

The first years of laser-induced breakdown spectroscopy

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The arrival of the laser in May, 1960 opened up new possibilities for microanalysis which were quickly explored. The origins of laser-induced breakdown spectroscopy (LIBS) begin in this era. Although several excellent historical overviews of LIBS have appeared in the past,^{1–3} as well as books,^{4–8} none have focused specifically on the details of the very first measurements. Here, we will do so. In January, 1962 G. C. Dacey, director of the solid state electronics research laboratory of Bell Telephone Laboratories, reported in a review in *Science* on the unpublished work of W. S. Boyle who had formed luminous plasmas on carbon and iron using a *ca.* 10 J, 1 ms ruby maser pulse.⁹ However, good plasmas were only formed in air, not in inert gases or nitrogen

Laser-induced breakdown spectroscopy (LIBS) has been developed since the invention of the laser in 1960. The push from analytical companies from the beginning started a huge scientific adventure combining analytical science, plasma physics engineering and commercialization that still is in place fifty years later. This historical review of the first five years of laser-based micro-spectrochemical analysis introduces the first measurements and systems for laser micro-analysis and its evolution to conventional LIBS.

and the long pulse duration, non-Q-switched lasers of the time deposited insufficient energy to reliably form vigorous plasmas. Fig. 1 shows the luminous plume obtained by Boyle on a carbon target.⁹ Stimulated by the Bell Labs results, Peter Franken at the University of Michigan began similar experiments with an interest in inducing nuclear fusion, using a ruby maser rented from a nearby instrument company, Trion Instruments.¹⁰ The idea emerged to develop an analytical method based on laser plasmas and Trion formed a relationship with Fred Brech, vice president for research at Jarrell-Ash. Early attempts proved unsuccessful. The occasionally luminous plume was too weak to provide usable spectra. Franken and Brech discussed using an auxiliary electrical spark for added excitation, and Franken then recorded a spectrum of iron obtained by focusing a 5 J ruby laser upon a razor blade mounted between two copper electrodes which were attached to a charged capacitor.^{11,12} Brech and Lee Cross of Trion Instruments reported on the method at the Xth

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cence, laser materials interactions and spectroscopic plasma diagnostics.

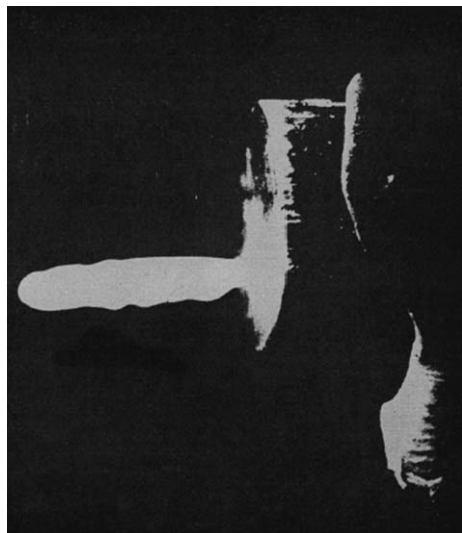


Fig. 1 Jet of self-luminous carbon vapor being ejected from the focal point of a ruby maser beam incident upon a carbon rod.⁹ (Reprinted with permission from AAAS. License # 3111900897722).

Colloquium Spectroscopicum Internationale at the University of Maryland on June 20, 1962. The paper was not published in the conference proceedings but an abstract appeared in *Applied Spectroscopy*¹³ and the work received wide attention.^{14–17} Brech's instrument was quickly commercialized by Jarrell-Ash and announced in the "What's New" column in *Applied Spectroscopy* in October, 1962.¹⁸ The technique was called "Micro-emission Spectroscopy" and marketed as a source unit which could be coupled to any spectrograph, using a ruby laser manufactured by Trion. The advertised cost, without the spectrograph, was \$12 000 (about \$100 000 in 2012 dollars). The laser produced a weak plume of vapor from the sample surface which initiated a breakdown between two electrodes mounted just above the laser focus. Thus this was a laser sampling technique with spark excitation. Fig. 2 shows the first model of the Jarrell-Ash instrument.¹⁹

