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Terahertz Beating of Laser Emission from an Organic Microcavity

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We present time-resolved studies of laser emission from an organic microcavity filled with a guest-host composite of tris-(8-hydroxy quinoline) aluminium (Alq_3) and 4-(dicyanomethylene)-2-methyl-6-(p-dimethylaminostyryl)-4H-pyran (DCM). A linesplitting of 0.18 THz between two perpendicularly polarized lines is observed, resulting from an optical anisotropy in the distributed Bragg reflectors. This particular behavior is attributed to oblique columnar structures shaped by off-axial preparatory conditions. By means of an up-conversion setup an oscillation at 5.5 ps period time and phase-coupling of the modes are observed. A rate equation-based approach is utilized for modelling the temporal behavior.

The development in information technology has befueled miniaturization and cost reduction of semiconductor lasers. Among these, vertical cavity surface emitting lasers (VCSELs) promise applications in fiber-optic communication and integrated optical circuits [1]. Interest in organic VCSELs (OVCSELs) has increased due to possible advantages in tunable emission and competitive fabrication costs. Lasing in organic thin film structures has been demonstrated for polymers as active medium [2], or mixtures of single molecules, e.g. DCM in Alq_3 [3, 4]. Relaxation oscillations were observed as modulation of the emission behavior of optically pumped thin films [5] and OVCSELs [6], with an oscillation period of the order of tens of picoseconds. Terahertz (THz) oscillations in emission from inorganic semiconductor microcavities were observed before with an angular dependency of the oscillation frequency [7]. Here, we report the observation of THz oscillations from OVCSEL emission, which are a consequence of the simultaneous emission of two perpendicularly polarized laser lines from a single organic microcavity laser.

Major advances have been made in the past few years in the control of photon modes in various types of microcavities. Simultaneous emission of two laser modes has been reported from coupled microcavities [8, 9]. However, the precise tuning of the two cavity layer thicknesses in a coupled microcavity complicates the fabrication process. Two mode laser emission from a single cavity has also been reported [10]. There, two perpendicularly polarized modes were generated by anisotropic molecular orientation in the active layer. We observe two mode laser emission leading to a THz oscillation in the temporal emission behavior due to anisotropy in the dielectric mirrors. The oscillation period corresponds to the spectral distance of both emitted modes.

The mirrors of our OVCSEL consist of alternating $\lambda/4$-layers of SiO_2 and TiO_2, with λ as the cavity design wavelength. Since both mirrors are distributed Bragg reflector (DBR) structures, we call the sample a double-DBR microcavity. The substrates were plates of optical glass, polished to minimize surface roughness. Reactive electron beam evaporation under high-vacuum conditions was used to deposit the mirror materials. This method reduces absorption in the inorganic layers. The substrates were mounted off-axially over the evaporation sources. The organic materials were deposited by thermal co-evaporation of the matrix material Alq_3 doped with 2 % of the emitter material DCM.

Optical pumping of our samples was performed with the frequency doubled output of a regenerative Ti:Sa amplifier system (SPITFIRE, Spectra-Physics), providing pulses of 200 fs length, pulse energy $75\,\mathrm{nJ}$ and $400\,\mathrm{nm}$ wavelength at a repetition rate of $1\,\mathrm{kHz}$. These pulses are focused to a spot size of $50\,\mu\mathrm{m}$. For studying emission dynamics, a gate pulse length of $200\,\mathrm{fs}$ at $800\,\mathrm{nm}$ wavelength in an up-conversion experiment provided a resolution of $400\,\mathrm{fs}$. These pulses were overlapped with the OVCSEL emission in a nonlinear crystal and the generated sum-frequency photons detected by a photomultiplier tube. The laser system also provides spectrally broad laser pulses from supercontinuum generation [11]. They are used for linear transmission studies and allow the determination of cavity quality factors.

