Highly efficient Tm$^{3+}$ doped germanate large mode area single mode fiber laser

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Abstract: We report the fabrication of a low numerical aperture large mode area, high concentration thulium doped germanate glass fiber using a rod-in-tube technique. The fiber core is 20 $\mu$m in diameter and operates in the single-mode regime in the 2 $\mu$m region. The Tm$^{3+}$ ion concentration is $3 \times 10^{20}$/cm$^3$ and the background fiber attenuation is 1.1 dB/m in the near infrared. We also demonstrate a high-power, high-efficiency fiber laser emitting at 1950 nm using a short (21-cm-long) length of the fiber. A slope efficiency of 55.9% with respect to absorbed pump power and a maximum output power of $\sim$1.52 W was achieved. This represents both the highest slope efficiency and the highest output power reported so far for Tm$^{3+}$ doped germanate single mode fiber using in-band core pumping. The large core area, short device length and high efficiency make this fiber attractive for the development of high peak power pulsed fiber amplifiers and single-frequency fiber lasers operating in the 2 $\mu$m region.

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1. Introduction

Fiber lasers operating in the eye-safe wavelength band of 2 $\mu$m have attracted a great deal of attention due to their potential use in applications such as LIDAR [1], laser surgery [2] and remote sensing [3]. Therefore, in recent years, considerable efforts have been devoted to the development and optimization of thulium and holmium doped glasses which emit at wavelengths around 1.95 $\mu$m and 2.1 $\mu$m respectively. In particular, substantial work has been dedicated to the development of effectively single mode, low numerical aperture (NA), large mode area (LMA) thulium doped fibers (TDFs) as a means to shorten active device lengths and hence to reduce the impact of optical nonlinearities in order to extend the performance limits of single-frequency lasers and pulsed fiber laser systems operating at wavelengths around 2 $\mu$m [4–6].

Having a large mode area is important for reducing the in-fiber pulse intensity and thereby increasing the threshold powers for nonlinear effects such as Stimulated Brillouin Scattering (SBS), Stimulated Raman Scattering (SRS) and Kerr based nonlinear phenomena such as Self Phase Modulation (SPM). Having a large core also results in a larger core/cladding overlap factor for cladding pumping which leads to an increased pump absorption per unit length, and hence shorter devices - which again serves to further increase the threshold for nonlinear effects. Likewise, increasing the rare earth ion concentration also reduces the length of fiber needed to
absorb the pump radiation and to achieve high levels of optical gain per unit length. This again serves to reduce the adverse effects of optical nonlinearities.

Only relatively low Tm-ion doping concentrations of $10^{18}$ - $10^{19}$ ions/cm$^3$ can be achieved in pure silica glass [7]. However, through the addition of dopants such as aluminum and phosphorous to the silica based core glass during the MCVD process, much higher levels of doping can be achieved ($\sim 10^{20}$ ions/cm$^3$) [8]. Unfortunately the addition of these dopants substantially raises the refractive index of the core glass relative to the surrounding silica (typical NA of $\sim 0.26$), and so these high Tm concentration core glass compositions are not directly compatible with LMA core designs unless a surrounding pedestal of passive doped silica glass (i.e. with no thulium doping) with a refractive index close to that of the thulium doped core is incorporated into the fiber cross-section. Whilst this can be done, and indeed excellent laser performance from meter scale devices can be achieved from fibers made this way [9], the need for a refractive index pedestal is undesirable and leads to increased fabrication complexity.