The first clear description of this instrument was provided by John Maxwell early in 1963.²⁰ By this time it included a Q-switched ruby laser head and the auxiliary spark source was arranged with a pair of carbon electrodes. The laser could be fired at intervals of one minute. Maxwell evaluated the system on a series of geological samples. Much of the early analytical work with the Jarrell-Ash system was carried out by Robert Rosan and various collaborators. Their first paper in *Science* greatly stimulated interest in the technique.²¹ Rosan quickly appreciated the advantages of simultaneous elemental analysis of microsamples from biological tissues and presented spectra of Mn, Fe, Be, Cu, Zn and Ca in 13 different tissue specimens. Several papers were presented at the 48th and 49th annual meetings of the Federation of the American Society for Experimental Biology, held in 1964 and 1965, respectively^{22,23} and at the 1st Annual Conference on Biologic Effects of Laser Radiation, held in 1964.²⁴ In the latter report a spectrum was shown which was taken without the use of the auxiliary electrodes, perhaps the first published spectrum by conventional LIBS. At

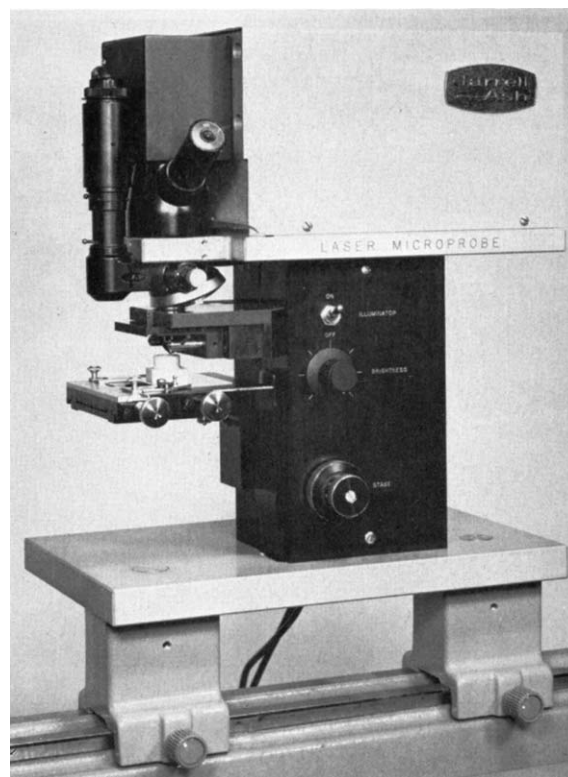


Fig. 2 The Jarrell-Ash Laser Microprobe.¹⁹ (Reprinted with permission from Elsevier. License # 3111900380812).

another of these conferences a 300% improvement in reproducibility was reported due to an improved Q-switch and data were shown from sampling of living tissue including a human finger "without harm".²³ Jarrell-Ash continued to advertise the microprobe, offering literature on the method.²⁵ Early applications included the micro-analysis estimation of 9 elements in bone²⁶ and calcified tissues.^{27,28} R. C. Rosan described various methods for laser microprobe sample preparation.²⁹

Meanwhile, interest continued to develop in using laser induced plasma without the auxiliary electrode excitation. Early in 1963 Runge and coworkers at Ford Motor Company studied the detection of Cr and Ni in stainless steels using a "giant pulse" ruby laser.³⁰ The work was reported in two subsequent papers, the first published reports of conventional LIBS, and included the first published calibration plot. They point out the inherent advantages: "With the laser source, samples can be analyzed that are: refractory in nature, non-conductors, sealed in evacuated or inert atmospheres, or are molten materials situated within a furnace".^{31,32}