We find a quality factor of $Q = 2900$ for our sample, corresponding to a cavity photon lifetime of $\tau_c = 0.95\,\mathrm{ps}$. The threshold for our system was found to be $0.74\,\mathrm{nJ}$, as previously reported [12]. Laser emission occurs perpendicular to the surface. The laser emission spectrum from our microcavity (Figure 1) shows two lines at $612.46\,\mathrm{nm}$ and $612.69\,\mathrm{nm}$, with linewidths of $\sim 45\,\mathrm{GHz}$ and $\sim 40\,\mathrm{GHz}$, respectively. This is of the order of the spectrometer resolution. Their spectral distance corresponds to $180.6\,\mathrm{GHz}$. Figure 1 also shows the polarization dependence of the emission. Both lines are perpendicularly polarized to each other. In contrast to previous reports of polarization splitting in microcavities [13, 14] we observe the splitting under a viewing angle of 0°. We ascribe the occurrence of the splitting to our evaporation conditions. The substrates are located off-axially over the evaporation source. Material from the source will then have oblique incidence on the samples. It has been shown that the dielectric materials SiO_2 and TiO_2 will then grow in oblique columnar structures during the evaporation process [15–17]. These columns in single dielectric layers have recently been resolved by scanning electron microscopy [18]. The layers then exhibit optical anisotropy in the deposited layers [16, 17, 19]. Using

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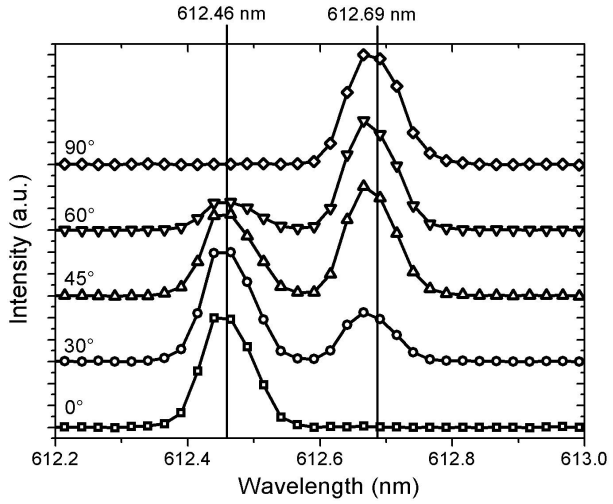

FIG. 1: Normalized microcavity laser emission spectra, dependent on polarizer angle. Two laser lines are observed, with either maximum at 90° and 0°.

transfer matrix methods, we can estimate the resulting birefringence in our DBR mirror as a difference of 10^{-3} between ordinary and extraordinary refractive index. This index difference is needed to obtain the observed polarization splitting of $\Delta\lambda = 0.23\,\mathrm{nm}$. Note that we still assume an optically isotropic organic cavity layer.

From the width of both lines and the length of the upconverted laser pulses, we can conclude that the pulse length of the laser emission is close to the Fourier limit of its spectrum. The superposition of two waves of different frequency will modulate the observed intensity pattern at a frequency corresponding to the frequency splitting. In our experiment, we observe this beating as a THz oscillation in the intensity of the up-conversion signal. The oscillation contrast depends on the amplitude ratio of both modes. Only the electric field in plane with the gate beam polarization is detected by up-conversion. Inserting a halfwave plate into the OVCSEL laser beam allows to rotate the respective mode polarization. Thus, the oscillation contrast can be tuned.

