HIGHLIGHTS
LAMBDA PHYSIK

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500 W XeCl – "Magic" Mark Nearly Attained

A team headed by Peter Oesterlin recently approached this goal (see Fig. 1) in an experimental set-up at Lambda Physik. The multi-hundred Watt project was supported by the German Ministry for Research and Technology. The laser is seen in Fig. 2. The laser channel and the beam output (4 cm x 5 cm) are at the small flange near the experimentalist's head. The tank is in fact a compact wind tunnel driven by two radial fans. 4 thyratrons mounted above the tank are connected in parallel to provide energy switching. Preionization is performed by UV sparks mounted behind a mesh electrode.
The set-up is an experimental one. It will also be used in the future for applied research within the Eureka EU 205 project, the European program for the investigation of multi-kilowatt excimer lasers.

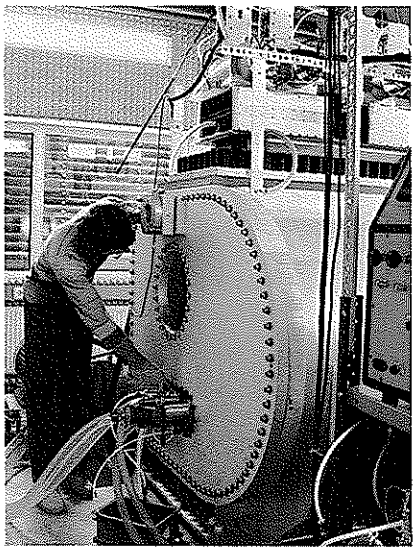

Fig. 2 Set-up of the 500 W laser

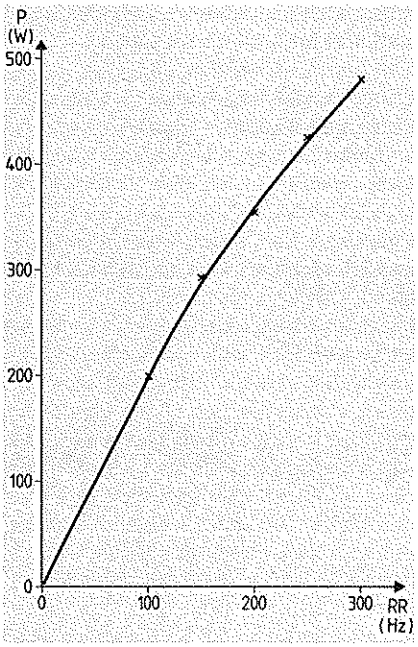

Fig. 1 Power versus repetition rate of the 500 W XeCl excimer laser

Polarized Excimer Laser Light

Frequency-narrowed excimer lasers (as EMG 150/160) emit polarized light. The broadband excimer lasers emit unpolarized light in their bandwidths given by the active excimer molecules (natural bandwidth).
Sometimes there are requests to have polarized light even for emission in natural bandwidth. Such applications are:
* Experiments which need a definite photon angular momentum as in nonlinear optics, frequency conversion and stimulated scattering. Introducing an additional quarter-wave plate leads to circular polarization.
* Molecular beam experiments or experiments in low-pressure gas cells needing aligned or oriented molecular excitation, e.g. by absorbing excimer laser photons at spectral positions of incidental coincidence.
* Low beam losses if only a small portion of the beam has to be coupled out by a tilted quartz plate for beam monitoring. The beam can be passed loss-free via Brewster cells.
The intracavity polarizing optics, shown in Fig. 1, has now been introduced for LPX lasers. Since it has been realized by polarization-dependent dielectric coating, it was tested firstly for one emission band, 308 nm; however, it is possible to reach similar results using coatings for 351 and 248 nm. For 193 nm, suitable coatings are not yet available.
The polarizer, which works for the 45° mounting, can be rotated around the laser beam axis to allow polarization plane rotation without beam displacement.
The degree of polarization was measured to be >100 for 130 mJ. There is no dramatic loss in total beam energy by inserting the polarizer: in the cavity configuration of Fig. 1 pulse energy rose to 140 mJ taking out the polarizer. Mounting the outcoupling mirror directly on the discharge unit (standard), energy rose to 160 mJ.
The polarizing optics will be available in summer 1988.

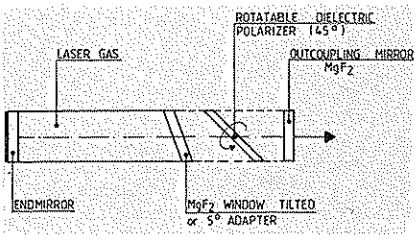

Fig. 1 Intracavity polarizing optics

New Results from the Lambda Physik Application Lab:

Excimer Laser Beam Coupling into Quartz Fibers

Laser beam-optical fiber coupling is performed in many laboratories. Especially excimer laser-fiber coupling contains pitfalls for the experimentalist. The consequence usually is a reduction in the pulse energy which can be reliably transmitted. In this article we communicate two aspects which may help to improve the performance of a laser beam-fiber interface.

Filling the fiber phase volume completely

The internal beam divergence of a standard excimer laser such as the LPX 120i is about 5 mrad (FWHM) horizontally and 1 mrad vertically. These values are far from the diffraction limit. However, even this is not sufficient for beam-fiber coupling.

The maximum amount of energy which can be reliably transmitted is limited by damage mechanisms. The most important is volume damage, which occurs when the local intensity in the fiber becomes too high, as a result of internal focusing.

Coupling a laser beam of low divergence into a fiber favours local intensity enhancement. This is immediately realized when one takes into account the fact that fibers are waveguides with well-defined wave propagation modes. The fibers used for power tranmission are of the multimode type, and accept a wide cone of radiation which corresponds, in the interior of the fiber, to higher order transverse propagation modes. Therefore, in order to transmit as much pulse energy as possible without the risk of damage, it is necessary to fill all propagation modes of the fiber. This can be achieved by filling the acceptance cone of the fiber at each point on the input face.

Liouville's theorem, which is valid for any beam guiding and beam shaping, states that the phase volume is preserved. In broad terms, this means that imaging to smaller dimensions leads to higher divergence and vice versa. Hence, suitable beam divergence on the fiber face (and corresponding filling of transverse modes in the fiber) can be achieved by imaging an aperture filled by the laser radiation onto the fiber's face at a reduced scale.

As a demonstration, an experiment was set up using a short (8 mm) quartz fiber with 600 μm core diameter, a variable aperture illuminated by the laser beam (308 nm), and a demagnification optics which used a field lens to avoid overfilling of the acceptance cone near the edge of the fiber face. In the tests, the aperture was imaged at the reduction ratios of 3.5, 8, and 11 onto the full entrance face of the fiber which meant that the aperture had to be adjusted in diameter according to the reduction of the imaging ratio. The beam illuminating the aperture was appropriately attenuated to keep the laser pulse energy transmitted through the aperture constant.

Fig. 1 a to 1 c show photographs of the laser beam shape obtained behind the fiber for the different imaging conditions.

For the demagnification ratios of 3.5, 8, and 11, a clear change from a locally inhomogeneous and filamentary pattern towards a more homogeneous one is observed. Consequently, the beam energy is better distributed in the waveguiding volume of the fiber. This result is independent of the applied excimer laser wavelength.

In a separate set-up, the fiber length was increased. It was found that longer fibers favour homogenization so that no significant differences were measured for different demagnification ratios. These observations are consistent with the experience that fibers usually break within the first few centimeters when energy is too high. Clearly, using a fiber of typical laboratory length and trying different imaging conditions, no significant differences would be observable in the beam shape at the output. Thus, when trying to find the optimum energy transport by a given fiber, a suitable procedure is to begin with short pieces according to Fig. 1 a-c.

Longer pulses are easier to transmit

Excimer lasers with longer pulse duration have been developed (e.g. LPX 605i: > 50 ns (FWHM), LPX 610i: 250 ns (FWHM), the successor to the EMG 602 described on p. 4 of *Highlights 10*). We report on tests at 308 nm which were designed to determine the reliable transmission of pulses through a polymer-cladded quartz fiber of 600 μm core diameter.

Measurements were made to determine how much energy can reliably transmitted in relation to pulse length. Fig. 2 shows a logarithmic plot of that quantity. A straight line fitted to the data by eye has a slope of 1/2, indicating that the pulse energy transmittable increases with the square root of the pulse length.

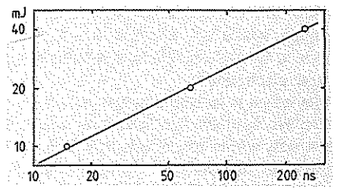

Fig 2 Reliably transmittable pulse energy at 308 nm for three different pulse durations

This result is consistent with that previously reported by Taylor et al. (R.S. Taylor, K.E. Leopold, S. Mihailov, R.K. Brimacombe, Opt. Commun. **63** (1987) p. 26) who also gave an explanation (CLEO 88) which, however, is still under discussion.