At this time work was also taking place in other countries. In particular, Debras-Guédon and Liodec had been studying the application of LIBS for different materials. Their communication to the French Academy of Science presented by Jean-Jacques Trillat³³ on September 21, 1963 and published later,³⁴ showed a laboratory LIBS setup consisting of a ruby laser from the Laboratoire Central des Communications (100 μ s, 100 J, focused to a 100 μ m spot, producing craters 5 \times larger). Optimized conditions for the spectral acquisition included placing

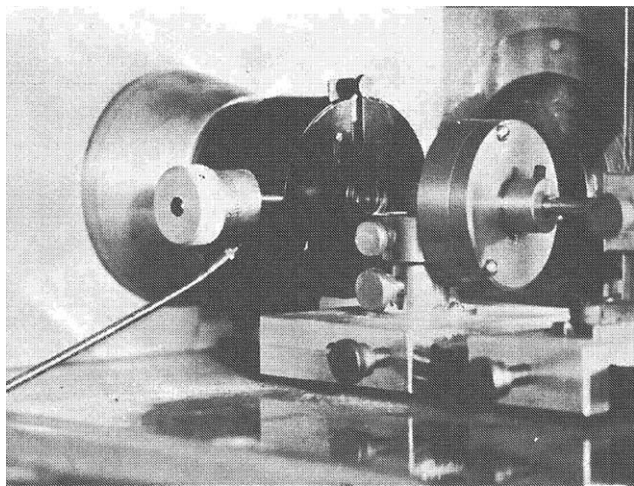


Fig. 3 The setup of Debras-Guédon and Liodec.³⁴ (Reproduced with courtesy permission of Société Française de Céramique, Centre Technique Industriel de la Céramique and Centre National d'Etudes et Recherches Céramiques).

the axis of the plasma parallel to the slit of the spectrometer and using an additional spherical mirror to collect the plasma emission opposite to the slit (Fig. 3). This paper listed the emission from 25 elements (Al, Ba, Ca, Ce, Cr, Cu, Fe, K, La, Mg, Mn, Mo, Na, Ni, Pb, Si, Sm, Sn, Sr, Ti, V, Y, Zn, Zr) and interestingly, as a footnote, mentioned the presence of diatomic bands in the spectrum from CN and AlO. These two molecules were detected during the analysis of carbonate and duralumin respectively, showing for the first time the presence of molecular formation in the plasma by reaction with the ambient atmosphere. The authors noted that the cyanogen bands were not present, for materials other than carbonates, thanks to the absence of graphite electrodes usually needed in spark excitation. They also mentioned that they could not detect Ag, B, Be, Bi, C, Cd, Co, Hg, Li, P or Sb. This was perhaps because of their low sensitivity in the ultraviolet region and the low signal-to-background due to the large unavoidable continuum emission. In their conclusion they mention that they were aware of the laser microprobe at the XIth Colloquium Spectroscopicum Internationale in Belgrad (October, 1963)³⁵ but they considered the use of additional electrodes to be a disadvantage and a further complication to the experimental set up. This French team later published a few quantitative results as well, such as the determination of Cr and Ni in steel.^{36,37}

By 1964, a great deal of fundamental work on laser-material interactions had been carried out. For example, Ready studied the behavior of a laser plasma formed on carbon using high-speed photography.³⁸ Some of the first time-resolved spectra were produced using a ruby laser focused on a wide variety of targets (Al, Cu, Zn, Ge, Sn, W, Au, Pb, glass and ruby).³⁹ The presence of highly ionized species, *e.g.*, Ge IV and Sn IV lines was also reported. Archbold and Hughes also made one of the first estimates of the electron temperature in a "laser-heated plasma" in 1964.⁴⁰ Using a Q-switched Nd:glass laser (10^{11} W cm^{-2}), on a graphite target in vacuum, they estimated a lower limit for the electron temperature of 1.1×10^5 K. This supported the theoretical estimates which had been made earlier by