Conventional rate equation models have been applied to our system [12, 20, 21]. Solving the corresponding set of rate equations allows to describe the laser dynamics in detail. Following [12], we use

$$\frac{dN_a}{dt} = -k_a N_a(t) - k_{\mathrm{tr}} N_a(t) \quad (1)$$

$$\frac{dN}{dt} = k_{\mathrm{tr}} N_a(t) - k_e N(t) - c\sigma_{\mathrm{SE}} q(t) N(t) \quad (2)$$

$$\frac{dq}{dt} = \left(c\Gamma\sigma_{\mathrm{SE}} N(t) - k_{\mathrm{cav}}\right) q(t) + \Gamma k_{\mathrm{e,r}} \beta N(t), \quad (3)$$

where k_a is the decay rate of the absorbing state, N_a the absorbing state population, $N(t)$ the population of the emitting level, k_{tr} the Förster-transfer rate, k_e the emitting level decay rate, σ_{SE} the cross-section for stimulated emission, $q(t)$ the photon density, c the vacuum speed of light, Γ the optical confinement factor, k_{cav} the cavity photon decay rate, $k_{\mathrm{e,r}}$ the spontaneous emission rate of the emitting state and β the spontaneous emission factor. Fig. 2 shows a comparison of measurement data with a numerical solution of Equations 1. In the lower part, measurement and calculation for the dynamics of a single mode laser pulse are presented. Different signal rise and fall times are visible. In order to describe the single mode temporal behavior as precisely as possible, we only fit the absorbed pump power and use all other parameters from [12]: $k_a^{-1} = 16\,ns$, $N_{a,0} = 1.51 \times 10^{-18}\,cm^{-3}$, $k_{tr}^{-1} = 19\,ns$, $k_e^{-1} = 1.3\,ns$, $\sigma_{SE} = 1.3 \times 10^{-16}\,cm^2$, $\beta = 6 \times 10^{-4}$, $\Gamma = 0.31$, $k_{cav}^{-1} = 1.5\,ps$, and $k_{e,r}^{-1} = 5\,ns$. We note that the OVCSEL is operated far above threshold to optimize the up-conversion signal. The result is plotted as model for 0° polarization angle in Fig. 2. We will henceforth assume both modes to be of equal temporal behavior and independent in their respective plane of polarization.

From solving Eq. (1-3), we obtain the evolution of the photon population in time $q(t)$. The photon number is proportional to the square of the optical electric field strength. Thus, we write an electric field $E(t)$ of arbitrary amplitude A_j and phase ϕ_j as the result of the existence of both modes as

$$E(t) = \sum_{j=1}^{2} \sqrt{q(t)} A_j \cos\theta_j \mathrm{e}^{i(\omega_j t + \phi_j)}, \quad (4)$$

where ω_j is the angular frequency of the corresponding mode, θ_j is the angle that each mode electric field has with respect to the polarization plane of observation. This is tuned by the $\lambda/2$-plate in the beam. It is a means of tuning the oscillation contrast as well, and with the condition $\theta_1 = \theta_2 + 90^\circ$, we obtain maximum contrast at $\theta_1 = -\theta_2 = 45^\circ$. In our calculations, we will consider A_2 and ϕ_2 as fit parameters and set $A_1 = 1, \phi_1 = 0$. Line positions and splitting are taken from the emission spectrum in Fig. 1. Spectrum and fitting procedure yield as parameters $A_2 = 0.2$,$\phi_2 = 0$,$\hbar\omega_1 = 2024.37\,\mathrm{meV}(\lambda_1 = 612.46\,\mathrm{nm})$,$\hbar\omega_2 = 2023.62\,\mathrm{meV}(\lambda_2 = 612.69\,\mathrm{nm})$. The amplitude square of the resulting electric field is plotted as up-conversion intensity model calculation for an angle of 45° in Fig. 2. The corresponding measurement data also shows the expected visibility of interference effects over a time of $20\,\mathrm{ps}$. We note that oscillation frequency in experiment and model do not exactly correspond. This is the result of different spots on the sample being used for emission spectrum and up-conversion. The spectrum shows a splitting of $\Delta\nu = 180.6\,\mathrm{GHz}$ in contrast to the observed $\Delta\nu = 181.8\,\mathrm{GHz}$ in the up-conversion measurements. This is within reason if considering the wedge of our sample, which is a result of the evaporation conditions. The sample emission can be tuned over a range of approximately $20\,\mathrm{nm}$, thereby also tuning the splitting magnitude.