More details of these experiments will be published in the August issue of *Laser und Optoelektronik* (U. Sowada, H. J. Kahlert, D. Basting, in German).

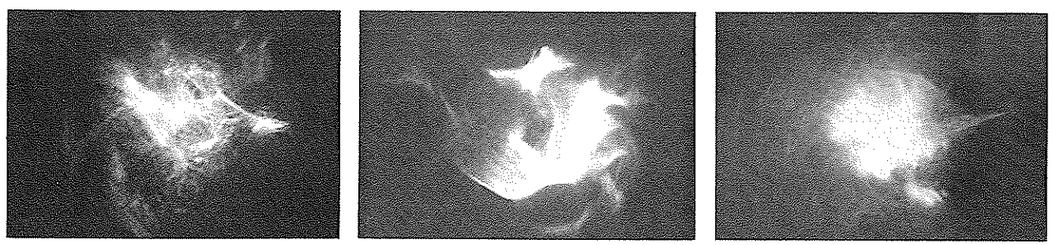

Fig. 1 Output of a short (8 mm) quartz fiber (∅ 600 μm) filled with excimer laser radiation at three different divergences: (a) low, (b) medium, (c) high divergence

Spectral Purity of a FL 3002 Dye Laser Measured with a High-Resolution Monochromator

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Introduction

The pulsed tunable dye laser has proven to be a versatile tool for the spectroscopist. In general, pulsed dye lasers do not emit exclusively at the selected frequency but show a spectrally broad background of spontaneous emission which is amplified by the high gain medium (ASE). Two kinds of ASE can be distinguished in an excimer laser-pumped dye laser: a first part is released immediately after excitation, a second part coincides with the build-up of optical feedback in the oscillator cavity. Although, due to high gain, most of the laser energy is concentrated in a narrow laser line, a low-power ASE background is always present.

The first part of the ASE can be easily suppressed in a dye laser, which consists of an oscillator and subsequent amplifiers, by an appropriate time delay in the optical excitation of the different stages. In order to suppress the second part, Lambda Physik introduced earlier the "Lambdapure" scheme for the FL 2001/FL 2002 dye lasers, in which the grating is used as an additional filter for the oscillator output.

Recently, Nogar and Keller [1] measured the resonant ionization mass spectrum (RIMS) of lutetium atoms using an excimer-pumped dye laser (Lambda Physik, FL 2002). The authors attributed the intense satellite lines (which appeared at 3 cm^{-1} intervals and covered a range of 15 cm^{-1} from the central peak) to weak sidebands in the output of the dye laser. The spectrum of the dye laser output was consistent with RIMS spectra showing sidebands with an intensity $< 0{,}5$ % of the central laser peak. In a technical note, Klauminzer [2] discussed the current Lambda Physik oscillator design and claimed that this design can produce a considerable ASE background in the central part of the tuning curve. Since in our experiments the spectral purity of the dye laser is of great importance, we decided to carry out more detailed studies on ASE background using the latest dye laser model from Lambda Physik, an FL 3002 in standard configuration.

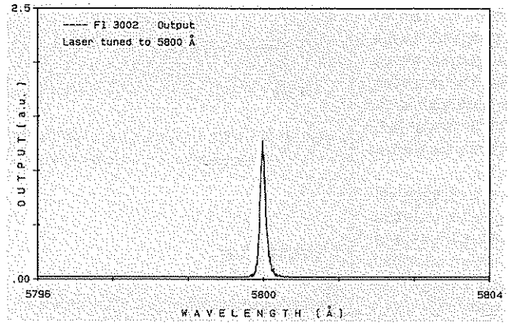

Fig. 2a FL 3002 dye laser line profile. The entire spectrum was recorded using aT=0.005 neutral density filter

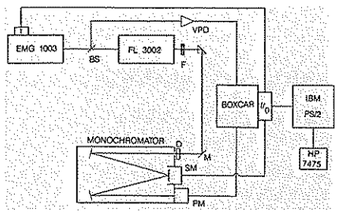

Fig. 1 Set-up for measurements of the dye laser line profile by the high-resolution spectrometer. T: external trigger of excimer laser, VPD vacuum photodiode (Hamamatsu), F: neutral density filter, D: diffuser, BS: beam splitter, M: mirror, SM: stepper motor, PM: photomultiplier

Experimental Methods

The experimental set-up used in these studies is shown in Fig. 1. The FL 3002, operating with an oscillator, pre- and main amplifier was pumped by the excimer laser (EMG 1003i, Lambda Physik). A high-resolution grating monochromator (1,5 m spectrometer Jobin Yvon, model THR) was used to analyse the dye laser output spectrum. The spectral resolution of this monochromator was about 0.01 Å at a slit width of 5 μm. The dye pulses were attenuated by neutral density filters and diffused by scattering. The output of the monochromator was recorded with a photomultiplier (1P28A, RCA). The photomultiplier signals were measured with the boxcar (Stanford Research System, Model SR 250) operating in the last-sample mode.

An IBM PS/2 computer was connected via an I/O interface with the boxcar, the excimer laser and the stepper motor of the monochromator. The computer was used to control the entire experiment and to process the data from the boxcar. The stepper motor and the excimer laser were simultaneously triggered. At a fixed position of the monochromator the desired number of pulses was recorded by the boxcar and averaged by the computer. The spectra presented in the next section were obtained by averaging 10 pulses and tuning the monochromator with a stepwidth of 0.08 Å. The width of the sample gate was set to 300 ns, and the laser pulse rate to 16 Hz. Rhodamine 6G in methanol was used in both the oscillator/preamplifier and amplifier dye cuvettes. All measurements were carried out at excimer laser pump energy of 100 mJ, which produced a dye laser pulse energy of about 15 mJ.

Result and Discussion

Fig. 2a shows the dye laser output recorded after an adjustment of the FL 3002 at the maximum of the dye tuning curve ($\lambda = 580$ nm), following a standard procedure. The laser beam was very strongly attenuated by the neutral density filters to avoid saturating the photomultiplier as the monochromator was scanned across the laser peak. In this recording, no satellite lines or structu-

res at the wings of the laser line were observed. The situation was quite different when the detection sensitivity was increased as shown in Fig. 2b. The left and right parts of the laser line were amplified by a factor of 200. This was accomplished by removing a T = 0.005 neutral density filter. At this intensity, conspicuous structures appeared at the wings of the laser line spectrum. The dotted line in Fig. 2b is a Lorentzian curve with the same halfwidth and peak height as the observed laser line. On the left side of the spectrum, the laser intensities show mostly higher values than the calculated Lorentzian curve. The wavelength period of the observed oscillations was, however, not the same as that reported by Nogar et al.[1]. The maximum height of the structures observed in the wings was less than 0.25 % of the laser peak height. This explains why the structures do not appear in Fig.2a.

In experiments where spectra of highly different peak heights have to be recorded, care has to be taken to avoid spurious lines. Hence, we decided to put more effort into the adjustment of the laser. The dye laser profile (λ = 580 nm) recorded after a very precise adjustment of the FL 3002, especially of the main amplifier stage is shown in Fig. 3. The spectral purity of the dye laser was significantly improved; both left and right wings of the laser line exhibited no structures. The Lorentzian curve was plotted with the measured linewidth of 0.07 Å ($\approx$ 0.2 cm^{-1}). The experimental values of the laser intensity at the wings of the laser line are well below the calculated Lorentzian curve. Both curves merge 4 Å away from the laser line. We calculated that 99% of the dye laser energy is concentrated in a range of 5 Å · $\Delta\lambda$ (5 times the linewidth) resulting in 0.35 Å at λ = 5800 Å.

