LASERFEST

History of Gas Lasers

Part 2: Pulsed Gas Lasers

In this second article of a two-part series, Colin Webb explores the origins of pulsed gas lasers, which made possible many critical applications of laser technology, including corneal reshaping and microlithography.

Ask anyone to name the most important application of lasers, and they will probably point to their clinical use—for example, lasers that reshape the cornea of the eye to correct vision defects, as explored in this month's cover article on "Lasers in Ophthalmology" (p. 28). The laser that has made corneal reshaping possible is the argon fluoride (ArF) excimer laser, which operates at 193 nm. It is a rare gas halide (RGH) excimer laser, a type that burst on the scene in 1975.

Because of its very short wavelength and its ability to propagate in air, the ArF laser has also become the key light source for microlithography—which allows scientists and engineers to produce features of size less than 100 nm on semiconductor chips. This is the largest application for this type of laser, and a driving force in electronic technology. Without it—or its KrF analogue—much of today's computer technology would not be possible.

But the excimer laser was not created in a vacuum. There is a considerable history of pulsed gas lasers that precedes it. Here, following Jeff Hecht's January feature on continuous wave gas lasers, I explore how pulsed gas laser systems came to be.

The nitrogen laser—a DIY project

Among the very earliest of the purely pulsed gas lasers is the molecular nitrogen system, which operates in the near ultraviolet on the C⟶B band of the N_2 molecule at wavelengths around 337 nm. H.G. Heard at Energy Systems discovered this laser in 1963. Its performance in terms of pulse energy (typically a few mJ), and pulse repetition frequency (typically 2-20 per second), are by no means impressive by today's standards. Nevertheless, it is of great significance in the history of lasers, not only because its output was (and still is) used to pump tunable dye lasers covering the near-ultraviolet and visible regions, but because it was so simple to construct and operate.

In fact, many thousands of these lasers have been built as science projects in colleges and domestic garages all around the world. *Scientific American*'s Amateur Scientist column published do-it-yourself instructions in 1974. This has been the inspiration for myriad designs. In 1973, Jim Piper and I built our own nitrogen laser to be used for atomic physics experiments at the Clarendon Laboratory at Oxford.

