DEVELOPMENT OF A TUNABLE, LONG-CAVITY, FEMTOSECOND TITANIUM-SAPPHIRE LASER; AND THE GROUP DELAY DEPENDENT LOSS OF DIELECTRIC MIRRORS

Theses of PhD dissertation

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Preliminaries and goals

I have been working on my PhD thesis in at the Laser Applications Department at the Research Institute of Solid State Physics and Optics of the Hungarian Academy of Sciences. My supervisor was dr. Róbert Szipőcs. I was dealing with two separate topics. The first one was the development of a long-cavity, broadly tunable femtosecond titanium-sapphire laser for nonlinear microscopy applications. The other one was the theoretical investigation of the relationship among the group delay, the absorptance and the electromagnetic energy storage of dielectric multilayer mirrors.

The most important types of femtosecond lasers include the titanium-sapphire lasers, mainly because their broad gain bandwidth allows the tuning of the laser wavelength in a hundreds of nanometers wide range and the generation of pulses shorter than 10 fs as well. The pulse repetition rate of common, commercially available titanium-sapphire lasers is around 80 MHz. However, this repetition rate and the other respective laser parameters (pulse energy, peak power) are not suitable for all applications. Significantly lower repetition rate is often needed, which can be obtained, the simplest way, by using a long laser cavity, since the repetition rate of a mode-locked laser is inversely proportional to the cavity length.

Most long-cavity lasers are built by extending the cavity of a ~80 MHz laser with a so-called Herriott-cell. This comprises two concave spherical mirrors (or one spherical and one plane mirror) facing each other, on which the multiple reflections of the beam ensures the long lightpath. By setting the number of reflections and the mirror distance correctly, it can be achieved that the building-in of the Herriott-cell does not change the laser mode parameters. The first long-cavity titanium-sapphire laser (which had a 15 MHz repetition rate) was also built this way. Another advantage of lengthening the cavity is that the pulse energy per average power or per pump power increases proportionally to the cavity length. That is why one important application of long-cavity lasers is the generation of high pulse energies (even beyond 500 nJ) directly at the laser output, without the use of laser amplifiers or cavity dumping. The high pulse energy can cause excess nonlinearity in the laser crystal inducing the instability of mode-locked operation which can lead to even splitting of the intra-cavity pulse. This can be prevented by lowering the peak power in the crystal either by enlargement of the laser mode waist diameter or by setting the intra-cavity dispersion to a high negative or a small positive value. However, positive intra-cavity dispersion infers strongly chirped output pulses that should be compressed by an extra-cavity pulse compressor.

Long-cavity femtosecond lasers have already been used in microscopy imaging applications as well. One of them is the fluorescence lifetime imaging microscopy (FLIM) where the distribution and the chemical environment of the different molecules in the sample (e.g. a cell) are imaged based on the measurement of their fluorescence lifetime. For the identification of certain long-fluorescence-lifetime molecules, low-repetition-rate lasers are needed. First of all, Nd:YVO_4 and Yb:KGW based types are reported to be used in former FLIM applications. The relatively high pulse energies of long-cavity lasers can also be advantageous when the excitation wavelengths are generated via nonlinear optical methods.

Another field of application is the nonlinear, particularly the multifoton microscopy. In this case, the femtosecond pulses are focused into the sample where the two- or three-photon absorption takes places only in the tiny volume of the focal spot where the intensity is sufficiently high. This way, high-resolution 3D imagery of tissues and cells is possible. However, the multiphoton excitation can cause chemical damage of the biological specimens (e.g. DNA damage). For specific specimens (e.g. skin) that have strong absorption in the infrared also the one-photon absorption can have a thermal damage effect, which can be, however, significantly mitigated by reducing the repetition rate of the laser. The minimization of the cell damaging effects of nonlinear microscopes is particularly important because of the high demand on the so called *in vivo* (i.e. inside a living organism) application of them in human medical diagnostics – skin cancer diagnosis, among others,. A human clinical application of a multiphoton microscope system has been recently authorized in the European Union.

