Two Level Metal Vapor Lasers with Thermal Creation of Population Inversion

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(Received 3 July 2005; published 30 March 2006)

We present a new technique for direct conversion of thermal energy into coherent radiation. The near-surface layer of evaporated excited atoms of rare earth metals is proposed to be used as a converter. There is an inverted population in this layer. Operation principles of two level lasers on the transitions to the ground state using Eu are considered. The fact of detachment of Sm atoms in the excited $4f^{N-1}5d6s^2$ state during the thermal evaporation process has been proved experimentally.

DOI: 10.1103/PhysRevLett.96.123902 PACS numbers: 42.55.Lt, 64.70.Hz, 71.20.Eh

The idea of using thermal excitation of a quantum system to generate coherent radiation was originally reported in [1] in 1959, where the possibility of the creation of population inversion in a three level system was considered. The suggestion concerning technical realization for direct conversion of thermal energy into coherent electromagnetic radiation in ruby was described in 1961 in the invention [2]. The first working device (gas-dynamic laser) where vibrationally excited CO_2 molecules were used as a source of radiation energy was first suggested by V. K. Konyukhov and A. M. Prochorov in 1966 [3]. The analysis of the above-listed methods for direct heat conversion into electromagnetic radiation in Ref. [4] showed that quantum generators with thermal excitation comply with the second law of thermodynamics and it is necessary to use both a heater and a cooler (heat machine) for their operation. For example, the population inversion in ruby sample appears during temperature leveling [2].

In this Letter we suggest to use the peculiarities of the thermal evaporation process of rare earth metals (REMs) for direct heat conversion into coherent radiation. And it should be noted that there is no need for a cooler to create population inversion.

As it was shown in Ref. [5], the thermal evaporation process from REM surfaces with an incomplete $4f$ shell is completely defined by neutral atoms in the excited $4f^{N-1}5d6s^2$ state (with the exception of Ce and Gd). It is reasonable to assume that these atoms detach from the surface only in the excited state.

During evaporation of these atoms in vacuum (provided that they do not return to the evaporated surface) the near-surface layer is formed; its thickness d (Fig. 1) is determined by their average thermal speed $\bar{\nu}$ and the lifetime τ of this excited state. The evaporated atoms in this layer are in the excited $4f^{N-1}5d6s^2$ state. The state under consideration could be used for laser creation where the $4f^N6s^2$ ground state is used as a lower laser level. The supporting evidence for such a possibility comes from ''spiking'' behavior corresponding to resonance transitions during dissociation of TmI_3 molecules after discharge initiation [6]. From our point of view, in case of bond breaking in TmI_3 molecule, Tm atom is found to be in the excited $4f^{N-1}5d6s^2$ state with subsequent conversion into the resonance state by electron impact.

Figure 1 shows schematically a metal vapor laser with thermal creation of population inversion. The resonator mirrors $M1$ and $M2$ are located coaxially to the active volume. The REM sample in the form of a bar having smooth upper surface is in close contact with the heater. The output windows $W1$ and $W2$ are heated to prevent vapor condensation. Buffer gas (He, for example) can also be used to protect the output windows.

Lasing into the ground state in the laser under consideration can be made directly on the transition from $4f^{N-1}5d6s^2$ to $4f^N6s^2$. We considered the operation of such a laser using Eu with singlet ground level as an example. In Eu the lowest level of the $4f^{N-1}5d6s^2$ configuration has energy $E_{5d} = 27\,852.90\ \mathrm{cm}^{-1}$ [7]. The wavelength $\lambda = 358.926$ nm corresponds to the transition to the ground state. The probability of the transition mentioned and the lifetime of this level, respectively, can be calculated using the oscillator strength $f_{10} = 0.0011$ [8]. The lifetime τ is 1.3×10^{-6} s. The average thermal speed of an evaporated atom calculated from the formula [9]