Dawson⁴¹ who had the foresight to recommend several interesting possibilities for such plasma including controlled thermonuclear fusion and confinement in a magnetic field. W. I. Linlor of Hughes Research Laboratories reported on plasma formed by a 200 mJ, 40 ns ruby laser on Al, Au, C and W targets. Absorption measurements in the plasma indicated that 94% of the laser pulse was absorbed.⁴² Pashinin, *et al.*, studied the breakdown in air of a 50 MW Q-switched ruby laser and reported that over 60% of the incident beam energy was absorbed by the plasma, forming electron densities of the order of 2×10^{18} cm^{-2} .⁴³ J. F. Ready at Honeywell Research Center modeled the interaction of laser pulses on various metals and predicted with reasonable accuracy the crater depth, *e.g.*, 840 μm in Al.⁴⁴ During the decade of 1960–1970 Ready published widely on fundamental aspects of laser-material interactions ultimately with the important application to laser machining and metalworking.

The other major instrument commercialization took place in Germany and was initiated in the laboratories of VEB Carl Zeiss, Jena beginning with the introduction of the LMA instrument at the Leipzig Spring Fair in 1965. This laser micro-analysis

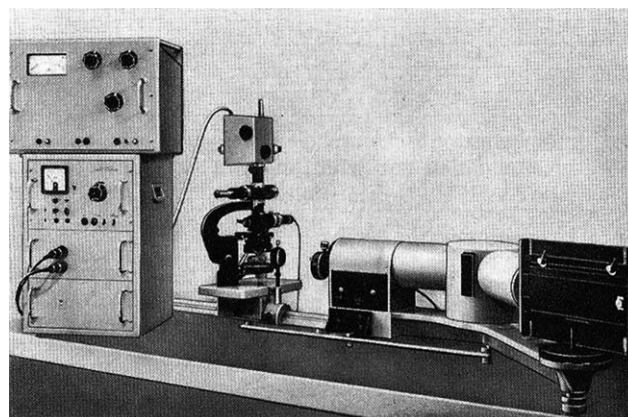


Fig. 4 the VEB Carl Zeiss, Jena LMA, 1964. (Reproduced from ref. 50).

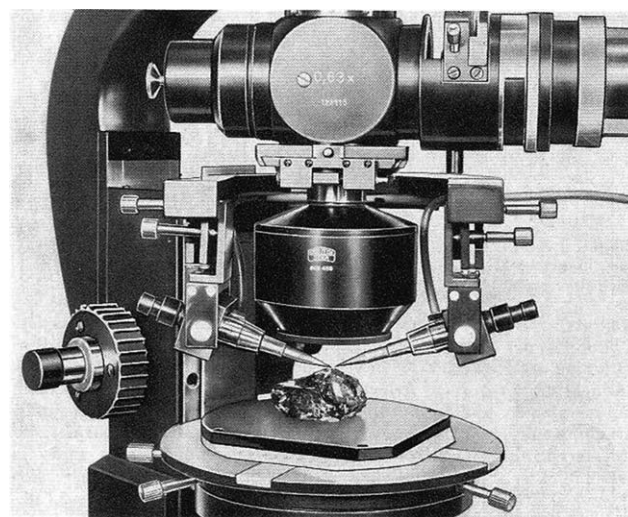


Fig. 5 LMA sample stage, December, 1964. (Reproduced from ref. 50).

