Abstract: Pulses as short as 260fs have been generated in an Er:Yb:glass laser by saturable absorber mode-locking using graphene as the only mode-locking mechanism. These novel saturable absorbers present a low-cost, ultra-broadband alternative to traditional SESAMs.

1. Introduction

Since the first report of single layer graphene a few years ago [1], this material has already revealed a wealth of interesting physics. One of these interesting properties is the non-linear optical behaviour, namely the wavelength independent saturation of optical absorption in graphene. It is its wavelength independence that makes this novel material particularly interesting for long-wave applications, where traditional semiconductor based saturable absorber mirrors (SESAM) are difficult to manufacture, or simply unavailable due to the lack of suitable materials. Multilayer graphene has recently been used to mode-lock fiber lasers [2], [3], and to generate picosecond pulses in a Nd:YAG ceramics laser [4]. While all these previous experiments utilized multilayer graphene to mode-lock lasers with very high round-trip gain, we use a single atomic layer of graphene yielding approximately 2.3% insertion loss. This low insertion loss paired with the low saturation intensity of graphene [4] might open up a way to monolithic, ultra-high repetition rate solid-state lasers that have long been aspired for robust, portable frequency comb generation and other applications that require ultra stable pulsed light sources.

2. Sample preparation

The absorbers used for this experiment are based on single layer graphene grown on copper foils using chemical vapour deposition (CVD). We chose to use copper over nickel due to its larger grain size, and low solubility of carbon which is believed to lead to near 100% single layer graphene [5]. The copper foils should first be cleaned in concentrated acetic acid and then annealed at 1000°C for 30 minutes in an argon and hydrogen atmosphere (500:50 sccm argon and hydrogen, respectively). This procedure can lead to larger grain sizes and it removes oxides from the copper surface. The sample shown in Fig. 1. was grown by flowing 200 sccm of argon, 50 sccm of hydrogen, and 65 sccm of methane at ambient pressure over the copper foil for five minutes. The copper was heated to 1000°C during this procedure. This forms a single layer of graphene on each side of the copper foil. After a rapid cool-down we removed the copper foil in an aqueous iron nitrate solution. The graphene layer is then cleaned in DI water and subsequently transferred onto a commercial, dielectric laser mirror. This procedure yields a large area, broadband saturable absorber well suited for mode-locking low-gain solid state lasers. Fig. 1 shows a photograph of such a laser mirror coated with three large-area, single layer graphene flakes. The transmission loss of the sample shown in Fig. 1 was measured at 980nm and is approximately 2.5% (with the exception of some localized areas that show ~5% insertion loss). This fits well to the predicted 2.3% loss of a single layer of graphene.

Fig. 1: Photograph of several single layer graphene flakes on a ½” diameter, broadband dielectric laser mirror (contrast artificially enhanced for clarity). The insertion loss of the graphene covered areas is approximately 2.5%.
3. Experimental setup and results

To test these absorbers we built a laser based on Er:Yb:glass (Kigre Inc.: QX/Er, 1%Er, 20%Yb, 1.6 mm plate under Brewster angle). The laser glass was directly diode pumped by a pigtailed single transversal mode and single wavelength 980 nm laser diode. The cavity was an astigmatically compensated, X-fold cavity with an additional focus on one of the end mirrors, which was replaced by the graphene-based saturable absorber mirror (see Fig. 2a). The chromatic dispersion of the cavity was solely determined by the anomalous second-order dispersion of the laser glass. All cavity mirrors were commercial, low-dispersion broadband mirrors. The pulse repetition rate was 90 MHz. At an output coupling ratio of ~0.4% and a pump power of about 130 mW stable mode-locking at 4.5 mW output power (~1.1 W intracavity power) at a center wavelength of ~1550 nm was obtained. The mode-locked spectrum is shown in Fig. 2b. From an autocorrelation measurement we inferred to a pulse duration of 260 fs, assuming a Gaussian pulse shape. The beam diameter on the saturable absorber was ~30 µm, resulting in a maximum peak-intensity of ~6 GW/cm².

The saturable loss of the graphene-based absorber is currently only ~20% of the insertion loss. The small saturable loss of the current absorber is not sufficient to self-start the mode-locking process. The large non-saturable loss likely originates from lattice defects in the current graphene samples. We confirmed this assumption with Raman spectroscopy. An improved CVD recipe already showed improvements in the Raman spectrum and we believe that further optimizations, along with better annealing of the copper will lead to a much more favourable saturable-nonsaturable loss ratio.

4. Conclusions

In conclusion we have demonstrated a single-layer graphene-based saturable absorber mirror with insertion loss as low as 2.5%. These novel saturable absorber mirrors enabled 260 fs pulses in a low-gain Er:Yb:glass laser. Further improvement in the graphene growth will likely enable pulses in the sub-100 fs regime. We gratefully acknowledge the support from Dr. Kaoru Minoshima, AIST/NMIJ, Tsukuba Japan, who supplied us the Yb:Er:glass sample that was used in this experiment.

5. References