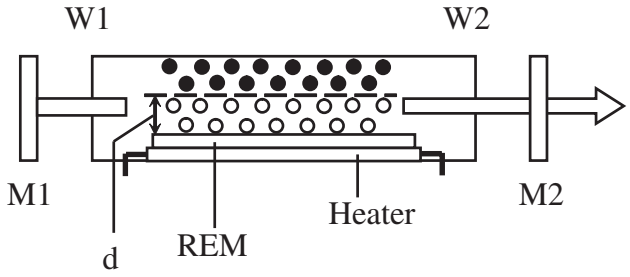

FIG. 1. Schematic diagram shows the REM vapor laser with thermal creation of population inversion. ○, atoms in the lowest excited of the $4f^{N-1}5d6s^2$ configuration; ●, atoms in the ground $4f^N6s^2$ state.

$$\bar{\nu} = 1.6\sqrt{\frac{RT_{\mathrm{ev}}}{\mu}}, \tag{1}$$

equals 3.92×10^4 cm/s, where R is the molar gas constant, μ is the molecular weight, and T_{ev} is the evaporation temperature (1090 K), corresponding to saturated-vapor pressure of Eu $P^0_{\mathrm{ev}} = 0.95$ Torr (Eu is still in solid state). For specified values of τ and $\bar{\nu}$, the thickness of the near-surface layer $d = 0.05$ cm.

The number of atoms passing through the level d per unit of time is estimated from evaporation rate (overall mass of atoms, evaporated from 1 cm^2 of Eu sample surface in 1 s) calculated with the formula [9]

$$W_{\mathrm{Eu}} = P^0_{\mathrm{ev}}\sqrt{\frac{\mu}{2\pi RT_{\mathrm{ev}}}}. \tag{2}$$

The value of evaporation rate is 2.15×10^{-2} g/(cm^2 s). This corresponds to evaporation of 8.7×10^{19} of excited atoms during 1 s from the surface of 1 cm^2. Since the energy of 1 quantum $h\nu$ is 5.3×10^{-19} J, the power of about 30 W (or 600 $\mathrm{W/cm}^3$) is released in monochromatic radiation in the active volume having the thickness of d and the area of 1 cm^2.

We estimated the efficiency of thermal energy conversion into monochromatic radiation using the energy losses of the sample heated in vacuum. Three main constituents of losses were taken into account: heated body radiation, kinetic energy of evaporated atoms, and electronic excitation energy of these atoms. The estimated efficiency exceeds 50%.

The two level laser under consideration is a transverse vapor flow laser [10]; i.e., the excited atoms enter the active volume from the surface of the metal sample. When interacting with self-field of induced emission the excited atoms emit quanta of energy and settle on a colder upper wall of the laser tube.

In the transverse vapor flow laser the thickness of stimulated emission layer d_{stim} (or the thickness of laser beam) is defined similar to the thickness of spontaneous emission layer $d (d = \tau\bar{\nu})$, i.e., via average thermal speed $\bar{\nu}$ and lifetime of excited level τ_{stim} in self-field of induced emission ($d_{\mathrm{stim}} = \tau_{\mathrm{stim}}\bar{\nu}$). Average thermal speed $\bar{\nu}$ is defined by temperature [see Eq. (1)] and hence $\bar{\nu}$ is equal for spontaneous and stimulated emission. To estimate τ_{stim} and beam thickness in our laser we used the relation between the Einstein coefficients for spontaneous (A) and induced (B) emission for Eu with $\lambda_{\mathrm{gen}} = 358.926$ nm:

$$B = \frac{A}{8\pi h(\nu/c)^3} = \frac{A}{8\pi h(1/\lambda)^3} = 3.7 \times 10^{12}\ \mathrm{A\,m^3/(J\,s)}, \tag{3}$$

where h is the Planck constant, c is the speed of light (*in vacuo*), $\lambda = \lambda_{\mathrm{gen}}$, $\nu = c/\lambda$ is the radiation frequency. Similar calculations were performed for $\lambda = 1.25$ cm, where estimated $B \approx 10^{25}$ A [11].