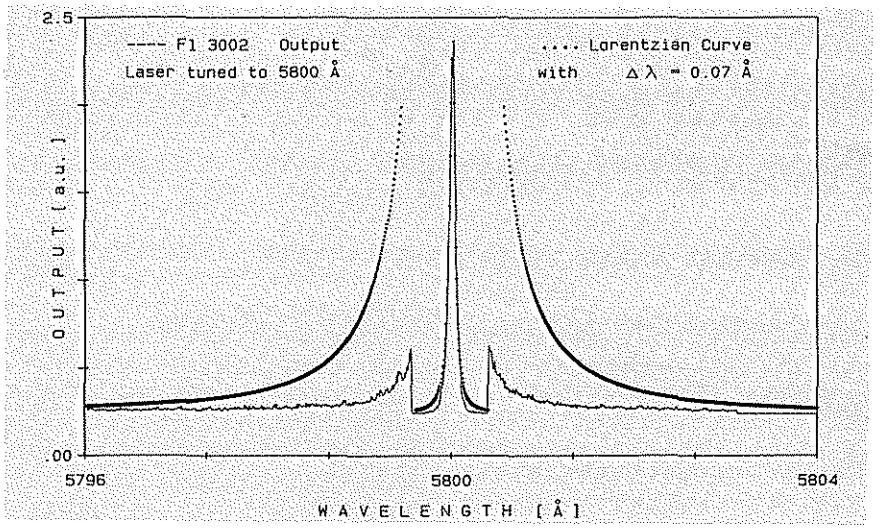

Fig. 3 FL 3002 dye laser line profile. The data was recorded as in Fig. 2b. A spectrum of this quality can only be achieved by a very careful adjustment of the FL 3002

Conclusion

We have demonstrated that in the center of the dye gain curve (see Fig. 3) there is no evidence of any ASE "hump" background when the system is very well adjusted. Even after coarse adjustment of the laser a "hump" ASE was not observed (see Fig. 2b). However, the laser output spectrum in this case revealed some similarities to the spectrum observed by Nogar et al. [1]. In Ref. [2] it was speculated that insertion of a highly reflective mirror near the dye cell causes a background hump. Our experiments have shown that this conclusion is not correct, but results from an oversimplified model. After optical feedback begins, the ASE and laser emission cannot be evaluated separately.

In conclusion, a very careful and precise adjustment of the FL 3002 is recommended when an extremely high spectral purity is required.

References

[1] N.S. Nogar and R.A. Keller, Analytical Chemistry 1985, **57**, 2992

[2] G.K. Klauminzer, "Aspects of pulsed dye laser design" in Technical Note No. 8 (1987) edited by Questek Inc., Billerica, MA, USA

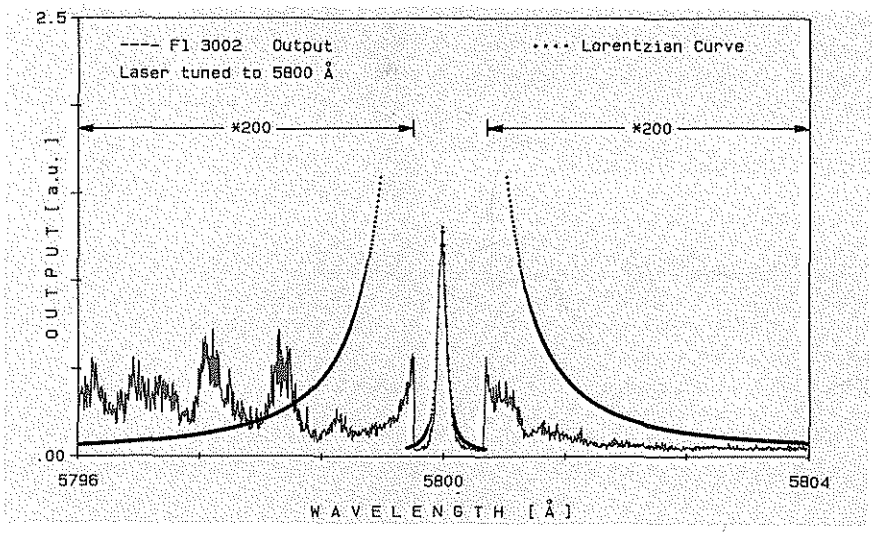

Fig. 2b FL 3002 dye laser spectrum, recorded without T=0.005 neutral density filter, except around the laser peak. The dotted line is a Lorentzian curve plotted using the measured laser linewidth

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HIGHLIGHTS
LAMBDA PHYSIK

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Excimer Lasers in High-T_c Superconductor Research

Only three years ago the scientific world was excited by the discovery of high-T_c superconductors. In the mean time, an estimated 50 000 scientists and engineers work in this field so that it is difficult to get an overview on the state of the art. Excimer lasers have become important tools in the production of superconducting thin films and in the analysis of the deposition process. This article, which is not necessarily comprehensive, is an attempt to report on recent progress in this special field.

Introduction

"At the extreme forefront of research in superconductivity is the empirical search for new materials", Karl Alexander Müller and Johannes Georg Bednorz began their Nobel Prize-honoured paper in 1986 [1], referring to M. Tinkham et al., who expressed the needs at that time at a workshop at Copper Mountain, Colorado, in August of 1983.

The "copper mountain" in superconductivity began to rise with Müller's and Bednorz' publication, and has apparently not yet reached its summit: *Fig. 1* [2] shows what happened with respect to the transition temperature of superconductors since 1911, when Kamerlingh-Onnes discovered superconductivity in mercury. Indeed, the compound Ba-La-Cu-O opened a new era; Cu-O was soon recognized to form a non-perovskite type phase of that compound responsible for superconductivity (as firstly communicated in a note added in proof in [1]).

Within a few months after the publication of [1], the transition temperature rose to 92 K, as achieved at the University of Houston (C.W. Chu and co-workers, 1987) by replacing lanthanum with the smaller yttrium ion. The importance of this result: the transition temperature was then raised to above 77 K, which is the temperature of liquid nitrogen at atmospheric pressure.

$YBa_2Cu_3O_{7-x}$ became the high-T_c compound mainly investigated until today, frequently called "one-two-three" for the stoichiometry of the first three atoms. "x" stands for a variable deficiency of oxygen. In other laboratories in Japan and the US, bismuth and thallium cuprates have been found meanwhile to exhibit even higher transition temperatures, and, following the opinion of experts, a definite end of this development is not yet to be seen.

The role of lasers in the game is twofold. Laser evaporation is very effective, very reliable, and fairly simple in the deposition of thin films of said superconductors. Furthermore, laser induced fluorescence is a convenient means to analyze molecular composition and details of the deposition kinetics.

Why thin films?

It turned out soon that high-T_c superconductivity in the bulk material suffered from low current density: if the current in a sample is increased, a breakdown of superconductivity occurs. Furthermore, since the superconductivity effect is strongly anisotropic, in a poly-crystalline bulk an unfavorable averaging of the superconducting properties of all crystal orientations occurs. On the other hand, once crystal growth is epitaxial, the supercurrent density can be increased over that of poly-crystalline material. *Fig. 2* [3] shows schematically the material deposited in the favorable c-orientation on a $SrTiO_3$

750 W XeCl reached at Lambda Physik R & D

A few weeks ago* 750 W output were reached with the XeCl laser which had already set landmarks earlier (cf. HL 11, June 1988). Fig. 1 shows the new curve 'average power versus repetition rate' now valid. Comparing it with that in HL 11 it is seen that the 50% higher average power is achieved by an increase of the repetition rate up to 500 Hz (300 Hz earlier). The repetition rate improvement was brought about by adding electric power. However, to take advantage of the higher electric power installed, UV preionization of the discharge had to be improved. The concept used earlier, UV emitting spark electrodes behind the mesh electrode, was given up in favor of a proprietary improved UV preionization. Dr. Elmar Müller-Horsche, head of the research team,

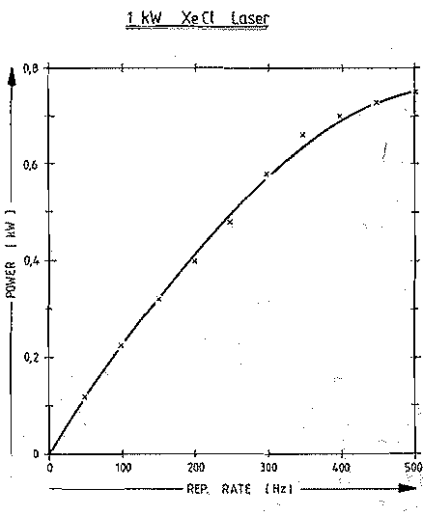

Fig. 1: Average power versus repetition rate

* First reported at the Eureka EU 205 meeting, October 9, 1989.

foresees further increase of average power of this type of laser using X-ray preionization. To reach 1 kW is the goal of the second phase of the Eureka EU 205 project which is to start in 1990.
Clearly, although under laboratory conditions, realizing average power in

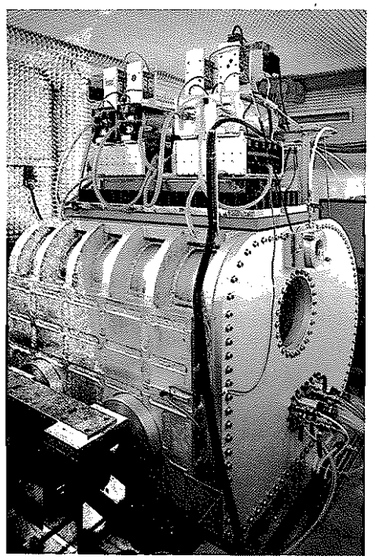

Fig. 2: 750 W XeCl laser at Lambda Physik

this order of magnitude provides a lot of experience for current product development, especially as the materials employed undergo rigorous testing. In addition, UV preionization, which is less costly than the X-ray technique, shows remarkable potential for medium to high-power lasers. Fig. 2 shows the laser in a view somewhat different from that in HL 8.

