Generation of strong short coherent terahertz pulses in 
ladder-Lambda and double-Lambda systems

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(Dated: October 5, 2018)

Abstract

We show that coherently driven atomic or molecular media potentially yield strong controllable short pulses of THz radiation. The method is based on excitation of maximal quantum coherence in a gas medium by optical pulses and coherent scattering of infra-red radiation to produce pulses of THz radiation. The pulses have the energies range from several nJ to $\mu$J and time durations from several fs to ns at room temperature.

PACS numbers:
TeraHertz (THz) radiation has unique potential for applications to material diagnostics including semiconductors, chemical compounds, biomolecules and biotissues [1, 2, 3]. However, despite significant progress in recent years [4, 5, 6], the methods of generation of THz radiation are still less developed than in the visible and near-infrared regions. The search for efficient, high-power, nonexpensive, portable, and room-temperature suitable methods of generation of THz radiation is one of the main topics in modern optoelectronics and photonics.

In this Letter, we suggest a way to generate short pulses of coherent THz radiation with high efficiency by using a coherently driven medium with excited maximal coherence induced by optical fields [7, 8, 9, 10, 11, 12]. This idea can be implemented in several schemes shown in Fig. 1 using atomic and molecular media involving rotational and vibrational molecular levels. Two optical fields with Rabi frequencies $\Omega_1$ and $\Omega_2$ propagate through a cell filled with a gas (the gases widely used for generation of THz generation are methanol (CH$_3$OH), CH$_3$F, H$_2$F$_2$, CH$_3$Cl, etc. [13, 14, 15]; simplified level structures are shown in Fig. 1) and prepare maximal coherence $\rho_{bc}$ between vibrational levels $b$ and $c$. Then, coherent scattering of IR radiation $\Omega_3$ produces THz radiation $\Omega_4$.

The interaction Hamiltonian for the system is given by

$$V_I = -\hbar[\Omega_2 e^{-i\omega_{ac}t}|a\rangle\langle c| + \Omega_1 e^{-i\omega_{ab}t}|a\rangle\langle b| + h.c.]$$

$$-\hbar[\Omega_3 e^{-i\omega_{cd}t}|d\rangle\langle c| + \Omega_4 e^{-i\omega_{db}t}|d\rangle\langle b| + h.c.]$$

(1)

$\Omega_i = \varphi_i E_i/\hbar$ is the Rabi frequency of the respective fields; $\varphi_{ab}$ and $\varphi_{ac}$ are the electrical dipole matrix elements between states $a$ and $b$, and $a$ and $c$; $\omega_{ab}$ and $\omega_{ac}$ are the frequencies of the electronic transitions; $\omega_{cd}$ and $\omega_{db}$ are the frequencies of the vibrational and rotational transitions; $E_i$ is the amplitude of the respective laser field.

The time-dependent density matrix equations are

$$\frac{\partial \rho}{\partial \tau} = -\frac{i}{\hbar}[H, \rho] - \frac{1}{2}(\Gamma \rho + \rho \Gamma),$$

(2)

where $\Gamma$ is the relaxation matrix. A self-consistent system also includes the field propagation equations

$$\frac{\partial \Omega_\alpha}{\partial z} = -\kappa_\alpha \Omega_\alpha - i\eta_\alpha \rho_\alpha,$$

(3)

where, index $\alpha = 1, 2, 3, 4$ indicates all fields and corresponding polarizations, $\eta_\alpha = \nu_\alpha N\varphi_\alpha/(2\epsilon_0 c)$, is the corresponding coupling constant, $\xi = \int F_{nm}(x, y)dxdy/S$ is the filling...
factor of the TE\textsubscript{nm} mode of the waveguide, \( F_{nm}(x, y) \) is the transverse field dependence\textsuperscript{17}, \( S \) is the area of the waveguide, \( \nu_{1,2,3,4} \) are the frequencies of the optical, IR, and THz fields; \( N \) is the density of medium, \( \epsilon_0 \) is the permittivity of the vacuum, \( c \) is the speed of light in vacuum, and \( \kappa_i \) are losses of the field in the cell because of scattering, diffraction, or nonresonant absorption. To avoid diffraction losses the cell may be placed in the waveguide for THz.

