Terahertz Pioneer: Frank C. De Lucia

"The Numbers Count"

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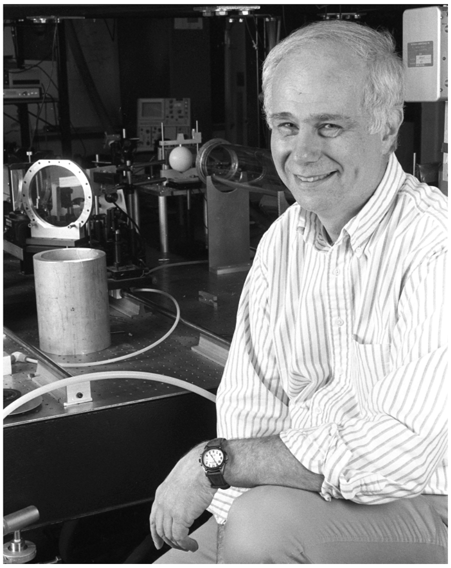
THE IOWA TOWN of Mt. Pleasant was home to a family run restaurant and hotel owned by Frank De Lucia Sr., a trained chemist who became a self-described "innkeeper and entrepreneur" after World War II. Perhaps because of his background, the restaurant was host to some notable science patrons, including Mt. Pleasant native son and personal friend, Jim Van Allen—long time Physics chair at University of Iowa, early rocketeer, and, with an instrument on Explorer I in 1958, discoverer of the charged particle belt around the Earth that bears his name.

Working in the kitchen, and wherever else he was needed, was young Frank Charles De Lucia Jr. A very hands-on teenager, Frank was interested in electronics (he would claim—in the electromagnetic spectrum), and he was an active HAM radio operator and tube tinkerer. He once took all the tubes left over from a tube swap in his family's electronic organ and strung them together in an attempt to generate enough multiplied power to break the local HAM radio club high frequency source record! Although not successful, he learned a valuable lesson in frequency scaling when he tried to make the resonant coils work at frequencies at which they were no longer lumped elements but significant fractions of a wavelength.

In high school, De Lucia was more interested in playing sports than in academics. However, by what would turn out to be a fortuitous reversal of fortune, he broke his leg during his sophomore year and lost the chance to keep playing. At this point, the Dean of local Iowa Wesleyan College, and a friend of De Lucia's father, offered Frank an opportunity to apply to the university rather than go back to High School. De Lucia enrolled in 1960, intending only to stay one year, and then to transfer to Iowa State to become an electrical engineer. He liked the school however and ended up staying through to his graduation. Van Allen was also a graduate of Iowa Wesleyan, and Frank fondly recalls a memorable commencement address by noted rocket scientist, and van Allen compatriot, Wernher von Braun.



FRANK C. DE LUCIA

At Iowa Wesleyan, De Lucia collaborated with their only physics professor, John Edie (who also doubled as the mathematics teacher). Edie was an enormous influence, and undertook a brutal class load (four courses plus labs each semester), but taught with great skill from top of the line textbooks. He also encouraged Frank to apply for a US National Science Foundation summer program for students at smaller colleges, that brought De Lucia to the University of Kansas, and exposed him to a much broader range of physics.

After finishing his degree at Iowa Wesleyan, De Lucia accepted a graduate fellowship in Physics at Duke University, Durham, NC. With some pangs about leaving the Midwest (to which he would later return), and perhaps with some regret about not attending University of Iowa, where Van Allen was teaching, De Lucia arrived at Duke in the fall of 1964. Having not had quite as many physics courses at Iowa Wesleyan as was customary for other graduates attending Duke, he spent a year immersed in Hamiltonians and the Schrödinger equation before he teamed up with pioneering microwave spectroscopist Walter Gordy [1].

Gordy, more than any other spectroscopist of his time, pushed the frequency limits of microwave technology in his quest to conquer the THz bands [2]. His first high frequency results appeared in 1953 and 1954, with W. C. King [3], [4]. Teaming with detector guru Charles Burrus, Gordy broke the 1 mm wavelength barrier with the observation of spectral absorption in OCS at 389 GHz using harmonics from a K-band (18–26 GHz) reflex klystron in a Burrus-made whisker contacted multiplier

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[1] F. C. De Lucia works and lectures tirelessly from his home base in the new Physics building at the Ohio State University campus in Columbus, OH, USA. His 4th floor corner office overlooks the sprawling west and north campuses of the university and takes in the enormous football stadium complex of the Ohio State Buckeyes. His labs are close by and fully occupied with current research projects. Professor De Lucia treated for breakfast and then graciously sat with me on an overcast Friday morning in August to conduct the interview that forms the basis for this article.

device [5]. Burrus later went on to Bell Laboratories and became one of their first millimeter-wave radio astronomy receiver specialist [6].

