Standoff detection of an electric field by bidirectional nitrogen lasing

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We report on standoff detection of dc electric field by bidirectional cavity-free lasing emission of neutral nitrogen molecules excited by intense circularly polarized femtosecond laser pulses. We observed that both the backward and forward 337.4 nm coherent lasing emission present a monotonous dependence on the strength of a remotely applied dc field up to $\sim$ 1 kV/cm field strength. Moreover, this method shows a dependence on the polarity of the external dc field, providing a sensitive method for remote characterization of the electric field amplitude and direction. We attribute the underlying mechanism of lasing signal modulation to the electric field induced electron acceleration and deceleration, which results in a variation of the kinetic energy of the free electrons and a modulation of the population inversion responsible for the nitrogen molecules' lasing. The polarity-sensitive detection anisotropy is interpreted by the symmetry breaking of the electron motion in the plane perpendicular to the laser propagation due to the injection of a weak second harmonic laser field produced in the quarter-wave plate for a circularly polarized pump laser. Numerical simulations based on the two-dimensional time-dependent Schrödinger equation for electron kinetic energy support our interpretation. This study provides a proof-of-principle method for standoff detection of electric fields based on nitrogen lasing, which can be potentially useful for atmospheric and metrological applications.

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I. INTRODUCTION

Air lasing refers to the cavity-free lasing action of the main components of air or its derivative under excitation by intense ultrafast laser pulses [1–7]. This lasing effect is unique in that the lasing medium can be generated in the atmosphere far away from the pumped laser thanks to the filamentation of high-power femtosecond laser pulses [8]. The plasma column produced during filamentation gives rise to both forward and backward coherent emission at a series of wavelengths determined by the species of the active elements. They include O and N atoms and neutral nitrogen molecules N_2, as well as ionic nitrogen molecules N_2^+. Due to its unique properties, the air lasing process holds great potential for optical remote sensing. For instance, it has been demonstrated recently that the forward lasing emission of N_2^+ can be used for the detection of trace elements in air [9], for the remote generation of optical radiation combs in the UV range [10], and for the construction of rotational wave packets of molecules [11]. On the other hand, the backward lasing signal can be used as an information carrier to convey atmospheric molecular information back to the ground observer, for which a single-end optical remote sensing can be conceived [12,13]. This could lead to revolutionary advances in optical remote sensing due to the intensity of the backward lasing signal being higher by orders of magnitude when compared to the fluorescence signal detected in conventional optical remote sensing.

Parallel to these efforts of spectroscopic studies with air lasing, it has been shown that the air lasing emission can be manipulated by an external electric field, which suggests that an air lasing signal can be used as a method for standoff detection of an electric field. In 2019, Clerici *et al.* reported the control of a nitrogen ion lasing signal using THz radiation [14]. This effect provides the basis for the application of nitrogen ion lasing for remote electromagnetic field detection. However, since the nitrogen ion emission signal is mainly emitted in the forward direction [15,16], backward detection is still impossible with this scheme.

In this work, we demonstrate standoff detection of a dc electric field based on bidirectional lasing of a neutral nitrogen molecule pumped by intense near-IR femtosecond laser pulses. Detection distance up to 12.5 and 3 m is achieved in the forward and backward direction, respectively. Significant modulation of both the forward and backward 337.4 nm signal is observed in the presence of an external electric field. More interesting, the 337.4 nm lasing signal presents a quasilinear dependence on the electric field for amplitudes $E < 1$ kV/cm. We attribute the lasing signal modulation by the external