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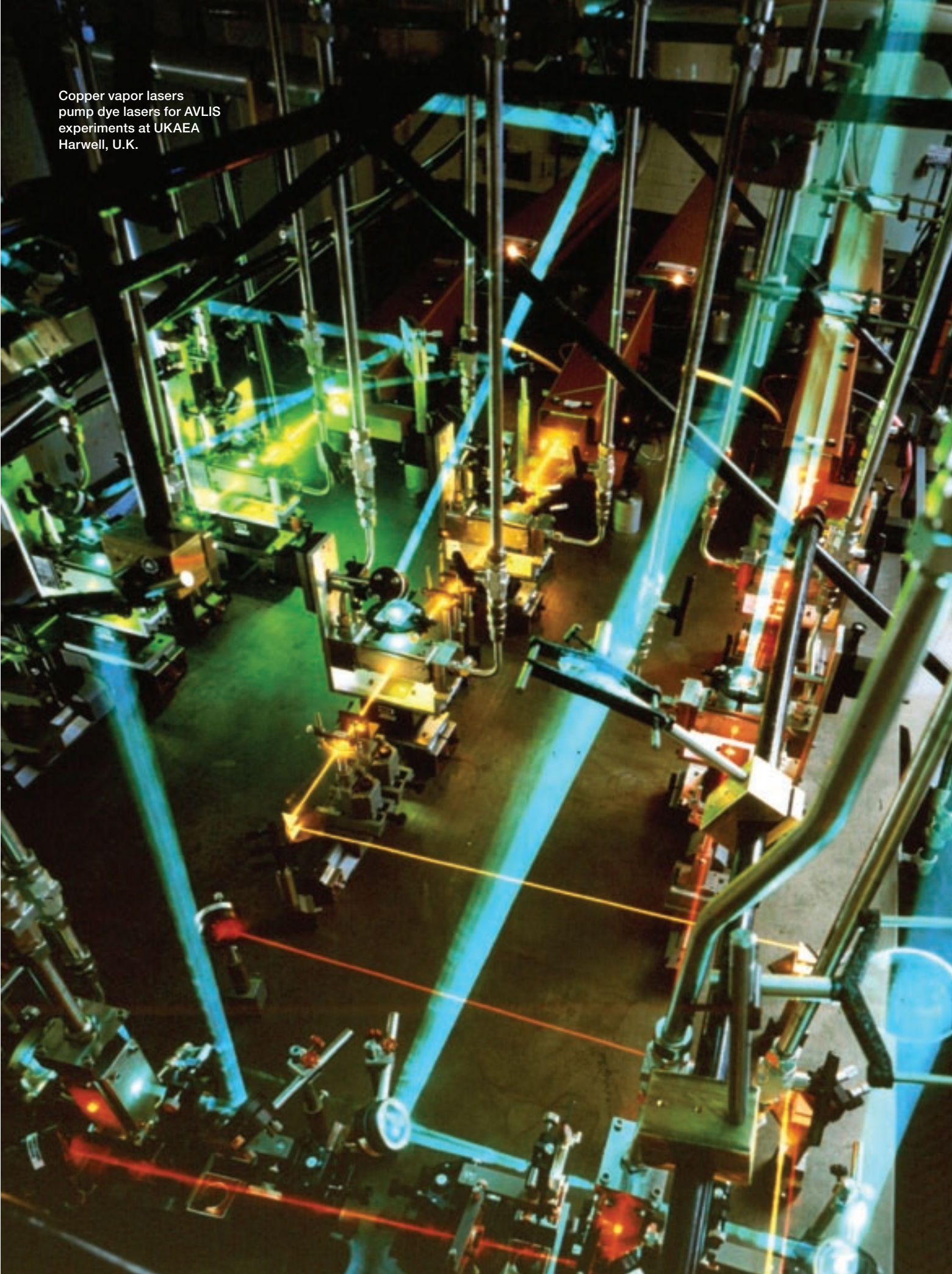
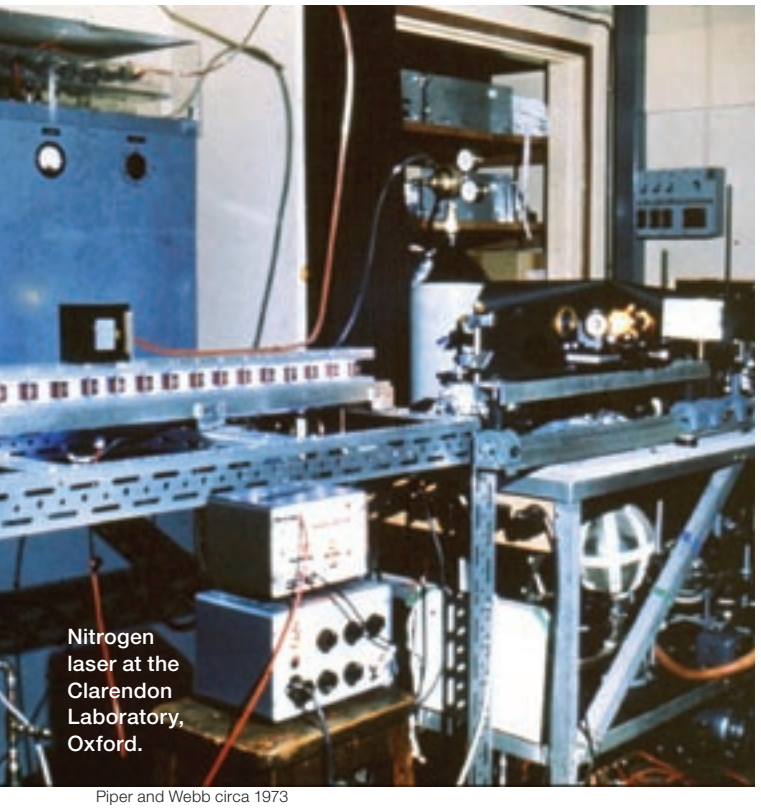


Copper vapor lasers pump dye lasers for AVLIS experiments at UKAEA Harwell, U.K.



Nitrogen laser at the Clarendon Laboratory, Oxford.

Piper and Webb circa 1973

The breakdown voltage of molecular gases at the pressures needed for laser operation tends to be much higher than in low-pressure atomic gases. For this reason, the discharge in the N_2 laser runs transversely across the width of the active volume rather than along its length. A capacitor bank charged to many tens of kilovolts and switched by a triggered spark gap (or, in more sophisticated devices, a thyratron) discharges its energy into the nitrogen at pressures from a few Torr to one atmosphere in a current pulse—which rises to several kiloamperes within 50 ns or less.

The output of the laser comprises a pulse that lasts some 10 ns for the lowest N_2 pressures or less than 1 ns in the case of atmospheric pressure operation. Whatever the pressure, however, the vibrational states of the lower laser level have no way to get rid of the population in timescales of interest, and the laser pulse is therefore self-terminating; longer excitation pulses do not result in longer laser pulses.

