Terahertz Pioneer: Fritz Keilmann

"RF Biophysics: From Strong Field to Near Field"

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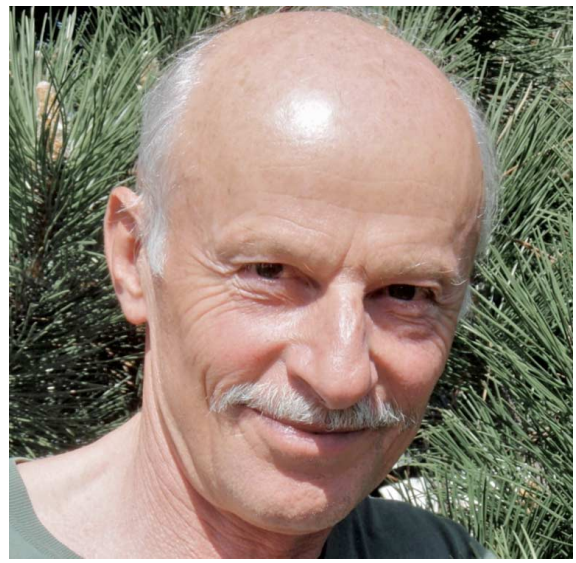
FRITZ Keilmann's[1] family home in the hills above Lake Ammer, a resort area of upper Bavaria, was filled with music. His father was a conductor and a conservatory music professor. His mother was a violinist and teacher. Fritz and his three siblings made up a quartette of flute (Fritz), piano, cello, and violin. As added income, the Keilmanns, along with several befriended professional musicians, gave formal home concerts, attracting 60 or more paying people on a monthly basis. This entrepreneurial family spirit was infused in Fritz at an early age and was to serve him well over his long career in science.

Despite the ravages of post-World War II Germany, Ammersee was far enough removed from nearby Munich to have offered Keilmann a sportive and musical Alpine upbringing—raising parallels in the interviewer's mind of the von Trapps and the *Sound of Music*. Fritz's great grandfather was an engineer, and perhaps some of his passion for precise measurements was passed down, along with the family gift for music. At age 14, Fritz convinced a representative from the Munich meteorological office to relocate the local village weather station to his own house, when he offered to take over from a recently deceased family friend as the resort area weather recorder. This experience launched his long-term interest in science. His high school thesis at Schondorf on Ammersee, was on thunderstorms, and for some time he had thoughts of an advanced degree in meteorology. However, Schondorf did not offer any formal science classes. He had two opportunities to dabble in science before he entered university; once for a six-week course in atomic physics with two other boys and a tutor at Schondorf, and again in a four month Fellowship at Radley College, Oxford, U.K., that he won when he was 16. It was at Radley that Fritz first experienced competition, as he worked his way through the Oxford curriculum along with 500 other young boys.

At 19, Fritz won a Bavarian Fellowship that allowed him the freedom to choose his university and his curriculum. He left



FRITZ KEILMANN

Ammersee for Ludwig Maximilians University (LMU) in Munich to study meteorology. Within a year he realized that becoming a meteorologist was not as interesting as he had envisioned as a boy of 14. He decided to capitalize on his brief exposure to physics, and switched to the Technical University of Munich in 1964. At TUM, Fritz became very interested in biophysics and completed his Diplom Physik (Masters level) on the microwave properties of hemoglobin [1] in 1967. He worked with an X-band Mach–Zehnder[2] interferometer to acquire the spectral transmission data on oxygenated and deoxygenated equine blood as well as human blood solutions. He "fondly" recalled the grisly work of having to acquire fresh samples from a local slaughterhouse, where he would personally have to hold the collection jar under the horse's neck.

The microwave blood measurements were actually conducted nearby at what was then still a private research institute that had been set up in 1961 by Werner Heisenberg and the Max Planck Society—the Institute for Plasma Physics in Garching. It was fully incorporated into the Max Planck Institutes in 1971. The Garching facility was home to many microwave physicists who were conducting experiments on plasmas as part of a large ongoing fusion energy program. After his Diplom, Keilmann was asked to stay on at the institute and work on plasma diagnostics employing his interferometer experience. He began using CO_2 (10.6 μm) [2] and HCN (337 μm) [3] laser

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[1]On a bleak and snowy day in March, I met a fit and vibrant **Fritz Keilmann** as he was just moving into his new laboratory in the Physics building at the very historic downtown campus of the Ludwig Maximilians University of Munich, founded in 1472 and at one time, home to the laboratories of Röntgen, Wien, Boltzmann and Sommerfeld. The afternoon was considerably brightened by our long and intense discussion, interrupted only briefly by a sinfully rich, and equally delicious piece of local pastry. The 20+ pages of notes and the many interesting stories that came between, constitute the bulk of this article on an individual career that is broad in scope, filled with innovation, and still going strong after 45 years.

