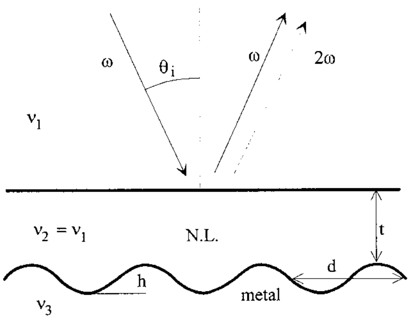
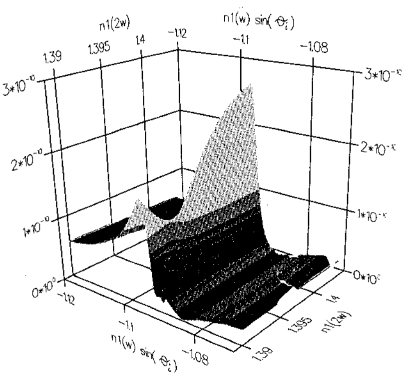
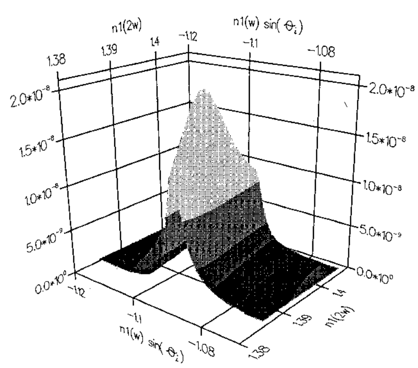



**CWL5 Fig. 1.** Schematical representation of corrugated nonlinear system. The nonlinear media extends from the grating surface to a plane at distance t from the groove top. The cladding and the nonlinear layer indices are equal. The metal is silver.



**CWL5 Fig. 2.** The amplitude of the nonlinear reflected signal for a system of Fig. 1. $n_1(\omega) = 1.534$, sinusoidal corrugation with $d = 4$ μm and $h = 15.4$ nm. TM polarized light with pump wavelength 1.06 μm. $\chi_{xxx} = \epsilon_0$, $\chi_{yyy} = 0$. $t = 0.01$ μm.



**CWL5 Fig. 3.** The same as in Fig. 2, except for $\chi_{xxx} = 0$, $\chi_{yyy} = \epsilon_0$.

second-harmonic reflectivity on these two parameters: angle of incidence (expressed in terms of $n_1(\omega)\sin(\theta_i)$) and $n_1(2\omega)$. The only non-zero component of the tensor of nonlinear susceptibility is $\chi_{xxx} = \epsilon_0$. The x-axis is along the grating surface and perpendicular to the grooves; y-axis is perpendicular to the grating plane. As already mentioned, the two plasmons (at ω and 2ω) have equal real parts of their propagation constants at $n_1(2\omega) = 1.3918$. Contrary from the intuitive expectations, the phase-matching then leads to a decrease of the second-harmonic reflectivity. This is due to existence of zeros in the close vicinity of the resonances.

The opposite effect is observed when $\chi_{yyy}$ is not equal to zero (Fig. 3): there is an enhancement of the signal with maximum value almost two orders of magnitude larger than in Fig. 2. The reason for the different behaviour is that the plasmon surface wave has a y-component of the electric field much larger than its x-component, so that second-harmonic generation through $\chi_{xxx}$ is much weaker than through $\chi_{yyy}$.

There are two direct conclusions from the comparison between Fig. 2 and 3.

(1) Phase-matching does not necessarily lead to enhancement of the second-harmonic generation.

(2) Depending on the geometry of the nonlinear media (non-zero components of the tensor of nonlinear susceptibility), the second-harmonic generation can vary significantly.