... Superconductors

crystal, supercurrents supposedly flowing in the Cu-O planes which extend parallel to the substrate.
In addition, from a practical point of view, thin films are the usual approach to designing advanced electronic circuits. This holds even more so as the production of superconducting wires raises problems, since the material is very brittle.
However, thin films need convenient substrates. To achieve epitaxial films, ideally the substrate should show a crystalline lattice favorable for the film

critical temperature, T_c	mean temperature at the transition from normal to superconductivity. The resistivity drop begins with the onset and ends with R=0 showing a narrow transition width.
critical current density	current density sufficiently high to destroy superconductivity
as-deposited	film properties (e.g. superconductivity) usable without any post-deposition processing
in situ	meaning here the same as 'as-deposited'
annealing	heating followed by slow cooling so that the crystal structure is modified
epitaxial	crystalline growth of material deposited on a substrate crystal
sputtering	sputtering of material yielding the atoms of the sputtered compound using the impact of energetic ions on the material; magnetron sputtering means the generation of sputtering ions in a magnetron-driven discharge.
isentropic expansion	is an adiabatic expansion keeping entropy constant. Practically, for the expansion of a molecular beam into high vacuum, this means rapid cooling by removal of the energy contained in the transverse, rotational and vibrational degrees of freedom of the beam molecules; simultaneously, an acceleration of the molecules up to supersonic speed (Mach numbers >1) occurs with drastic narrowing of the velocity distribution.

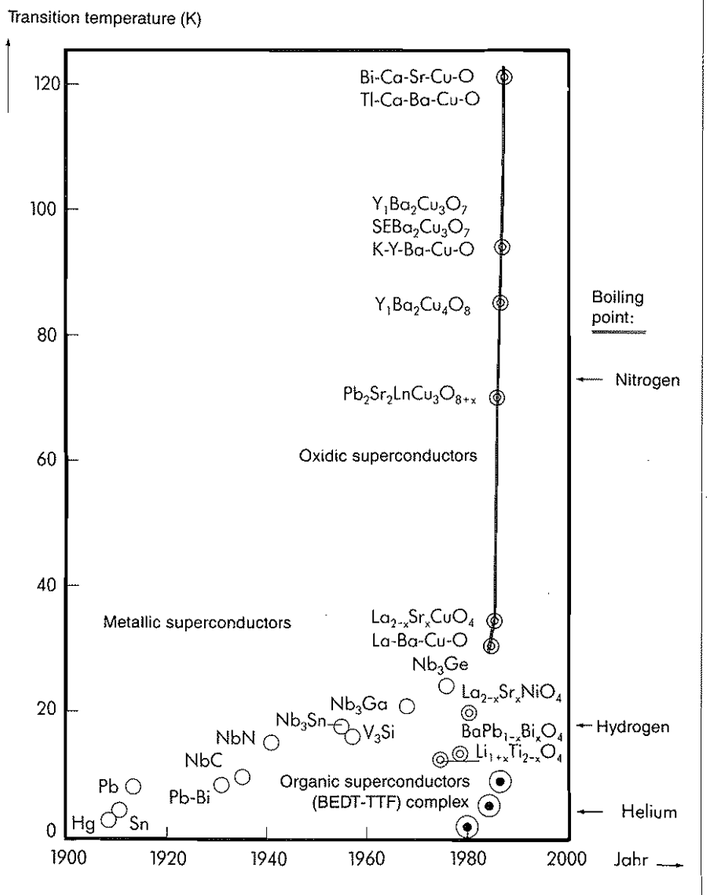

Fig. 1 History of superconductivity: the transition temperature is plotted versus time. On the right scale the boiling points of helium, hydrogen, and nitrogen are marked [2].

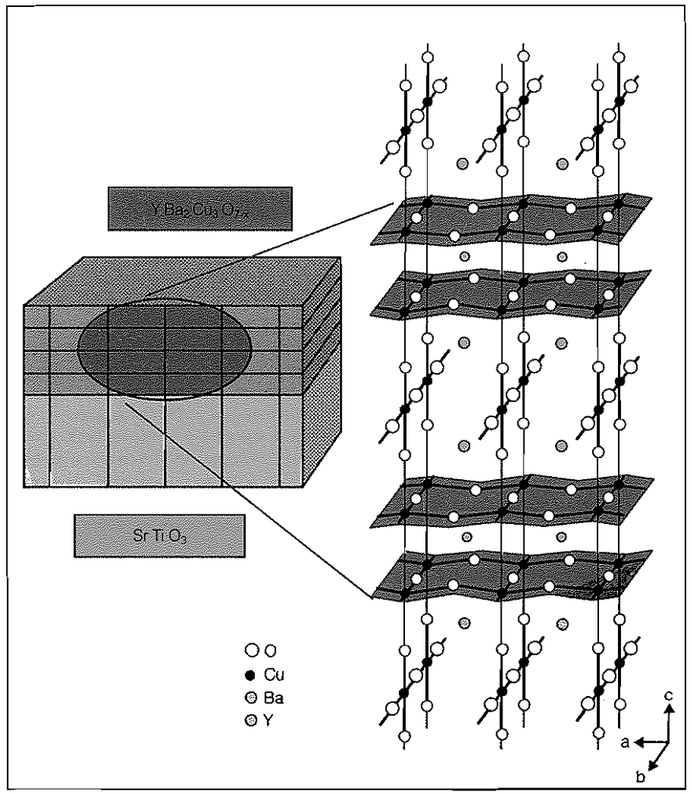

Fig. 2 Scheme of a $YBa_2Cu_3O_{7-x}$ film deposited with the c–axis normal to the substrate surface [3]

atoms to arrange. This is the reason that $SrTiO_3$ is frequently chosen as the substrate for experimental investigation. However, for any practical application in electronics circuits, Si would be much more convenient as the substrate.

Depositing thin films

Producing thin films of multi–element composition can be performed by simultaneous vapor or molecular beam deposition using e.g. three different ovens in an oxygen atmosphere at low pressure. Also metal–organic vapor deposition is used. These procedures are difficult to control and costly.
Therefore, the superconducting compounds are mixed in advance in an almost correct stoichiometric ratio. Targets prepared in this way are then sputtered onto a substrate. Also, flash-evaporation of the premixed material was tried. The difficulty is maintaining the stoichiometric ratio during evaporation.
Another well-proven deposition technique is magnetron sputtering.
The aim of any deposition technique for high–T_c films is to obtain the required stoichiometry in the film including the oxygen content, to achieve almost unidirectional growth in the required crystalline structure, and to avoid impurities. Frequently, these goals cannot be achieved simultaneously so that an annealing process of the as–deposited film in oxygen may be necessary.

Depositing high-T_c layers: annealing or not?

Excimer laser ablation was firstly used at Bellcore by Venkatesan et al., as *Highlights* reported in its October 1987 issue (HL 7). Generally, pulsed laser radiation can be applied in a well-controllable way to the target. Pulsed UV radiation is very effective in the ablation of non-volatile and ceramic material. As a consequence, in the film, almost the same stoichiometry as in the bulk can be ascertained. However, as reported in HL 7,1987, in these early experiments annealing in oxygen at 900 °C and cooling down over several hours was still necessary. Heating of the substrate during deposition was required providing thermal energy for the deposited atoms and molecules to migrate on the surface and to arrange themselves in the energetically favored lattice. Post-deposition annealing is not only time-consuming for any practical application. Also, high-temperature annealing favors interdiffusion from the target to the superconductor, thus spoiling the superconducting properties of the film. Due to a careful control of the deposition process this situation has now changed dramatically, as will be described below.
A recent magnetron sputtering experiment should be mentioned [4]: it allowed a moderate temperature (645 °C) at the substrate, and did not require annealing, but a careful cooling-down at a rate of 30 K/min. Critical current density was 2×10^5 A/cm² at 77 K which is not as high as that reached by the laser techniques.

UV wavelengths are favorable

Infrared lasers have also been investigated for laser evaporation. Recently, films close to the stoichiometry of the target were obtained [5] using a long–pulse (about 1 ms) 50 J Nd:glass laser, providing a fast deposition rate (100 nm/pulse); however, 850 °C-annealing was still necessary. Deposition by frequency-doubled short-pulse (10 ns) Nd:YAG laser radiation, 532 nm, required 725 °C at the substrate [6]; the same group reports that frequency-tripled radiation (355 nm) produces much smoother films [6]. A similar result is reported in [12].
Obviously, Nd:YAG laser users recognized that short wavelengths are more favorable. In principle, this would not be surprising, considering the background of excellent ceramics material processing capabilities demonstrated in many laboratories for excimer lasers. However, very recently, this trend has been questioned again by a paper published by the same IBM group [7], who reports that in-situ layers epitaxially grown on $LiNbO_3$ do not show superconducting properties different when deposited using Nd:YAG fundamental, frequency-doubled or tripled radiation.