When a group at a rather large and well known industrial laboratory published a paper in 1966 [7], claiming to have observed harmonically produced signals up to 1.2 THz, Gordy was notably upset at being usurped in his quest to be first to do THz spectroscopy. It was De Lucia, as a second year graduate student, who realized that the observed spectral signatures were not actually of THz origin, but were in fact, much lower frequency alternate mixing products produced in the instrumentation. This realization and subsequent publication [8], endeared De Lucia to Gordy, and was the beginning, for De Lucia, of a life long career in THz science.

In these early days of THz spectroscopy there were no commercial components and everything in the lab was put together by hand (*not so very much has changed today! Ed.*). Sources were generally composed of semiconductor Schottky diode or p-n junction point contact frequency multipliers driven by reflex klystrons in the microwave bands [4], [5]. Detectors at the time were also composed of point contact rectifiers (whisker-contacts), but were much less robust than multipliers (smaller contact areas and purer barriers were needed for high sensitivity). Detector performance was dependent on a number of uncontrollable parameters having to do with junction area, contact pressure and Schottky barrier purity—which degraded with use. In Gordy's lab at the time, the detector life was measured in hours! The idea of spending the equivalent of months looking through a microscope and tediously assembling point contact rectifiers in order to complete measurements on a few spectral families, was not appealing to De Lucia, who decided he might graduate sooner if he spent his time working on other potentially more robust detector technologies.

MASERs (microwave amplification by stimulated emission of radiation), had been demonstrated at 24 GHz using gaseous ammonia in the early 1950's [9], predating the demonstration of their optical equivalent—LASERs by more than five years [10], [11]. They were being used as low noise microwave and millimeter-wave amplifiers for spectroscopic purposes since their demonstration as such by Gordon, Zeiger, and Townes [12] in 1955.

Using the molecular beam MASER principles, De Lucia and Gordy published their first >100 GHz MASER paper in 1969 [13], doubling the previous MASER record at 88 GHz set in 1961 [14]. It was based on the $J = 2 - 1$ transition of HCN at 177.2 GHz, and they used it both in oscillator and amplifier mode to measure spectral parameters of HCN and DCN. Shortly afterwards, De Lucia and Gordy demonstrated a submillimeter-wave MASER, based on a transition of D_2O at 316 GHz. They described it in a classic paper [15], in what became the first well publicized Submillimeter Wave conference, held at Brooklyn Polytechnic Institute, NYC in April 1970. All the Duke University high frequency MASER results were later summarized by Garvey and De Lucia in [16].

As the experimental data were coming out, accompanying theoretical work with Duke colleague Bob Cook (co-author of the popular Gordy spectroscopy text [17]), led to the widespread use of angular momentum theory for describing nuclear hyperfine structure [18]. This became De Lucia's most requested reprint—he personally sent out more than 500 copies to colleagues who wrote in.

In the mid-1960s, infrared researchers like Rollin and Kinch [19], Putley [20], and Kimmitt [21], [22], had discovered Indium Antimonide, InSb, photodetectors and were applying them at longer wavelengths as helium cooled bolometric detectors and later as heterodyne receivers [Phillips, [23], [24]]. For De Lucia, these detectors were miraculous! Not only did they have sensitivity equal to, or better than, the point contact rectifiers, they performed consistently through measurement after measurement. He successfully lobbied Gordy to purchase one of the new detectors, and after seeing how it worked, then made his own—right down to the glass helium dewar. By combining the InSb bolometers with the tunable microwave klystrons and the high harmonic multipliers, De Lucia quickly developed a robust, flexible, millimeter and submillimeter-wave spectrometer [25]. This combination of components became a mainstay for the THz gas spectroscopy community that persisted for more than two decades [26]. It continues to be used today with only minor variants in the components [27].

