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Observation of saturated dispersion resonances of methane in a two-mode Cr^{2+}: ZnSe/CH_4 laser

M.A. Gubin, A.N. Kireev, V.I. Kozlovsky, Yu.V. Korostelin, V.A. Lazarev, A.B. Pnev,
Yu.P. Podmar'kov, D.A. Tyurikov, M.P. Frolov, A.S. Shelkovnikov

Abstract. **Solid-state cw Cr^{2+}: ZnSe laser has been used for Doppler-free spectroscopy of lines of the vibrational – rotational $\nu_1 + \nu_4$ band of methane. Saturated-dispersion resonances on the components of the R(2) line near $\lambda = 2.36$ μm are revealed. The parameters of the saturated-dispersion resonance obtained upon cooling intracavity methane cell to a temperature of 77 K confirmed good prospects of developing an optical master oscillator with a high (10^{-15} – 10^{-16}) short-term frequency stability based on a Cr^{2+}: ZnSe/CH_4 laser.**

Keywords: *Cr^{2+}: ZnSe laser, IR lasers, tunable lasers, solid-state lasers, two-mode lasers, optical frequency standards, Doppler-free resonances, laser spectroscopy.*

A necessary element of actively developed microwave and optical quantum frequency standards based on cold atoms and ions in optical and electromagnetic traps (with expected relative instability in the range of 10^{-16} – 10^{-18}) is a master oscillator (MO) with a narrow emission spectrum. The MOs that have been developed to date for sensing ultranarrow (~1 Hz) optical resonances are characterised by short-term frequency stability: $\sigma_y \sim 1 \times 10^{-15}$ (averaging time $\tau = 1$ s). Intense studies aimed at achieving a stability $\sigma_y \sim 3 \times 10^{-16}$ ($\tau = 1$ s) are under way (see [1 – 3] and references therein). These oscillators are based on stabilisation of the frequency of laser or microwave radiation using optical and microwave cavities with an ultrahigh (~10^{10}) Q factor, carefully isolated from environment. An example is cooling sapphire microwave cavity to liquid-helium temperatures. We develop another approach to designing MO with a short-term stability of frequency $\sigma_y = 10^{-15}$ – 10^{-16}, which is based on lasers stabilised using Doppler-free saturated-absorption (SA) and saturated-dispersion (SD) resonances in low-pressure gas cells [4]. These systems impose less stringent requirements on the external conditions and can additionally provide a high middle-term stability, since they use active spectral-line stabilisation and exhibit no frequency drifts (beginning with $\tau \geqslant 10$ s), which are characteristic of passive-cavity MOs. A promising laser – absorption cell system, which is considered here, consists of a cw solid-state Cr^{2+}: ZnSe laser and a methane absorbing cell.

All previously developed methane MOs and optical frequency standards (OFSs) traditionally used a He – Ne laser with a working wavelength of 3.39 μm; its lasing frequency is close to those of the F_2^2 and E components of the P(7) line belonging to the ν_3 band [5 – 8]. The development of cw tunable solid-state lasers on crystals of A_2B_6 compounds doped with divalent ions of transition metals [9] solved the problem of choosing lines. For example, the wavelength tuning range of cw Cr^{2+}: ZnSe lasers includes the $\nu_1 + \nu_4$ absorption band of methane (2.3 – 2.5 μm) [10], and one can choose any line within this band.

The main parameters determining the MO frequency stability are the width of the reference resonance and the signal-to-noise (S/N) ratio. The methane absorption cross section in the range of 2.3 – 2.5 μm is two to three orders of magnitude smaller than on the lines of the ν_3 band. This circumstance reduces the SA and SD resonance signals in comparison with those used in He – Ne lasers (3.39 μm) but prevents resonances from large field broadening. The saturation parameter of the laser transition in the Cr^{2+}: ZnSe crystal is ~15 kW cm^{-2}, which is three orders of magnitude larger than that in gas lasers, while the characteristic output powers are several tens of milliwatts and several tenths of milliwatt for the Cr^{2+}: ZnSe and He – Ne/CH_4 lasers, respectively. For the same reason the limiting frequency noise of the single-mode Cr^{2+}: ZnSe laser, which is determined by the contribution of spontaneous emission, is lower than in the He – Ne laser by a factor of about $\sqrt{10^3}$ (provided that the cavities have the same Q factor). A Cr^{2+}: ZnSe laser with a frequency noise at a level of 0.03 Hz Hz$^{-1/2}$ was described in [4]; at a lasing frequency of 125 THz this noise level corresponds to a relative spectral density of frequency fluctuations of 3×10^{-16} in a 1-Hz band. This fundamental limit is obscured by technical laser frequency fluctuations caused by the cavity length instability; to suppress them, one needs a stabilisation system based on a quantum discriminator: a narrow SA or SD resonance on one of methane spectral lines.