The most fruitful applications of long-cavity femto- or picosecond lasers can be the FLIM and multiphoton microscopy. Since there are several different fluorescent molecules in biological samples having different excitation wavelengths, another important requirement for these light sources is the broadly tunable wavelength. However, I have found no reports on wavelength-tunable ultrashort-pulse lasers in the literature. Accordingly, the aim of my work was the development of a broadly tunable, long-cavity femtosecond titanium-sapphire laser with approximately 20 MHz repetition rate. Due to the long cavity, a reduction of the pump threshold for pulsed operation is expected at the same time which could facilitate the use of cheaper pump lasers compared to the ~80 MHz lasers.

The other topic of my dissertation is a theoretical work associated with dielectric multilayer mirrors. These mirrors can exhibit higher (nearly 100%) reflectance in the wavelength range around a given design wavelength than metallic mirrors. This high reflectance is of primary importance in the case of lasers and other light sources having a resonator, like optical parametric oscillators (OPO-s) since these devices can only work efficiently with low cavity loss. Therefore these resonators comprise appropriate multilayer mirrors and in most cases these mirrors are used for extra-cavity beam steering as well. The not-reflected part of light incident on a mirror is partly transmitted and partly absorbed and scattered in the mirror's volume. The absorbed energy raises the temperature of the mirror which can lead to deformation of the mirror surface and, consequently, to wavefront distortion of the reflected beam, hereby degrading e.g. its focusability. In the case of high power lasers, laser amplifiers, OPO-s and optical parametric amplifiers (OPA-s), the heating due to absorption can even cause mirror damage. This heating effect of one-photon absorption is, in particular, critical in the case of systems having pulses longer than 50 ps because for shorter pulses, mainly multiphoton absorption and avalanche ionization are responsible for mirror damage. Apart from reflectance, absorptance and damage threshold, another important parameter of mirrors is their bandwidth, is that the width of their high-reflectance wavelength range. In the case of laser and optical parametric light sources used in e.g. spectroscopy and in the above mentioned microscopy applications, broadband cavity mirrors are needed for wavelength tunability. Additionally, broadband laser operation is crucial for ultrashort, i.e. pico-, femto- or attosecond pulse generation.

In the case of ultrashort pulse sources, the intra- or extra-cavity dispersion compensation of the pulses is required. By multilayer mirrors with appropriate structure, much greater group delay dispersion (GDD) can be attained than with common quarter-wave mirrors. The first mirrors whose dispersion function could be set relatively freely even in higher than second orders were invented by my supervisor, Róbert Szipőcs, in the early '90s – those were the so called chirped mirrors in which the frequency dependent penetration depth of light is the source of dispersion. In the present days they are worldwide used, particularly in ultrashort pulse lasers. There are also single- or multi-cavity Gires-Tournois interferometer type dispersive mirrors operating based on standing-wave resonances by which even -2000 fs^2 GDD can be attained per reflection. The use of chirped mirrors is essential also in long pulse or even in CW systems when the aim is the broadly tunable wavelength since a reflection bandwidth wider than 200 or even 300 nm can only be obtained with these chirped structures (ultrabroadband chirped mirror, UBCM).

One can ask if the phase shift of a mirror influence the amount of energy absorbed in the multilayer structure. This question, as far as I know, was first dealt with by a paper published in 1992, where the authors demonstrated analytically, specifically for quarter-wave mirrors that under certain conditions (low refractive index difference of the layers and other restrictions) the absorptance is proportional to the group delay (GD) at the central wavelength of the mirror bandwidth. In a paper from 1993 my supervisor pointed out for another specific mirror structure, based on numerical calculations, that there is proportionality between the GD and the absorptance in a wide wavelength range. There are also some papers that mention the relationship between the GD and the electromagnetic energy stored in the layer structure of the mirror. Based on the continuity equation of energy flow it can be calculated analytically for a general one dimensional photonic barrier (e.g. a mirror) that

