Standoff spectroscopy via remote generation of a backward-propagating laser beam

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In an earlier publication we demonstrated that by using pairs of pulses of different colors (e.g., red and blue) it is possible to excite a dilute ensemble of molecules such that lasing and/or gain-swept superradiance is realized in a direction toward the observer. This approach is a conceptual step toward spectroscopic probing at a distance, also known as standoff spectroscopy. In the present paper, we propose a related but simpler approach on the basis of the backward-directed lasing in optically excited dominant constituents of plain air, N_2 and O_2. This technique relies on the remote generation of a weakly ionized plasma channel through filamentation of an ultraintense femtosecond laser pulse. Subsequent application of an energetic nanosecond pulse or series of pulses boosts the plasma density in the seed channel via avalanche ionization. Depending on the spectral and temporal content of the driving pulses, a transient population inversion is established in either nitrogen- or oxygen-ionized molecules, thus enabling a transient gain for an optical field propagating toward the observer. This technique results in the generation of a strong, coherent, counter-propagating optical probe pulse. Such a probe, combined with a wavelength-tunable laser signal(s) propagating in the forward direction, provides a tool for various remote-sensing applications. The proposed technique can be enhanced by combining it with the gain-swept excitation approach as well as with beam shaping and adaptive optics techniques.

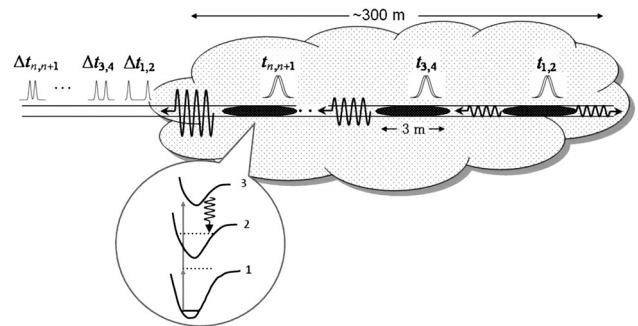air laser | atmospheric surveillance | threat detection | Raman

Fig. 1. SOS. Multiple pairs of pulses are generated such that the spacing between pulses in each pair is decreasing. The second pulse in each pair has a higher velocity because of atmospheric dispersion. The first pair of pulses overlaps near the back of the cloud, creating a small region of gain. Subsequent pairs overlap at closer and closer regions of the cloud, producing a swept-gain amplifier that lases back toward the observer.

The continuous monitoring of the atmosphere for traces of gases and pathogens at kilometer-scale distances is an important and challenging problem, with applications in environmental science and national security. Measurements using light detection and ranging (LIDAR) techniques coupled with differential absorption LIDAR have been reported (1, 2). These techniques provide valuable tools for the measurement of trace impurities in the atmosphere.

However, to enhance sensitivity and information content retrieved by the return signal, a different technology is needed. In ref. 3, we presented standoff spectroscopy (SOS) technique for detection of trace impurities in the atmosphere via gain-swept superradiance (4). In that scheme, we first pump the molecules of interest, e.g., nitric oxide or phosgene, at some prearranged distance, say, 300 m. These molecules decay spontaneously to a lower state via a specific radiation frequency. Then the impurity molecules at, say, 290 m, are excited at a later time that is delayed from the first pulse by $\Delta\tau = \frac{10m}{c} \cong 3 \times 10^{-8}$ s, so that some gain is realized in the second region. Subsequent regions of inversion are generated by later pulse pairs as in Fig. 1. In ref. 3, which will be briefly reviewed in *Gain-Swept Lasing of Impurities in Air*, the impurity molecules themselves constitute the lasing medium. In the present paper, we propose a simpler alternative approach that is based on transient backward-directed lasing in atmospheric gases such as N_2 and O_2 on the far side of the region to be probed.

The basic idea behind the remote generation of a back-propagating laser probe beam is illustrated in Fig. 2. This technique relies on the creation of a "seed" plasma channel or series of chan-

nels via controlled prechirping of intense femtosecond laser pulses at the launch site. These pulses self-compress through natural dispersion of air as they propagate. At a predetermined remote location, the peak power of these pulses exceeds the threshold for self-focusing in the air. The pulses collapse transversely and form laser filaments, leaving weakly ionized plasma channels in their wake. A high-energy, nanosecond laser pulse that immediately follows the seed pulse deposits energy into the plasma channel and creates conditions for optical gain in the backward direction.