At the same time as De Lucia was setting up the Duke spectrometer, University of Reading, UK, theoretical chemist James Watson [28] (later at Herzberg Institute of Astrophysics, Ottawa, Canada, and Royal Society Fellow), was working to refine quantum mechanical models for molecular rotors to get accurate spectral line predictions in the presence of asymmetries. Before this time, models had more fitting parameters than were determinable from observed energy levels, and would generally blow up. Watson developed a reduced Hamiltonian that better matched the number of observational free parameters to the amount of information contained in a rotational spectrum. Bob Cook, then a post-doc at Duke, was quantifying Watson's models and developing a computational program for accurately predicting spectral line features. As Cook and De Lucia worked with, and extended the Watson models, they had to add higher and higher terms to the Hamiltonian to match up with observations. Above the sixth order, they had no physical basis for the added constants and resorted to numerology (looking at trends), to establish the highest order coefficients. At one point De Lucia coined the term "dectic" to articulate the tenth order coefficient. He thought it would be good to check the term in the Oxford English Dictionary first, did not find it, and decided to use it. Apparently it never caught on, because the term is still absent from the OED!

Before this period, most submillimeter wave spectra were measured without detailed frequency foreknowledge. This required a rough line frequency prediction followed by a "hunt and peck" type search for the actual absorption dip. Armed with these new methods [25], [26] and a robust submillimeter spectrometer, it was possible to generate accurate full-family absorption line catalogs of many small fundamental molecules at submillimeter wavelengths. Along with Paul Helminger, Cook and De Lucia worked out and measured the submillimeter-wave spectra of the hydrogen halides [29] and a wide range of light asymmetric rotors including the various isotopes of water and hydrogen sulfide [30]–[33], as well as prototypical internal rotor structures such as methanol [34] and hydrogen peroxide [35].

By the early 1970s, after the Duke team had well-established instrumentation and good theoretical models, De Lucia started to feel that the submillimeter-wave molecules of interest would all be rapidly explored, and he might have to find a new field to specialize in. At about this time, University of Illinois offered De Lucia a faculty position in Electrical Engineering, which he was excitedly preparing to accept. However Gordy, now in his 60's, intervened. Unwilling to lose his prized student, he offered to turn the spectroscopy laboratory over to De Lucia in stages leading up to his, Gordy's, retirement. De Lucia decided to stay at Duke, and indeed, became director of the Microwave Laboratory in 1979, while still an Associate Professor.

In the late 1970s, after the pioneering work of Claude Woods (see discussion in [36]), De Lucia and other microwave spectroscopists became very interested in ions, many of which were then being discovered in the interstellar medium by submillimeter wave astronomers [36, p. 267]. Unfortunately, ions are very difficult to produce in the laboratory, with the most favorable, HCO^+, occurring at only 1 part in 10^6 in low-pressure gas discharges, and were therefore extremely hard to detect.

Like many spectroscopists, De Lucia was using gas discharge lasers to generate submillimeter wave signals. He started using his spectroscope to study the discharge dynamics, and particularly the temperature distributions in the gas [37], [38]. He noticed that as the discharge voltage was increased in the cavity, the lasing decreased, rather than increased, and he determined, through his submillimeter-wave diagnostics, that this was due to a temperature rise in the gas itself. Molecular absorption is extremely sensitive to temperature (goes as $T^{5/2}$) and so the ion signal was dropping rapidly as the pump power increased. De Lucia also noted that the temperature was not uniformly distributed throughout the discharge tube and was higher in the center than at the walls.

Accordingly, the Duke team thought that they could reduce the ion temperature by going to a smaller diameter discharge tube, and by adding a magnetic field to keep the ions from hitting the tube walls. They constructed a narrower discharge tube with a surrounding solenoid to generate the magnetic field. In their first experiments, while graduate student Wayne Bowman [39], [40] ran the spectrometer, De Lucia controlled the discharge, and they watched the detected ion signal on the oscilloscope. As the magnetic field was increased, there was a Eureka moment when the detected ion signal scaled off the scope, increasing by more than 100×!

As it turned out, this was not due to the smaller diameter tube. Rather, it was because the ions were being generated not in the long positive column of the discharge tube, but rather in the normally short negative glow region of the exciting cathode. Without the magnetic field present, the electrons in the negative glow region were scattered away as they entered the positive column. With the field, they were guided along until the negative glow extended the full length of the discharge tube. The enhanced interaction created an enormous signal gain—resulting in more than a 10,000 fold decrease in integration time (integration time decreases as the square of the signal power). In a classic methods paper with Eric Herbst (now at University of Virginia), Grant Plummer (now at Enthalpy Analytic Inc., Durham, NC) and graduate student Geoff Blake (now at Caltech, Pasadena, CA) [41], De Lucia and company described their new ion generation, confinement and enhancement technique, that is still the preferred ion spectroscopy method used today.