$$R\tau_{gr} + T\tau_{gt} = \frac{U}{P_i} \tag{1}$$

where τ_{gt} and τ_{gr} are the transmission and reflection group delays, R and T is the reflectance and transmittance, respectively, U is the energy stored in the volume of the barrier and P_i is the incident light power. This calculation can be found in a paper of H. G. Winful from 2006. It is written also in this report that for the lossless and stationary case, the τ_c (cavity-)lifetime of stored energy is equal to the right hand side of Eq. (1):

$$\tau_c = \frac{U}{P_i} \tag{2}$$

Based on the above mentioned results and on the above equations I assumed that there is a general relationship, an approximate proportionality among the reflection group delay, the absorptance and the energy stored during reflection within the mirror bandwidth if the absorption loss is as low as in most practical situations. The aim of my work was to verify this assumption and to check under which conditions, how high loss these relationships hold. The connection between the group delay and absorptance can have significant practical consequences for some laser applications when it is about to select, to design the proper mirrors and about the technical realization of the laser system. The relationship of these two physical quantities, however, can be understood based on the one between the GD and the stored energy.

Applied methods

To build the tunable, long-cavity laser, I started with a ~71 MHz repetition rate, astigmatically compensated, linear titanium-sapphire laser resonator that was designed previously by others. The gain medium was a strongly doped titanium-sapphire crystal with a 4 mm lightpath length. I extended this resonator with a Herriott-cell properly designed to attain a repetition rate around 20 MHz. I operated the laser with negative intra-cavity dispersion and used a prism pair for dispersion compensation in which the prisms can be translated perpendicularly to the beam to make the dispersion precisely adjustable. The tuning of the laser wavelength is carried out by rotating a birefringent filter. The pump source was frequency-doubled Nd:YVO_4 operating at 532 nm. I have built two different setups. In the first one the mode-locking was initiated with an acousto-optic modulator operated with the regenerative feedback method, however, the femtosecond pulse duration was achieved with the soft-aperture Kerr-lens effect. The cavity mirrors included ones made via electron-beam evaporation and ion-beam sputtering, the most reflection, 7 on each mirror, were on the Herriott-cell mirrors made via the former technology. In this setup, the repetition rate was 19.6 MHz and the wavelength could be tuned over a 115 nm wide range. In the second setup I exchanged the cavity mirrors to ion-beam sputtered (thus low-loss) ultrabroadband chirped mirrors (except for one for technical reasons) and I used another Herriott-cell configuration with mirrors with two reflections on each. I did not use the acousto-optical modulator here and mode-locked the laser with the hard-aperture Kerr-lens technique. This laser version had a repetition rate of 22.2 MHz and the wavelength could be tuned over a 170 nm wide wavelength range. By further optimization of the geometric parameters of the resonator the width of the tuning range could be increased to 185 nm. I have measured the average output power of the built lasers as a function of wavelength during tuning and determined the pulse duration by second order autocorrelation measurement. I have carried out a multiphoton microscope experiment where I used, first, a common 76 MHz titanium-sapphire laser and then the 22.2 MHz laser built by myself, as a light source. I have found that the same two-photon absorption fluorescence signal can be generated by a 1.82 times lower average excitation power than by the 76 MHz laser when the other output parameters are identical for the two lasers. For the 22.2 MHz laser, I measured the pulse duration in the focus of the microscope objective by second order autocorrelation measurement using two-photon absorption to produce the autocorrelation signal that is proportional to the square of the intensity. For the microscopy measurements, I used fluorescent microbeads as specimen and, in one case, a biological specimen as well (a piece unhaired mouse skin).