Self-terminating VUV lasers—The Franck-Condon Loop

This self-terminating feature is shared by many important gas laser systems, where the lower laser level is long-lived. It limits pulse length to about the upper-level lifetime—which, in the case of the H_2 laser operating on transitions of the VUV Lyman bands of molecular hydrogen around 160 nm, implies rise-time requirements that are even faster than those of the N_2 laser.

In 1926, James Franck of the University of Göttingen, Germany, suggested that, because the masses of the nuclei of a diatomic molecule are so much larger than the mass of an electron, the instantaneous positions of the vibrating nuclei cannot be significantly affected when an electron changes from one excited state to another by absorbing or emitting a photon. Thus, electronic transitions can be represented as vertical lines on a diagram of potential energy vs. internuclear separation. Within a few weeks, Franck's principle was put on a sound quantum mechanical basis by Edward Condon of the University of California, Berkeley. He showed that, in addition, the most probable transitions occur at internuclear separations for which the overlap between the vibrational wavefunctions of the upper and lower level is maximized. This is known as the Franck-Condon principle.

When a fast moving electron impacts a molecule, the probability of excitation mirrors the optical transition probability. Thus, to a first approximation, electron impact excitation conforms to the Frank-Condon principle. As suggested by Bazhulin and colleagues at the Lebedev Institute in Moscow, a scheme based on the Frank-Condon loop can be used to create transient population inversion in molecular gases. To illustrate this scheme in the case of H_2, high-energy electrons in the discharge preferentially excite H_2 molecules in the ground vibrational level ($\upsilon'' = 0$) of the ground electronic level $X^1\Sigma_g^+$ to vibrational levels ($\upsilon' = 4–8$) of the electronic level $B^1\Sigma_u^+$. The selective excitation produces short-lived inversions with respect to excited vibrational levels ($\upsilon'' = 11$-14) of the ground electronic level, which are themselves far too high in energy to be populated at thermal energies. On much longer timescales, the population in these states eventually returns to the ground state by thermal collisions before the start of the next discharge, thus completing a "loop" in the energy diagram.

As suggested by Bazhulin and colleagues at the Lebedev Institute in Moscow, a scheme based on the Frank-Condon loop can be used to create transient population inversion in molecular gases.

In 1970, R.T. Hodgson at the IBM Watson Research Center first demonstrated VUV laser action in H_2. He observed oscillation on several transitions in the Lyman bands around 160 nm. This was followed in 1971 by Waynant's observation of oscillation on two transitions of the Werner bands at 116 and 123 nm using a traveling wave Blumlein excitation circuitry developed by Waynant, Shipman and colleagues at NRL Washington.

The molecular hydrogen lasers provided early evidence that gas lasers would be possible in short wavelength regions of the spectrum where most solid-state materials have limited optical transmission. Despite that, these systems never seem to have been

exploited commercially. This may be because their output pulse energy and repetition rate are very limited, or it could be because there has been—up to now—little demand for such short wavelength lasers. Whatever the reason, these systems remain today as scientific curiosities.

Pulsed CO_2 lasers—TEA anyone?

The pulsed CO_2 laser system is certainly more than a curiosity. It provides high-power pulses at many discrete frequencies in the mid-infrared bands around 10.4 and 9.4 μm. As Jeff Hecht described in his January feature, at low gas pressures, the CO_2 laser is basically a continuous wave system that depends for its excitation on resonant energy transfer in collisions of N_2 molecules in the first vibrationally excited level (υ = 1) with ground state CO_2 molecules:

$$N_2(\upsilon=1) + CO_2 \text{ (ground state)} \rightarrow N_2(\upsilon=0) + CO_2 \text{ (upper laser level)} + \Delta E \text{ (small)}. \quad (1)$$

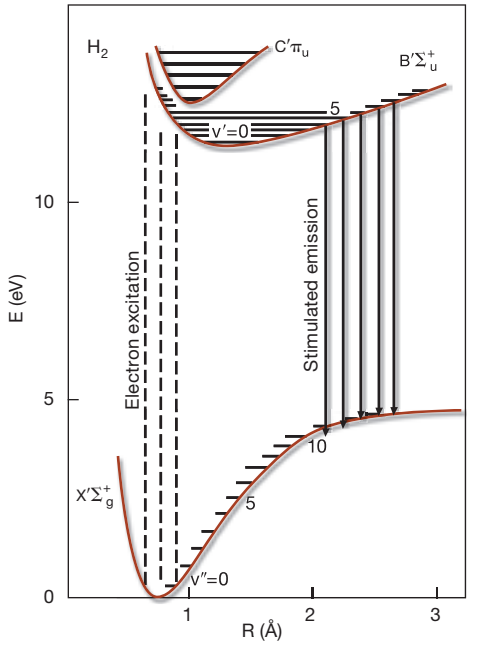
The N_2 molecules are excited by collisions of electrons in the discharge run in a gas mixture containing He, N_2 and CO_2.