An alternative to pure or doped silica, glass systems do though exist which can in principle be doped with even higher rare-earth ion concentrations, typically up to $10^{21}$ ions/cm$^3$ [10]. These can be more readily and directly index-matched to suitable glass cladding materials to obtain simple LMA structures, thus raising the possibility of ultrashort fiber devices ($\sim$ a few to a few 10's of cm scale) to help mitigate nonlinearity. In particular, oxide glasses in the silicate [11] or germanate [6] families offer much higher levels of rare-earth ion solubility than silica glass and have thus attracted much attention in recent years. However the high phonon energy of silicate glasses ($\sim 1100$ cm$^{-1}$) [12] results in fast multiphonon relaxation, which can lead to an undesirable reduction in quantum efficiency and in turn to problems with thermal management (and ultimately damage) within a fiber laser cavity. By contrast, a germanate glass host offers a comparatively low phonon energy ($\sim 845$ cm$^{-1}$) [10] and hence a higher quantum efficiency, whilst providing excellent Tm$^{3+}$ ion solubility, and higher IR transparency. Multi-component germanate glass fibers with high Tm$^{3+}$ doping concentrations have now been developed [13–16] and a gain per unit length of 3.6 dB/cm at 1.95 $\mu$m was obtained for Tm$^{3+}$-doped germanate glass SM fibers [15]. However, aside from a few works reported using thulium-doped germanate glass double-clad single mode fiber [14,16], most germanate single-cladding fibers reported to date have typically exhibited relatively low laser efficiencies ($< 50\%$) and have only been operated at modest output power levels ($< 800$ mW) because of their relatively high background fiber attenuation, which directly affects the related laser performance. However, both high output power (104 W) and high slope efficiency (68%) [16] have now been reported for thulium-doped double-clad germanate glass fiber lasers, results that serve to highlight the potential of germanate glass and the importance of glass quality in realizing high performance laser systems in the 2 $\mu$m eye-safe waveband.

In this research we report on the development of high quality germanate glass materials suitable for the fabrication of LMA fibers doped with Tm$^{3+}$, Ho$^{3+}$ or Tm$^{3+}$/Ho$^{3+}$. In this initial work we concentrate on germanate glass fiber doped only with thulium which allows for easier laser experiments and simpler comparison with the large body of previous work. Specifically, we present recent results on the development of novel high concentration Tm$^{3+}$ doped, low-NA, LMA germanate fibers for fiber amplifiers and lasers operating in the 2 $\mu$m wavelength region. We report a high Tm$^{3+}$ concentration ($3 \times 10^{20}$ ions/cm$^3$), large core diameter (20 $\mu$m) low NA (0.07) single mode fiber and use it to realize an efficient single mode fiber laser at 1952 nm operating at an average output power of 1.5 W with a slope efficiency of 55.9%.

2. Fabrication and characterization of the Tm-doped germanate glass

The glass samples used in this work were synthesized by the conventional melting-quenching technique using high purity chemicals (more than 99.9%). The core and cladding glasses (labeled as Ge-01 and Ge-02, respectively) were produced from the chemical composition
of 58GeO$_2$-15PbO-13ZnO-4Nb$_2$O$_5$-7Na$_2$O-1.5SiO$_2$-1.5Al$_2$O$_3$ and the core glass was doped with Tm$^{3+}$ ions at a concentration of $3\times10^{20}$ ions/cm$^3$. The detailed Ge-01 and Ge-02 glass compositions were designed to provide both a low refractive index contrast for LMA single mode fiber operation and compatible thermo-mechanical properties for fiber drawing. For each glass composition, well-mixed precursor materials were melted at 1230 °C for about 3 hours in a platinum crucible under a dry O$_2$/N$_2$ atmosphere to reduce the OH$^-$ content. Thereafter, the molten liquid was cast into a stainless steel mold preheated to a temperature around the glass transition temperature, $T_g$. The cast glass was then annealed at $T_g$-10 °C for 4 hours to relieve internal stresses before it was cooled to room temperature. After the annealing process, glass samples (12 mm in diameter and 4 mm thick) were cut and polished for optical and spectroscopic characterization.

Differential thermal analysis (DTA) was carried out using a PerkinElmer Diamond TG/DTA instrument at a heat rate of 5 °C/min from 40 °C to 1350 °C under a N$_2$ atmosphere, in order to assess the glass transition temperature ($T_g$) and the crystallization temperature ($T_x$). The glass transition temperatures were measured to be 485 °C and 489 °C for Ge-01 and Ge-02, respectively. Using the DTA data we can calculate the stability parameter $\Delta T = T_x - T_g$, which gives an indication of the stability of the glass and its tendency of devitrification [17]. The difference between the crystallization temperature and the glass transition temperature was 260 °C for the core glass. Such high transition temperatures and high thermal stability, as compared to other reported germanate glass compositions, makes the fabricated glasses suitable for sustaining the multiple thermal processes involved with fiber fabrication and for handling high average powers without incurring crystallization or devitrification [18]. The coefficient of thermal expansion (CTE) was measured using a PerkinElmer Diamond TMA instrument at a heat rate of 5 °C/min from 40 °C to 1350 °C with a constant compressive force of 5 mN. As summarized in Table 1, the CTE of the core and cladding glasses was 7.18×10$^{-6}$ / °C and 6.66×10$^{-6}$ / °C, respectively and the relatively small difference between the two glasses ( ~ 7.2%) is sufficient to enable us to draw the fiber with negligible residual stresses at the core-cladding boundary. The refractive indices of both the core and cladding glasses were measured using ellipsometry (MC05-Woolham-Ellipsometer) and were 1.8025 and 1.8012, respectively, at 1.7 µm. The NA was thus as low as 0.07 ± 0.005, a value that allows single mode behavior at wavelengths around 2 µm in a fiber with a core diameter as large as 20 µm.