[2]Ludwig Zehnder was a student of Wilhelm Röntgen and co-invented the interferometer that bears his name with Ludwig Mach in 1891 (von L. Zehnder, "Ein neuer Interferenzrefraktor," *Zeitschrift für Instrumentenkunde*, Verlag von Julius Springer, Berlin, pp. 275–284, Jan. 1891).

sources to develop IR interferometers that could characterize electron density by variations in the refractive index [4], [5]. He also developed a novel Schlieren[3] technique, in which he used the plasma as a beam splitter and imaged the spatially distorted IR signal and reference rays onto a photographic (liquid crystal) detector to directly "see" the electron density variations [6]–[8]. This work served as his PhD thesis, which he completed at Garching in 1970.

At this time sensitive point contact diodes began opening up the submillimeter-wave and infrared spectrum. Keilmann was excited about replacing his doped Germanium cryogenic detectors with these new room temperature devices, which he had read about in papers from a laser group at MIT [9]. He wanted to learn more about these devices (which were to become very important for his later work on near-field imaging) and was anxious to do so by attending his first major international event, the *Sixth International Quantum Electronics Conference (IQEC)*, being held in Kyoto, Japan, in September 1970. He also hoped to give a report there on his plasma diagnostic experiments. Unfortunately his paper was rejected, and Max Planck was not able to fully fund his trip without a paper to present. However he was told that if he could raise half his travel funds, Max Planck would give him the remaining portion. Always the entrepreneur, Keilmann found out that if he wrote up short articles on leading laser scientists at the conference, a journalist sponsor would pay him for each line of published text he could produce, and more for photographs. With camera in hand, he attended the conference and interviewed many attendees, including Ali Javan (co-inventor with William Bennett of the gas laser in 1960), Charles Townes (1964 Nobel Prize in Physics), Nicolaas Bloembergen (1981 Nobel Prize in Physics), and other notables. As a result of these interviews, and his interest in "whisker" diode detectors, Keilmann not only paid for his travel, but also was invited by Javan to work as a post-doc at MIT in 1972/1973.

At MIT, Keilmann and the Javan team were working on direct frequency measurements of IR lasers at wavelengths as short as 3 μm, amongst other topics [10]. A key experiment was a more precise measurement of the speed of light. However, before they could accomplish the measurement, a horrible accident—the death of a student by electrocution from a gas laser power supply[4]—closed the lab for three months. The group's competitors—Ken Evenson, Joe Wells, and others—at National Institute of Standards and Technology, Boulder, Colorado, ended up beating them to the chase, with their ground breaking measurement of a 3.39 μm methane-stabilized HeNe laser in 1972 [11].

In yet another MIT story, the laser lab was having a visit from some high level Air Force people who were looking for interesting research projects to invest in (*if only it were still so easy…voc. Ed*). Keilmann suggested they try an experiment to lase humid air using optical pumping [12]. Benjamin Lax (head of the MIT Francis Bitter Magnet Lab) had an unopened pulsed CO_2 laser that no one yet had time to set up. With help from the Bitter Lab's, Ken Button (later editor of the book series and journal, and chair of the long running conference series on Infrared and Millimeter Waves – now *IRMMW-THz*), Keilmann unpacked the laser, which could produce several joules/pulse, and ran it through a single pass gas cell containing low pressure deuterated water vapor. As he watched the gas fluoresce, he held up a homemade whisker-coupled point-contact diode detector on the output side, and gleefully observed submillimeter-wave pulses [13]! These early observations of optically pumped D_2O were reported at the *First International Conference on Submillimeter Waves and Their Applications* [14] and ended up in Coleman's well-read review paper on IR lasers in 1974 [15, ref. [10]].

Keilmann returned to Germany in 1973, where he was offered a position at the newly established Max Planck Institute for Solid-State Research in Stuttgart. His interests were shifting away from plasma physics and towards basic infrared solid-state physics. Ludwig Genzel, the founding director, already had a thriving FTIR spectroscopy group at Stuttgart, and Keilmann set himself apart by working with coherent high power optically pumped gas laser sources[5] (carried over from his MIT experiences). He began by focusing on bulk nonlinearities in liquids and solids, which he could excite with his very strong pulsed fields. With a 5 m long CO_2 cavity and a 10 m long gas cell, he was able to cover almost the entire submillimeter-wave region with high coherent power.

Early experiments at Stuttgart involved generating submillimeter-wave acoustic phonon pulses (280, 407 and 670 GHz) on superconducting films and measuring their scattering properties [16]. With his high field lasers, Keilmann was able to perform the first infrared saturation spectroscopy[6] experiments on solids, observing "hole-burning" spectral dips in germanium [17]. He verified for the first time, the existence of a coherent saturation dip predicted by Chebotaev [18], on top of the Lamb dip. At the time, femtosecond lasers did not exist, and this nonlinear frequency-domain technique was the only way to get at the ultra-short population difference (T_1), and dipole (T_2) lifetimes of holes in Ge, of 600 *fs* and 12 *fs*, respectively [19]. These experiments secured him a permanent research position at Max Planck in 1977.

[3]Indirect visualization of density or index variations through distortions of collimated light passing through a transparent medium. Generally attributed to German physicist August Toepler who developed the technique to identify defects in glass lenses.