In 1969, R. Dumanchin and J. Rocca-Serra in France and A.J. Beaulieu in Canada recognized that, by increasing the gas pressure of the discharge from 5-20 Torr (the range characteristic of the CW systems) to pressures on the order of one atmosphere (760 Torr), the population inversion density could be increased enormously. This is because the number of target N_2 molecules that electrons could hit, and the number of target CO_2 molecules that excited N_2 molecules could hit, would increase correspondingly. However, there is a problem in running longitudinal discharges at such high pressures. The voltages required to cause breakdown and to sustain a discharge are unacceptably high, and so Beaulieu turned to the solution of running the discharge transversely across the width of the discharge volume rather than along its length.

Thus, the TEA (transversely excited atmospheric) CO_2 laser was born. In order to stop the discharge from collapsing into a few isolated arcs along the length of the electrodes, Beaulieu used distributed ballasting in the form of a closely spaced array of 1 kΩ resistors. Each one was connected at one end to a common rail; the free ends constituted the segmented cathode on one side of the discharge gap, and the anode side comprised a solid rod of conductor. If, in the early stages of the collapse from a spatially extended glow to a constricted arc, the current through one resistor should show an increase above the ambient value, the increased voltage drop across that resistor would stifle the current growth and quench the incipient arc. This is how the ballasting action comes about.

Although Beaulieu's device established the principle of TEA laser operation, the discharge volume was composed of a thin array of spatially separate discharges, which did not easily lend itself to scaling the discharge volume. The next step demanded the creation of a discharge volume of approximately square cross-section; this was done by using uniform field electrodes

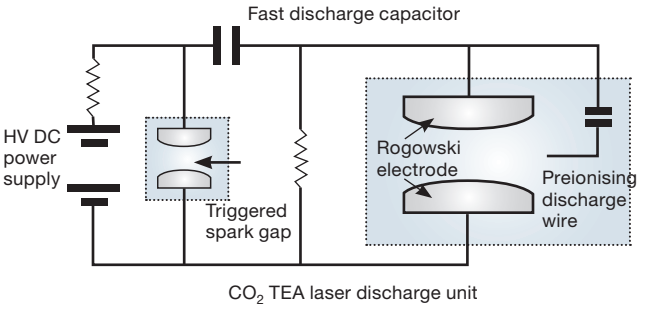
[The Franck-Condon Loop]



The energy level scheme of Hodgson's VUV hydrogen laser.

Reprinted with permission from R.T. Hodgson. Phys. Rev. Lett. **25**(8), 494-7 (1970). ©1970 American Physical Society

[Lamberton and Pearson's TEA CO_2 laser]



CO_2 TEA laser discharge unit

From Wikimedia Commons, TEA Laser

that were carefully shaped to Rogowski or Bruce profiles. Several groups, including H.M. Lamberton and P.R. Pearson working at the U.K.'s Services Electronics Research Laboratory in Baldock, Hertfordshire, experimented with such designs and found that pre-ionizing the discharge in the He:N_2:CO_2 mixture would allow uniform excitation of a large volume for a few microseconds.

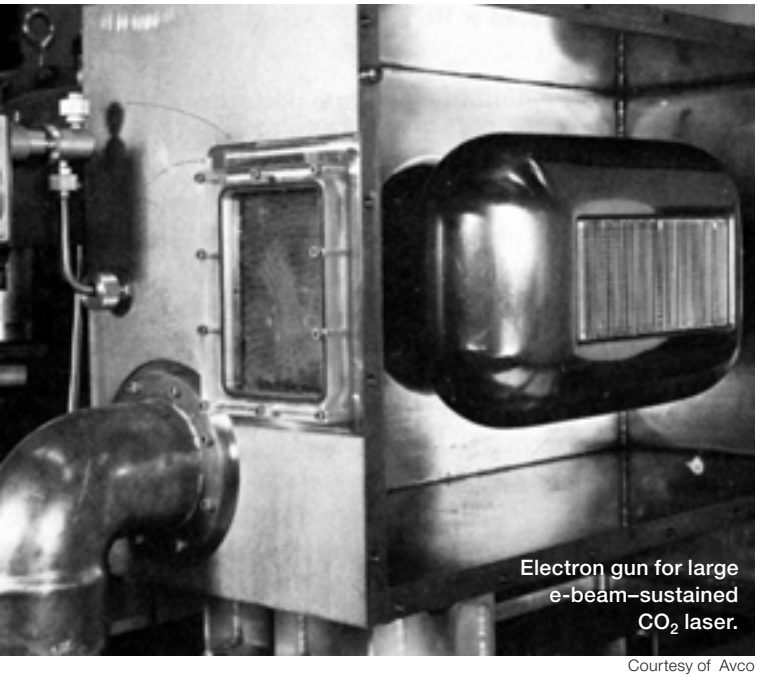
These early TEA lasers gave output pulse energies on the order of 5J from a discharge volume of 2.5-cm width, 3.2-cm height and 100-cm length. Pre-ionization of the main discharge volume was provided by the photo-ionizing effect of deep UV emitted by a corona discharge around wires situated

