



Novel techniques for high precision refractive index measurements, and application to assessing neutron damage and dose in crystals

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ABSTRACT

In this work we present novel techniques for high precision index of refraction measurements for transparent crystals, and demonstrate a change from neutron irradiation. Radiation damage affects the structure of material, which can be read out nondestructively in transparent crystals. There is some difference in gamma-ray and neutron interactions which may be useful in characterization. Ionization from gamma rays produces color centers in the material, producing distinct spectral absorption, and some small shift in the index of refraction. Neutrons produce atomic recoils and, while the recoils do some ionization, they have a much greater efficiency for lattice displacement than do gamma rays, and these displacements can have a greater effect on the index of refraction. Using CaF_2 crystals exposed to neutron radiation, together with a new high precision technique of detecting changes of index of refraction, we establish proof that this type of measurement can be used to monitor neutron exposure. This can provide a basic study of material changes with radiation and, with calibration of material in known neutron fields, this may even find application to neutron dosimetry.

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1. Introduction

Radiation damage to materials may be used to detect the presence of or exposure to radioactive sources. Depending on the radiation field type (e.g., neutrons, electrons, gamma rays), dose rate, and the presence of impurities, it is expected that certain materials may exhibit a characteristic response. Optical properties of the materials may show the radiation damage, allowing optical assessment of the radiation. In addition, optical crystals can find applications where other modes to detect radiation may not work, they may be selected for harsh or corrosive environments.

It has been established that irradiation of crystals and glasses by neutrons [1], gamma rays [2], electrons [3], and heavy ions [4] produces long term electron displacements and color centers, which can be observed in the absorption spectrum. Long lived excited states have also found use in thermoluminescent dosimetry and optically stimulated luminescence, and are well characterized for certain materials. Work on all these effects has been used for optical fiber based dosimetry in high radiation environments [5].

Radiation also affects the index of refraction of crystals and glass as is well known, e.g., in space applications [6]. Changes in refractive index are reported from gamma-ray [7,8], proton [9], and neutron [10] radiation. According to the Kramers–Kronig relations, there is a small change in refractive index associated with color center absorption [9]. These are formed from ionization effects, and the

index change saturates at a lower level than the index change resulting from heavy particle interaction with the material [11]. Heavy particles are much more efficient than gamma-rays at lattice displacements and material density change, which has been shown to cause change in the refractive index [10,12]. A clear conceptual example of refractive index differences due to lattice differences may be seen in a comparison of the indices for materials with the same components but different internal structures such as crystal-line quartz and amorphous quartz glass [12]. Protons and other directly ionizing charged particles have a high efficiency for ionization in addition to producing atomic displacements, complicating the interpretation. Both for this reason and the importance of neutrons in nonproliferation work, reactors, and other applications, neutron radiation is used for the current work.

In this work we present a novel technique for high precision measurements of the index of refraction in transparent crystal samples. The technique uses analysis of the interrelation of a nested frequency comb within a mode-locked laser with its own frequency comb. As a proof of concept, CaF_2 crystals with and without neutron irradiation are compared. These crystals were selected for research on ionic crystals and color center formation, but were found useful for the current proof of concept measurements.

2. Theory

We have developed a unique mode-locked laser consisted of two nested combs, by insertion of a small Fabry–Perot etalon (FPE), as in

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Fig. 1. In this configuration two radio frequencies are generated, a high frequency RF (f_{HF}) that is inversely proportional to the optical path (distance and index) in the Fabry–Perot etalon and a low frequency RF (f_{LF}) inversely proportional to the optical path in the laser cavity. In the current application, the Fabry–Perot etalon may be a crystal sample that is irradiated, or an unirradiated standard for comparison. By measuring the ratio between the high frequency and low frequency RF for the irradiated and for the unirradiated Fabry–Perot etalon crystals we are able to determine the indices of, and the differences of indices between, the two samples. The reasons for the exceptional precision of the index measurements are (1) the high accuracy of frequency measurements and (2) the fact that the low and high frequencies are locked together in a ratio directly proportional to the index to be measured.

The frequency combs which are represented in the high and low frequency pulses may be understood in terms of resonances in the system. The parallel mirrors of a laser cavity give specific resonance modes, based on an integer number (N_L) of half wavelengths spanning the length between the mirrors (L). This is expressed as

$$N_L \lambda / 2 = L, \quad (1)$$

or with explicit reference to the phase index of refraction in the cavity, n_{pL} , and vacuum wavelength, λ_0 , as

$$N_L \lambda_0 / (2n_{pL}) = L. \quad (2)$$

Rewritten

$$2\omega L n_{pL} / c = 2\pi N_L, \quad (3)$$

this resonance condition determines the optical frequency ω that can oscillate in the cavity.

