文章编号:1001-9014(2013)02-0108-05 DOI:10.3724/SP.J.1010.2013.00108

Efficient THz generation via stimulated Raman adiabatic passage

SHI Hua-Feng[1,2], YU Wen-Long[1,2], YU Fei[1,2], JIN Lei[1,2]
(1. Shenzhen Institutes of Advanced Technology, Chinese Academy of Sciences, Shenzhen 518055, China;
2. Key Lab for Biomedical Informatics And Health Engineering, Chinese Academy of Sciences, Shenzhen 518055, China)

Abstract: A THz generation via stimulated Raman adiabatic passage in three-level Λ system was demonstrated. The basic model and theory had been analyzed in three levels coherent with pump laser and THz generation. The application also had been mentioned in D_2O molecules optically pumped by a TEA CO_2 laser. It produced THz radiation at the wavelength of 385 μm. The power of the THz pulse as a function of the vapor pressure, and a maximum THz power had been obtained. It was displayed that this technology is a robustness method of THz source.
Key words: THz generation; pump laser; three level system; stimulated Raman adiabatic passage
PACS: 42.72. Ai, 41.20. -q

经由受激拉曼绝热过程高效产生太赫兹

时华峰[1,2], 于文龙[1,2], 余 菲[1,2], 金 雷[1,2]
(1. 中国科学院深圳先进技术研究院 生物医学与健康工程研究所 生物光子学研究中心, 广东 深圳 518055;
2. 中国科学院生物医学信息与健康工程学重点实验室, 广东 深圳 518055)

摘要:论证了在三能级系统中经由受激拉曼绝热过程产生太赫兹,分析了基本模型和泵浦激光与太赫兹产生相干的三能级的理论,并应用在横向激励二氧化碳激光光学泵浦重水分子中,可以产生波长为385 μm的太赫兹辐射.产生的太赫兹脉冲功率表现为蒸汽压的函数,并得到了最高太赫兹功率.该技术提供了一种鲁棒性非常好的产生太赫兹的方法.
关 键 词:太赫兹生成;泵浦激光;三能级系统;受激拉曼绝热过程
中图分类号:O452;TN99 **文献标识码**:A

Introduction

Terahertz (THz) is preliminary defined as the wavelength region of 1 mm ~ 30 μm (frequency of 0.3 ~ 10 THz) in the electromagnetic spectrum. In recent works, several new techniques in THz radiation sources have been reported, and various applications have been shown[1-3]. The stimulated Raman adiabatic passage (SRAP) technology is an established technique [4], especially in some special applications. An efficient but inexpensive practical THz sources are needed, which can work at room temperature. The SRAP technology offers many advantages. For example, the excitation efficiency can be made relatively insensitive to the pump lasers, and the THz pulse duration can be as fast as pump pulse, not restricted by relaxation rates [5].

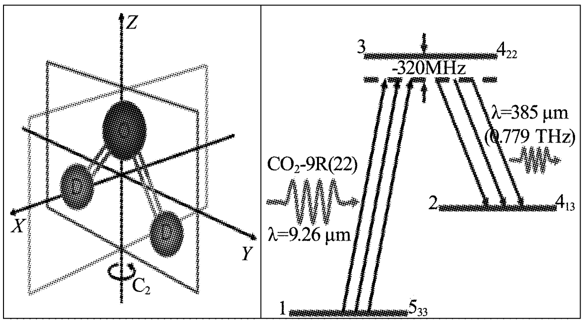
Adiabatic passage is a way of affecting complete population transfer, which happened between selected states and their coherent superposition. The stimulated

Received date: 2011-11-28, **revised date**: 2012-07-28 **收稿日期**:2011-11-28, **修回日期**:2012-07-28
Foundation items: Supported by the Comprehensive Strategic Project Cooperation with the People's Government of Guangdong Province and Chinese Academy of Sciences (2009B091300011).
Biography: SHI Hua-Feng(1981-), male, master. Research area is the practical terahertz source and the application of THz. E-mail: hf. shi@ siat. ac. cn

emission pumping is a popular successful method for the transfer of population in a three-level system [6]. It is independently implemented in the experiment by the SRAP technology[7] and can be used for the THz generation. The theory model based on a three-level Λ system of D_2O molecule has been shown. It is described by a semi-classical method, assuming classical pump fields and quantized molecule states. The experiment and results prove that it is practicable for THz generation.