Excimer-laser deposition: defining the process parameters

Avoiding post-deposition annealing

First results in producing excimer laser-deposited 1-2-3 layers in-situ without annealing ("one-step process") were soon reported by the Bellcore group in 1988 [8]:

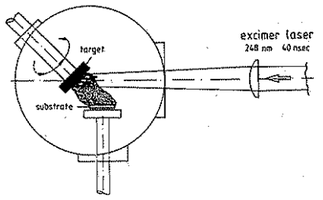

Fig. 3 Typical excimer laser deposition scheme [10]

they improved their technique by applying an oxygen jet directly to the substrate and observed chemiluminescence when the plume of laser-ablated atoms appeared: apparently, oxidizing of metal atoms occurred already in the gas phase. In this way, in-situ films of about 2000 Å thickness on $SrTiO_3$ and Al_2O_3, superconducting as-deposited, could be achieved, needing fairly low substrate temperatures of 650 °C and 580 °C. Cooling down at 250 Torr oxygen pressure took about 1 h. A short time later, a Siemens group also succeeded in circumventing annealing. However, the substrate temperature was still high (750 °C) and, again, cooling-down as long as 1 h was necessary [9].

It was soon recognized that lengthy post-deposition annealing could be avoided by using moderate substrate temperatures only, with appropriate oxygen admission, or by switching off the heater as early as possible after deposition:

A typical set-up for laser deposition is shown in *fig. 3* as realized by B. Stritzker and co-workers at KFA Jülich, FRG [10]. They use a 248 nm excimer laser (LPX), slightly focussing the beam. As will be described below, only a few J/cm² are necessary to obtain good results. As reported also by other groups, the repetition rate has to be in the range of 5 to 10 Hz, otherwise the films do not grow in the required crystalline structure. Due to the short wavelength, sputtered material as well as ambient gas is excited to fluorescence or chemiluminescence *(fig. 4)* (compare also HL 7). In a recent experiment [11], Fröhlingsdorf, Zander, and Stritzker achieved 500 Å thick films within 5 min (including heating, ventilation, and removal from the stage) which showed a T_c of 92 K (R=0) and a critical current density of 1.5 x 10^6 A/cm². In this experiment the substrate (single-crystalline $SrTiO_3$ or random–ZrO_2 as well as YSC–ceramics) was kept at an elevated temperature (optimum at 780 °C for $SrTiO_3$) during deposition, the heating switched off immediately after deposition, and oxygen used for ventilation during the cooling-down period. No further annealing was necessary.

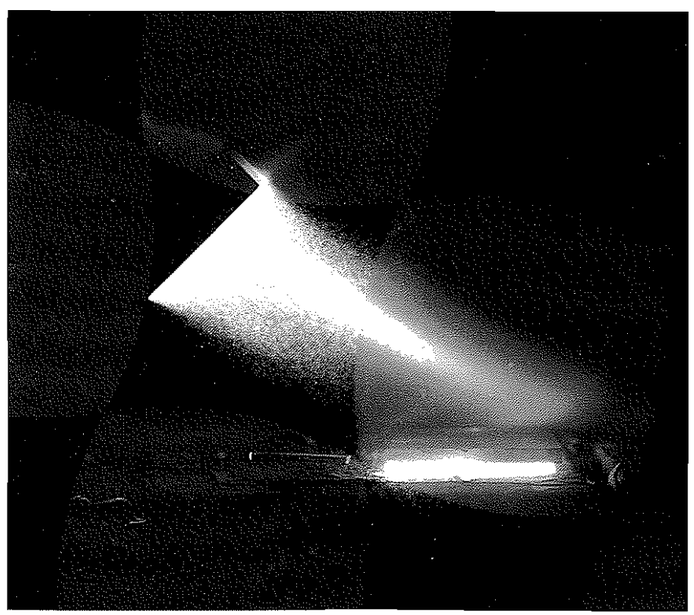

Fig. 4 Light emitting plume due to excimer laser ablation [3]. The deposition geometry is that of fig. 3.

The deposition temperature

In this experiment, the deposition temperature was found to be fairly critical: *Figs. 5a, 5b* show the optimal transition to superconductivity (a) achieved in this experiment, and (b) a plot demonstrating the importance of a careful choice of the substrate temperature in order to achieve a narrow transition width: the T_c onset (R=100%) temperature is plotted against substrate temperature as well as temperatures belonging to fractional resistivities including R=0. Clearly, at 780 °C the curves most nearly approach each other, showing that the transition from ohmic resistance (100%) to superconductivity (R=0) is very steep when this temperature at the substrate had been chosen. The transition width (defined as the 10%-90% resistance interval) was 1K only.

It should be mentioned here that the resistance curves are generally measured with the standard four-probe technique, the critical current densities up to which superconductivity survives are obtained by applying increasing current through a well defined bridge in the film (suitably prepared by excimer laser ablation) and measuring the voltage/current characteristics until ohmic behaviour appears.

Congruent evaporation: dependence on fluence

The dependence of the film composition on laser pulse fluence for a given 1-2-3 target was systematically investigated by Roas, Schultz, and Endres [12] of Siemens, Erlangen, using a 308 nm laser. *Fig. 6* shows a triangular at-% composition diagram for Y-Ba-Cu. To use this diagram, start at a given point within the triangle, projecting a line parallel to the Ba axis onto the Cu axis to find the copper percentage. Then rotate counterclockwise until parallel to the Y axis in order to find the Ba %, then parallel to the Cu axis to find the Y %. The ideal 1-2-3 stoichiometry is given by the intersection of the three lines (verify the 50 : 33,2 : 16,6 percentage ratio). The experimental points, for different fluences, refer to the compositions found on the film (on the substrate) and, correspondingly, on the target at the spot after laser evaporation. Clearly, the circles (4.5 J/cm²) and the triangles concentrate both for target (full circles) and film (open) close to 1-2-3 stoichiometry indicating the most congruent evaporation at this fluence. Fortunately, fluences in this order of magnitude are easily achieved by current excimer lasers.

The layers obtained in this way were found to have grown epitaxially (on $SrTiO_3$) with excellent superconducting properties: critical current was measured to be 5.2 x 10^6 A/cm² at 77 K, at 2 Tesla still being 3 x 10^6 A/cm². Also, at 4.2 K, where classical superconductors work, superconductivity up to a current density of 10^7 A/cm² was found, about an order of magnitude higher than that in commercial Nb_3Sn wires, the authors report.

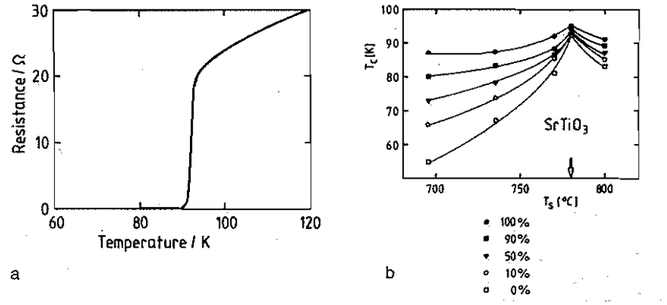

Fig. 5a Optimum transition to superconductivity achieved in [11] with 780°C at the $SrTiO_3$ substrate showing a 10-90% width of 1 K. R=0 is reached at about 92 K.

Fig. 5b Plot showing the transition widths of $YBa_2Cu_3O_{7-x}$ films on $SrTiO_3$ prepared at various substrate temperatures: R(T_c onset)=100%, 90%, 50%, 10%, and 0% (R=0) [11]. At 780°C the steepest transition is obtained (cf. fig. 5a).

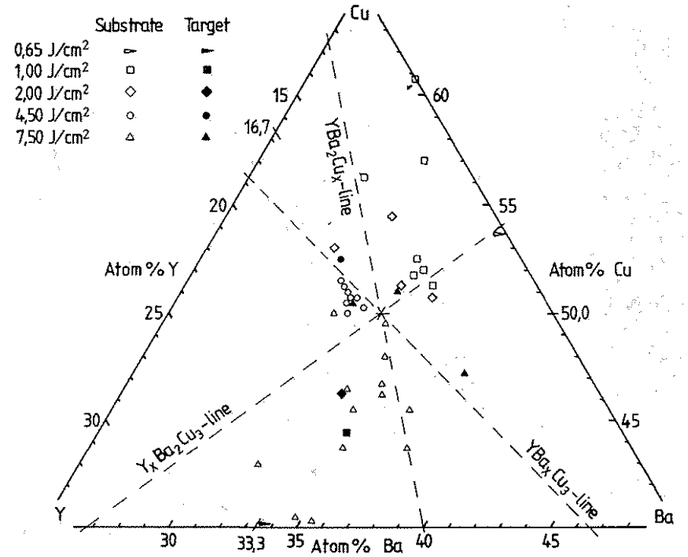

Fig. 6 Composition of deposited films and target beam spots after laser evaporation for various laser beam fluences [12]. See text for further explanation of this diagram.