As shown in \[9, 10\], vibrational coherence \( \rho_{cb} \) can be excited as much as \( 0.1 - 0.5 \). The field \( \Omega_4 \) at the output of the cell is given by

\[
\Omega_4 = \xi \int_0^L dz e^{i\delta k z} \eta \tau \rho_{cb} \Omega_3^* = \xi \frac{\sin(\delta k L)}{(\delta k L)} \eta \tau \rho_{cb} L \Omega_3^*
\]

where \( L \) is the length of the cell, and the pre-factor describes phase-matching \((\delta k = k_4 - k_3 - k_1 + k_2), \xi \simeq 1\).

The THz radiation can be generated in a cell with a waveguide filled with a atomic or molecular gas by applying 3 parallel coaxial beams (two visible pump beams to prepare vibrational coherence, and an IR probe beam to produce THz radiation). There are two pulsed regime scenarios to generate THz radiation. First, we apply two short optical laser pulses (for example, femtosecond) to excite quantum coherence\textsuperscript{12} and then a short IR pulse to obtain \( \Omega_4 \) as a result of coherent scattering of field \( \Omega_3 \) on this coherence (simulations are shown in Fig. 2a,b,c).

The second scenario allows us to obtain a pulse of THz radiation shorter than the IR pulse by applying two optical pulse in the counter-intuitive pulse sequence\textsuperscript{18}. Large coherence is excited during stimulated adiabatic passage (STIRAP), and the THz radiation occurs via coherent scattering during the time of the existence of maximal quantum coherence (simulations are shown in Fig. 2d,e). The efficiencies for both these processes are the same and given by

\[
\epsilon = \frac{I_{4}^{L}}{I_{3}^{0}} = \xi^2 \text{sinc}^2(\delta k L) \left( \frac{4\pi^2 \varphi_v \varphi_j N \rho_{cb} \tau L}{\lambda h} \right)^2,
\]

where \( I_{3}^{0} \) and \( I_{4}^{L} \) are the intensities of IR at the input and THz radiation at the output of the cell correspondingly, \( \varphi_v \) and \( \varphi_j \) are the dipole moments of transitions between vibrational and rotational levels correspondingly, \( \tau \) is the duration of the THz pulse, \( \lambda \) is the wavelength of THz radiation, and \( L \) is the length of the cell.

In the case of methanol, the energy of the THz pulse can be as much as 6 nJ (using commercially available IR CO\textsubscript{2} laser, 6 kW, 15 ns. On the other hand, with a better source of
IR radiation, pulses of THz radiation with energies of 10 µJ can be obtained, for example, by using the CO$_2$ laser with 5 mJ energy and a duration of 500 ps \cite{15}. The estimated efficiency can be as much as $\epsilon \simeq 1$ (for parameters $\tau = 10^{-12}$ s, $L = 10$ cm, $\varphi_v = \varphi_j = 1D = 10^{-18}$ esu cm, $N = 5 \times 10^{16}$ cm$^{-3}$, $\lambda = 100$ µm). Note here that Eq.(5) does not take into account depletion of the probe field. The overall efficiency have to be estimated by Manley-Rowe relations which are different for different proposed schemes. For example, for the double-$\Lambda$ scheme, it is $n_1 - n_2 + n_3 - n_4 = 0$, where $n_i$ is the number of photons in the $i$th beam. The maximum $\epsilon$ one can obtain is given by $n_3 = n_4$. But for the ladder-$\Lambda$ scheme, $n_1 - n_2 - n_3 - n_4 = 0$ and $n_3$ and $n_4$ both grow during nonlinear transformation, efficiency as defined by Eq.(5) can be even bigger than 1: the energy is coming from optical fields. Also it is important that this nonlinear transformation is not dependent on population inversion, so this efficiency can be obtained at room temperature.