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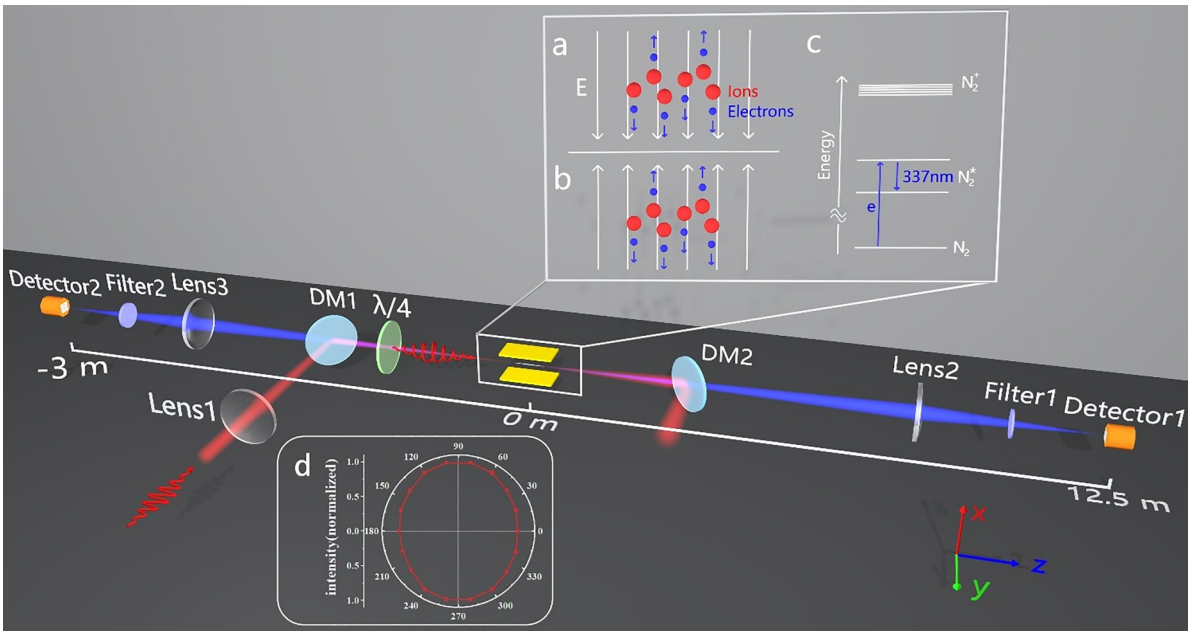

FIG. 1. Schematic diagram of bidirectional nitrogen lasing for detection of external electric field. We observed both the backward and forward stimulated 337.4 nm emission of neutral nitrogen molecules in the presence of an external electric field. In the inset, *a* and *b* present the electron motion influenced by the external field, leading to increase or decrease of the electron kinetic energy. In *c*, excitation of nitrogen molecules by electron collision and the subsequent lasing at 337.4 nm ar shown. In *d*, the polarization measurement of the pump laser is presented.

electric field to the fact that the kinetic energy of the free electrons is changed by the electric field. The dependence on the polarity of the electric field is explained by the symmetry breaking of the electron motion in the plane perpendicular to the laser propagation direction by a weak linearly polarized second harmonic pulse generated in a thin quarter-wave quartz plate. We believe that standoff polarity-sensitive detection of an electric field by backward nitrogen lasing can be applied to exploration of the atmospheric environment, weather forecasting, high-voltage transmission, and so on.

II. EXPERIMENT RESULTS

In the experiment, a Ti: sapphire laser system (Coherent Legend DUO) delivers 35 fs, 1 kHz pulses at a central wavelength of 796 nm with a maximum energy of 11 mJ. The laser pulses were focused by an $f = 750$ mm lens into a chamber filled with 1 bar pure nitrogen, as schematically shown in Fig. 1. We used a quarter-wave plate made of crystalline quartz to change the linearly polarized pulse to circular polarization. Around the filament plasma inside the gas chamber, a pair of parallel electrodes (10 cm $\times$ 10 cm) was installed to apply a dc electric field in the vertical direction, as shown in Fig. 1. The backward and forward lasing signals at 337.4 nm produced by the plasma were filtered out by a bandpass filter and detected by a spectrometer (Avantes, Avaspec-3648). By changing the voltage and direction of the electric field, we observed the variation of the forward and backward 337.4 nm lasing signal. We noticed that both the forward and backward emission signals are very sensitive for the polarization of the pump laser. With a slight azimuthal rotation of the quarter-wave plate by $\pm 5^\circ$, the signals decreased by orders of magnitude, which demonstrated that the detected signals are not fluorescence but lasing signals [17,18].

In Figs. 2(a) and 2(b), the power spectra of the 337.4 nm lasing signal detected 1 m away from the plasma column for both the forward and backward directions in the presence of a dc electric field are presented. The presence of the external electric field results in a substantial enhancement of the lasing signal. We then tried to detect nitrogen molecular lasing at increased distances in both forward and backward directions. We have managed to detect the forward 337.4 nm signal 12.5 m away from the plasma filament. By changing the strength of the electric field, a variation of the forward 337.4 nm signal at 12.5 m was observed. The experimental results are shown in Fig. 3(a). For positive electric field E pointing in the x-axis direction ($E > 0$), the intensity of 337.4 nm emission first increases and then tends to saturate with the increase of the electric field. Here we increased the external electric field