Based on the specific features of two-mode nonlinear laser spectroscopy [8] and the results of [4], we established that the SD resonances observed in the beat frequency of a two-mode laser with a methane cell provide in the best way the necessary S/N ratio. In contrast to the SA resonances, the SD signal is in fact independent of the output power, and one can use a

M.A. Gubin, A.N. Kireev, V.I. Kozlovsky, Yu.V. Korostelin, D.A. Tyurikov, A.S. Shelkovnikov P.N. Lebedev Physics Institute, Russian Academy of Sciences, Leninsky prosp. 53, 119991 Moscow, Russia; e-mail: kireev@okb.lpi.troitsk.ru;
V.A. Lazarev, A.B. Pnev Scientific and Educational Center 'Photonics and IR Technique', Bauman State Technical University, Vtoraya Baumanskaya ul. 5, 105005 Moscow, Russia;
Yu.P. Podmar'kov, M.P. Frolov P.N. Lebedev Physics Institute, Russian Academy of Sciences, Leninsky prosp. 53, 119991 Moscow, Russia; Moscow Institute of Physics and Technology (State University), Institutskii per. 9, 141700 Dolgoprudnyi, Moscow region, Russia; e-mail: frolovmp@x4u.lebedev.ru

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cavity with a small (~1%) loss on radiation output, which, in principle, makes it possible to obtain spontaneous laser noises at the millihertz level in Cr^{2+}:ZnSe/CH_4 OFS, with a simultaneous decrease in the optical pump power of the crystal.

The scheme of the developed experimental setup is similar to a great extent to that described in [4]. An additional element is a methane cell 20 cm long, placed in the laser cavity. The design of this cell made it possible to cool gas in it to liquid nitrogen temperature. The laser wavelength was tuned using a diffraction grating (600 lines mm^{-1}, efficiency 80%). The grating zeroth order served to couple out radiation. Fine selection was performed using an intracavity Fabry–Perot interferometer (free spectral range 15 GHz), tuned by a piezoelectric transducer.

The laser generated two neighbouring axial modes (intermode distance ω_{12} = 130 MHz); with a synchronous change in the lengths of cavity and intracavity interferometer, it could be continuously (without jumps between modes) tuned in a range ~150 MHz wide. To choose the central frequency in the smooth tuning range, we used additionally an external reference cell with methane at a pressure $p_{CH_4} \sim 5$ Torr. A diffraction grating was applied to tune the laser frequency to the center of Doppler profile for a chosen line.

When detecting the intermode frequency, we obtained SD resonances at room and liquid nitrogen temperatures (Fig. 1). We chose the E component of the R(2) line in the $\nu_1 + \nu_4$ band ($\lambda = 2.36$ μm), which was not affected by the magnetic hyperfine structure and, therefore, was of greatest interest for OFSs. The total width of SD resonances was ~500 kHz (it was estimated from the transmission peak of the external interferometer), and the total peak-to-peak amplitude was ~2 kHz at a temperature T = 300 K and 20 kHz at T = 77 K. When the cell was cooled, the methane pressure was reduced by an order of magnitude (to ~1 mTorr instead of 10 mTorr at room temperature) to exclude lasing suppression.

