

Laser-assisted vibrational control of precursor molecules in diamond synthesis



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ABSTRACT

Control of chemical reactions is the essence of chemistry, producing designed outcomes while suppressing unwanted side products. Laser-assisted molecular vibrational control has been demonstrated to be a potential approach to influencing the outcome of a chemical reaction. In this article, we reviewed recent progress in the laser control of diamond synthesis through vibrational excitation of precursor molecules in a laser-assisted combustion chemical vapor deposition process. Significantly promoted diamond deposition rate (139 $\mu\text{m}/\text{h}$) and crystalline quality were achieved by resonantly exciting the Q-branch ($\Delta J = 0$) of the CH_2 -wagging mode (ν_7 mode 949.3 cm^{-1}) of C_2H_4 molecules. Resonant excitation of the fundamental vibrational modes is more effective in promoting diamond growth than random vibrational excitation. Control of diamond crystallographic orientation was also realized by resonantly exciting the R branch ($\Delta J = 1$) of the CH_2 -wagging mode of C_2H_4 molecules and resulted in the preferential growth of $\{100\}$ -oriented diamond crystals. Nitrogen-doped diamond films with a nitrogen concentration of $1.5 \times 10^{20}\text{ atoms}/\text{cm}^3$ were synthesized by resonantly exciting the rotational–vibrational transition ($J = 5 \rightarrow J' = 6, K = 0$) of the N–H wagging mode (ν_2 mode) in ammonia molecules. The findings demonstrate the feasibility of laser-assisted vibrational control in steering chemical reactions and controlling reaction outcomes.

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Introduction

The essence of chemistry is achieving precise control of chemical reactions towards anticipated outcomes while suppressing unwanted side products [1–6]. Conventionally, chemical reactions can be influenced either dynamically, varying external parameters such as temperature, pressure, and concentration, or kinetically, such as utilizing catalysts [1–6]. Since the invention of lasers, laser intervention has been a conceptually appealing tactic to steering chemical reactions and influencing final outcomes. Laser-matter interactions can be either a thermal process or a non-thermal process [1–6]. For a thermal process, incident photons are used primarily as a heating source, in which laser energy is generally deposited into specific reactants and dispersed rapidly to other molecules achieving an equilibrium thermal heating. Similar to conventional thermal driven chemical reactions, the laser thermal process is short of reaction selectivity [1–6]. For a non-thermal process, laser

control of chemical reactions can be realized via a number of mechanisms, including (1) controlling molecular vibrational states; (2) photo-isomerization of reactant molecules; (3) exciting electrons to states of different bonding properties; and (4) regulating electron populations at different states [1–6].

Laser-assisted vibrational excitation of molecules is an intuitively attractive approach realizing laser control of chemical reactions [1–6]. In 1972, Polanyi introduced the concept of mode-selective chemistry [7–10], in which vibrational excitation along the reaction coordinate would be more effective than translational motion in promoting endoergic reactions, e.g. the mode of internal excitation could control the reaction outcome. It is expected that the excitation of fundamental vibration or overtone vibration of higher levels would selectively localize energy in a specific bond or a motion along the reaction coordinate, lead to the preferential cleavage of the corresponding bond, and achieve reaction pathway control [7–10]. Extensive investigation on simple molecules, such as HOD [11] (an isotopic variant of water), $\text{HCN} + \text{Cl}$ [12], and $\text{ND}_3 + \text{NH}_3$ [13], has confirmed the validity of the conjecture. Crim reported selective cleavage of chemical bonds and achieving chemical reaction control via vibrational excitation [2,11,12,14]. In the

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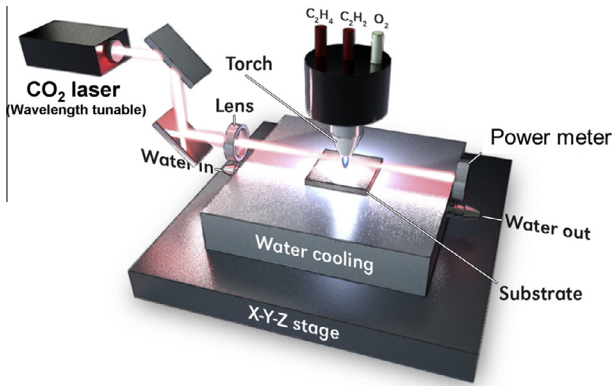


Fig. 1. A schematic experimental setup of the LA-CCVD system.