The above approach is particularly plausible if applied to lasing in atmospheric nitrogen at the 337-nm wavelength (Fig. 3). N_2 lasing has been previously demonstrated at atmospheric pressure by using electrical discharge and microwave pumping and detailed analysis shows that such a filamentation N_2 air laser is feasible. Another possibility is to utilize various visible and near-infrared transitions in the ionized oxygen, although this route is more challenging. Previously, oxygen lasing has been demonstrated only at gas pressures substantially lower than atmospheric pressure. However, oxygen lasing in realistic open-air conditions has now been demonstrated (5). An additional degree of control over optical excitation of atmospheric gases can be offered by spatial beam shaping and adaptive optics.
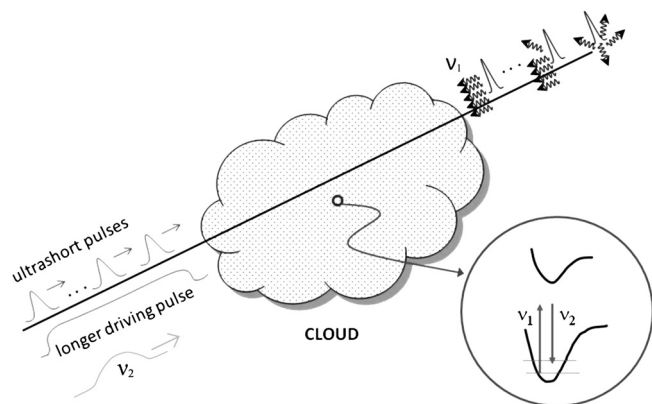
The utilization of the strong backward-propagating optical probe in remote spectroscopy applications will result in a dramatic improvement of the detection sensitivity compared to the standard LIDAR techniques. LIDAR is based on the detection of the nearly isotropic scattering of a forward-propagating probe

Fig. 2. An ultrashort laser pulse or sequence of pulses is prechirped in such a way that they become compressed by the air dispersion at a prearranged distance behind the cloud. Self-focusing collapse of these pulses results in the generation of weakly ionized seed plasma channels. The plasma density in these seed filaments is increased by several orders of magnitude by the application of a longer drive pulse. The properties of the pulses are tailored to produce population inversion in the ionized N_2 and O_2. The air-laser radiation at frequency ν_1 is combined with an interrogation pulse at ν_2 to identify trace amounts of gas, such as NO_2, in the cloud.

beam. The component of this scattering that propagates toward the observer is a priori weak. On the other hand, the sensing approach proposed here employs a bright and highly directional backward-propagating probe, thus increasing the signal-to-noise ratio of the detected signal dramatically. Potential schemes of remote spectroscopic analysis using a back-propagating coherent optical probe are described in *Experimental Plan*.

Gain-Swept Lasing of Impurities in Air

Let us briefly summarize the ideas of ref. 3, which provides the conceptual background for the present work. As depicted in Fig. 1, the key idea is the generation of a gain-swept excitation via pulse "catch up" due to atmospheric dispersion. Consider the initial pulse pair that overlaps at the far side of the cloud. We arrange such conditions that the first pulse in the pair is at a higher frequency (e.g., $\lambda_1 = 400$ nm), and the second pulse is at a lower frequency (e.g., $\lambda_2 = 580$ nm). The two pulses have different group velocities because of the dispersion of air. The initial delay time Δt_{12} between the first two pulses determines the distance z at which the faster-moving lower-frequency pulse catches up with the slower-moving higher-frequency pulse.

The group velocity in air is given by

$$v_g \cong c\left(1 + \lambda\frac{dn}{d\lambda}\right),$$ **[1a]**

where n is the refractive index of air whose dependence on the wavelength is given by the following formula:

$$n(\lambda) \cong 1.0007566 + \frac{1.34}{\lambda^2[\text{nm}]} + \frac{3.777 \times 10^4}{\lambda^4[\text{nm}]}.$$ **[1b]**

For example, the difference in group velocities of the two pulses for the case when $\lambda_1 = 400$ nm and $\lambda_2 = 580$ nm is $\Delta v_g \approx 4 \times 10^5$ cm/s. Thus the pulses will overlap at a time τ such that the initial pulse separation $\Delta L = \Delta v_g \tau$. Hence, for $\Delta L = 1$ mm, $\tau \cong 2 \times 10^{-5}$ s; i.e., the pulses will overlap at a distance of about 5 km from the transmitter.