1 Basic theory model

D_2O molecule is a light asymmetric-top molecule as shown in Fig. 1(a). The structure consists of a tiny V-shape. It is symmetric with two mirror planes (the blue plane and red one) and a 2-fold axis of rotation (axis z), which is called point group C_{2v}. The molecular dipole moment is from the center of negative charge (Deuterium atoms, green) to the center of positive charge (Oxygen atom, red). This is equivalent to a unit negative charge separated from positive charge.



(a) (b)

Fig. 1 The basic theory model in a three-level system of D_2O molecule (a) The heavy water molecule model, the deuterium atoms partially positively charged (green), while the oxygen atom partially negatively charged (red), (b) A simple three-level system adopted for the D_2O molecule to generate THz pulse

图 1 D_2O 分子三能级系统的基本理论模型 (a) 重水分子模型,氘原子部分带正电(绿色),氧原子部分带负电(红色),(b) D_2O 分子产生太赫兹脉冲采用的简单三能级系统

In D_2O molecule, the states $|1>$, $|2>$, and $|3>$ are the molecular energy levels of $(0, 5_{33})$, $(\nu_2, 4_{13})$, and $(\nu_2, 4_{22})$, respectively. Their eigenvalues are E_1, E_2, E_3, which have been measured by Mellau and co-workers [8-9]. The pump pulse is the CO_2-9$R_{(22)}$ line with a wavelength of 9.26 μm ($\lambda_p = 1/\omega_p$), and the emission THz wave is the λ_s ($1/\lambda_s = \omega_s = E_3 - E_2$). The frequency of the pump pulse (ω_p) is lower by about 320 MHz than the resonance frequency of the absorption ($\omega_{13} = E_3 - E_1$). It implies that the existence of the off-resonant pumped and stimulated Raman adiabatic passage. At t = 0, the molecules are in the thermal equilibrium states, and obey the Boltzmann distribution:

$$\frac{\rho_{ii}^0}{\rho_{jj}^0} = e^{-(E_i - E_j)/kt} (i, j = 1, 2, 3). \quad (1)$$

And the odds of the molecules at three levels equal to 1. Then it can be obtained that the probability measured for the distribution of the D_2O molecules occupied each of the states are $\rho_{11}{}^0 = 98.85\%$, $\rho_{22}{}^0 = 0.61\%$, and $\rho_{33}{}^0 = 0.54\%$. It means that most of the molecules are at the ground state and only few (less than one percent) at the excited states.

Figure 2 shows how the THz pulse is generated in D_2O molecules. The process can be identified distinctly three different intervals I, II, and III. At first, the molecules is interacted with the THz pulse, and then with the pump laser. It seemed counterintuitive at first glance, but is the natural choice when the consequences of coherence are considered. Initially, the THz pulse was coupled with the two empty states, which created a coherent superposition in the two unpopulated states. When the pump laser is coupled with the coherent superposition and the ground state, there is a trapped state [10]. In region I, the Rabi frequency of the pump pulse was zero, all three eigenvalues are degenerate. Most of the molecules are in the ground state (level $|1>$). In region II, the Rabi frequency of the THz pulse is reduced, while the Rabi frequency of the pump laser increases to its maximum value. The coupling of the Rabi frequencies of the pump laser and THz pulse is strongest, and both radiation fields contribute to it. The molecules transfers from the ground state, level $|1>$, into the excited state, level $|3>$, forming inverse population. And then the molecules dump into the lower state, level $|2>$, generating THz pulse at the wavelength of 385 μm (0.779 THz). This motion results in complete population transfer if the state vector evolves adiabatically. If the Rabi frequency of pump laser is too small, i. e. the pump laser density is not high enough, the coupling is insufficient, and the transfer process from the ground state into the excited states would be incomplete. Less molecules would take part in the inverse population, and so the generating THz pulse between the levels |3 > and |2 > would be less. In region III, the Rabi frequency of the THz laser is zero, the number of molecules in the ground state |1 > is the smallest, most of the molecules transfer interim of excited state, level |3 >, and then dump into lower state, level |2 >. This also can be seen clearly in Kalugin's work [11].