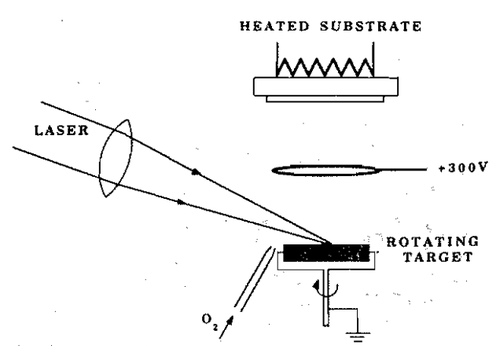

Fig. 7 Scheme of the plasma-laser deposition according to [14]

Oxygen admission: the key for high quality films

As already mentioned above, an extremely important process parameter is the oxygen partial pressure during deposition and cooling down. Since Cu-O areas are believed to be essential for high-T_c superconductivity, the control of the oxygen admission is crucial. This holds the more as the oxygen atom sticking coefficient is less than that of the metal atoms, and oxygen may be ejected.

An investigation on the oxygen admission was recently performed by Ying, Kim, Shaw, and Kwok of New York State University, Buffalo [13] who used an 193 nm excimer laser (EMG) with a fluence of 4 J/cm² at 8 Hz. They examined, at 600 °C substrate temperature, the oxygen admission parameters leading to films superconducting as-deposited (with 5 mTorr oxygen), and the parameters required for producing films (0.5 mTorr oxygen) which showed superconductivity only after a subsequent 850 °C annealing procedure. To do this, they measured the resistivity during laser deposition and oxygen admission after deposition. At the deposition temperature at the substrate (ZrO_2 (100)) there is, of course, no superconductivity, but the resistivity results were strongly dependent on the amount of oxygen used during deposition. The resistivity in $Y_1Ba_2Cu_3O_{7-x}$ is known to depend strongly on the oxygen deficiency x which, at the same time, determines whether the tetragonal phase (non-superconducting) or the orthorhombic phase (O, superconducting) prevails.

The authors concluded that at higher substrate temperatures (as 600 °C), outward diffusion of oxygen leads to a tetragonal (T) phase instead of the orthorhombic (O) phase unless oxygen background pressure is not rather high (this method is used by the Jülich group [11]). The O phase is recognized to be responsibel for good high-T_c superconductivity. Therefore, the authors conclude, true in-situ superconducting films can be obtained only with substrate temperatures lower than 350 °C. However, in this case, the activation of the substrate surface which is needed for epitaxial film growth is critical.

To achieve this, Witanachchi, Kwok, Wang, and Shaw introduced plasma-assisted laser deposition for high-T_c layers [14]. *Fig. 7* shows the scheme of this technique: a low-pressure (10^{-4} T) O_2 discharge is sustained, generating oxygen atoms, ions, and molecular ions. The substrate was at floating potential. The authors report that only O_2^+ ions formed by electron impact ionization between the ring electrode and the substrate were effective in the improvement of the deposition (ion-assisted deposition). Also, O_2^+ ions

are believed to enhance the oxygen content of the deposited film, thus improving its superconducting properties. Very recently, Bormann and Nölting [15] pointed out that favorable O_2 conditions are in accordance with the O_2 vapor pressure of the material in dependence of temperature.

Homogeneous velocity of atoms in supersonic beams

In a recent publication by the Buffalo group [16] the influence of the oxygen background on the velocity distribution of the target atoms released by the laser pulses was investigated in more detail. Using an optical time-of-flight method (TOF) (see *fig. 8*) to analyze the velocity distribution of individual species in the plasma plume at 10^{-5} Torr background pressure, the distributions of Cu, Y, and Ba atoms were measured (ascertaining that TOF spectra were independent of the atom's emission line monitored) at a distance of 7.2 cm from the target. The distributions, averaged from over 200 laser shots, could be fitted almost perfectly with a distribution to be obtained in an isentropic supersonic beam expansion. Thus, considerable energy and speed was transferred to the atoms:

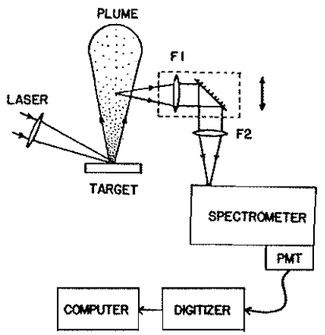

Fig. 8 Set-up for optical time-of-flight analysis of ablated species [16]

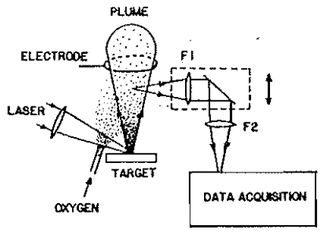

Fig. 9 Optical time-of-flight measurement of ablated species including oxygen admission [17]

Species number	Most probable speed (10^4 m/s)	Mean kinetic (eV) energy	Mach
Cu I	1.12	41.35	4.05
Y I	0.97	43.40	2.3
Ba I	0.82	47.92	1.2
Ba II	1.12	86.40	0 (Maxwellian)

Table 1: Supersonic expansion parameters obtained in ArF laser–generated ablation with 5.3 J/cm², focused to a spot of 0.5 x 1 mm [16]

The remarkable result is that a supersonic molecular beam is formed very similar to beams obtained in hydrodynamic expansion of a (fairly) high-pressure gas via a narrow nozzle. As a consequence, the most probable velocity values do not differ so much due to equalization in the isentropic expansion. In an accompanying paper [17], the influence of the oxygen flow onto the velocity distributions was studied (cf. *fig. 9*). Again, an EMG ArF laser was used to collimate the beam via the oxygen jet onto the target, providing a fluence of 5.3 J/cm² which was found most convenient for the generation of superconducting films. The oxygen jet was observed to emit a red glow indicating efficient interaction of the 193 nm radiation with O_2 molecules. A DC voltage applied downstream even enhanced radiation emission and changed its color. Oxygen admission was adjusted such as to provide as-deposited superconducting films. Under these conditions, the velocity distributions of the atoms Cu I, Y I, Ba I, Ba II, and O I were measured as a function of distance from the target. *Fig. 10* shows the result both for the Mach number and the mean velocity. Clearly, at a distance of about 7.2 cm, equalization has occurred including the Ba and oxygen atoms which have been accelerated. This distance was also found to be optimal for high-T_C as-deposited films under these experimental conditions!

The authors conclude that, for as-deposited superconducting films, it is important that the atoms travel at the same speed so as to impinge on the substrate at the same rate. In addition, the energetic oxygen atomic beam with a velocity of 6 km/s is believed

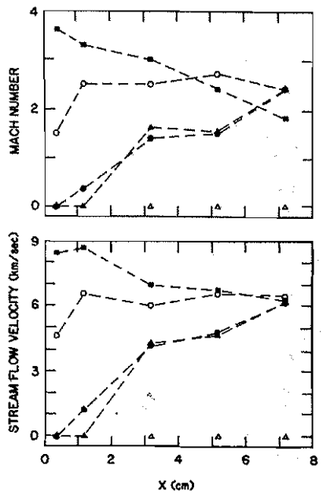

Fig. 10 Dependence of the Mach number and v_o on distance from the target for various species: (■) Cu I, (○) Y I, (▲) Ba I, (●) O I, (△) Ba II.

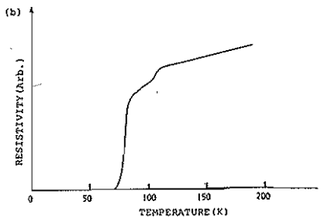

Fig. 11 Resistivity versus temperature of a Bi-Sr-Ca-Cu-O film on MgO(100) annealed at 885 °C in air for 1 minute, showing decrease of resistivity in two steps [21]

to allow the low temperature at which as–deposited superconducting films can be formed.

Using an EMG KrF laser (248 nm) at a fluence of 4 J/cm² and time-resolved spectroscopy, C. Girault, D. Damiani, J, Aubreton, and A. Catherinot of Faculté des Sciences, Limoges, France [18] found similarily energetic atomic beams, mean velocity components perpendicular to the target being almost higher than 10^4 m/s with rather narrow distribution which also is valid for the diatomic molecules CuO, BaO, YO. The authors attribute their result to the ablative nature of the decomposition.