The efficiency for $\tau \sim 100$ fs can be increased by increasing the density of molecules to $10^{17-18}$ cm$^{-3}$. For ultra-fast pulsed optical systems ranging in their pulse duration from 15 fs to 150 fs, one is able to obtain the same duration for THz radiation as well. At the same time, using optical pumping beams with up to nanosecond pulse duration, one can generate efficiently an intense THz pulse of a nanosecond duration. Efficiency of generation of THz pulses with longer durations is limited by the relaxation of molecular vibrational coherence.

We perform simulations for laser pulse duration $\tau = 150$ fs, optical beams 1 and 2 have 30 µJ with corresponding Rabi frequencies $0.3 \times 10^{15}$ s$^{-1}$, $\varphi_v = 0.1D$, $\varphi_j = 1D$ ($CH_3F$), IR correspond CO$_2$ laser line 9P20, wavelength 500 µm ($j = 12, K$) - ($j = 11, K$), K=1-5, density $10^{17}$ cm$^{-3}$, relaxation rate due to pressure broadening is chosen to be 250 MHz, detuning 300 MHz. The results are shown in Fig.2.

It is worth comparing the estimate above with the currently achieved parameters by a variety of methods already considered as successful for generation of short THz pulses. THz pulses with ns durations were achieved in THz semiconductor lasers (quantum cascade lasers, p-Ge and n-Si lasers \cite{19, 20, 21, 22, 23}, limited by the need for cryogenic cooling). In p-Ge lasers \cite{20, 21, 22}, in the mode-lock regime, 20 ps-short pulse durations have been achieved with peak power up to a few Watts in a few-µs train of pulses.

Two other optical-pumping-based approaches giving subpicosecond THz pulses are the photocurrent method using the Auston-switch technique \cite{5, 6, 24, 25, 26} and optical rectification \cite{6, 26}. These methods allow one to generate short THz pulses with subpicosecond
durations, but with rather low intensities. The nonlinear resonant mixing in semiconductor quantum-well systems, can provide short pulses, but also with very low conversion efficiency, and it works in the mid-IR rather than in THz region [27].

The most impressive results in generation of short THz pulses were achieved by using free electron beam based sources: free-electron lasers (FEL) [28, 29] and synchrotrons [30]. Recently, the substantially higher power of coherent broadband THz radiation pulses produced by synchrotron emission were obtained from the electron beamline [30, 31], up to 100 \( \mu \text{J} \), hundreds of femtoseconds-short half-cycle THz pulses.

Our results, obtained in the example of methanol and CH\(_3\)F show that using intensive enough pumping beams, one can generate the short THz pulses with intensities, already comparable with intensities of synchrotron or FEL-based THz sources and with very high efficiency. As was already shown above, the proposed method allows us to go beyond currently reached shortest durations of the THz pulses [6, 30, 31, 32], i.e. to reach fs-durations. In addition to that, in our method, the duration of THz pulse can be controlled by the durations of both the pumping and drive beams. Besides, it works in either generating single pulses defined by the durations of the pump and drive beams, or in generating a sequence of relaxation oscillation processes-defined pulses if to pump/drive a gas cell by long pulses.

Moreover, the approach elegantly developed in [9, 10, 11] can be applied to THz radiation as well. Strong dispersion of molecular gas in THz region of vibrational-rotational resonances create additional possibilities [11] to obtain short THz pulses.

In conclusion, we have suggested an efficient method of generation of short THz pulses using coherent scattering of drive radiation in a coherently prepared atomic or molecular gas. Our work opens up a new way of using molecular gas sources for highly efficient generation of short intensive THz pulses. Applications of the obtained results have a broad range: from molecular spectroscopy to imaging, monitoring of environment and diagnostics of liquid and solid materials.