to one side of the discharge volume or behind a mesh electrode. Devices based on this technology have found widespread applications, including the date-marking systems for consumer products that have been used since the 1970s.

Throughout the 1960s, 1970s and 1980s, defense contractors worked on scaling CO_2 lasers to still-higher pulse energies. In 1969, W.L. Nighan and J.H. Bennett at Avco made calculations that showed that, in order to excite N_2 molecules to the first few vibrational levels some 0.3 to 1.2 eV above their ground state in a gas discharge, the optimum value of E/N—the electric field to gas number density ratio—must be far smaller than that needed for a discharge in the same CO_2:N_2:He gas mixture in order to be self-sustaining.

By 1992, Livermore had established a copper laser MOPA (master oscillator-power amplifier) array that provided an average power of 7 kW to a dye laser MOPA array with 2.5-kW average power for 24 hours per day, 7 days per week.

However, by using an external electron beam source to ionize the gas mixture, researchers could run a discharge at an E/N ratio chosen to be optimum for exciting the vibrational levels of N_2, which then go on to excite the CO_2 by the reaction of equation (1). Typical of this approach is the device reported in 1976 by J.D. Daugherty of the Avco Everett Research Laboratory; it employed a cold cathode electron gun to provide a beam of 300 kV electrons 10 cm high × 25 cm wide. Square pulses of discharge current lasting 80 μs could be sustained by the e-beam, leading to output pulses of some 55 μs duration, with energies of several kJ. By far the largest part of the input energy is supplied by the discharge.



Electron gun for large e-beam–sustained CO_2 laser.

Courtesy of Avco

Iodine—a cure for all ills?

The photodissociation iodine laser discovered by J.V.V. Kasper and G.G. Pimentel at the University of California, Berkeley, in 1964 represents a very different type of gas laser. It operates on the magnetic dipole transition (1.315 μm) between the fine structure levels $^2P_{1/2} \rightarrow {}^2P_{3/2}$ of the ground state of atomic iodine. The upper laser level is populated selectively when iodine-bearing compounds such as CF_3I or CH_3I are dissociated by intense radiation from a flashtube.

In this form, iodine lasers are self-terminating pulsed devices, since there is no way for the population to exit from the $^2P_{3/2}$ lower level (the ground state) in relevant time scales. Lasers of this type were developed progressively via a series of devices known as Asterix for laser-target studies at the Max Planck Institute in Garching, Germany. The technology was later transferred to Prague in the Czech Republic, where today the Prague Asterix Laser System can deliver pulses of up to 1 kJ at 1.315 μm.