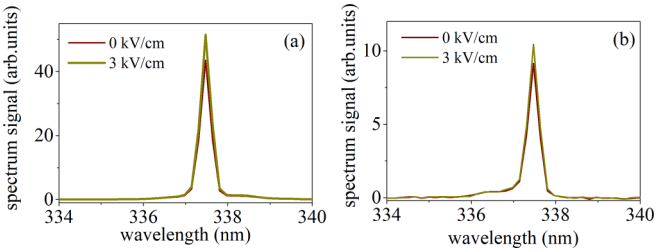

FIG. 2. The influence of the applied electric field on the 337.4 nm signal measured 1 m away from the plasma. (a,b) respectively show the forward and backward 337.4 nm spectral signals for different strengths of electric field. The spectra were averaged over about 3000 laser shots.

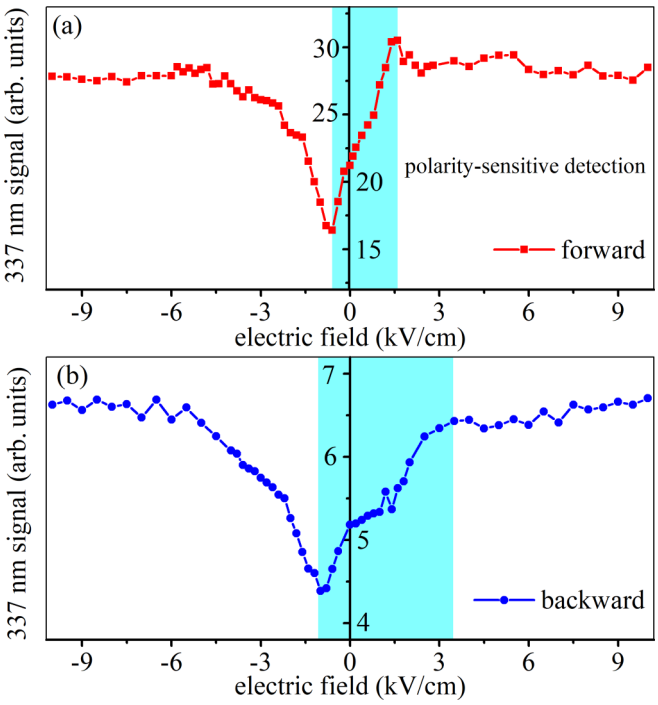

FIG. 3. Effect of electric field on 337 nm signal. (a) The forward 337.4 nm signal measured at a distance of 12.5 m from the plasma. (b) The backward 337.4 nm signal measured at 3 m from the plasma. The signals are results averaged over about 3000 laser shots.

up to 10 kV/cm. We have also tried to detect the backward nitrogen molecule emission far away from the filament. Figure 3(b) shows the dependence of the backward emission on the electric field measured 3 m away from the plasma. A significant saturation behavior was observed for both forward and backward signals for electric field strengths exceeding ~ 2 kV/cm, which can be due to the lasing signal being driven into the saturation amplification regime.

Interestingly, for the negative electric fields ($E < 0$), it was found that the 337.4 nm signal first decreases when the external electric field is varied from zero to -1 kV/cm. Therefore, this sensitive dependence of the bidirectional 337.4 nm lasing on the electric field polarity provides a method to determine the direction of the standoff electric field, not just its amplitude. The results of the backward 337.4 nm signal for negative electric field ($E < 0$) measured 3 m away are also shown in Fig. 3(b). A dependence similar to that in Fig. 3(a) was found, except that the electric field strength for lasing signal saturation was slightly higher, around ~ 6 kV/cm.

How should we understand the modulation effect of the lasing signal by the electric field and the polarity-sensitive detection property? It is well known that the 337.4 nm lasing signal of nitrogen molecules originates from the transition from the third excited state $C^3\Pi_u$ to the second excited state $B^3\Pi_g$ of the triplet state of the nitrogen molecule. Circular polarization of the pump laser is necessary so that the free electrons obtain enough kinetic energy, up to $2U_p$, in the circularly polarized laser field, where $U_p = e^2/c\varepsilon_0 m_e \times I/2\omega_0^2$ is the free electron ponderomotive energy [17,19,20]. For a pump pulse intensity of $I \sim 10^{14}$ W/cm^2, the peak of the electron energy distribution is about ~ 16 eV, which is above the threshold (~ 11 eV) for impact excitation of ground state nitrogen molecules to the $C^3\Pi_u$ level [17,19,20]. This leads to a rapid population inversion between the $C^3\Pi_u$ and $B^3\Pi_g$ states and optical amplification occurs. The population inversion is established by inelastic collisions between nitrogen molecules in the ground state and high-energy electrons, along the following scheme: $N_2X^2\Sigma_g^+ + e = N_2(C^3\Pi_u) + e$. We assign the modulation of the lasing signal to the electric field induced electron acceleration and deceleration inside the plasma. In fact, it has been reported previously that the plasma fluorescence can be enhanced by a terahertz electric field and the effect has been exploited as a method for characterization of THz field [21,22].