Then an excited atom will emit a photon after reaching the self-field of induced emission (even for energy density of the radiation $\rho = 1$ J/m^3) in a time

$$\tau_{\mathrm{stim}} = \frac{1}{\rho B} = \frac{1}{3.7 \times 10^{12}\rho}\frac{1}{A} = \frac{\tau}{3.7 \times 10^{12}} \approx 3.5 \times 10^{-19}\ \mathrm{s}, \tag{4}$$

where $\tau = 1/A$ is the lifetime of the excited level of an Eu atom ($\tau = 1.3 \times 10^{-6}$ s). The value of energy density of radiation $\rho = 1$ J/m^3 was used only as an example, though actually $\rho \gg 1$, and therefore τ_{stim} will be much lower than calculated value in (4).

It is well known from quantum dynamics that the laser beam thickness cannot be less than radiation wavelength. Under these conditions the time required for an excited Eu atom to travel the distance of wavelength λ_{gen} from the surface is

$$t_\lambda = \frac{\lambda_{\mathrm{gen}}}{\bar{\nu}} \approx 9.2 \times 10^{-10}\ \mathrm{s}. \tag{5}$$

It follows from Eqs. (4) and (5) that $\tau_{\mathrm{stim}} \ll t_\lambda$; i.e., an atom emits a photon before it passes the distance equal to layer thickness of λ_{gen}. Considering the above we suppose that the thickness of the laser beam can be in the range from the wavelength λ_{gen} to spontaneous emission layer d.

We estimated the power of laser radiation for the Eu sample surface area of 1 cm^2 using the following equation:

$$P = \frac{1}{2}\varepsilon\frac{W_{\mathrm{Eu}}}{m_a} \approx 23\ \mathrm{W}, \tag{6}$$

where $\varepsilon = h\nu$ is the photon energy and m_a is the Eu atom mass. The power density of laser radiation for the sample surface area S_{surf} and laser beam thickness d_{beam} can be calculated from the following equation [using Eqs. (2) and (6)]

$$I_{\mathrm{Eu}} = \frac{\alpha S\varepsilon}{2m_a}P^0_{\mathrm{ev}}\sqrt{\frac{\mu}{2\pi RT_{\mathrm{ev}}}}, \tag{7}$$

where $S = S_{\mathrm{surf}}/(1\ \mathrm{cm}^2)$ and $\alpha = (1\ \mathrm{cm})/d_{\mathrm{beam}}$. The maximum possible power density of laser radiation, corresponding to $d_{\mathrm{beam}} = \lambda_{\mathrm{gen}}$ and $S_{\mathrm{surf}} = 1\ \mathrm{cm}^2$, is 6.4×10^5 $\mathrm{W/cm}^2$. But so high power density and some nonlinear effects would prevent the laser beam from forming. For more real beam thickness, for example, $d_{\mathrm{beam}} = 0.01$ cm the power density $I_{\mathrm{Eu}} = 2.3 \times 10^3$ $\mathrm{W/cm}^2$.

To prove the possibility to realize such lasers we studied experimentally the emission spectrum of evaporated REM atoms. Sm was chosen because its radiation on the transition from the excited $4f^{N-1}5d6s^2$ state to the $4f^N6s^2$ ground state is in visible spectrum region. The lowest level of Sm $4f^{N-1}5d6s^2$ configuration has energy $E = 18\,075.67\ \mathrm{cm}^{-1}$, but the ground state consists of 6 suble-

vels [7]. From the excited $4f^{N-1}5d6s^2$ state into the ground state in such level system the transitions are found for 2 sublevels with energies $E_1 = 292.58$ cm^{-1} and $E_2 = 811.92$ cm^{-1} and wavelengths $\lambda_1 = 562.179$ nm, $\lambda_2 = 579.091$ nm, respectively [8]. A bar with the active surface of 4×7 mm^2 was cut out from available metallic Sm sample with purity of 99.83% and placed in the laser cavity (Fig. 1) without resonator mirrors $M1$ and $M2$. To prevent vapor condensation on the output windows the previously vacuumed laser cavity was filled with He as a buffer gas at the pressure of 0.5 Torr. The radiation was detected by a monochromator MDR-23 (LOMO PLC, St. Petersburg) with 600 grooves/mm diffraction grating, a photomultiplier tube FEU-62, and an oscillograph S1-55 with high-impedance input. After the sample was heated to 600 °C, the single spontaneous line with $\lambda_1 = 562.179$ nm was detected. The radiation with given wavelength was also observed visually (yellow-green glow along the sample surface). As it is well known, the evaporation rate and, accordingly, the concentration of evaporated atoms over the Sm surface highly depend on the surface condition. As the surface of the sample was not specially processed, there was no way to evaluate the concentration of evaporated atoms over the surface. However, we consider the fact of evaporation of atoms in the excited state from Sm surface to be proved.