A series of three core/clad batches was also manufactured. The corresponding standard deviation on the core refractive index value was 0.0002, while the standard deviation (SD) on the numerical aperture value was 0.01. These low SD values illustrate the control and reproducibility of the glass manufacturing process, and it gives us confidence in the prospect of achieving core/clad glass compositions offering even lower NA values than that reported here.

The absorption spectra of the core glass, doped with Tm$^{3+}$ ions at a concentration of $3\times10^{20}$ ions/cm$^3$, was measured using a UV-visible NIR double beam Agilent Cary 500 spectrometer in the wavelength range from 200 to 2000 nm. Note that although higher thulium concentrations have been reported in germanates ($8\times10^{20}$ ions/cm$^3$ [15]), this initial result already represents a high thulium concentration and we have every confidence that higher thulium concentrations will be possible in our glass matrix with further optimization and this will be a topic of future study. As shown in Fig. 1, strong absorption peaks were observed at 660, 792 and 1210 nm and a broadband peak around 1652 nm. The fluorescence emission was measured at room temperature.

### Table 1. Properties of the glasses used in this work

|                | Density (g/cm$^3$) | $T_g$ (°C) | $T_x$ (°C) | $\Delta T$ (°C) | Refractive index @ 1.7 µm | CTE (10$^{-6}$/°C) |
|----------------|-------------------|------------|------------|-----------------|--------------------------|-----------------|
| Core (Ge-01)   | 5.38              | 485        | 745        | 260             | 1.8025                   | 7.18            |
| Cladding (Ge-02)| 5.11              | 489        | 734        | 245             | 1.8012                   | 6.66            |
under 790 nm pump laser excitation through a multimode silica glass fiber (200 µm core diameter and 0.22 NA). The measured fluorescence spectrum can be used to estimate the emission cross section ($\sigma_e$) by using the Füchtbauer-Ladenburg (FL) equation as below [19]:

$$\sigma_e^{FL}(\lambda) = \frac{A_{ij}^5 I(\lambda)}{8 \pi n^2 c \int A I(\lambda) d\lambda}$$

(1)

where $A_{ij}$ is the spontaneous emission probability, $I(\lambda)$ is the emission intensity, $\lambda$ is the wavelength, $n$ is the refractive index and $c$ is the speed of the light. Figure 2(a) shows the calculated absorption ($\sigma_a$) and emission cross section ($\sigma_e$) of the core glass with a peak value of $4.1 \times 10^{-21}$ cm$^2$ at 1650 nm and $7.06 \times 10^{-21}$ cm$^2$ at 1875 nm, respectively. Note that the core glass has a larger $\sigma_e$ than that of previously reported Tm$^{3+}$ doped silicate glass ($3.89 \times 10^{-21}$ cm$^2$) [20], ZBLAN ($2.4 \times 10^{-21}$ cm$^2$) glass [21], silica ($4.6 \times 10^{-21}$ cm$^2$) glass [22] and other germanate ($5.95 \times 10^{-21}$ cm$^2$) glasses [10]. Based on the calculated $\sigma_a$ and $\sigma_e$, the wavelength dependence of the net gain was analyzed to estimate the gain properties as a function of population inversion. The net gain coefficient was expressed using the following equation [23]:

$$G(\lambda) = N [p \sigma_e(\lambda) - (1 - p) \sigma_a(\lambda)]$$

(2)