The important advance of our current work is that we employ the directional stimulated lasing beam as the information carrier, instead of the fluorescence signal emitted in the 4π solid angle, which facilitates remote detection. Also, the lasing signal modulation method is more sensitive than the technique based on the modulation of the fluorescence signal of nitrogen molecules [21,22], due to the fact that the lasing signal depends exponentially on the population in the $C^3\Pi_u$ state, while the fluorescence depends linearly on the $C^3\Pi_u$ state population.

Now we turn our attention to the fact that different directions of the electric field lead to an increase or decrease of the lasing signal. When the circularly polarized pump laser ionizes nitrogen molecules, the free electrons are distributed isotropically in the x-y plane. Therefore, one expects no difference for the lasing signal whatever the direction of the external electric field. However, we noticed that the quarter-wave plate always gives rise to a parasitic second harmonic radiation at 400 nm. We have measured the spectrum and polarization of the parasitic second harmonic and the results are presented in Fig. 4. In our current experiments, the second harmonic is almost linearly polarized in the horizontal direction. We measured the energy of this second harmonic and found it to be ~ 0.1 μJ for the 800 nm pump pulse with 10 mJ pulse energy.

The presence of a second harmonic may explain an enhancement and anisotropy of the emission at 337 nm as follows. The linearly polarized 400 nm second harmonic pulse superimposes with the circularly polarized fundamental 800 nm pulse in the plasma. It is well known that the two-color field, consisting of the fundamental and second harmonics field, breaks the symmetry of electron motion in the x-y plane and results in a net transverse residual photocurrent in the wake of the femtosecond laser pulse. The direction of the net electron current is determined by the polarization and relative phase $\Delta\varphi$ of the two optical fields [23,24] [see Eq. (4) below]. A brisk macroscopic electron motion in a privileged direction in the x-y plane excites plasma oscillations with a period on the order of 0.1 ps defined by the density of free electrons. These oscillations decay on a few picoseconds timescale due to electron collisions with the nitrogen molecules and ions. The characteristic electron collision time is 1.4 ps in atmospheric air [25]. During this time, the electron motion is anisotropic, and it results in a change of electron energy in the dc electric field, which can be estimated as follows. Assuming the mean energy of a free electron to be ~ 10 eV, its mean displacement during the plasma oscillations

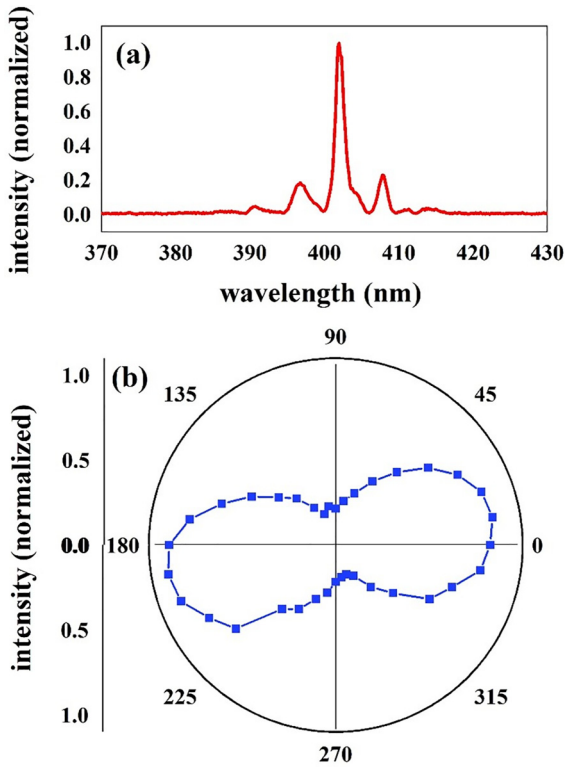

FIG. 4. Measured spectrum (a) and polarization (b) of the parasitic second harmonic generated on the quarter-wave plate.

would be about 1 μm, which corresponds to the energy gain or loss of 0.3 eV in the dc field of 3 kV/cm, depending on the orientation of the dc field with respect to the direction of the electron current. This electron energy variation by $\sim 3\%$ can lead to a change of population inversion between the $C^3\Pi_u$ and $B^3\Pi_g$ states resulting in a change of emitted signal by 30%–50%, assuming an exponential signal amplification with gain 10–12.