**On leave from the Institute of Solid State Physics, Bulgarian Academy of Sciences, Sofia, Bulgaria*

**CWL6 1815**

## Cerenkov second-harmonic radiation from organic calix[4]arenes thin films

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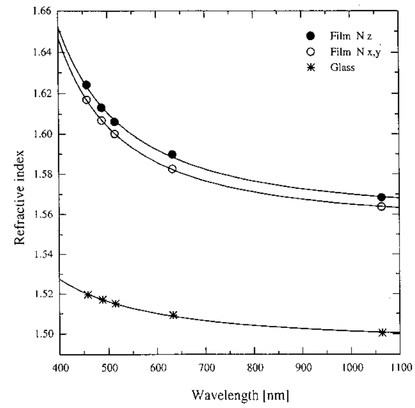
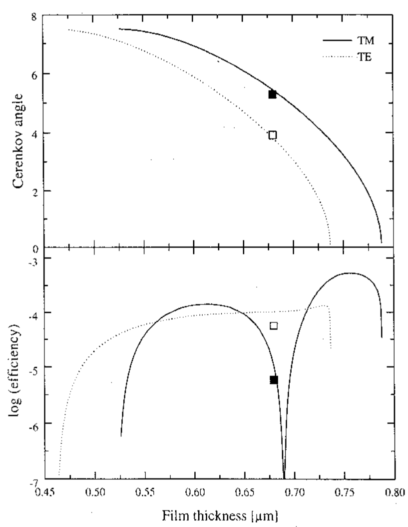
Nitro-calix[4]arenes are a novel type of organic molecules. In a thin film the material shows high nonlinear optical properties combined with good temporal stability at room temperature.[1,2] Furthermore, a nitro-calix[4]arenes film is transparent down to 400 nm, which makes it suitable for frequency doubling of laser-diodes.

The calix[4]arenes can easily be made into a thin waveguiding film by spin-coating it onto a glass substrate. In order to have films sufficiently thick for waveguiding a mixture of the calix[4]arenes with a polymer (PPMA) is used (78 wt. % calix[4]arenes). The thickness of the film was chosen such that the criterion for Cerenkov second harmonic (SH) was met, meaning that the effective refractive index of the guided fundamental beam is below the index of the glass for the SH-wavelength.

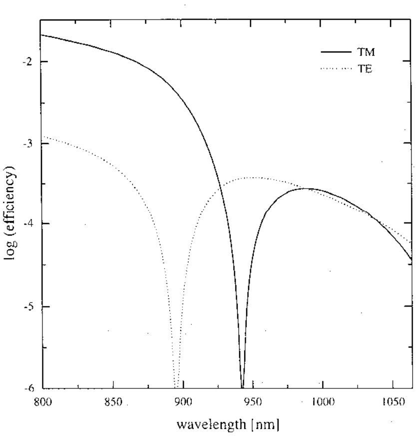
The calix[4]arenes in the film were oriented by the corona poling technique. The film had only a few non-zero d-values: $d_{33}$ and $d_{31}$ ($=d_{32}=d_{24}=d_{15}$). They were measured with a makerfringe experiment, comparing the SH with that from a quartz reference. The values were 6.3 and 1.9 pm/V, respectively, and were measured after the initial decay of 35% in 10 days. (After this decay previous measurements showed stable d-values for at least one year.) The refractive indices, birefringence, and dispersion were measured with the prism coupling technique and are shown in Fig. 1. The thickness of the film was 680 ± 20 nm.




**CWL6 Fig. 1.** Refractive indices and birefringence of the 78 wt. % nitro-calix[4]arenes—PPMA thin film after corona poling and of the glass substrate.




**CWL6 Fig. 2.** Measured and theoretically calculated Cerenkov angle and second-harmonic efficiency from the corona-poled thin film. λ = 1064 nm.




**CWL6 Fig. 3.** Cerenkov second-harmonic efficiency predicted by coupled mode theory vs fundamental wavelength. Film thickness is 550 nm.

For the Cerenkov experiment a Nd: Yag laser beam (1064 nm, 10 ns, 10 Hz) was coupled into the waveguide for both TE and TM polarization. The peak power inside the waveguide, estimating a coupling efficiency of 5%, was approximately 5 kW. The beamwidth was 1 mm and the interaction length 5 mm. The generated second-harmonic and its angle of radiation into the glass substrate (Cerenkov angle) were measured. The values are shown in Fig. 2 and appear to be in good agreement with the theoretical values, which were calculated vs film thickness by means of a coupled mode theory using the measured linear and nonlinear optical properties. Unfortunately the conversion for the TM fundamental beam was almost in a minimum of the curve. Theory predicts an efficiency of $5 - 10^{-4}$ for a 750-nm-thick film. The efficiency can be even further increased by changing the fundamental beam to shorter wavelengths as is shown in Fig. 3. (The enhancement of the d-values for shorter wavelengths is taken into account: $d_{33}$ up to 25 pm/V for 800 nm). An efficiency of $2 \times 10^{-2}$ is predicted for 800-nm fundamental wavelength.