where $N$ is the concentration of Tm$^{3+}$ ions and $p$ is the population of the upper laser level. Figure 2(b) shows the calculated gain coefficient of the core glass for $p$ ranging from 0 to 1 with a step of 0.1. From this calculation, we expect that with $p$ higher than only 0.1 we should already have a
positive gain in the wavelength range from 1900 to 2000 nm, and that a relatively low population inversion (i.e. low pump power) should thus be required for 2.0 µm laser operation in our Tm\(^{3+}\) doped germanate fiber. Furthermore, the maximum simulated gain coefficient reaches 2.11 cm\(^{-1}\) at 1878 nm, which is larger than that of Tm\(^{3+}\)-doped silicate glass (1.5 cm\(^{-1}\)) [12], showing GE-01 to be a promising gain host material for efficient 2.0 µm fiber laser development.

The fluorescence lifetime of the \(^3\)F\(_4\) level of Tm\(^{3+}\) ions was measured at room temperature using a modulated 793 nm laser diode with a 6-ms-duration square pulse. The fluorescence decay from the glass was collected using a multimode fiber connected to an oscilloscope equipped with an InGaAs photodetector (PDA10D-EC 1200 nm-2600 nm). As shown in Fig. 3, unlike the fluorescence decay of Tm\(^{3+}\) doped silica fiber (fitted with a double exponential function [24]), our Tm\(^{3+}\) doped germanate fiber shows a better fit with a single exponential decay function. The measured lifetime for our fiber with a Tm\(^{3+}\) doping concentration of 1.84 wt% (3×10\(^{20}\) ions/cm\(^3\)) was ∼960 µs. This is considerably longer than the lifetime of other glass types with similar dopant concentration, e.g. 420 µs measured in a 1.44 wt% Tm\(_2\)O\(_3\) silica fiber [22] and 460 µs measured in a 1.46 wt% Tm\(_2\)O\(_3\) silicate fiber [12]. Note that a long lifetime is a favorable property in achieving the population inversion required to realize an efficient fiber laser with a low laser threshold. The longer lifetime in our fiber stems from its “low” OH content (∼700 dB/m at 3 µm), compared to the 1300 dB/m we measured before improving our glass preparation process, and from a reduced number of extrinsic quenching centers, and it thus highlights the overall quality of the glass developed. We expect that even better lifetime results could be achieved with additional improvements in glass preparation to further reduce the content of OH\(^-\) groups, which are responsible for the quenching of the energy transfer process between Tm\(^{3+}\) ions. Compared to other multi-component glasses reported in the literature, the absorption coefficient of 700 dB/m (∼1.5 cm\(^{-1}\)) is higher than that of another Tm\(^{3+}\) doped germanate glass (0.39 cm\(^{-1}\)) [10] but lower than that of Tm\(^{3+}\) doped lead silicate glass (1.7 cm\(^{-1}\)) [25].

![Fig. 3. Measured fluorescence lifetime of the core glass using a modulated 793 nm pump LD.](image-url)

3. Fabrication and characterization of the Tm-doped germanate LMA single mode fiber

For the fabrication of Tm\(^{3+}\) doped germanate LMA fiber, the Ge-01 glass rod (120 mm long and 11.5 mm in diameter) was first drawn into a cane of 1.3 mm diameter. The Ge-02 glass billet was then extruded into a tube of 9.5 mm outer diameter and 1.7 mm inner diameter. To extrude the cladding glass, a ram speed of 0.05 mm/min was used when glass first flowed out of the die and 0.1 mm/min after a few centimeters had emerged. The temperature was around 575-615°C in order to have a force of 1.1-2.5 kN. Both speed and temperatures were chosen in
order to avoid bending. The Ge-01 glass rod and Ge-02 glass tube were then co-drawn into an optical fiber using a rod-in-tube technique, at a speed of 5.4 m/min. The fiber core and cladding diameters were $20 \pm 0.5 \, \mu m$ and $110 \pm 1 \, \mu m$, respectively. As shown in Fig. 4, there are no signs of residual stresses or bubbles at the interface between the core and cladding, which demonstrates the compatibility of these glasses and their CTE and the quality of the drawing process.