III. SIMULATION AND DISCUSSION

To verify the above interpretation, we perform a numerical solution of the two-dimensional time-dependent Schrödinger equation (TDSE). Under the single-electron approximation for the N_2 molecule, the TDSE is written as (Hartree atomic units are used unless stated otherwise)

$$i\frac{\partial}{\partial t}\psi(x,y,t) = \left[\frac{p_x^2}{2} + \frac{p_y^2}{2} + V(x,y) + V_L(x,y,t)\right] \times \psi(x,y,t), \quad (1)$$

where $p_{x/y}$ is the electronic kinetic momentum operator, and the Coulomb interaction between electron and atomic core is assumed as

$$V(x,y) = -\frac{1}{\sqrt{(x-x_l)^2 + (y-y_l)^2 + sf}} - \frac{1}{\sqrt{(x-x_r)^2 + (y-y_r)^2 + sf}}. \quad (2)$$

Here $x_{l/r} = \mp R/2$ and $y_{l/r} = 0$ denote the two nucleus displacements. R is the internuclear distance, which is fixed at $R = 2.07$ a.u. in all simulations [26]. The softcore factor $sf = 4.58$ a.u. guarantees that the first ionization potential I_p of N_2 is equal to -0.573 a.u. (15.58 eV) in such a two-dimensional model [27–29]. The interaction between electric field and electron is expressed in the dipole approximation

$$V_L(x,y,t) = xE_x + yE_y, \quad (3)$$

where $E_{x/y}$ includes 800 and 400 nm laser electric fields together, which are expressed as

$$\boldsymbol{E}(t) = E_{800}F(t)[\cos(\omega_{800}t)\hat{\boldsymbol{e}}_x + \sin(\omega_{800}t)\hat{\boldsymbol{e}}_y] + E_{400}F(t)\cos(\omega_{400}t + \Delta\varphi)\hat{\boldsymbol{e}}_x, \quad (4)$$

where $E_{800/400}$ and $\omega_{800/400}$ are their amplitudes and angular frequencies and they have the same laser envelope $F(t) = \sin^2(\pi t/\tau)$; τ is the duration of the whole laser pulse. The field parameters are fixed as $E_{800} = 0.0844$ a.u. (2.5×10^{14} W/cm^2), $E_{400} = 0.0189$ a.u. (5×10^{12} W/cm^2), and $\tau = 40$ fs, which correspond to our experimental parameters. The photoelectron momentum wave functions $\psi(p_x, p_y)$ are calculated by the Fourier transform of the final wave functions $\psi(x, y, t_{\text{end}})$ excluding the bound states. Then the photoelectron momentum distribution is propagated based on Newtonian classical mechanics in the presence of a dc field. Note that here the Coulomb potential is neglected since the ionized electron is far away from the nuclei after the femtosecond pulse finishes. Finally, the photoelectron kinetic energy spectrum is obtained by

$$P(E_k) = |\psi(p_x, p_y)|^2/\sqrt{p_x^2 + p_y^2},$$

where the kinetic energy $E_k = p_x^2 + p_y^2$.

In the simulation, we calculated the electron momentum distribution in the presence of an external electric field with a combined 800 and 400 nm optical field as the excitation pulse. Figure 5(a) shows the calculated photoelectron momentum spectrum without the dc field. For the combined optical fields of the above-mentioned 800 and 400 nm, the electron momentum distribution is asymmetric in the x-y plane and the electrons acquire a macroscopic drift motion in the direction of $\phi \approx 225°$, where ϕ is the azimuthal angle with respect to the vertical x axis. This distribution is determined by the circularly polarized 800 nm and linearly polarized 400 nm pulses in the horizontal direction. When the external dc field is applied in the direction $\phi = 45°$ the electrons will obtain additional kinetic energy. In contrast, if the dc field is applied at $\phi = 225°$, the external electric field decreases the electron kinetic energy. Therefore, one can expect an enhancement or suppression of the lasing signal depending on the direction of the electric field. With the electric field in these directions we indeed observe a significant increase or decrease of the electron energy for $E_e > 11$ eV, as presented in Fig. 5(b). In contrast, for an electric field applied in the direction of 135°/315°, the energy distribution of the electrons is similar due to the reflection symmetry with respect to the plane $x = y$. This explains the polarity-sensitive detection feature observed in Fig. 3. The anisotropic dependence of the electron energy on the external field provides a method to measure the