Regarding the multilayer mirrors, I carried out a theoretical study. I took Eq. (1) as a starting point that implies that the reflection group delay (GD) and the stored energy are proportional to each other in the lossless case within the high-reflectance ($R \approx 1$) region of the mirror, provided that $T\tau_{gt} << R\tau_{gr}$. I assumed, intuitively, that this remains approximately valid also for low loss. Based on Eq. (2) it is also expected that, for low loss, the group delay and the absorptance of the mirrors are approximately proportional as well. From the above relations it naturally follows that the stored energy and the absorptance are assumed to be proportional as well. I verified these assumptions by numerical calculations performed mirrors of four different types. These four included a quarter-wave (QW) mirror, an ultrabroadband chirped mirror (UBCM) and two multi-cavity Gires-Tournois interferometer mirrors. For the QW mirror and the UBCM the GDD, i.e. the frequency dependence of the GD is determined by the penetration depth of the electromagnetic field, while for the MCGTI mirrors it is determined by the combination of penetration depth and the resonances appearing in the spacer layers located in different depths. The QW mirror has the lowest GDD, the UBCM has a higher and the MCGTI mirrors have the highest GDD. Consequently, the QW mirror has the lowest and the MCGTI mirrors have the highest GD values. The two MCGTI mirrors have the same thickness and the same layer materials and number of layers. They have been designed to have similar group delay functions and nearly identical maximum GD in the relevant wavelength range where the GDD is nearly constant. The difference between these two mirrors is that their resonant spacer layers have different amount of material loss. It turned out, however, that the absorptance of these two mirrors does not differ significantly. I calculated the different quantities (e.g. absorptance, stored energy, group delay) for normal incidence and by using the transfer-matrix method commonly used in thin-film technology. By investigating the normalized relative variations of the ratios of the above mentioned quantities, I analyzed the effect of the Fabry-Perot type resonances (that cause high transmission group delay) on the proportionality of these quantities. To evaluate the degree to which these physical quantities are proportional to each other, I fitted straight lines crossing the origin on the datasets that show the dependence of one quantity on another one. I also checked to which extent unconstrained straight lines fit on these datasets. I used the adjusted R-square values to characterize the goodness of the fits. I carried out the analysis first for each mirror's usable band, i.e. the band with high reflectance and proper dispersion, for the wavelength dependent GD, stored energy and absorptance and for several cases of different loss. I assumed that the different layers have either identical or differing extinction coefficients that I varied within the 10^{-5} – 10^{-3} interval, which are the common values in practice (in the near infrared). Although I assumed wavelength-independent extinction coefficients, the loss is determined by the absorption coefficient that is inversely proportional to the wavelength. Thus I also analyzed the proportionality between the GD and the absorptance at a given wavelength by taking into account the values of these two quantities belonging to mirrors with different structure but with the same layer materials. I obtained a series of the new mirror structures needed for this analysis by rescaling the physical layer thicknesses of the initially used mirrors.

New results (list of theses)

T1. I have developed a long-cavity femtosecond titanium-sapphire laser that is wavelength tunable over a 170 nm wide range and has a repetition rate of 22.2 MHz that is approximately the quarter of the common titanium-sapphire lasers' ~80 MHz repetition rate. I used the Kerr-lens method for mode-locking. The laser has negative intra-cavity dispersion and generates nearly transform-limited, typically 180 – 300 fs pulses. In pulsed mode it operates stably with 2.6 or lower pump power and can produce a maximum of 311 mW average output power. By further optimization of the cavity, the width of the tuning range could even be extended to 185 nm.

T2. Starting with theoretical considerations, I examined through numerical examples the physical relationships among the reflection group delay, the absorptance and the electromagnetic energy storage of highly reflective dielectric multilayer mirrors, in a detailed way. I carried out the analysis for different mirror types and for different amounts of loss. I found that within the high-reflectance band of the dispersive mirrors (and in the absence of Fabry-Perot type resonances that would cause high transmission group delay values), the stored energy and the reflectance are practically proportional to each other and, furthermore, there is a strong positive correlation among these two physical quantities and the absorption loss as well.