We thank A. Chugreev, J. Kono, A. Nazarkin, and O. Portugal for helpful discussions. We also gratefully acknowledge the support from the Office of Naval Research, the Air Force Office of Scientific Research, the Defense Advanced Research Projects Agency, and
the Robert A. Welch Foundation.

[1] D. Grischkowsky, Soeren Keiding, M. van Exter, and Ch. Fattinger, J Opt. Soc. B 7, 2006 (1990).
[2] X.-C. Zhang, Physics in Medicine and Biology 47, 1 (2002).
[3] Encyclopedia of Chemical Physics and Physical Chemistry, edited by J. H. Moore and N. D. Spencer, (Institute of Physics, London, 2001).
[4] N. G. Kalugin, in Handbook of semiconductor nanostructures and nanodevices, edited by A. A Balandin and K. L. Wang (American Scientific Publishers, Los Angeles, 2005).
[5] Sensing with Terahertz Radiation, edited by D. Mittleman (Springer, New York, 2003).
[6] B. Ferguson and X.-C. Zhang, Nature Mater. 1, 26 (2002).
[7] S. E. Harris, Phys. Today 50, No. 7, 36 (1997).
[8] A. B. Matsko, Y. V. Rostovtsev, M. Fleischhauer, and M. O. Scully, Phys. Rev. Lett. 86, 2006 (2001).
[9] A. V. Sokolov, D. R. Walker, D. D. Yavuz, G. Y. Yin, and S. E. Harris, Phys. Rev. Lett. 85, 562 (2000).
[10] S. E. Harris and A. V. Sokolov, Phys. Rev. Lett. 81, 2894 (1998).
[11] A. Nazarkin and G. Korn, Phys. Rev. Lett. 83, 4748 (1999).
[12] M. O. Scully, G. W. Kattawar, P. R. Lucht, T. Opatrny, H. Pillof, A. Rebane, A. V. Sokolov, and M. S. Zubairy, Proc. Natl. Acad. Sci. U.S.A. 9, 10994 (2002).
[13] T. Y. Chang, T. J. Bridges, and E. G. Burkhardt, Appl. Phys. Lett. 17, 249 (1970).
[14] T. Y. Chang, T. J. Bridges, and E. G. Burkhardt, Appl. Phys. Lett. 17, 357 (1970).
[15] E. Mueller, Wiley Encyclopedia of Electrical and Electronics Engineering, Vol.20, edited by J. G. Webster (John Wiley & Sons, New York, 1999), p.597.
[16] H. Houtman, J. Mayer, J. Appl. Phys. 61, 843 (1087).
[17] Max Born and Emil Wolf, Principles of optics , (Cambridge, UK; Cambridge University Press, 1997).
[18] K. Bergmann, H. Theuer, and B. W. Shore, Rev. Mod. Phys. 70, 1003 (1998).
[19] R. Koehler, A. Tredicucci, F. Beltram, H. E. Beere, E. H. Linfield, A. G. Davies, D. A. Ritchie, R. C. Iotti, and F. Rossi, Nature 417, 156 (2002).
[20] E. Gornik and A. A. Andronov, Opt. Quantum Electron. **23** (1991), and references therein.

[21] V. I. Gavrilenko, N. G. Kalugin, Z. F. Krasil’nik, V. V. Nikonorov, A. V. Galyagin, and P. N. Tsereteli, Semic. Sci. Technol. **7** B649 (1992).

[22] A. V. Muravjov, R. C. Strijbos, C. J. Fredricksen, H. Weidner, W. Trimble, S. H. Withers, S. G. Pavlov, V. N. Shastin, and R. E. Peale, Appl. Phys. Lett. **73**, 3037 (1998).

[23] S. G. Pavlov, R. Kh. Zhukavin, E. E. Orlova, V. N. Shastin, A. V. Kirsanov, H.-W. Huebers, K. Auen, and H. Riemann, Phys. Rev. Lett. **84**, 5220 (2000).

[24] X.-C. Zang and D. H. Auston, J. Appl. Phys. **71**, 326 (1992).