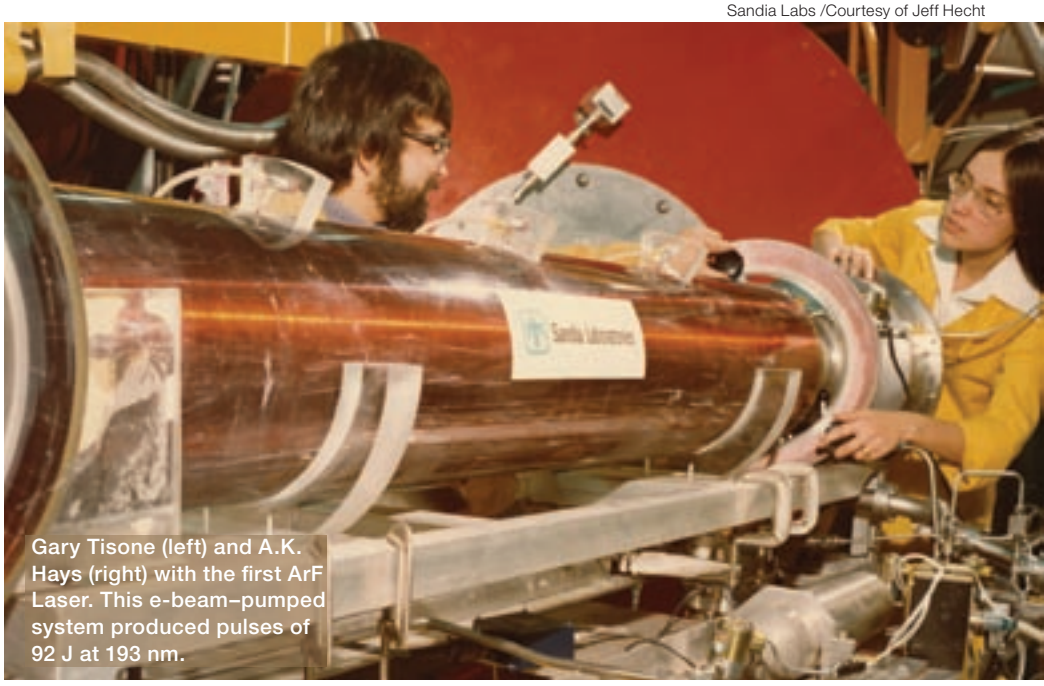
In recent years, the iodine laser system has morphed into a purely chemically excited device (the chemical oxygen–iodine laser, or COIL), which does not require any electrical input to produce long pulses of high power radiation; rather, it uses collisions of excited oxygen molecules in the metastable $^1\Delta$ state to excite the ground-state iodine atoms from the lower $^2P_{3/2}$ lower laser level to the $^2P_{1/2}$ upper laser level. The $O_2(^1\Delta)$ molecules are produced as the result of chemical reactions of Cl_2 molecules with basic hydrogen peroxide solution. In this form, the iodine laser is effectively a continuous wave device capable of very long pulses. Such a device is currently in development as Northrop Grumman's Airborne Laser.

Copper—an atomic laser

In 1966, W.T. Walter et al. at TRG Inc. in Melville, N.Y., U.S.A., reported oscillation on the $^2P_{3/2} \rightarrow {}^2D_{5/2}$ and $^2P_{1/2} \rightarrow {}^2D_{3/2}$ transitions of the neutral copper atom at 511 and 578 nm, respectively. Metallic copper that was vaporized at high temperatures and excited in a longitudinal pulsed discharge formed the basis of their copper vapor laser. A.A. Petrash and colleagues at the Lebedev Institute in Moscow showed that, by repetitively pulsing the discharge at a high repetition rate, the device becomes self-heating, obviating the need for an external oven. I. Smilanski at the Nuclear Research Centre Negev in Israel showed that it was possible to scale the output power with the tube diameter.

These lasers definitely belong to the self-terminating class of pulsed gas lasers, since the $^2D_{5/2}$ and $^2D_{3/2}$ lower laser levels are fully metastable, and collisional deactivation on the time scale of the laser pulse is not possible. However, they are relatively

Sandia Labs /Courtesy of Jeff Hecht



Gary Tisone (left) and A.K. Hays (right) with the first ArF Laser. This e-beam–pumped system produced pulses of 92 J at 193 nm.

efficient since the electron collision cross-section for exciting the $^2P_{3/2}$ and $^2P_{1/2}$ upper laser levels from the $^2S_{1/2}$ ground state dominates electron inelastic loss processes so that the copper system is the laser analogue of the sodium lamp. Typical commercial copper lasers (Oxford Lasers Ltd.) offer average powers of up to 45 W at 511 and 578 nm combined with pulse lengths of 25 ns, and pulse repetition frequencies adjustable in the 10-30 kHz range.

Throughout the 1970s and 1980s, many laboratories were developing copper laser systems for dye laser pumping for atomic vapor laser isotope separation applications. The Lawrence Livermore Laboratory in the United States, for example, was heavily involved in this area. By 1992, Livermore had established a copper laser MOPA (master oscillator-power amplifier) array that provided an average power of 7 kW to a dye laser MOPA array with 2.5-kW average power for 24 hours per day, 7 days per week. However, with the collapse of the former Soviet Union and the consequent availability of diluted weapons-grade uranium for reactor fuel on the world market, the momentum to develop new methods of enrichment itself evaporated.

The pure rare gas excimers—promise unbounded

The word "excimer" is a contraction of the words "excited dimer." Technically, rare gas halide molecules should not belong to this class because the word "dimer" applies to a molecule comprising two nuclei of the same atomic species. Still, the word excimer has come to apply to any type of laser involving a transition from the bound excited state of a molecule to a lower level that is unbound.