Oxide formation in the plume

Metal oxides in the plume were spectroscopically studied by T. Venkatesan, E.W. Chase, C.C. Chang of Bellcore and coworkers of different institutions [19], in correlation with the oxygen partial pressure. It was shown that the formation of oxides already in the plume, enhanced by a high oxygen partial pressure, is essential for the production of higher-quality superconducting films.

"Tailored" films of Bi-Sr-Ca-Cu-oxides by excimer laser ablation: Higher T_C by lattice engineering?

As already mentioned at the beginning of this article, higher T_c values were observed, e.g. in Bi-Sr-Ca-Cu-oxides (BSCCO) [20] which appeared to contain two superconducting phases with T_C's at 85 K and 105-115 K (*fig.11* [21]). Clearly, to analyze the site of superconducting phases and the influence of the constituents on the film is a major challenge. Investigations focussed to this aspect are being done at Osaka University, Japan, by the group of Professor Kawai using an EMG ArF excimer laser [22]. In order to control the deposition of multi-element films, the researchers used several targets ("multitarget") exposing them sequentially in a N_2O gas atmosphere as the oxidizer. Targets were sintered disks of $Bi_7Pb_3O_y$, $Sr_1Cu_1O_y$, $Ca_1Cu_1O_y$, $Ba_1Cu_1O_y$, and $Y_1Cu_1O_y$. Irradiation was performed in different sequences in repeated operation as to form 300Å-films of different compositions in which Sr atoms were partially replaced by Ba in a BSCCO fim, and Ba, Sr or Y atoms were substituted for Ca layered in BSCCO. Post-deposition annealing at 800 °C in air

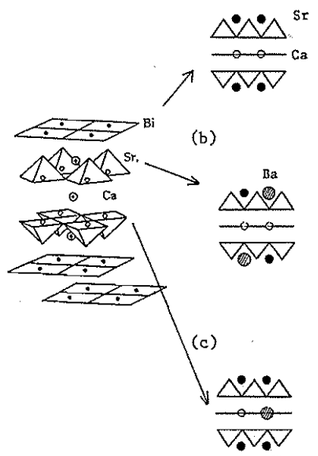

Fig. 12 Partial substitution of Ba at the Sr and Ca site. (a) BSCCO film without substitution, (b) partial substitution for Sr, 30-40%, (c) partial substitution for Ca, 30-40% [21]

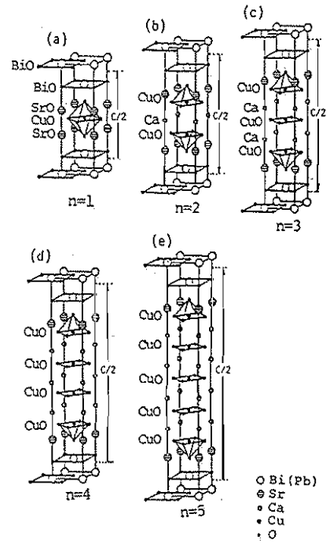

Fig. 13 "Tailored" layers of $(Bi(Pb)O)_2Sr_2Ca_{n-1}Cu_nO_{2n+2}$ in which n is 1-5 representing the number of CuO_2 layers between adjacent $Bi(Pb)_2O_2$ layers [23]

was applied for 30 min. *Fig. 12* shows schematically the modification of the crystal lattice due to the substitutions. The authors used X-ray diffraction for crystal analysis. In this figure, the pyramids mean the cuprates.

With such "lattice engineering", the c-axis length can be changed and correlated to the superconducting (or non-superconducting) properties. E.g. *fig. 13* shows lattices with different numbers of Cu-O layers between the BiO layers [23] which were obtained at low substrate temperature and reaction with N_2O.

Surely, these investigations are in an early stage. However, even if no industrial application would emerge – due to complexity of the process or other restrictions – the merits obtained for an understanding of the high-T_C phenomenon are most important.

Resumé: High-tech pushes high-tech

This short survey of the work going on with $Y_1Ba_2Cu_3O_{7-x}$ and other compounds is by no means complete. Tremendous efforts are being made around the world to define the processing parameters and to correlate them with good results for high-T_C films. Obviously, most parameters are interdependent so that comparisons are somewhat difficult. However, considering all the results as a whole, a preliminary view arises:

- *Stoichiometry is most critical especially with respect to the oxygen content.*
- *Epitaxially grown films in single phase are most important.*
- *Reproducible production is to be achieved.*
- *Long-term stability has to be guaranteed.*
- *Lattice structures leading to higher T_C should be explored.*

All the different experimental attempts discussed here have these goals in mind. Whichever technique will prove best is presently open. This is certainly also true for techniques competing with laser deposition methods.

Furthermore, for a technical application in electronic circuits, deposition of high-T_C films on Si or other semiconducting materials as well as the combination with metallic conductors will be most important. In this respect research is at the early beginning. First results of in-situ preparation of c-axis oriented Y-Ba-Cu-O and Bi-Sr-Ca-Cu-O films on Si were presented by Krebs and Kehlenbeck [24]; using an LPX 110i they produced films showing superconductivity by applying oxygen in a narrow pressure range in agreement with [15]. In this young field, experimental physics is in the exciting stage of being far ahead of a theoretical understanding of the phenomena. Therefore, techniques which allow researchers to vary parameters almost independently are welcome. With respect to magnetron sputtering and other techniques, laser deposition has an immediate advantage due to its flexibility in varying experimental parameters, hence precise interaction control, and quick response. Thus, new high technology gets its best support from established high-tech!

We thank Martina Kehlenbeck and Professor Freyhardt, University of Göttingen, for critical reading of this manuscript and valuable suggestions, and the authors of papers discussed here for sending information.

Uwe Brinkmann

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Chinese Scientist guest at Lambda Physik R & D

張育川

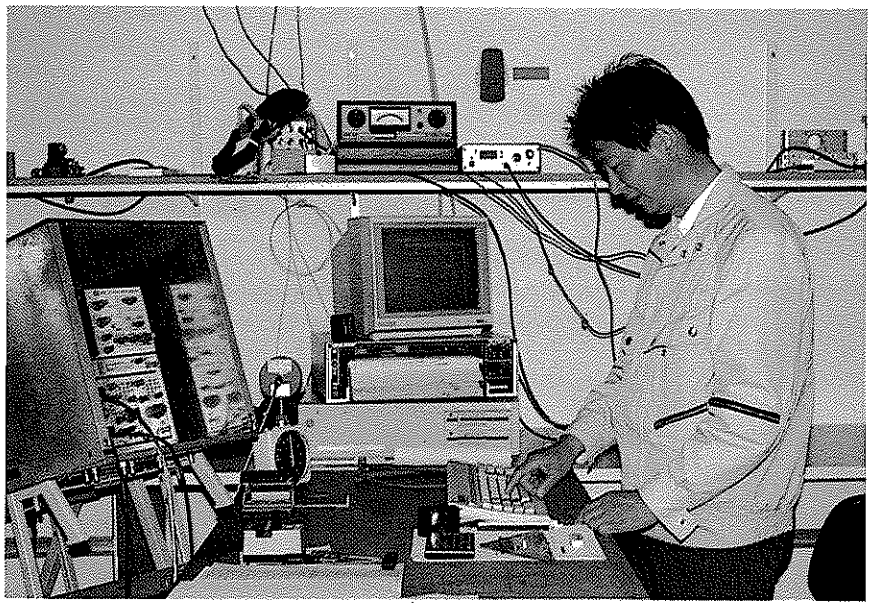
Zhang Yuchuan, Senior Engineer and Deputy Director at the Beijing Institute of Opto-Electronic Technology, Beijing, Peoples Republic of China, was at Lambda Physik for 11 months, working at the R & D division in Göttingen. His stay was a good example for the fruitful cooperation possible across borders which, until recently, were believed to be unsurmountable for a long time. At his Institute he mostly fulfilled management functions, so he enjoyed laboratory work, having sufficient time to do practical things. For example, using a laboratory set-up, he investigated the excimer laser beam pattern under various conditions, extending the analysis to the farfield pattern. Such time-consuming measurements are important for laser application and instrument improvement.

In 1977 Zhang Yuchuan first took notice of Lambda Physik, still in the "nitrogen laser era", when he investigated at his university a nitrogen laser he had built himself. At that time, Dirk Basting visited that laboratory so that the personal contact came about.

Yuchuan took advantage of his Stay travelling through different parts of Germany. When asked what he liked the most he appreciated very much Göttingen, "the city is so quiet – in comparison to the densely-populated big Chinese cities". In this respect, Munich seemed to him more comparable to Shanghai!

He noted a high efficiency in the daiöy work, and he was especially impressed by the skill of the technicians working at Lambda Physik. On coming home, he intends to apply his new experiences, as much as possible, to his work at the Institute.

However, first of all, he looked forward to seeing his family again.