It is apparent that the oscillation contrast in the experiment is reduced, corresponding to an unequal contribution of the two modes ($\mathrm{A_2{:}A_1} = 0.2$) to the up-conversion signal.

Finally, the measurement data raises the question of mode-coupling. All data is obtained from averaging multiple pulses, but the observation of interference implies that the phases of both modes are reproducibly coupled. If the respective mode phases were arbitrary each time the sample is pumped, av-

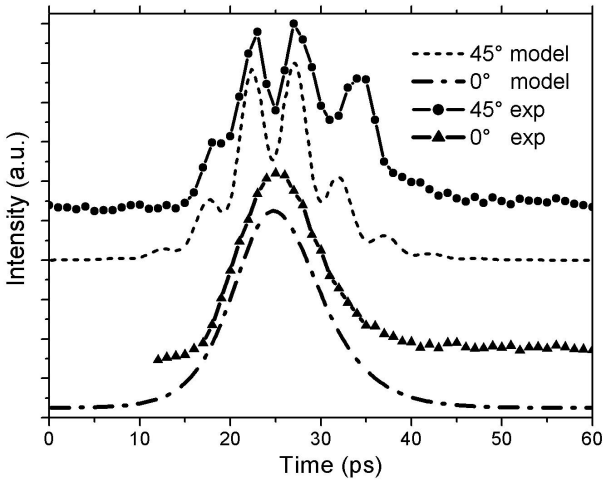

FIG. 2: Model vs. Experiment - From top: Measurement with two lines contributing to the up-conversion signal with clearly visible interference oscillation at 5.5 ps period time, model calculation for the same situation, measurement with one mode and corresponding model calculation by rate equation approach.

eraging the up-conversion signal over a multitude of pulses would yield the signal of only one mode. As possible explanation, we suggest a coupling mechanism that is based on the mirror anisotropy. In accordance with YARIV [22] and coupled-mode theory, anisotropic media consisting of mutually detuned anisotropic layers exert coupling of perpendicularly polarized modes by repeated rotation of the mode polarization. The two modes will exchange energy based on the dielectric material response and if a resonance phase-matching condition is met, a finite energy transfer occurs.

From fitting the interference pattern, we obtain a phase difference of $\Delta\phi = 0$ between both modes. We interpret this as indicator that both modes stem from the same initial event. One mode would start lasing and, while travelling through the anisotropic mirrors, transfer energy to the other mode, without loss of phase information. Afterwards both modes are amplified in the active medium and coupled out by the mirrors. First estimations of our system anisotropy and mode-coupling efficiency yield a finite exchange. Further experiments on this hypothesis are still in progress.

It is worthwhile to note that the unique features introduced to OVCELs by optical anisotropy in the mirrors make these structures promising and attractive candidates for the THz emitter devices based on photomixing techniques [23, 24]. The phase coupling of two emitting modes will in general lead to high monochromaticity of THz radiation since the spectral quality of radiation generated by photomixing is determined by the quality of the optical beat. Furthermore, the lasing at two modes from the same cavity leads to good spatial overlap of the two modes on the optical switch, allowing a potentially very compact THz generator.

In this letter, we have shown emission of two perpendicularly polarized laser modes from a single planar Fabry-Perot microcavity. Up-conversion experiments demonstrate possible interference at an oscillation frequency of $f = 0.18\,\mathrm{THz}$. Extended rate equations explain the observations accordingly. A phase-coupling of the two modes is observed and suggested to be the consequence of our anisotropic mirror structures. Possible applications may lie in optical driving configurations and compact bulk-devices for terahertz-modulated light. The authors gratefully acknowledge funding by the EU Commission through 5th framework Research Training Network 'HYTEC' (HPRN-CT-2002-00315) and by the Deutsche Forschungsgemeinschaft via the Leibniz award.

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