The idea that transitions from the bound excited state of an excimer to the unbound ground state would make good candidates for laser molecules dates back to 1960. F.B. Houtermans proposed that such transitions from excited states of mercury dimers to the ground states would easily produce inversions, since the ground state is unstable and easily breaks apart into two separate Hg atoms in timescales of a few picoseconds. The ground state is therefore virtually unpopulated, so lower-level bottlenecking could not limit the pulse energy or duration.

Although Hg_2 does not appear to have produced a viable high-power laser system to date, it stimulated the idea that other excimers—notably those of the rare gas molecules (Xe_2, Kr_2 and Ar_2)—could produce lasers. Lasers based on rare gas dimers were reported in 1970, when N.G. Basov et. al. demonstrated gain at 175 nm in electron-beam-pumped liquid xenon. And the next several years brought the first operation of a pure rare gas excimer laser with xenon (1972), krypton (1973) and argon (1974).

From that time on, many large labs around the world began scaling the output of Xe_2, Kr_2 and Ar_2 lasers to high-pulse energies. However, as C.W. Werner and E.V. George pointed out in the 1976 book *Principles of Laser Plasmas* (G. Bekefi, ed.), a barrier appeared to be set by the fact that the laser photon in these systems has enough energy to cause the photo-ionization of states of the excimer populated by collisional transfer from the upper laser level. According to this explanation, photo-ionization would provide an increasing source of internal loss as the intensity of pumping increased. In contrast, the rare gas halide's laser photon energy is not enough to photo-ionize the upper laser level (or states nearby); thus, one major source of internal loss is absent.

The rare gas halide excimers—promise fulfilled

The first member of the class of rare gas halide (RGH) lasers to be successfully demonstrated was the xenon bromide (XeBr) laser. In May 1975, S.K. Searles and G.A. Hart of the Naval Research Laboratory (NRL) in Washington D.C. first reported operating this laser at 282 mm. The idea that RGH molecules would make good candidates for powerful short-wavelength lasers was put forward by D.W. Setser and J.E. Velazco of Kansas State University. The previous year, Setser had been working in the chemistry laboratory at Cambridge University in the United Kingdom, in the group headed by B.A. Thrush. That team had observed UV emission from the bands identified as belonging to the argon chloride molecule excited when Cl_2 was titrated into a flowing afterglow in argon.

ELECTRA KrF laser at the Naval Research Laboratories in Washington D.C., U.S.A.

Courtesy of John Sethian and F. Hegeler

The reason why Searles could so quickly test the suggestion that XeBr would lase in the UV is that he was already working on pure rare gas excimer (Xe_2) laser research at NRL, and he had an electron-beam-excited device available for this work. Thus, he only needed to change the mirrors and introduce a small amount of bromine vapor into the laser chamber along with the xenon to see if XeBr would lase.

At an internal seminar held at NRL in March 1975, Searles' work on XeBr was communicated to representatives of Avco, Northrop and Livermore and other U.S. laboratories working on excimer laser research. J.J. Ewing and C.A. Brau at Avco Everett lost no time in acting on the information, and, at the June 1975 Conference on Lasers and Electro-Optics, they announced lasing in KrF (249 nm), XeF (354 nm) and XeCl (308 nm). Then, in 1976, Hoffman, Hays and Tisone at Sandia Labs announced laser action on ArF (193 nm), also in an e-beam–pumped cell.

These advances stimulated intense research. An important strand of this work was aimed at emulating the combination of electron-beam and discharge pumping, which had so successfully been applied to the CO_2 laser system. In the case of the RGH lasers, the same approach did not yield such rewards in the fraction of power that could be usefully applied as discharge excitation. Not much more than half the energy input to an electron-beam-sustained RGH laser could be applied as discharge pumping.

The gas mixture of an e-beam-pumped RGH laser such as KrF is made up of three components:

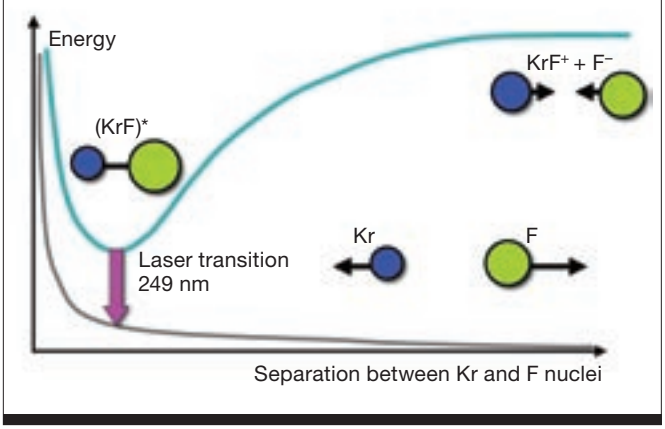
(1) the rare gas Kr at some tens of Torr pressure, (2) a few Torr of the halogen donor (F_2), and (3) two or three atmospheres of argon or neon. The argon or neon is needed to scatter the incoming electron beam so that it deposits its energy in the gas cell instead of sailing straight on through. A second important function of this background gas is the provision of an abundant supply of "third bodies" for the three body reactions involved in the formation kinetics of the excimer KrF* via the ion recombination reaction of positively charged Kr^+ ions and negatively charged fluorine ions F^- :