Summarizing the above results, we have drawn the following conclusions: (i) using Sm, the fact of excited atom detachment in the lowest $4f^{N-1}5d6s^2$ state with subsequent emission during thermal evaporation was established. (ii) In the near-surface layer of evaporated excited REM atoms in vacuum there is the active medium. There is inverted population in this medium. The near-surface layer is determined by the average thermal speed of these atoms and the lifetime of the excited lowest level of the $4f^{N-1}5d6s^2$ configuration. (iii) Contrary to the conclusions made in [4], a cooler is not required for the process of thermal creation of population inversion (cooler acts only as a condenser, preventing evaporated atoms from returning to metal surface). The population inversion takes place in the two level system. In this system the role of lower laser level is played by the ground state of REM atom. (iv) The laser beam thickness d_{beam} of two level lasers with thermal pumping using Eu as an example can be in the range from the wavelength λ_{gen} to spontaneous emission layer d. And the maximum possible power density I_{Eu} of continuous laser radiation from the evaporation area of 1 cm^2 can reach the order of 10^5 W/cm^2 in the ultraviolet region of spectrum. (v) The presence of excited atoms in the near-surface layer could be used to reduce the number of photoionization stages in the process of laser separation of isotopes.

We cannot exclude the possibility of nonradiative release of excitation energy of evaporating atoms, for example, via energy transfer to a crystal or into additional kinetic energy of evaporated atoms [12,13]. To investigate these ways in more detail, it is essential to have more pure conditions for carrying out the experiment. After these experiments we will be able to draw conclusions concerning the prospects for such lasers.

We are thankful to Professor V. G. Bagrov and Professor G. F. Karavaev for interest in this work and critical remarks. This work was supported in part by the programme of the President of Russian Federation ''Leading Scientific Schools of Russian Federation'' (Grant No. NSh 373.2003.5).

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[1] H. E. D. Scovil and E. O. Schulz-DuBois, Phys. Rev. Lett. **2**, 262 (1959).
[2] E. O. Schulz-DuBois and H. E. D. Scovil, U.S. Patent No. 3015072 1961.
[3] V. K. Konyukhov and A. M. Prokhorov, JETP Lett. **3**, 286 (1966).
[4] V. K. Konyukhov and A. M. Prokhorov, Usp. Fiz. Nauk **119**, 541 (1976).
[5] V. A. Gerasimov and V. V. Gerasimov, JETP Lett. **78**, 339 (2003).
[6] M. Neiger, W. Kaesler, and H.-P. Popp, Appl. Phys. B **37**, 73 (1985).
[7] W. C. Martin, R. Zalubas, and L. Hagan, Nat. Bur. Stand. Ref. Data Ser. **60**, 174 (1978).
[8] N. P. Penkin, V. N. Gorshkov, and V. A. KomarovskiĬ, Zh. Prikl. Spektrosk. **41**, 533 (1984).
[9] Saul Dushman, *Scientific Foundations of Vacuum Technique* (Wiley, New York, 1962).
[10] C. M. Ferrar, IEEE J. Quantum Electron. **9**, 856 (1973).
[11] J. J. Turner in *Vibrational Spectroscopy. Modern Trends*, edited by A. J. Barnes and W. J. Orville-Thomas (Elsevier, New York, 1977), Vol. A, Chap. 2.
[12] A. M. Bonch-Bruevich *et al.*, Sov. Phys. JETP **65**, 161 (1987).
[13] A. M. Bonch-Bruevich *et al.*, Sov. Phys. JETP **70**, 993 (1990).