De Lucia continued to take advantage of both the instrumentation and modeling expertise at Duke and along with long time colleagues Eric Herbst, Paul Helminger and students Todd Anderson and Wayne Bowman, began to make measurements and construct accurate models of ions and molecules that were important to astronomers—who were now dominating the THz spectroscopy applications area. They were the first to theoretically model the spectrum of methanol to experimental accuracy [42]. Similarly, they accurately modeled hydrogen peroxide [43] and other relevant molecules with internal rotors [44]. These more accurate quantum mechanical models, coupled with experimental verification, allowed complete catalogs of lines to be built up for the first time.

Although these quantum mechanical models were instrumental in allowing astronomers to focus their submillimeter-wave observations at key frequencies, they did not usually predict the very large numbers of excited vibrational states that can show up in the measured astronomical spectra. If these uncataloged transitions in the excited vibrational states occur at the frequency of a target species, they can be misidentified, leading to erroneous "discoveries." It would take many more years (until just recently), and a combination of extremely accurate swept frequency measurements with temperature dependent intensity and line shape fitting to do significantly better at modeling all the excited state interaction signatures—but we are jumping ahead.

By the mid-1980s De Lucia was again looking for some other interesting problems to tackle. Atmospheric chemists were extremely interested in the effect of molecular collisions on spectral line shape, because the line width was used to back out the surrounding pressure and hence the altitude of the observed signal. However it was very difficult to model collisional broadening to reasonable accuracies in complex, or even in simple molecules. Most of the data used in the atmospheric models was acquired by a very tedious set of laboratory measurements over varying temperature and pressure.

Astronomers on the other hand, had the inverse problem. They were often dealing with an environment at low temperature and low pressure. Individual state-to-state collision rates were hard to model and no laboratory data were available to compare with the calculations. In addition these cold collisions were of significant interest to physical chemists because the number of energetically "open" collision channels was relatively small and it was possible to do "exact" quantum scattering calculations.

De Lucia figured that one way to get at the molecular interactions directly, was to cool the gas spectral chamber with liquid helium, fill it with gaseous helium, and then inject trace amounts of warm, spectroscopically active molecules. In theory the molecules would collide several thousand times with the cold helium gas before hitting the chamber walls. Since they would cool off within the first hundred or so collisions, the collision effects could be recorded. Having a well calibrated temperature and pressure environment to work in allowed De Lucia and a favorite graduate student, Dan Willey (now at Allegheny College, PA), to accurately measure quasi-bound states and interstate hopping as the injected gas cooled through direct collisions [45]–[48]. This very effective *collisional cooling* technique has been rediscovered in recent times and is often referred to by the atomic physics community as *buffer gas cooling*.

In the late 1980's De Lucia was appointed Chair of the Physics Department at Duke and became more acutely aware of the role of Physics in the larger research picture at the university. As a result in 1990, when Ohio State University came looking for someone with a greatly expanded vision of the role of a Physics department it its future, De Lucia was interested. While he insisted that he preferred to be a faculty member only, the lure of the Midwest—the environment of his youth; the substantial new resources that would be at his disposal (including a new Physics building); and the promise of significant administrative support in his position as Chair; convinced him to uproot the Gordy Microwave Lab at Duke and bring it *en masse* to OSU.

He planned the move so carefully and completely (*a De Lucia trait, Ed.*), that when Duke colleague and Physics Professor Bobby Guenther told De Lucia the relocation would cost him at least a year of research, he responded by sending Guenther a spectra from Columbus two weeks after packing up his instrument in Durham. As anyone who has moved or set up a large lab knows, this is a very impressive feat. By the fall of 1990, the Ohio State University could boast of having one of the world's premier submillimeter wave spectroscopy labs with significant historical links back to the early 1950's.

At OSU, De Lucia felt he had better facilities for instrument construction and a much wider range of collaborative research opportunities as well. As THz technology became more available and robust, he began to focus more on applications. Whereas in earlier times, THz laboratory spectroscopy was a necessary precursor to observational instruments, by the 1990's astronomical receivers at mountain top observatories such as the Caltech Submillimeter Observatory [24], high altitude balloon and aircraft platforms like the Kuiper Airborne Observatory, and the first of several heterodyne receiver based space satellite instruments [49] began to reverse this trend. Spectral data was now being collected that contained many uncataloged lines, and spectroscopists needed to go into the laboratory to try and assign these lines to known, or unknown, molecules [36].