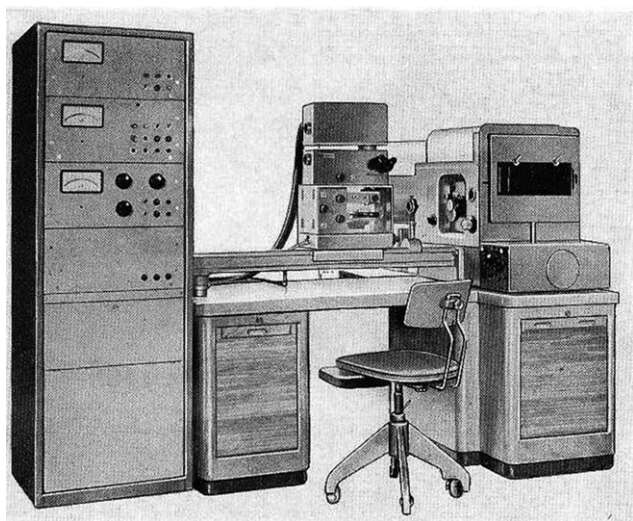


Fig. 6 VEB Carl Zeiss, Jena Model LMA 1, 1965. (Reproduced from ref. 50).

instrument (Fig. 4) used either a Nd:glass or ruby rod laser at 1060 and 694.3 nm, respectively. The first published results appeared in 1965.^{45,46} Fig. 5 shows a close up of the LMA sample stage with a geological specimen and the auxiliary electrodes. By the end of 1965, the improved LMA 1 was issued (Fig. 6)

featuring a maximum laser energy of 1 J, a repetition rate of 3 pulses per minute and a 2 m focal length Ebert plane grating spectrograph.⁴⁷ Numerous studies and practical applications appeared in the following several years. Geological applications naturally attracted much attention and included a semi-quantitative analysis of the meteorite "Pawel" which fell in 1966. This study listed 12 elements in concentrations ranging from 10^{-3} to 3%.⁴⁸ By this time enough work had been carried out to justify the first book on laser microanalysis, by Moenke and Moenke-Blankenburg.⁴⁷ This was followed by a second edition in 1967, a Russian edition⁴⁹ in 1968 and a revised and enlarged English edition in 1973.⁵⁰ The latter edition included results and applications in the fields of mineralogy, geology, metallography, silicates and glasses, archaeology, biology and medicine and forensic science. The chapter on quantitative analysis is instructive even today: "A clear warning must be given not to practice quantitative analysis uncritically..." (p. 204). The importance of well characterized homogeneous standard reference materials was strongly emphasized. Naturally, such work with photographic plate detection was especially challenging. Relative limits of detection, ranging from 10–1000 ppm were given for 44 elements. Absolute limits of detection ranged from 0.1–1000 pg.

During this same period of time an instrument was designed and constructed at the Institute for Spectrochemistry and