$$Kr^+ + F^- + Ne \rightarrow KrF^* + Ne\ . \qquad (2)$$

The main reason why the RGH lasers and CO_2 differ so much in their division of labor between sustaining the ionization rate and exciting the upper laser level precursor species is that the energy required to create the Kr^+ ions needed for reaction (2) is the same or very close to that required for creating electron-ion pairs to sustain the discharge.

Recent research efforts to develop high-pulse-energy excimers for potential applications as drivers for thermonuclear fusion reactors have been focused on purely e-beam-excited systems. The ELECTRA KrF laser at NRL currently produces pulses of 250 to 700 J at repetition rates of 1-5 pulses per second. One advantage that gas laser systems have over their solid-state counterparts is their ability to shed waste heat

[Formation of excited KrF molecules]



rapidly from the active medium by circulating the gas mixture in a closed loop containing heat exchangers.

Throughout the 1980s and 1990s, researchers looking to develop commercial RGH excimer lasers focused on purely discharge-excited systems without the e-beam generators—which tended to restrict their repetition rate capability and increase their complexity and cost. Lumonics and Lambda Physik were prominent developers of systems for scientific applications; sometimes they used part of the excimer laser's output to pump dye lasers for pump-probe spectroscopy. Another company, Cymer, supplied the market for UV lasers for photolithography, where the short wavelength of the ArF laser also gives it an advantage. Companies that supplied the ophthalmologic markets, including Summit and Visex, set out to supply the market for ArF systems for corneal-reshaping applications.

Another system that can be operated in the same type of discharge-excited device as ArF or KrF is the molecular fluorine laser. It provides pulses of a few nanoseconds' duration at the even deeper ultraviolet wavelength of 157 nm. Since radiation at this wavelength is strongly absorbed in air, the transport of an F_2 laser beam necessarily involves the use of vacuum (or helium-filled) light paths, which may make it less attractive for many potential applications.

Discharge-excited excimer lasers typically produce pulses in the range 0.5-2.5 J at repetition rates of 0.1-1 kHz. R.S. Taylor and K.E. Leopold at NRC Ottawa have reported pulse lengths as long as 1.5 μs at 308 nm for specially optimized XeCl lasers, but, for KrF discharge lasers, the longest pulses are around 800 ns, with typical values of 10 to 50 ns.

This raises at once an interesting question. The lower laser level of an excimer laser transition is unstable against dissociation into neutral atoms (Kr and F in the case of KrF)—so why are the pulses so short? These lasers cannot be self-terminating like the N_2 and H_2. However, like the CO_2 system, they have an energy-level scheme that one might expect would lend itself to CW operation. A bright young Scottish graduate student, Julian Coutts, who was working in my group at Oxford in 1986, provided insight into this. He noted, for example, that, in the KrF laser, the loss rate of electrons is controlled by the dissociative attachment reaction:

$$F_2 + e^- \rightarrow F^- + F\,. \qquad (3)$$

If now the halogen donor (F_2) density in some small region should decrease from its ambient value, the electron density will grow more rapidly in that region. Hence, the concentration of a neutral halogen donor would be further diminished locally. This is clearly a positive feedback mechanism, which, if unchecked, will lead to the formation of a column high in electron density, ultimately forming an arc that disrupts the uniformity of the gain region. Such an arc or array of arcs can cause the volume of uniform gain medium to decrease rapidly and, in extreme conditions, can result in the voltage across the transverse electrodes collapsing.

Andrew Kearsley in February 1978 with the first commercial-discharge-pumped KrF laser in the United Kingdom.

Courtesy of Colin Webb

Julian Coutts' theory predicts the length of time after which an initially uniform laser discharge becomes unstable due to halogen donor depletion. The theory has been tested successfully by many groups around the world. Coutts died last year aged only 45, after losing his battle with cancer. Perhaps this article can serve as a small memorial to him.

OSA Member **Colin Webb** (colinwebb9@sky.com) is a professor emeritus at the University of Oxford, United Kingdom, and chairman of Oxford Lasers.

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