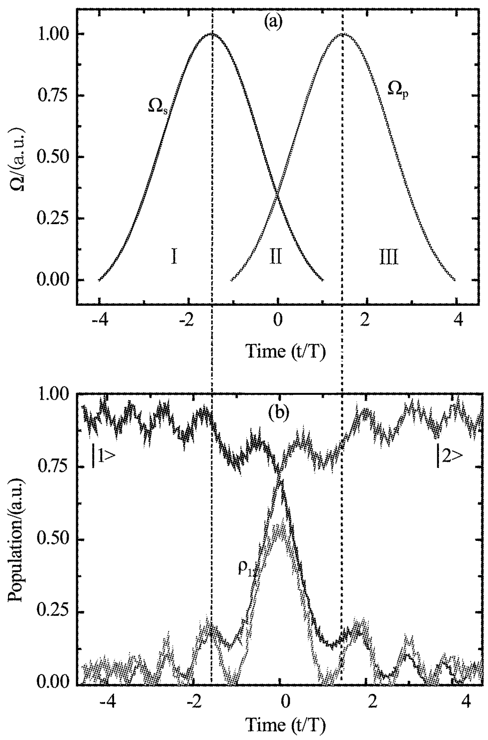


Fig. 2 T is the pulse duration, the coherence ρ_{12} between these two states (|1 > and |2 >) is maximal while the transformation is taking place (a) Time evolution of the Rabi frequencies of the pump laser and THz pulse in SRAP configuration, (b) The population of the initial level (|1 >) and the final level (|2 >)
图2 T是脉冲周期（a）在受激绝热通道中泵浦激光和太赫兹脉冲的拉比频率的时间演化,(b) 最初能级1和最终能级2的布居

2 Experimental setup

Figure 3 shows a sketch of our experimental set-

Table 1 Parameters of D_2O molecule and pump laser
表1 D_2O 分子和泵浦激光的参数

Parameter	Symbol	value	unit
Asymmetry[a]	A	462278.8077	–
Asymmetry[a]	B	218038.2178	–
Asymmetry[a]	C	145258.0022	–
Molecular level[a]	E_1	267.53083	cm^{-1}
Molecular level[a]	E_2	1321.41375	cm^{-1}
Molecular level[a]	E_3	1347.39375	cm^{-1}
Dipole moment[b]	μ_{13}	0.0323	Debye
Dipole moment[b]	μ_{32}	0.68	Debye
Relaxation time[b]	τ	2.5e–8	$s*Torr^{-1}$
Wavelength of pump laser[c]	λ_p	9.2605258	μm
Length of cell	L	0.97	m

[a] From J. Mol. Spectrosc., 224, 32 (2004)
[b] From J. Mol. Spectrosc., 206, 41, (2001)
[c] From Opt. Quant. Electron., 4, 215, (1972)

up. A grating-tunable transverse excited atmospheric (TEA) CO_2 laser was used as the pump laser. The dashed square shows the metal shielded room, which was used to avoid the THz generation from EMI (Electromagnetic Interference). The maximum output power of the CO_2 laser was about 0.3 J/pulse for TEM_{00} mode with a pulse width of about 100 ns measured with a photonic drag detector. Thereby the maximum output peak power density could be up to 3.0 MW/cm^2. Tuning grating (150 grooves/mm), more than 60 laser lines could be gotten covering 9 ~11 μm, and a desired line could be selected. Combined with the energy levels of heavy water molecule, the 9R(22) line was picked out as the pump laser.

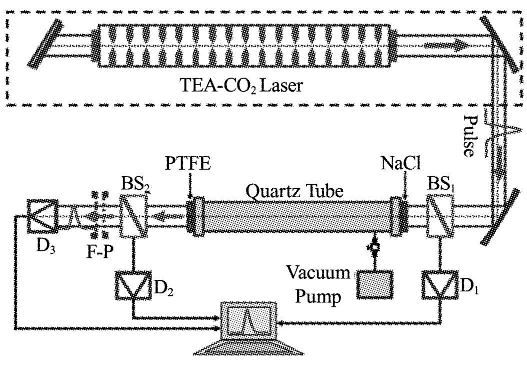


Fig. 3 Schematic experimental setup of the THz pulse generation optically pumped by TEA CO_2 laser. F-P is a Fabry-Perot interferometer and D3 is a pyroelectric detector
图3 横向激励二氧化碳激光器光学泵浦产生太赫兹脉冲的实验装置示意图. F-P 是法布里·泊罗反射仪;D3 是热释电探测器