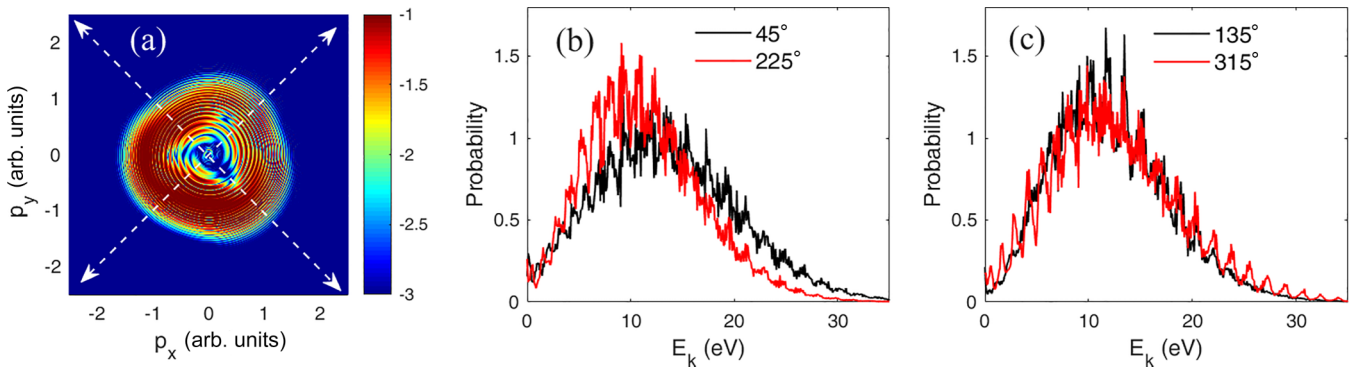

FIG. 5. (a) The photoelectron momentum spectrum without the dc electric field. In (b,c), the photoelectron kinetic energy distributions are presented. In (b), the black line denotes that the dc electric field points to 45° and the red line refers to an opposite direction (225°) to the black one. In (c), the black (red) line corresponds to the electric field at 135° and 315°. The calculating parameters: $E_{800} = 2.5 \times 10^{14}\,\mathrm{W/cm^2}$, $E_{400} = 5 \times 10^{12}\,\mathrm{W/cm^2}$, $\tau = 40$ fs, $E_{\mathrm{dc}} = 3$ kV/cm, and effective dc electric field duration of 4.8 ps before plasma screening.

direction of an external unknown dc field. With a separated linearly polarized 400 nm beam whose polarization direction can be rotated, the direction of the electron current in the transverse plane can be manipulated, which may serve as a "compass" for the dc field under study. By rotation of the compass with respect to the external field, one can record the different behavior of the field-dependent lasing signal, which tells the direction of the dc field.

IV. CONCLUSION

In conclusion, we demonstrated a proof-of-principle method for standoff detection of a dc electric field based on the bidirectional cavity-free lasing emission of neutral nitrogen molecules. Polarity-sensitive detection of the electric field has been achieved thanks to the presence of a linearly polarized weak residual second harmonic laser field superimposed on the circularly polarized fundamental 800 nm laser field. We have detected the forward 337.4 nm lasing signal at 12.5 m and the backward 337.4 nm lasing signal at 3 m. We attribute the enhancement or suppression of the 337.4 nm signal to the fact that the electric field accelerates or deaccelerates the movement of free electrons in the $x - y$ plane. As a result, the kinetic energy of the electrons is changed due, leading to increased or decreased electron-molecule collision excitation and population inversion between the $C^3\Pi_u$ state and the lower $B^3\Pi_g$ state. These results lay the basis for application of nitrogen molecule lasing for standoff detection of an electric field. A self-consistent model will be developed in the future to establish the quantitative relationship between the modulation of the lasing signal and the strength of the electric field. Also, we would like to point out this method can be applicable for standoff detection of ac electric fields, such as THz electric transients.

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