The following pulse pairs are to be delayed so as to generate a population inversion swept back toward the sender. The relaxation time of the population inversion caused by collisions in the

air is in the nanosecond range. Therefore, each pair of visible pump pulses should follow the previous pair by the time T_{rep} on the order of 1 ns.

This swept-gain configuration yields a beam of exponentially amplified spontaneous radiation that is gain-guided in the backward direction. Because, in principle, the gain path can be made arbitrarily long, it is possible to generate a strong signal from only parts per million concentration of the impurity in the atmosphere.

Lasing of N_2 and O_2 in Air

The gain-swept approach described above turns the analyte to be sensed into a wavelength-selective active optical medium. This approach is very powerful because it provides for the ultimate control over temporal and spatial distributions of optical gain throughout the sensing region. However, the practical implementation of this technique will be extremely challenging because of the overall complexity of the approach. Each particular analyte gas will have its own unique lasing parameters. Therefore, the pulse-shaping apparatus involved in the generation of adequate pulse sequences will either have to be dedicated to sensing a particular gaseous substance at a particular range or be excessively complex. A simpler SOS scheme is needed to make the overall approach more practical. This simpler alternative that we propose here relies on using major constituents of air, such as nitrogen and oxygen, as gain media for the generation of a backward-propagating coherent optical probe. This probe will be used in combination with a tunable forward-propagating laser beam(s) for a remote Raman-type spectroscopic analysis of trace amounts of various gases in the atmosphere.

In the gain-swept scenario, the extremely low concentration of the analyte that at the same time was acting as the laser gain medium was translated into the very low per-unit length value of gain. Consequently, a long gain path was required. In the case of the dominant constituents of the ambient air, the gas concentration is orders of magnitude higher, and a much shorter gain path (of the order of 1 m or even less) would suffice. In fact, there are a variety of commercial gas lasers, including a molecular nitrogen laser (6), that are compact and efficient devices. These lasers can serve as models, which we will attempt to emulate in ambient air. The main challenge in such emulation will be to replace the discharge excitation mechanism used in the overwhelming majority of practical gas lasers with a remote optical excitation using laser beams. In addition, contrary to commercial gas lasers, there will be no freedom in optimizing the gas pressure and composition. The conditions for the creation of population inversion will have to be realized in plain air under atmospheric pressure, whereas most commercial gas lasers operate at much lower pressures and utilize particular selected mixtures of gases.

Central to the alternative SOS strategy, on the basis of lasing in the major constituents of the ambient air, is the process of femtosecond laser filamentation (7–9). When a light beam with peak power exceeding the self-focusing threshold ($\sim$3 GW = cm^2) propagates in the air, its transverse intensity profile shrinks because of self-focusing. In the absence of any defocusing mechanism, the self-focusing would result in a collapse of the beam profile into a singularity. What stops the collapse is the defocusing action of plasma generated on the beam axis via multiphoton ionization. The peculiar object that is composed of the hot and intense core and the generated plasma on the beam axis is termed the laser filament. Laser filaments in air are typically about 100 μm in diameter. They have been shown to propagate over distances orders of magnitude longer than the Raleigh range corresponding to their transverse dimensions.

The light intensity in the filament core is clamped to the level approximately equal to the multiphoton ionization threshold, about 10^{13}–10^{14} W/cm^2 in the air. The combination of this high level of intensity and the extended propagation distance facilitates efficient nonlinear-conversion processes in the filament

core, which result in the bright emission of broadband supercontinuum light (10). This emission is a signature phenomenon of filamentation; it makes femtosecond laser filaments potentially useful as light sources in LIDAR, but it cannot be used in our SOS scheme because the supercontinuum emission by filaments is forward-propagating.