![Microscope Image](image)

**Fig. 4.** Optical microscope image of the fabricated Tm doped Germanate LMA single mode fiber.

The fiber loss was measured to be $\sim 1.14 \, dB/m$ at 980 nm by a cut-back method over a length of 5.3 m. This is substantially lower than that of previously reported Tm$^{3+}$ doped lead silicate glass (7 dB/m) [25] and germanate glass (3 dB/m) [26] fibers, and it confirms the absence of crystallization during the fiber fabrication. The background fiber loss was also confirmed using a White Light Source (WLS) and an Optical Spectrum Analyzer (OSA) as it is shown in Fig. 5. For both measurements the fiber was manually coated with high refractive index paint (Graphite).

As shown in Fig. 5, due to the high absorption of the doped core glass, the fiber at 793 nm and at 1565 nm was too high to be measurable in that experiment. We did however perform an additional cutback, from 4.2 cm to 2.8 using a more powerful Supercontinuum (SC) source, to cross check the pump absorption value at 1.7 um (inset of Fig. 5). The measurement confirmed the same $\sim 500 \, dB/m$ in the fiber as was measured for the bulk glass. Figure 5 shows also a comparison between the measured loss in the fiber and in bulk, where the bulk measurement is inaccurate due to Fresnel reflections and excess optical losses in the spectrometer. From this we can estimate that the absorption of the fiber at 793 nm and 1565 nm will be not too far from the 1051 dB/m and 62 dB/m measured in bulk.
Fig. 5. Loss superposition curves of the core bulk glass (UV-Visible NIR spectrometer) and the thulium-doped germanate fiber (WLS+OSA). The inset shows a cutback over a very short piece of fiber using a supercontinuum source to infer the pump absorption at 1700 nm.

4. Fiber laser performance

4.1. Experimental set-up for the Tm$^{3+}$ doped GeO$_2$ fiber laser

To check the laser performance of the fiber, we tested it in a simple laser configuration. A schematic diagram of the Tm$^{3+}$-doped germanate fiber laser is depicted in Fig. 6. An in-house built erbium-ytterbium co-doped fiber laser was employed as a pump source and a 21 cm length of the in-house fabricated Tm$^{3+}$-doped germanate glass fiber was used as the gain medium. The fiber laser cavity was constructed by splicing a highly reflective fiber Bragg grating (FBG) at one end of the cavity, centered at 1952 nm with a FWHM of 0.12 nm, and a flat cleaved germanate fiber at the other, which served as a partially reflective output coupler with a Fresnel reflection of 8.18%. Note that the FBG is a wavelength selective device allowing the 1565 nm pump light to couple into the fiber without introducing additional loss whilst locking the laser wavelength at 1952 nm. The laser output beam was collimated using an aspheric lens with a focal length of 15 mm and split from the residual pump light using two dichroic mirrors (high reflectivity at 2 µm and high transmission at 1565 nm). The laser output was measured using a thermal power meter (Ophir 3A-FS) and an optical spectrum analyzer (Yokogawa, AQ6375) with a resolution of 0.1 nm.

Fig. 6. Schematic diagram of the Tm-doped germanate fiber laser.
4.2. Laser output performance

The laser output power was measured as a function of pump power and the results are shown in Fig. 6(a). The threshold pump power was about 0.3 W and the maximum output power was $\sim 1.52$ W with a slope efficiency of 38.7% with respect to the launched pump power. When taking into consideration the residual pump power and 1.1 dB splice loss between the germanate fiber and the conventional single mode fiber (SMF28) in which the grating was written, the effective slope efficiency with respect to the absorbed pump power was 55.9%. This is to the best of our knowledge the highest slope efficiency and output power so far reported for an in-band pumped Tm$^{3+}$-doped germanate fiber in a core-pumped configuration. The side mode suppression ratio (SMSR) of the laser output was higher than 55 dB and the 3 dB laser bandwidth was less than 0.2 nm. Based on the approach in [27], we numerically modelled the laser using the commercial software “RP Fiber Power”. As shown in Fig. 7(a), when pumped at a wavelength of 1565 nm, the simulated slope efficiency reached 57.2%, in good agreement with the experiment. Although J. Wu et al [5] achieved a slightly higher laser efficiency (58%) using a different pump scheme (808 nm laser diode), our results was obtained for power output nearly two orders of magnitude higher with no particular sign of thermal roll over which suggests again the high quality of the glass material and the developed fiber.