The experimental and analytic work that produced precise quantum mechanical models had to be appended to include all the excited vibrational states that were now being detected by these much more sensitive astronomical instruments. De Lucia began to tackle this problem experimentally by taking advantage of his own laboratory experience and the enhanced fabrication facilities at Ohio State. He constructed a spectroscopy instrument capable of producing the entire complex rotational and excited state spectra of gases of interest to astronomers, as well as many other physical chemists. He called the instrument FASSST for Fast Scan Submillimeter Spectroscopy Technique [50], [51]. The early FASSST technique could rapidly record extremely high resolution, well calibrated spectra of isolated or mixed gases over wide spectral bandwidth by electronically sweeping narrow line width backward wave tube oscillators, and detecting with a very sensitive helium cooled InSb bolometer. The backward wave oscillators were not phase locked, and although they drifted, they did so relatively slowly and their spectral purity was very good. The strategy was to scan very rapidly, and simultaneously record the spectrum and the fringes of a very long Fabry–Pérot cavity. One could then use a few chosen lines to calibrate the cavity and use software to calibrate the rest of the spectrum.

FASSST had a major impact on the accurate analysis, and ultimately fingerprinting of unknown gas mixtures, whether recorded in the cool regions of space that surrounds newly forming stars [52]–[54] or generated in the laboratory for diagnostic purposes [55]. With the addition of absolute intensity calibration and variable temperature measurement, De Lucia calls this analysis approach "*Complete Experimental Spectra*". While experimentally challenging, it eliminates the extremely difficult quantum mechanical assignment of vibrational modes, and the detailed analysis of the perturbed excited vibrational states. It also provides "3-D"—frequency, intensity, and lower state energy—as a basis for quantum mechanical mode assignment spectroscopy [56].

As solid-state sources and more sensitive room temperature detectors became available, De Lucia took advantage of these components to expand the available bandwidth; substantially reduce the cost, power and volume footprint; and greatly improve the reliability of the FASSST instrumentation [57], [58]. At the same time large quantities of new astronomical and atmospheric data were pouring in from the Herschel Space Telescope, the Atacama Large Millimeter Array and the Aura Microwave Limb Sounder. Potential THz applications for chemical threat detection and security also pushed their way into the public eye. These application areas were a perfect fit for De Lucia's *Complete Experimental Spectra* approach to THz line analysis. He very effectively demonstrated the accuracy of the technique on recent data from the ALMA telescope array [54].

In addition to his work on THz spectral techniques, De Lucia has long been interested in THz imaging. He was the first person to produce a high resolution THz image [58], back in 1987, by using a helium bolometer to record a now famous (at least in the THz community) passive image of Donald Bittner (now at Schafer Livermore Laboratory, CA). He is also well aware of the many pitfalls of trying to use THz frequencies for imaging applications and has long been an outspoken advocate of not overselling the inherent capabilities of this wavelength range (see for example [59]–[61]). His most recent work in this area nicely summarizes and compares THz imaging techniques [60] and introduces new methods to improve contrast [61].

As De Lucia's interests have spread out from cataloging lines and developing detailed quantum analysis techniques, he has focused more and more on THz instruments and spectral techniques that have commercial or civilian, rather than just science applications.

For the past ten years De Lucia has been very involved with various US agencies in trying to sort out methods for chemical fingerprinting, with application to security and threat detection, including remote sensing at atmospheric pressure [62]. However, he is careful to apply his very analytic scientific methods as he begins to propose, evaluate or tackle these often over publicized THz applications. It is not enough to simply demonstrate that some new instrument can detect a THz spectral signature. The real question is whether it can do it better, or at least with the same level of quantitative performance as existing instruments. "*Numbers count*."

The same mathematical rigor and careful experimental verification that allowed De Lucia and his colleagues to first solve the asymmetric molecular rotor, must also come into play when applying THz signals to imaging, material fingerprinting, remote sensing or any other application where the detailed physics does not precede the measurement. De Lucia's many presentations at the OSU International Symposium on Molecular Spectroscopy, and invited talks at other conferences and workshops appear on his web page [63] along with tutorials on terahertz science, techniques and applications, historical data and classroom notes. It is a great place to start learning about the field.

In discussing some lessons learned and advice to young THz researchers, De Lucia offers the following: Attracting new people to a field adds more opportunities, rather than making the field more competitive. THz has plenty of room for more good ideas. Although you do need to sell your ideas, don't over do it, and be honest! Hold to quantitative standards, not qualitative ideas.

As we concluded our interview, and Professor De Lucia showed me his extensive laboratory facilities as well as several of his current projects, I could not help being impressed by how his approach to THz science permeated his approach to life: Careful, thorough, thoughtful and analytic. Never too concerned about pursuing a course that might clash with convention—like applying to college without a high school diploma. Always remembering to apply the numbers, because *the numbers count*.

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