Because oxygen has the lowest ionization potential of all constituents of air, the overwhelming majority of ionized species inside laser filaments constitutes singly ionized oxygen molecules and freed electrons. This plasma is very dilute. Although reported experimental data on the plasma density inside filaments vary by about 2 orders of magnitude, it is certain that less than 0.1% of all oxygen molecules are ionized. Thus, ultrafast laser filaments themselves cannot serve as practical gain media for efficient oxygen or nitrogen lasers. The ionization fraction of oxygen is too low, and that of nitrogen is even lower. Simple scaling of the pulse energy is not going to help because filamentation is a self-consistent process, and increasing the pulse energy typically results in the creation of multiple filaments inside the transverse profile of the laser beam, not in a higher plasma density in a single filament. However, high plasma density is needed for the lasing action in the ambient air to occur.

The solution that we propose to this problem is the use of a dual-pulse excitation scheme. In this approach, the femtosecond pulse with peak power above the self-focusing threshold but limited pulse energy of the order of tens of millijoules (the igniter) creates the filament, thus providing seed electrons that are liberated from oxygen molecules by multiphoton ionization. A second, much longer pulse with joule-level pulse energy (the heater) accelerates the seed electrons and initiates an electron avalanche that ionizes a substantial fraction of nitrogen and oxygen in the air. A sequence of multiwavelength heater pulses may have to be applied in order to select a particular excitation pathway in the air molecules and thus affect the wavelength of the backward-generated probe beam.

In order to initiate an electron avalanche, the duration of the heater pulse needs to exceed the electron collision time in air, ~350 fs (11). In addition, the complete ionization of air inside a 1-m-long filament with a diameter of 100 μm will require pulse energy of the order of 1 J. A standard Q-switched lamp-pumped Nd:YAG laser and its harmonics easily satisfy both of these requirements.

The igniter–heater scheme described above has been used previously for the creation of centimeter-long dense plasma channels in laser wake-field tabletop accelerators and Raman lasers (12, 13). We extend this approach to meter-scale plasma channels in ambient air.

Nitrogen, the most abundant constituent of air, offers the most straightforward choice for the application of this approach. Nitrogen is one of the few gases that have been shown to produce efficient lasing in the UV range at atmospheric pressure. In fact, stimulated emission in the backward direction from atmospheric nitrogen excited by an ultrafast laser filament has been reported (14). The femtosecond pulse was the only source of excitation in that work, and no heater pulse was used. Consequently, the density of the ionized nitrogen molecules was extremely low, and the emitted radiation was too weak to be practical or even to be properly characterized in terms of power, pulse energy, and precise spectral content. Furthermore, remote excitation of atmospheric nitrogen by microwave pulses has been shown to result in quite efficient transient lasing (15). The backward-propagating laser pulse in that case had three spectral lines at 316, 337, and 358 nm, and the total generated pulse energy was in tens of microjoules. Commercial nitrogen lasers that operate at about one-tenth of atmospheric pressure and utilize electric discharge excitation produce UV pulses at the 337-nm wavelength, in Fig. 3, with energy in the hundreds of microjoules (6). This level of performance can be achieved by using our dual-pulse excitation

approach. A potential complication may be related to the presence of oxygen, which provides for an alternative, preferential pathway to ionization with a lower ionization potential than that of the nitrogen. For that reason, oxygen is typically viewed as being toxic in nitrogen lasers with discharge excitation. At the same time, it is possible that smart application of coherent-control techniques may turn this disadvantage into a benefit and utilize nitrogen-oxygen collisions as a mechanism for emptying the lower lasing level in the nitrogen laser.

The second major constituent of air is oxygen. Because the ionization potential of oxygen is the lowest among air constituents, oxygen is the only air species that becomes appreciably ionized through ultrafast filamentation. Under electric discharge excitation, oxygen has been shown to lase in various spectral lines throughout the UV, visible, and near-IR parts of the spectrum, but in the past lasing was demonstrated only at pressures much lower than atmospheric pressure (16, 17). Moreover, we have now made oxygen lase under optical filament excitation at atmospheric pressure (5). Technical realization of neon-oxygen and argon-oxygen lasers offers insights into possible routes to inversion (18, 19). These lasers belong to the general class of dissociative excitation transfer lasers. Specifically, inversion is produced by the collision of the excited-state neon and argon atoms with molecular oxygen. This process leads to the production of two oxygen atoms—one in the ground and the other in the excited state. Two inversion mechanisms are possible depending on the energy of the excitation transfer from the respective noble gas. For the case of neon in the $^3P_2$ excited metastable state, the resonant energy transfer of approximately 16.6 eV leads to oxygen dissociation and production of oxygen atoms in the ground and 3^3P states. In this case, lasing occurs in the fine structure on the 3^3P-3^3S transition at 844.6 nm as well as on the 3^5P-3^5S transition at 777.3 nm. For the case of the inelastic collisions with excited-state argon, atomic oxygen in 2^1S and 2^1D can be further excited by electron impact in an electric discharge to yield emission from the same transitions discussed above. The intensities of the femtosecond laser pulses in a laser filament are on the order of 10^{13}–10^{14} W/cm^2. This intensity level suggests a possibility of the nonlinearity that is sufficiently strong to mimic inelastic collisional excitation through multiphoton excitation by the laser pulses at the 800-nm wavelength commonly used for filament generation. Multiphoton excitation of molecular oxygen to the states accessible by the collisional energy transfer from neon and argon atoms, primarily in the Schuman–Runge continuum above the $^3\Sigma_u$ state, can yield excited-state atomic oxygen and create conditions for lasing.