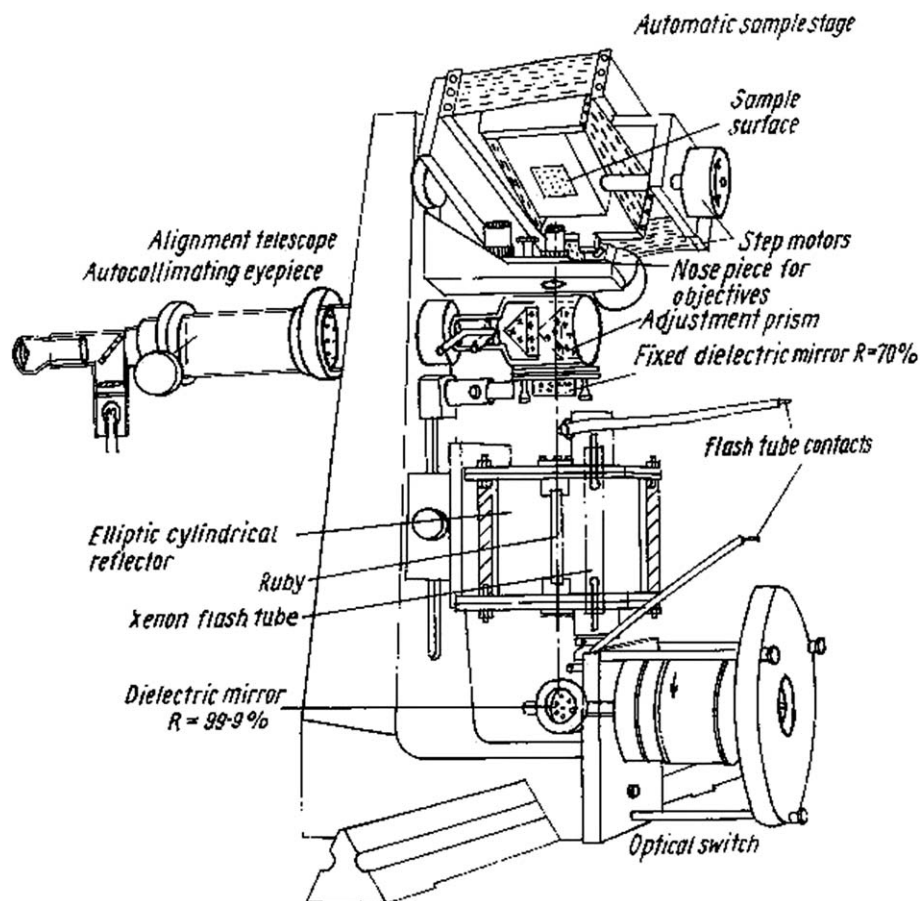


Fig. 7 ISAS Laser Microanalysis Instrument, 1965. (Reproduced from ref. 50).

Applied Spectroscopy (ISAS) at Dortmund, first described by Laqua *et al.* in 1965.⁵¹ Fig. 7 shows a schematic of the design which used a 100 mJ ruby laser operating at 12.5 Hz. No auxiliary electric spark was used. Vanadium, copper, manganese, silicon, molybdenum, nickel and chromium were all determined in steels using an iron line as an internal standard. Detection limits of the order of 0.1% were obtained.

It is remarkable that LIB and LIBS-spark instruments were so rapidly commercialized in these early years. In the 1973 English edition of their book, Moenke and Moenke-Blankenburg provide a list of all commercial laser-microprobe instruments available at that time.⁵⁰ By 1968 three companies had commercial versions: VEB Carl Zeiss (LMA 1), Jarrell-Ash (Model 45) and LOMO, Leningrad (MSL-2) while Shimadzu, CISE and JEOL had developed prototypes.

In 1981, Loree and Radziemski from the Los Alamos National Laboratory introduced the acronym "LIBS" by referring to the breakdown of air by laser pulses during the plasma creation.⁵² This rebirth of the technique brought more attention and initiated a resurgence of research which has continued to the present. However, many of the problems related to accurate quantitative analysis remain unsolved. Standard protocols for routine analytical methodology remain elusive. Only in the last ten years have several companies focused on the development of new LIBS systems, taking advantage of the development of compact lasers, spectrometers and detectors. Very recently large analytical instrument companies such as Thermo Fisher Scientific (with the Blast Hole Xpert Online Analyzer) and TSI have become interested in the development of commercial LIBS instrumentation. LIBS is now included in the ChemCam Instrumentation Suite on the Curiosity mission on Mars.⁵³ Its role is to classify rocks using spectral libraries, evaluate sample heterogeneity (at the millimeter scale), depth profiles in the rocks, identify possible organic materials (*via* the presence of P and N or even CN molecular bands), identify water on surfaces (*via* OH emission) and any eventual hazardous elements (Cd, As, Pb and Be). Fifty years later, where does LIBS stand? A major driving force for the continued study of the technique has always been the need for rapid micro-analytical measurements. This was the primary focus of the early commercial instruments. However the more successful applications have emphasized other unique capabilities of LIBS which provide for stand-off analysis, portability, measurements in harsh environments and direct analysis of samples which are difficult to dissolve. Given the success of laser ablation-ICP-MS for micro-analysis, the applications of LIBS in this realm may very well diminish while applications in a wide range of specialized areas continue to grow.

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