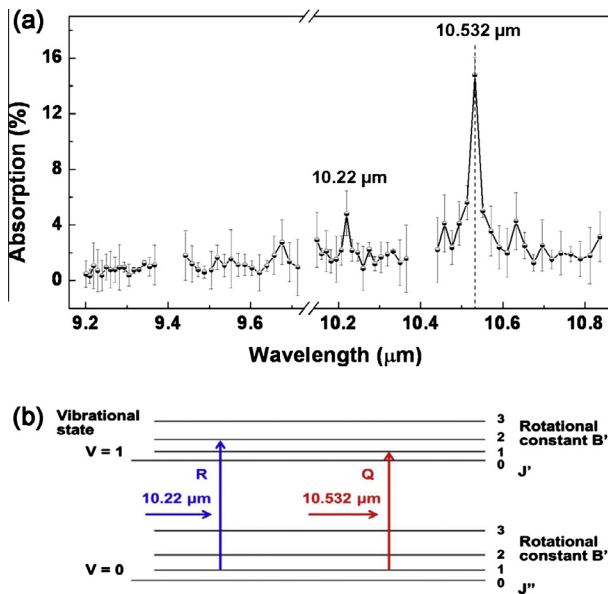


Fig. 2. (a) A typical absorption spectrum of the $C_2H_2/C_2H_4/O_2$ flames as a function of laser wavelength from 9.2 to 10.9 μm . (b) Vibration–rotation transitions of an ethylene molecule at 10.22 and 10.532 μm , respectively.

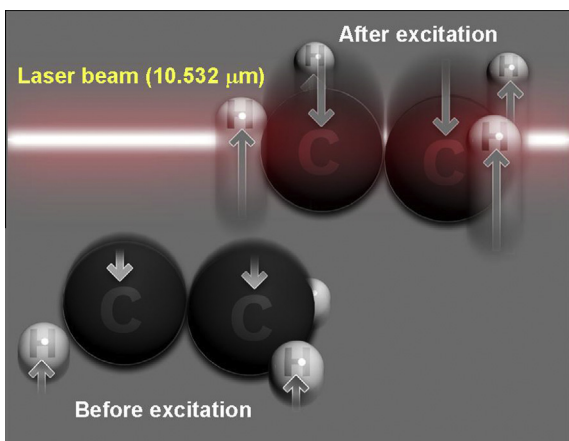


Fig. 3. A schematic of the resonantly excited CH_2 -wagging mode (a type c fundamental mode, v_7) of an ethylene molecule.

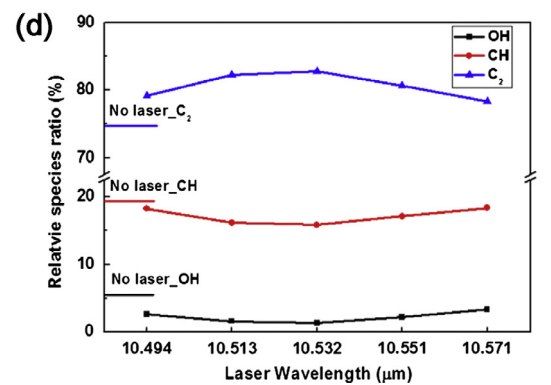
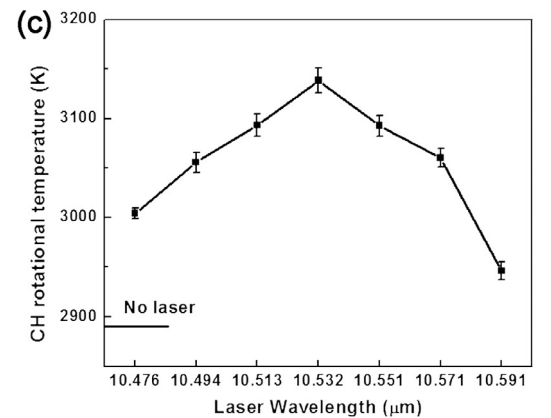
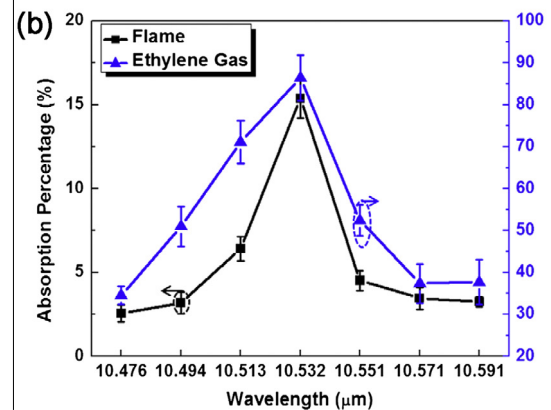
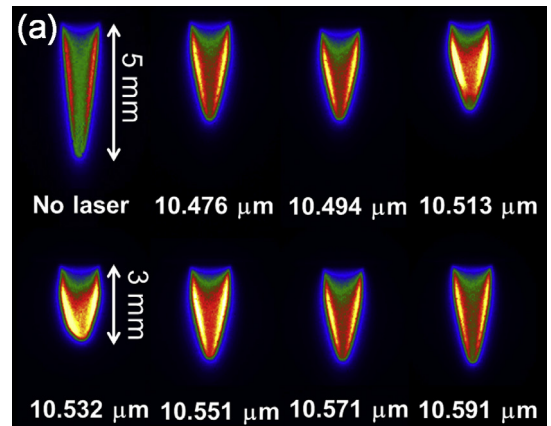


Fig. 4. Influence of the laser wavelength: (a) optical images of the acetylene–ethylene–oxygen flames; (b) laser energy absorption rates of the flame (left) and ethylene gas (right); (c) flame temperature; and (d) relative species ratios as the functions of the laser wavelength.

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