It is a property of a mode-locked laser to emit a train of pulses separated in time by

$$\tau_{rt} = c / 2n_{gL} L, \quad (4)$$

where n_{gL} is a group index describing the transit of each pulse. In frequency, the laser output is a comb of delta functions of equal spacing [13],

$$1/\tau_{rt} = 2n_{gL} L / c, \quad (5)$$

a property shown to be due to a correction of the dispersion of the cavity by Kerr modulation [14]. The comb – in particular the group index n_{gL} – is modified considerably by insertion of a parallel plate window (Fig. 2), acting as the Fabry–Perot etalon. Because of the low reflectivity of the glass–air interface ($0.04 = 1/25$), one would expect the second transmitted pulse reduced in intensity by a factor $25^2 = 625$, and the third by a factor of $(625)^2$ and so on, and no coherence relation between these successive pulses. Instead a remarkable comb of tens of discernible pulses with an approximately Gaussian envelope is observed which allows the analysis presented here. The reason for the extended comb is that the laser cavity itself establishes coherence between the successive Fabry–Perot reflections. Each of the successive pulses shown in Fig. 2 feeds energy to the next later one, resulting in a considerable reduction of

the cavity group velocity which is represented in an increase of the cavity group index n_{gL} . The coherence between successive pulses established through the laser cavity also implies that the optical frequency has to be resonant with the glass plate, or

$$2\omega d n_{pFP} / c = 2\pi N_{FP}, \quad (6)$$

which is the classical resonance condition for a Fabry–Perot etalon [15]. Combining the resonance conditions of the etalon and laser cavity, it is possible to find an expression for the ratio of the low frequency and high frequency of the nested combs [16]:

$$\frac{f_{HF}}{f_{LF}} = \frac{n_{gL} n_{pFP} N_L}{n_{gFP} n_{pL} N_{FP}} \quad (7)$$

where N_L and N_{FP} are the resonance integers for the laser cavity and the FPE, respectively. An experimental trace of the high frequency and low frequency pulse trains can be seen in Fig. 3, bottom. In Eq. (7), the group indices are determined by the laser gain and saturation properties, as well as the reflectivity of the glass faces. For the gains, losses, and laser phase index kept equal, the ratio is simply proportional to the phase index of the glass or crystal inserted in the cavity.

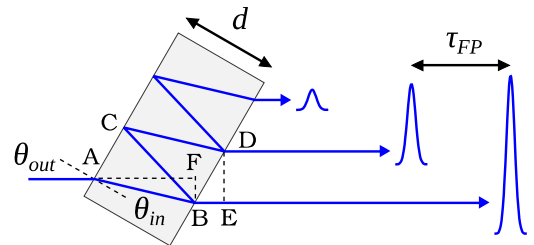


Fig. 2. Schematic of light pulses through the Fabry–Perot etalon at an angle to the incident light. Due to different path lengths, the internally reflected pulses have a time lag of T_{FP} , leading to a high frequency pulse packet.

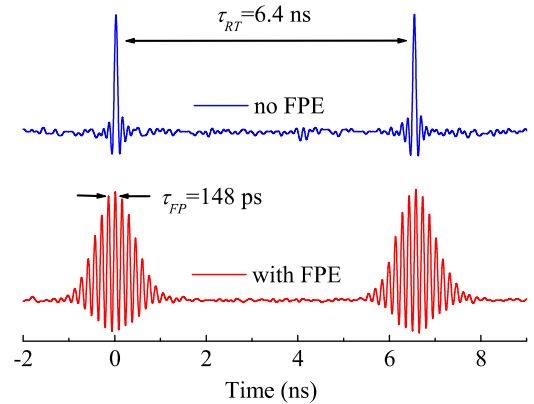


Fig. 3. Oscilloscope traces of the low repetition rate from resonances within the full laser cavity length L with a repetition time of T_{RT} (top), and the addition of a Fabry–Perot etalon with higher frequency resonances with shorter repetition time T_{FP} (bottom).

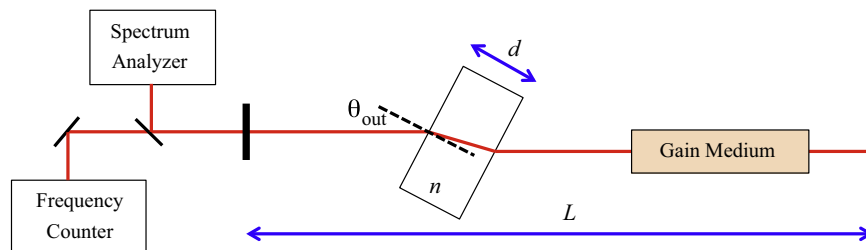


Fig. 1. Schematic of laser cavity of length L with sample crystal inserted as a FPE at angle θ (θ_{out} here) to the laser cavity light path.

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