The main component of the experimental setup is a superradiation structure. It was consisted of a sample tube, input window, and output window. The sample tube was a quartz tube about 97 cm in length and 5 cm in diameter. The input window was a piece of NaCl crystal (6 mm thickness), which was good for 9 ~ 10 μm signal transmission. The output window was a polytetrafluoroethylene (PTFE) plate (4 mm thickness), which could be effectively opaque to 9 ~ 10 μm signal and allow the THz pulse signals (including 385 μm) to pass efficiently. The D_2O vapor (specified isotopic purity: 99.8% D) filled the sample tube at a controllable pressure monitored by a pressure meter. The THz pulse was measured by a pyroelectric detector and recorded by a digital storage oscilloscope.

3 Results and discussion

The THz pulse generation dependence on the vapor pressure was investigated. It should be noted that the vapor pressure is an important but not the only parameter of the system. There are other decisive parameters such as relaxation rates, power of the pumped laser, and so on.

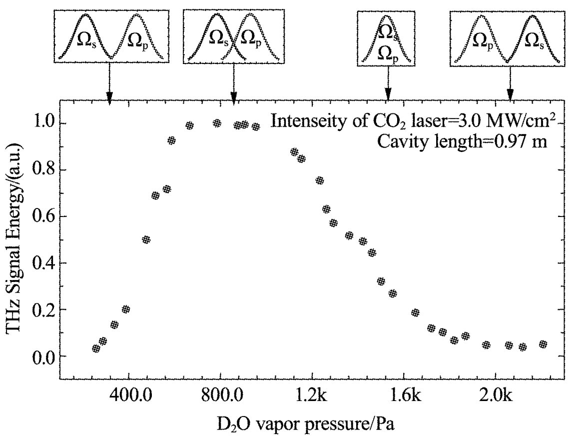


Fig. 4 Experiments on SRAP in D_2O molecules. The output power of THz pulse profiled as a function of the vapor pressure at room temperature. The red dots are the sampling datum and the overlap between pump laser and THz pulse are shown on the top

图4 D_2O 分子受激绝热通道的实验. 在室温下太赫兹脉冲的输出功率曲线为蒸汽压的函数。红色点是样品数据，泵浦激光和太赫兹脉冲的重叠显示在顶部

As shown in Figure 4, the pressure was varied over a wide range from 0.2 up to 2.3 kPa, while other operating parameters were kept constant. The output of the THz pulse signal power reached a maximum (about 1 mW, output power measured by bolometer,) by properly tuning the vapor pressure. The corresponding vapor pressure is called the optimum vapor pressure. The dynamics of this phenomenon is very complicated. When the vapor pressure is relatively low, it is insufficient to exchange the energy between the pump laser and medium molecules. The axis of the THz pulse is shifted far upstream compared with the pump laser, so that there is no overlap between them, (see in Fig. 4 top). The THz pulse does not participate in the process of the transfer, because there is not enough energy from THz pulse and pump laser simultaneously interacting with the molecules. Increasing the vapor pressure by filling more vapor, there are more D_2O molecules in the ground state. Thus there are more molecules transiting from the ground state to the higher excited state via absorbing energy from the pump laser, dumping into lower state, and generating THz radiation. This is accompanied by the movement toward each other between the axes of the pump laser and THz pulse. The Rabi frequencies overlap gradually, resulting in dramatic increase of the THz pulse power to the maximum value. At the point of top value, the vibration relaxation of the D_2O molecule is much faster compared with the duration of the pump laser. A sufficiently large vibration population can be excited and the overall ground state population is depleted or simply bleached, i. e. an equivalent vibration saturation or "bottleneck" occurs. Rising of the vapor pressure continually, although the axes of the pump laser and THz pulse are coincided, essentially there are less molecules stayed on the excited level. Self-absorption of medium increases which can not be ignored [12-13]. As a result, the output of the THz pulse power comes down instead of rising, as shown clearly in Fig. 4. This can be seen in the works of M. Fleischhauer and P. Král [14-15].