Conclusions

I have built, for the first time, a long-cavity femtosecond titanium-sapphire laser that *has a tunable wavelength*. For this reason and because of the much lower repetition rate than that of the ~80 MHz types, this laser could be a proper light source for certain microscopy imaging applications. One example is FLIM where the investigation of such specimens that contain molecules with long fluorescence lifetime is only possible with low-repetition-rate pulses. In multiphoton microscopy, the risk of thermal damage of the specimen can be mitigated due to the low repetition rate. This is particularly critical for specimen having strong one-photon absorption at the near infrared wavelength of the titanium-sapphire laser, such as skin that contains melanin or brain slices labeled with fluorescent dye. The wavelength tunability ensures that the different fluorescent molecules present in biological specimens can be excited always at maximum efficiency or at all detect them. The laser built by myself has a much lower pump threshold (in pulsed operation) than the ~80 MHz lasers, which enables the use of cheaper pump lasers as well. There is a chance that because of it advantageous properties, the laser could be utilized in *in vivo* human medical diagnostics, however, appropriate test should be carried out to determine the extent of the chemical cell and DNA damage it causes during multi-photon microscopy investigations. These tests are in progress.

My work related to dielectric multilayer mirrors revealed that the light power absorbed by the dispersive mirrors is dependent on the reflection group delay, namely, there is a strong positive correlation between them. This relationships can be important, in some cases, when the mirrors are design and when the proper mirror should be chosen for a given application. In particular, when the use of dispersive mirrors having high group delay values, e.g. in the case of short pulse lasers or broadly tunable laser sources. It turns out, for example, that chirped mirrors that ensure broad tunability range, are suggested to be designed so that they have an increasing GD towards longer wavelengths (where the material loss is lower), even if they are not used for dispersion compensation. When using dispersive mirrors in high-power systems, one should take into account that the wavefront of the reflected beam could be distorted more because of the heating of the mirror than for low-dispersion mirrors. If the pulse duration is exceeds 50 ps, a higher reflection group delay implies a higher risk of mirror damage. This is particularly important in the case of high-power, broadband or broadly tunable laser systems, optical parametric oscillators or amplifiers operating in the ultraviolet, visible or near infrared spectral range for which the use of broadband, chirped dielectric mirrors is necessary.

Publications that support the theses

For thesis T1:

[1] P.G. Antal, R. Szipőcs, "Tunable, low-repetition-rate, cost-efficient femtosecond Ti:sapphire laser for nonlinear microscopy," Appl. Phys B, DOI 10.1007/s00340-011-4830-7, *published online* 2011. Nov. 25.

[2] R. Szipocs, P. G. Antal, A. Szigligeti, A. Kolonics, "Tunable, Low Repetition Rate, Femtosecond Pulse Ti:Sapphire Laser for In Vivo Imaging by Nonlinear Microscopy," *Novel Techniques in Microscopy*, OSA Technical Digest (CD) (Optical Society of America, 2011), paper JTuA12.

For thesis T2:

[1] P. G. Antal, R. Szipőcs, "Relationships among group delay, energy storage, and loss in dispersive dielectric mirrors," Chin. Opt. Lett., *accepted for publication.*

[2] P. Gy. Antal, R. Szipőcs, "Relation between Group Delay and Energy Storage in Dispersive Dielectric Mirror Coatings," Advanced Solid-State Photonics, OSA Technical Digest Series (CD) (Optical Society of America, 2010), paper AMB17.

[3] P. Antal, R. Szipőcs, "Relation between Group Delay, Energy Storage and Absorbed/Scattered Power in Highly Reflective Dispersive Dielectric Mirror Coatings," Optical Interference Coatings, OSA Technical Digest (Optical Society of America, 2010), paper FB3.

[4] Antal Péter Gyula, Szipőcs Róbert, "A csoportkésleltetés, a tárolt energia és az abszzorpciós/szórási veszteség kapcsolata diszperziós dielektrikum tükrökben," *Fizikus Vándorgyűlés 2010*, Collection of Abstracts, page 1.

Other publications in the topic

- P. Dombi, P. Antal, "Investigation of a 200-nJ chirped-pulse Ti:Sapphire oscillator for white light generation," Laser Phys. Lett. **4**, 538-542 (2007).
- P. Dombi, P. Antal, J. Fekete, R. Szipőcs, Z. Várallyay, "Chirped-pulse supercontinuum generation with a long-cavity Ti:sapphire oscillator," Appl. Phys. B **88**, 379-384 (2007).