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for André

Influence of hydrogen on the characteristics of an excimer XeCl* laser

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(Submitted July 24, 1987)
Kvantovaya Elektron. (Moscow) **15**, 112–113 (January 1988)

The characteristics of an electric-discharge XeCl* laser were improved by the addition of hydrogen to an He–Xe–HCl mixture. The energy of the output radiation pulses increased by 15% and the operation of the laser became stabilized. A study was made of the dependence of the output energy on the hydrogen concentration.

Beginning from the seventies the popularity of excimer lasers as effective sources of visible and ultraviolet radiation has increased. The best characteristics among excimer lasers have been reported for rare-gas halides KrF*, XeCl, etc. Further forced energy deposition in the medium is causing difficulties, so that it would be highly desirable to consider improvement of the excimer laser characteristics by optimization of the composition of the gas mixture.

The present paper reports an increase in the efficiency of an electric-discharge XeCl* laser on addition of hydrogen to an He–Xe–HCl mixture. We shall give the dependence of the output energy of the laser on the hydrogen concentration.

The active medium was excited by a transverse discharge preceded by ultraviolet preionization. The electrical part of the apparatus was in the form of the circuit shown in Fig. 1. A storage capacitor C_0 consisted of three K-15-10 capacitors connected in parallel and 10 nF each. A high-pressure spark gap G was used to switch on the discharge. The dimensions of the main discharge gap were $60\times0.5\times2$ cm (where 2 cm was the distance between the electrodes). The voltage across the electrodes was $U = 18$ kV, the duration of the discharge at midamplitude was $\tau_d \approx 50$ ns, the duration of the laser output pulses was $\tau_l \approx 30$ ns. The output energy was measured with a IKT-1M calorimeter and the duration of the output pulses was determined with an FK-22 photocathode.

Preliminary ionization of the medium was provided by a discharge on the surface of an insulator. The preionization system[2] was located axially along the electrodes. The discharge on the insulator surface was characterized by a high spectral brightness in the short-wavelength range, which ensured effective preionization of the gas mixture.

The influence of the addition of hydrogen on the output characteristics of the laser was investigated as follows: first, the composition of the He–Xe–HCl mixture was optimized in respect of the output energy. It was found that when this composition was He:Xe:HCl = 1000:10:1, the energy reached 21 mJ, which corresponded to an efficiency of $\eta = 1.5\%$ relative to the stored energy. Then, hydrogen was added to this optimized He–Xe–HCl mixture. The dependence of the output energy on the hydrogen concentration was determined (Fig. 2). In the case of a mixture with the He:Xe:HCl:H_2 = 1000:10:1:1 composition the energy of the radiation pulse is increased by 15% reaching 25 mJ; moreover, the stability of operation of the laser improved. Lasing occurred at wavelengths $\lambda_1 = 307.87$ and $\lambda_2 = 308.0$ nm. An improvement in the reproducibility of the energy from one pulse to another was clearly due to recovery of the HCl molecule by the reaction

$$H_2 + Cl_2 \rightarrow 2HCl. \qquad (1)$$

Since an increase in the energy deposited in the mixture resulted in a practically linear increase in the output energy, it was concluded that the HCl donor was not significantly "depleted" during one pulse. Therefore, the increase in the energy of the first pulses in the series could not be explained by the reaction (1). The improvement in the output characteristics of the laser was clearly due to the following factors:

1) cooling of electrons due to excitation of vibrations of the H_2 molecule:

$$H_2 + e \rightarrow H_2(v) + e, \qquad (2)$$

which increased the rate of excitation of vibrations of the HCl molecule[1] in accordance with the reaction

$$HCl + e \rightarrow HCl(v) + e; \qquad (3)$$

2) transfer of the vibrational excitation from the molecules

$$H_2(v) + HCl \rightarrow HCl(v) + H_2.$$

Therefore, when hydrogen was added to the He–Xe–HCl mixture, there was an effective increase in the

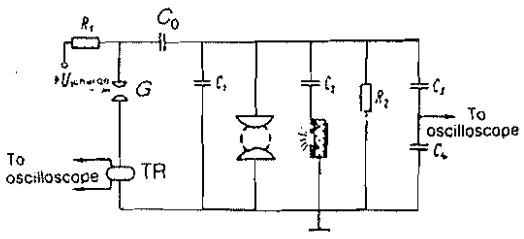

FIG. 1. Electrical part of the apparatus: R_1 and R_2 are resistors in the charging circuit; C_0 is a storage capacitor; G is a discharge gap; C_1 is a peaking capacitor; C_2 is a decoupling capacitor in the preionization circuit; C_3 and C_4 are capacitors in the voltage divider circuit; TR is the current transformer.

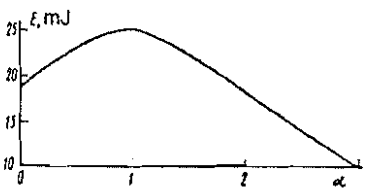

FIG. 2. Dependence of the output energy on $\alpha = [H_2]/[HCl]$ for a mixture of the He:Xe:HCl = 1000:10:1 composition with $[He] = 8\times10^{19}$ cm^{-3}.

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number of vibrationally excited $HCl(v)$ molecules and of active $XeCl^*$ molecules as a result of the multistage process $HCl(v) \rightarrow Cl^- \rightarrow XeCl^*$.

When the hydrogen concentration was $[H_2] > 1.5[HCl]$, there was a strong reduction in the output energy of the laser. This was due to quenching of the $XeCl^*$ molecules by hydrogen and due to absorption of laser radiation by the H_2 molecules.

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Lasing due to impurity color centers in yttrium aluminum garnet crystals at wavelengths in the range 1.35–1.45 μm

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Low-threshold wide-band lasing was achieved for a YAG crystal with optically and thermally stable color centers. The absorption and luminescence spectra were determined, and the gain was measured in the range of wavelengths corresponding to the color-center luminescence. The cross section of the lasing transition was estimated (8×10^{-19} cm^2) and the efficiency of conversion of the pump radiation from a neodymium laser into the output radiation was determined (10%).

Solid-state tunable lasers operating in the near infrared have some important applications. Much of the work on these lasers has been concerned with the spectral and lasing characteristics of crystalline materials doped with transition-metal ions V^{2+}, Ni^{2+}, and Co^{2+} and with lasers made using these materials and emitting in the range 1.2–1.8 μm (Ref. 1). However, lasers of this type are not used widely because the active medium has to be cooled to cryogenic temperatures. Considerable progress has recently been made on the construction of color-center lasers using alkali halide crystals, which can operate efficiently at room temperature.[2,3] Unfortunately, the instability of the parameters of active elements of lasers of this type under the influence of elevated temperatures or of ultraviolet and visible optical radiation limits seriously the potential applications of these lasers.

We shall report lasing observed in the range 1.35–1.45 μm for optically and thermally stable impurity color centers in yttrium aluminum garnet (YAG) crystals. Impurity color centers in crystals with the garnet structure can be formed by doping with chromium and oxidation of this impurity to the quadrivalent state. These color centers have strong absorption bands in the visible and near-infrared parts of the spectrum. Figure 1 shows the absorption spectrum recorded at 300 K for a YAG crystal containing color centers. Excitation of such a crystal in the absorption band of the color centers produced luminescence in the wavelength range 1.25–1.7 μm, the spectrum of which is shown in Fig. 2. The luminescence decay kinetics observed on pulsed excitation of the color centers with a Q-switched and neodymium laser was exponential with a time constant 4.6 μs at 300 K. Bleaching of the crystals with color centers was observed in a band at 9000–12 000 cm^{-1} when they were subjected to the same neodymium laser radiation and the density of the absorbed energy was ~40 mJ/cm^3, which indicated that a population inversion was attained for the main transition in the color centers. Bleaching of YAG crystals with color centers in the absorption band at moderate densities of the absorbed energy and the wide spectrum of the color-center lumines-

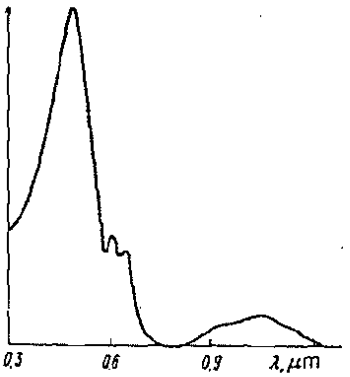

FIG. 1. Absorption spectrum of a YAG crystal with color centers.

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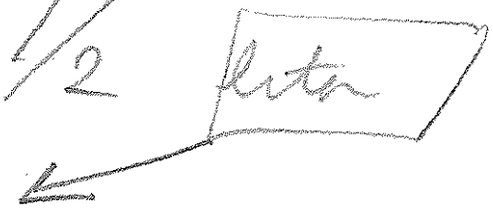

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