[25] G. Zhao, R. N. Schouten, N. Van der Valk, W. Th. Wenkebach, P. C. M. Planken, Rev. Sci. Inst. **73**, 1715 (2002).

[26] A. Rice, Y. Jin, Z.-C. Zang, D. Bliss, J. Larkin, and M. Alexander, Appl. Phys. Lett. **64**, 1324 (1994).

[27] D. S. Pestov, A. A. Belyanin, V. V. Kocharovsky, Vl. V. Kocharovsky, and M. O. Scully, J. Mod. Opt. **51**, 2523 (2004).

[28] W. Chin, J.-P. Dognon, C. Canuel, F. Piuzzi, I. Dimicol, M. Mons, I. Compagnon, G. von Helden, and G. Meijer, J. Phys. Chem. **122**, 054317 (2005).

[29] X. G. Peralta, S. J. Allen, M. C. Wanke, N. E. Harff, J. A. Simmons, M. P. Lilly, J. L. Reno, P. J. Burke, J. P. Eisenstein, App. Phys. Lett. **81**, 1627 (2002).

[30] G. R. Neil, G. L. Carr, J. F. Gubeli, K. Jordan, M. C. Martin, W. R. McKinney, M. Shinn, M. Tani, G. P. Williams, and X.-C. Zhang, Nuclear Instruments and Methods in Physics Research A **507**, 537 (2003).

[31] Y. C. Shen, P. C. Upadhya, E. H. Linfield, H. E. Beere, A. G. Davies, Appl. Phys. Lett. **83**, 3117 (2003).

[32] J. Bromage, S. Radic, G. P. Agrawal, C. R. Stroud, P. M. Fauchet, R. Sobalevski, Opt. Lett. **22**, 627 (1997).

[33] V. P. Kalosha and J. Herman, Opt. Lett. **26**, 456 (2000).

[34] R. A. Bartels, T. C. Weinacht, N. Wagner, M. Baertschy, Chris H. Greene, M. M. Murnane, and H. C. Kapteyn, Phys. Rev. Lett. **88**, 013903 (2002).
Figure caption

Figure 1.

Level schemes for THz generation. $\Omega_1$ and $\Omega_2$ are optical fields in two-photon resonance to excite atomic or molecular coherence between levels $b$ and $c$. $\Omega_3$ is the IR field, and $\Omega_4$ is generated THz radiation (Double-$\Lambda$ shown by dashed lines, Ladder-$\Lambda$ by solid lines). Atomic medium (a). For example, Rb levels are $a = 5S_{1/2}$, $b = 10P_{1/2,3/2}$, $c = 6P_{1/2,3/2}$, $d(d') = 8D_{3/2,5/2}(9D_{3/2,5/2})$. Molecular medium (b). Level $b$ is the ground state, vibrational quantum number $v = 0$ and angular momentum $j$; $c$ is the level with $v = 1$ and $j'$, $d$ is the level, $v = 0$ and $j + 1$; $d'$ is the level $v = 1$ and $j' + 1$. Levels of CH$_3$F shown in (c).

Figure 2.

Results of simulations for double-$\Lambda$ scheme in CH$_3$F for two temporal scenarios. Scenario 1. Fields $\Omega_1$ and $\Omega_2$ and prepared by them coherence $\rho_{bc}$ are shown in a). Probe IR field $\Omega_3$ and generated THz field $\Omega_4$ are shown in b). Efficiency of nonlinear transformation is shown in c). Scenario 2. Fields $\Omega_1$ and $\Omega_2$ in STIRAP configuration and prepared by them coherence $\rho_{bc}$ are shown in d). Probe IR field $\Omega_3$ and generated THz field $\Omega_4$ are shown in e). Parameters used in simulations are in the text.
Figure 1, N.G. Kalugin, et al., “Generation of strong short coherent terahertz pulses in a ladder-Lambda system”
Figure 2, N.G. Kalugin, et al., “Generation of strong short coherent terahertz pulses in a ladder-Lambda system”