In conclusion, the nitro-calix[4]arenes are very well suitable for use in second-harmonic generative waveguides due to their high and stable d-values and ease of fabrication into thin films. The generated Cerenkov SH is in good agreement with theory and can be enhanced by changing the thickness of the film and/or the pump wavelength.

The authors thank E. Kelderman and D. N. Reinhoudt for making available the calix[4]arenes.

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## CWM 1700
Room A

## Nuclear and Free-Electron Lasers

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### CWM1 1700

### Fast manipulation of the gain medium of an infrared free electron laser

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A free electron laser is similar to regular lasers in the sense that a light pulse is amplified on multiple passes through an optical cavity. Although the pump and gain processes are completely different, manipulation of the stored field is possible using similar tricks as in regular lasers, for instance phase locking by means of an intracavity interferometer.[1,2] In addition, however, a free electron laser has the unique feature that also the properties of the gain medium (a beam of relativistic electrons) can be manipulated on a time scale essentially down to the cavity roundtrip time.

Two examples of rapid manipulation of the laser medium, as investigated with the free electron laser for infrared experiments Felix, will be presented. The first involves making ultrafast wavelength scans by ramping the electron energy during each "macropulse" (i.e., the on-time of the accelerator). In exploratory experiments, a frequency chirp of 2% in 4.5 μs was obtained. This technique conceivably permits to maintain resonance in multiple-photon dissociation of molecules having an anharmonic vibrational ladder, thus maximizing the dissociation efficiency.

The second example is related to the fact that the optical group velocity increases as the stored field approaches saturation. This permits variation of the gain per pass, the saturated power, and the optical pulse length, by straightforward adjustment of the cavity length. We have found that a suitable manipulation of the electron bunch repetition frequency during each 10-μs macropulse results in simultaneous operation at the highest gain per pass and the highest saturated power. In addition, we will present indications that this "dynamic cavity desynchronisation" also eliminates the limit-cycle oscillation of the laser power which was reported earlier for a fixed cavity desynchronism.[3]

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### CWM2 1715

### Amplification on uranium lines in a discharge in contact with an electrolytic solution

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The work presents results of spectroscopic measurements which are indicative of amplification at the wavelength of 2.6 μm in a gaseous medium containing uranium ions.

The generation of the uranium ions in the discharge is accomplished by the so-called "explosive electrolysis." The phenomenon is produced by discharging a capacitor on a cell containing an electrolytic solution. The explosive electrolysis disrupts the flow of current through the electrolyte and generates metal ionic species in the neighbourhood of the surface of the liquid. Under conditions corresponding to the generation of the surface discharge, the current density in the electrolyte layer surrounding the small dimension electrode in contact with the surface is in excess of $10^3$ A/mm$^2$. The energy deposited in unit volume, proportional to $j^2$, will be enough for superheating of this liquid. Consequently, a violent vaporization of the solution surrounding the electrode will take place. This "explosive electrolysis" generates a great number of ionic species in the neighbourhood of the surface of the solution near the electrode. The potential difference between the electrode and the surface sets these ions into oriented motion, thus initiating the surface discharge. Spatial confinement of the discharge is in a capillary with a square 1-mm$^2$ cross-section. With 10 kV on the 200-nF capacitor, the discharge in atmospheric air is up to 5 cm long. The length can be doubled if two identical capacitors are discharged simultaneously on identical cells. The discharge has characteristics of discharges at high pressure in solid capillary channels[1] as well as characteristics of discharges on liquid surfaces.[2]

The light coming out of the system in the longitudinal direction is monitored via a monochromator. We measure the intensity of the light of the particular wavelength in three situations: separately for each of the two discharges and then with simultaneous discharges.

The results show that in the latter case the intensity at 2.6 μm is in excess of the sum of the two intensities generated with single discharges.

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### CWM3 (Invited) 1730

### Compact free electron lasers in the far infrared-millimetre region

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Conventional free electron lasers (FELs) suffer from some drawbacks related to the large size of most electron beam (e-beam) accelerators that implies high costs, system complexity, and shielding requirements. However, in the far infrared (FIR) and millimetre (mm) wave region the FEL can meet the demand of compactness. For example, the low e-beam energies required (<10 MeV) allow the use of small size accelerators like radio-frequency (rf) linacs or microtrons. The increased performance in terms of gain at wavelengths in the FIR and mm also requires a shorter length of the interaction region, thus allowing the use of short period undulators and a small number of periods which results in a better efficiency. Moreover, the presence of a dispersive element inside the undulator, like a waveguide needed for operation at

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