In summary, ultrafast laser filamentation can be applied to mimic physical processes involved in the creation of population inversion in the constituents of ambient air by electrical discharge. The possibilities of the interplay between constituents of the medium, in this case molecular nitrogen, oxygen, and their atomic dissociation products, as well as the electron impact in the plasma provide numerous possibilities for pumping inversion in any of these constituents. Whereas filament configuration is predominantly defined by the properties of the initial ultrashort pumping laser pulse, the excitation of the medium to states enabling stimulated emission at the specified wavelengths can be achieved and optimized via intelligent or adaptive pulse shaping of the filament pulse itself or via the application of auxiliary laser pulses.

Experimental Plan

Our immediate experimental plan is as follows. We will first study cavityless air lasers in a controlled laboratory setting. We will use a single millijoule-level femtosecond igniter pulse to produce a relatively short (meter-scale) plasma filament, approximately 0.1 mm in cross-section. This low carrier-density plasma will then be heated by a joule-level nanosecond laser pulse timed with

respect to the igniter. We will determine optimal conditions (pulse shapes, frequencies, and relative timing) that result in denser plasma and investigate various nitrogen and oxygen lasing schemes discussed above. At that step we expect to demonstrate a single efficient "open-air" bidirectional laser (lasing simultaneously forward and backward). The main difference of our air laser from other easily constructed air lasers will be the plasma preparation mechanism (using a pump laser beam instead of, for example, an electric discharge). We expect the probability of success at this first step to be high.

We will then verify that the distance from the pump laser source to the filamentation and lasing air region can be readily controlled by a negative prechirp of the igniter pulse. There is sufficient prior work to give us confidence that this can be done (20, 21). A further improved control over the distance at which filamentation occurs can be achieved by adjusting the igniter beam focusing geometry, as was also demonstrated in the past (22).

After an efficient open-air bidirectional laser is attained, and after we demonstrate an ability to control its position (with respect to the source laser), we will move to the next step, which will be to demonstrate production of multiple laser gain regions at controlled distances with controlled timing, so as to achieve the swept-gain scheme and demonstrate the preferentially backward lasing. We will work toward this more difficult task in a laboratory setting first. An extension to a backward swept-gain laser many kilometers away will pose additional challenges (due to, for example, beam distortion by air turbulence) that could be addressed through an adaptive optical technique, such as was recently applied in order to enhance filaments at large distances (23).

SOS via Raman Spectroscopy

We next briefly discuss remote-sensing scenarios using our coherent backward air-laser beam that produce a strong species-specific signal at the detector. This back-propagating signal is in contrast to the conventional (1, 2) (e.g., LIDAR) weak signals that are based on incoherent scattering with no preferred direction.

We consider spectroscopic configurations that use the remote air laser to interrogate the suspect "cloud" and detect trace amounts of gas (as alluded to in Fig. 2). Linear absorption spectroscopy will clearly be of little use, because the air laser will typically operate at a "single" wavelength, producing a relatively narrow spectral band without the possibility for wavelength tuning. An obvious route is to do nonlinear spectroscopy and supplement the backward-generated light by additional laser pulses (multiwavelength, tunable, or broadband) sent in the forward direction. The end goal is to induce a coherent, directional, signal beam
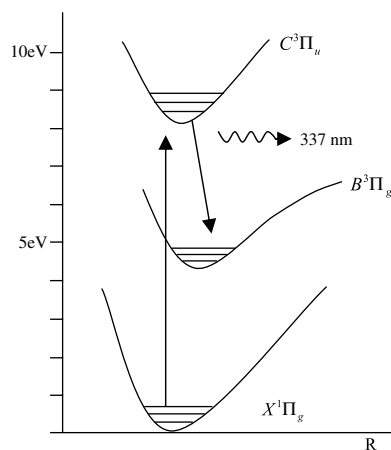
that carries the molecular fingerprint straight back toward the detector.