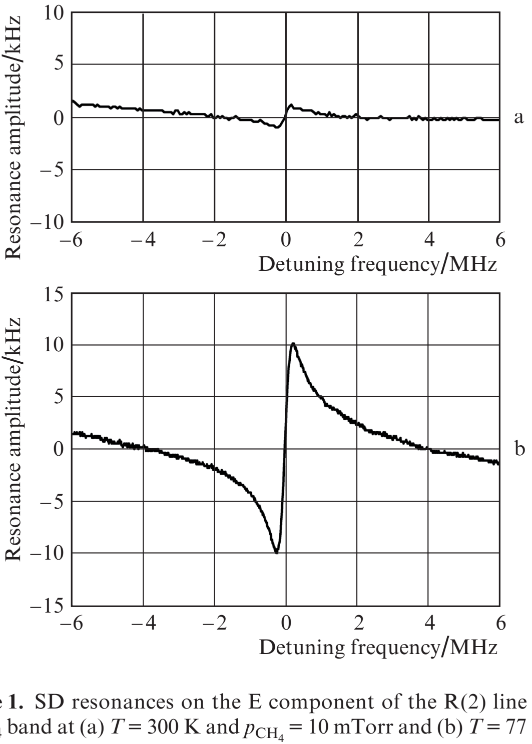

Figure 1. SD resonances on the E component of the R(2) line in the $\nu_1 + \nu_4$ band at (a) T = 300 K and p_{CH_4} = 10 mTorr and (b) T = 77 K and p_{CH_4} = 1 mTorr.

Thus, when the cell was cooled, the signal increased by a factor of almost 100 due to the increase in the number of absorbing molecules at the rotational level with $J = 2$, which compensated for the same decrease in the dipole moment for the lines belonging to the $\nu_1 + \nu_4$ band in comparison with the lines in the traditionally used ν_3 band. The main contribution to the observed resonance width is from the technical component of the Cr^{2+}:ZnSe laser emission spectrum, which is caused by insufficient passive stability of its cavity. Without this contribution the full width of the resonances at half maximum, γ_Σ, which is determined by collisions, finite time of flight, and field broadening, should not exceed, according to estimates, 150 kHz (T = 77 K). It is not difficult to reduce γ_Σ to ~50 kHz with conservation of the resonance amplitude $\Delta\omega_{12}$; in this case, the resonance slope $\Delta\omega_{12}/\gamma_\Sigma$ is ~0.4.

Thus, we observed for the first time Doppler-free SD resonances on the singlet E component of the R(2) line in the $\nu_1 + \nu_4$ band of methane ($\lambda = 2.36$ μm). An increase in the resonance amplitude by two orders of magnitude upon methane cooling to liquid nitrogen temperature due to the use of line with a small rotational quantum number was revealed. The obtained resonance parameters confirm the estimate [4] of the possibility of attaining short-term frequency stability $\sigma_y = 10^{-15}$–10^{-16} (τ = 1 s) in Cr^{2+}:ZnSe/CH_4 OFSs.

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References

1. Millo J. et al. *Phys. Rev. A*, **79**, 053829 (2009).
2. Vogt S., Lisdat C., Legero T., Sterr U., Ernsting I., Nevsky A., Schiller S. *Appl. Phys. B*, **104**, 741 (2011).
3. http://www.uliss-st.com/technology.html.
4. Gubin M.A., Kireev A.N., Kozlovsky V.I., Korostelin Yu.V., Pnev A.B., Podmar'kov Yu.P., Tyurikov D.A., Frolov M.P., Shelestov D.A., Shelkovnikov A.S. *Kvantovaya Elektron.*, **42**, 509 (2012) [*Quantum Electron.*, **42**, 509 (2012)].
5. Hall J.L. *Laser Phys.*, **4**, 306 (1994).
6. Bagaev S.N., Chebotaev V.P. *Usp. Fiz. Nauk*, **148**, 143 (1986).
7. Rastorguev Yu.G., Titov A.N. *Kvantovaya Elektron.*, **18**, 383 (1991) [*Sov. J. Quantum Electron.*, **21**, 348 (1991)].
8. Gubin M.A., Protsenko E.D. *Kvantovaya Elektron.*, **24**, 1080 (1997) [*Quantum Electron.*, **27**, 1048 (1997)].
9. DeLoach L.D., Page R.H., Wilke G.D., Payne S.A., Krupke W.F. *IEEE J. Quantum Electron.*, **32**, 885 (1996).
10. Brown L.R. et al. *J. Quant. Spectrosc. Radiat. Transfer*, **82**, 219 (2003).