic crystals than for covalent crystals.

Sheldon et al [8] carried out a thermal lensing experiment using liquid in a parallel-sided cell. They derived the theory for the temperature rise, as a function of the position across the beam, versus time. This experiment showed that the refractive index of the liquid changed during heating because of a decrease in density with increasing temperature.

The case of a flat metal mirror surface where the thermoelastic deformation process leads to local variations in the geometry of the surface has also been extensively researched [9]. Three cases were considered:

- a) When the laser energy absorbed was small and the temperature deformation was elastic the surface expanded non-uniformly in a convex fashion. When the beam was switched off, the mirror relaxed back to its original conformation.
- b) At higher energies, the mirror deformation showed up as a residual expansion which appeared as a wavy surface structure.
- c) When the thermally induced stress exceeded the material tensile strength the mirror surface underwent microcracking which in turn resulted in higher energy loss/greater absorption.

In all instances, the mirror surface lost its high-quality optical shape before the incident light intensity was sufficient to melt the reflecting surface material. This was the reason why the maximum flux permissible for a given reflecting surface is lower than the flux associated with surface melting. Figures 13 and 14 show the calculated displacement at the centre of a metal mirror versus time when it is irradiated with 10,6 μm CO_2 cw laser radiation. Figure 13 shows the magnitude of the expansion at the centre of an aluminium mirror when irradiated with a 760 W cw beam as a function of the beam radius. Figure 14 contains lines for Ag, Al, Cu, Mg, Si/Au and steel/Au mirrors when irradiated with a 800 W, 0,9 cm diameter beam.

4 Refractive index and birefringence

4.1 General

The refractive indices of materials are temperature dependent. This means that the optical shape of a material changes as the material heats up or is irradiated. The change of refractive index with temperature, for a range of transmitting laser window materials is included in Table 1. It will be noticed that the variation of refractive index with temperature is usually greater than the coefficient of linear expansion and it should also be noticed that although linear expansion always increases with temperature there are both positive and negative