Molecular excitations produced by pairs of photons—one from the backward air-laser beam and the other from the forward interrogation laser—provide a sensitive molecular sensing mechanism. Examples include two-photon absorption (TPA) and stimulated Raman scattering (SRS). Ordinary TPA and SRS (Fig. 4 A and B) can be directly used for remote sensing utilizing the backward laser, because these mechanisms do not require phase matching (or, alternatively, one may say that they are always automatically phase-matched). Then the concentration of trace gasses can be quantified by measuring absorption of the backward-propagating beam as a function of wavelength of the interrogation laser. For gas molecules, both TPA and SRS will show species-specific (fingerprint-like) structure of vibrational and rotational energy levels. For gas-molecule fingerprinting, each (TPA and SRS) will have its pros and cons. SRS will benefit from smaller collisional dephasing of the ground-electronic-state molecular oscillations, whereas TPA will have the advantage of a nearly Doppler-free configuration (when nearly the same wavelengths are used for the two counterpropagating laser beams). For remote spectroscopy of small particles and aerosols, SRS will be preferred because TPA will typically be relatively featureless because of large electronic broadening in the condensed phase.

Ordinarily, TPA and SRS will suffer from shot-to-shot fluctuations of the air laser, which is because of the fact that measurements will have to be made by comparing signals obtained from different laser shots at different interrogation laser wavelengths. In principle, this difficulty can be circumvented if the air laser can be made to operate at multiple wavelengths. We also note that traditional coherent anti-Stokes Raman scattering (CARS) cannot be used in scenarios involving both forward and backward laser beams because of phase-matching constraints (Fig. 4C). Even if an additional Stokes laser beam is injected in the forward direction (and the air laser is used to provide a probe beam), k-vector mismatch for "backward" CARS will be reduced but will still remain substantial and will prevent backward anti-Stokes generation.

Finally, we note that higher-order nonlinear processes can be considered as alternatives to TPA and SRS. Those higher-order processes will require larger laser intensities and will generally produce weaker signals, while possibly allowing higher molecular selectivity by addressing several molecular levels at once.

Summary and Conclusion

The present paper focuses on generating laser action in O_2 and N_2 in air, as a backward-propagating spectral probe. The basic idea involves production of plasma filaments by chirped high-power laser pulses that are further heated by longer high-energy pulses in order to produce population inversion. By varying the chirp, individual gain regions can be sequentially prepared in a

Fig. 3. Sketch of the energies of N_2, as a function of the internuclear separation R, that are most relevant for the nitrogen laser.
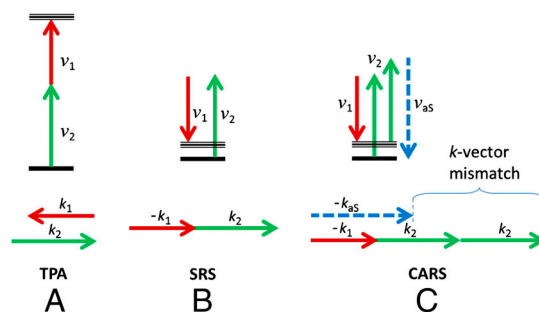
Fig. 4. Energy level and k-vector diagrams for TPA (A), SRS (B), and CARS (C) with counterpropagating beams at frequencies v_1 and v_2 as shown in Fig. 2. Whereas TPA and SRS are automatically phase-matched, CARS is not.

gain-swept superradiant mode. Such gain-swept superradiance is discussed at length in ref. 3.

Weak backward lasing from excited nitrogen molecules inside femtosecond laser filaments has been observed by Chin and coworkers at the University of Laval in Canada (14). Recent theoretical work also supports the concept of N_2 lasing through filamentation. Furthermore, recent experiments by Miles and coworkers at Princeton show that remotely initiated lasing in atmospheric O_2 is feasible (5).

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