4 Conclusions

An efficient THz generator has been developed, which works at room temperature with less cost and simple operation. The CO_2 laser is a better choice as the pump laser with high power and suitable wavelength. The stimulated Raman adiabatic passage process happened with THz generating. Heavy water worked as the gain medium in the experiment, but not the only one, such as ammonia, formaldehyde, and so on. The THz pulse and continuous THz wave can be generated depending on the operation of CO_2 laser. It can work at many different frequencies, ranging from less than 0.3 THz to more than 10 THz. This kind of THz generation can be applied as a practical THz source in many fields, such as THz imaging, biomedical effect, different material detection, and spectroscopy.

Acknowledgment

We acknowledge informative discussions with Baojun Li, Zhihong He, and Xizhang Luo. This work was supported by the comprehensive strategic project cooperation with the People's Government of Guangdong Province and Chinese Academy of Sciences (2009B091300011).

REFERENCES

[1] Carr G L, Martin M C, McKinney W R, *et al.* High-power terahertz radiation from relativistic electrons [J], *Nature*, 2002, **420**: 153 - 156.

[2] Borak A. Toward bridging the terahertz gap with silicon-based lasers [J], *Science*, 2005, **308**(5722): 638 - 639.

[3] Imeshev G, Fermann M E, Vodopyanov K L, *et al.* High-power source of THz radiation based on orientation-patterned GaAs pumped by a fiber laser [J], *Opt. Express*, 2006, **14**(10): 4439 - 4444.

[4] Bergmann K, Theuer H, Shore B W. Coherent population transfer among quantum states of atoms and molecules [J], *Rev. Mod. Phys.*, 2003, **70**(3): 1003 - 1025.

[5] Ye C Y, Sautenkow V, Rostovtsev Y, *et al.* Fast optical switching via stimulated Raman adiabatic passage [J], *Opt. Lett.*, 2003, **28**(22): 2213 - 2215.

[6] Hamilton C E, Kinsey J L, Field R W. Stimulated emission pumping: New methods in spectroscopy and molecular dynamics [J], *Annu. Rev. Phys. Chem.*, 1986, **37**: 493 - 524.

[7] Gaubatz U, Rudecki P, Schiemann S, *et al.* Population transfer between molecular vibrational levels by stimulated Raman scattering with partially overlapping laser fields. A new concept and experimental results [J], *J. Chem. Phys.*, 1990, **92**: 5363 - 5376.

[8] Scully M O, Zubairy M S. *Quantum optics* [M], Cambridge University Press, Cambridge, UK, 1997, Chap. 5.

[9] Mellau G, Mikhailenko S N, Starikova E N, *et al.* Rotational levels of the (000) and (010) states of $D_2{}^{16}O$ from hot emission spectra in the 320-860 cm-1 region [J], *J. Mol. Spectrosc.*, 2004, **224**(1): 32 - 60.

[10] Arimondo E. V coherent population trapping in laser spectroscopy [J], *Prog. Opt.*, 1996, **35**: 257 - 354.

[11] Kalugin N G, Rostovtsev U V. Efficient generation of short terahertz pulses via stimulated Raman adiabatic passage [J], *Opt. Lett.*, 2006, **31**(7): 969 - 971.

[12] Yu B L, Yang Y, Zeng F, *et al.* Terahertz absorption spectrum of D2O vapor [J], *Opt. Commun.* 2006, **258**(12): 256 - 263.

[13] Rønne C, Åstrand P, Kerding S R. THz spectroscopy of liquid H_2O and D_2O [J], *Phys. Rev. Lett.* 1999, **82**(14): 2888 - 2891.

[14] Fleischhauer M, Imamoglu A, Marangos J P. Electromagnetically induced transparency: Optics in coherent media [J], *Rev. Mod. Phys.*, 2005, **77**(2): 633 - 673.

[15] Král P, Thanopulos I, Shapiro M. Colloquium: Coherently controlled adiabatic passage [J], *Rev. Mod. Phys.*, 2007, **79**(1): 53 - 77.