At a fixed pump power of 4 W we used the model to estimate the optimum length of germanate fiber. As shown in Fig. 7(b), lasing is possible down to a minimum length of 4.8 cm and the output power gradually increases as the fiber length is increased from 4.8 cm to 41 cm. The maximum achievable output power is predicted to be $\sim 2.02$ W at the optimum fiber length of 41 cm. Beyond this length, the output power decreases because of the quasi-three-level behavior between the $^3H_6$ and $^3F_4$ levels.

In order to characterize the beam quality of the laser output, the $M^2$ factor was measured at a laser output power of 1 W. An optical attenuator was included after the dichroic mirror (DM2) and $\sim 50$ mW of optical power was used to avoid any damage to the beam profiler Nanoscan (NS2s-Pyro9/5-STD) and beam-profiling camera (Pyrocam III). The $M^2$ factor was measured by focusing the laser beam with a 100 mm aspheric lens and by measuring the beam diameter (or beam divergence) around the focus. As shown in Fig. 8, a hyperbolic fit to the beam width data is used to extract the $M^2$ value for each direction. The measured $M^2$ was 1.28 and 1.12 along the two perpendicular directions. These close-to-unity values demonstrate the high quality SM beam of the as-drawn fiber and are a further indirect measure of the quality and control of both glass and fiber fabrication processes.
5. Conclusion

In summary, we have reported a significant technological step towards the realization of practical active fibers for the realization of compact single frequency lasers and amplifiers for ultrashort pulses operating in the 2 µm region. We have developed two thermally compatible germanate glasses in the GeO$_2$-PbO-ZnO-Nb$_2$O$_5$-Na$_2$O-SiO$_2$-Al$_2$O$_3$ system which can be co-drawn into a fiber. The ability to reproducibly control the index difference between these glasses down to an NA of 0.07 has allowed us to produce a single mode fiber (M2 ~1.2) with a core diameter as large as 20 µm. As a result of meticulous additional attention to the optimization of the glass material quality and to the control of the fiber fabrication process, the fiber has a background loss of ~1 dB/m, the lowest value reported to date for germanate fibers. The core glass was doped with Tm$^{3+}$ ions at a concentration of 3×10$^{20}$ ions/cm$^3$ (1.84 wt%), which is high but by no means the highest that can be achieved with this glass system. Therefore further optimization of thulium concentration will be our priority in the further development of our newly developed germanates glass and fibers. A single mode fiber laser operating at 1952 nm was then demonstrated using a 21 cm length of Tm-doped germanate fiber, with in-band pumping at 1565 nm. The maximum achieved output power of ~1.5 W (limited only by the available pump power) and the slope efficiency of 55.9% are to the best of our knowledge both record results for in-band pumping in this particular glass system. Note that without any optimization of the dopant concentration, we have already achieved similar levels of pump absorption as in state-of-the-art alumino-silicate fibers [8,9] which have been optimized over many years. Our solution has the added advantage that a much simpler step-index refractive index profile can be achieved, without the need of a pedestal which is undesirable in many practical applications. These results reveal the high quality of the in-house fabricated Tm$^{3+}$ doped germanate glasses and LMA fiber and demonstrate that such Tm$^{3+}$ doped germanate LMA fiber represents a very promising solution to reduce nonlinearities in short pulse amplifiers and single frequency lasers. Significant further improvements are expected from optimization of the doping concentration of Tm$^{3+}$ and Tm$^{3+}$/Ho$^{3+}$ for the longer wavelength windows in the 2 µm region, from further reduction of the OH content in the glass and also from developing new fiber design like the hexagonal or the D-shaped inner cladding in the case of a double cladding Tm-doped Germanate glass fiber. Note that for a double cladding fiber, cladding-pump scheme becomes possible using commercially available high power 790 nm laser diodes. This enables exploitation of the cross-relaxation process (2-for-1), which in theory could double the achieved slope efficiency (up to 82% with this glass). Having said that, we believe that a symmetric inner cladding structure (e.g. hexagonal), could lead to improved slope.

Fig. 8. Measured beam quality of the output beam. The far field beam profile is shown in the inset.
efficiency and output power as compared to an asymmetric structure (D-shape) due to a lower expected splicing loss to conventional fibres.

Funding

Engineering and Physical Sciences Research Council (EPSRC) (EP/P030181/1); H2020 European Research Council (ERC) (682724).

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