[4]S. K. Singh was a starting graduate student in Physics at MIT, when this tragic accident occurred in mid-October 1972. Keilmann and others found him lying on the floor of the lab upon their return from lunch. Apparently he had touched the 4000 V electrode while performing an adjustment on the gas laser. *Boston Globe*, pg. 36, October 18, 1972.

[5]The first optically (CO_2) pumped far-IR lasers were pioneered by T.Y. Chang and T. J. Bridges at Bell Telephone Laboratories in the late 1960's. Chang actually came to Stuttgart in 1979 under a Humboldt Fellowship to work with Keilmann and his very broad coverage high power optically pumped far IR lasers. While he was in Stuttgart, Chang received a call to come back immediately to Bell where his lab was being disassembled in his absence. Unfortunately upon his return he found he had been transferred to another division and his research on optically pumped lasers came to an untimely end.

[6]Saturation spectroscopy involves a strong pump beam and a weak probe beam superimposed in the sample. The pump produces a population change which, in inhomogeneous media, effects a "spectral hole-burning" that can be measured by the probe as a spectral dip (*Lamb dip* in gas lasers). Its shape and width yield lifetime information on both phase (T_2) and energy (T_1) relaxation processes. The technique is generally applicable to gases and was applied to a solid for the first time in these experiments.

In 1975, Herbert Fröhlich[7] came on a visit to Max Planck Stuttgart, where he had an honorary directorate position. Fröhlich's biophysics lean [20] struck a chord with Keilmann, who had spent his early years in Munich working on hemoglobin. After helping Keilmann understand some of the high dipole moment properties of holes in germanium, Fröhlich, who was aware of Keilmann's background in RF, interested Keilmann, in a series of experiments on the biophysical effects of microwaves to test his, Fröhlich's, conjectures on vibrational condensation [21][8]. These started with some spectral measurements of biomolecules in the far-IR [22] and later moved on to experiments to look for low power microwave resonance effects [23].

Keilmann was fortunate to link up with former student colleague and friend Werner Grundler, who was at the National Research Center for Environment and Health in Munich, doing very careful radiation exposure measurements on yeast colonies. Together they started a series of extensive measurements on the effects of microwave exposures on yeast growth rates [24]–[29]. A comprehensive symposium [30] on the subject of biological coherence (and later a book [31]), chaired by Fröhlich and sponsored by IBM Germany, was held in Bad Neuenahr in 1982. At this meeting Grundler and Keilmann presented results on resonant effects in yeast colonies at 42 GHz with a frequency specificity of only 8 MHz [27].

Although Keilmann concluded that these experiments demonstrate genuine non-thermal effects in biological systems, some observations pointed to a two-level system saturation rather than to Fröhlich's conjecture of direct resonance. Keilmann formulated an alternative hypothetical mechanism [32], [33] wherein microwaves influence enzymatic reactions by populating zero-field-split spin sub-states, an hypothesis which is still being investigated[9]. He and Grundler's last paper on the subject [34], is particularly well cited.

[7]Herbert Fröhlich was a German born, U.K. theoretical physicist and a student of Arnold Sommerfeld at Ludwig Maximilians University. He was a pioneer in the physics of dielectrics and metals, and in the nonlinear dynamics of nonequilibrium systems, and thus, interested in fundamental biophysics. As a Jewish scientist, he had fled Germany in 1933 for the Soviet Union and then had to flee again to U.K. in 1935 at the start of Stalin's Great Purge. He worked for Neville Mott at Bristol and then became chair of the physics department at University of Liverpool, however he held several visiting appointments at universities and laboratories in Europe, including Max Planck. At this time, Fröhlich was most influential for his theories of quantum coherence. This is what interested Keilmann in beginning a microwave biological study.

[8]The idea that groups of excited molecules can act together to trigger a ground vibrational state similar to a Bose–Einstein condensate.

[9]Since the key paper is not available on line, a brief explanation from Keilmann of the hypothesized mechanism for explaining the yeast observations is as follows. It is based on the ubiquitous transient presence of triplet spin states during biochemical reactions. For molecules that contain two unpaired electrons there are three spin orientations with respect to the molecular frame that would normally occur in equal populations because their energy differences, which fall in the microwave region, are below the thermal background. Any resonant microwave pumping from one state to another has no net effect. However, immediately after being formed, a triplet molecule will have one favored state, the one whose spin orientation has the greatest similarity with the parent molecule. By energy relaxation, the other substates would become equally populated. If we consider that under external microwave excitation, the reaction occurs faster than the substate relaxation time, the microwave energy can pump the preferred population into one of the other two states resulting in a changed overall reaction rate. The resulting resonance would be as sharp as allowed by the substate relaxation time, which is generally long for spin systems. Without knowing which molecule is involved in such a mechanism, the observation of an 8 MHz wide resonance in yeast activity under low power microwave excitation at 42 GHz is therefore plausible.

During this period of intense biophysical investigations, Keilmann continued to use his unique high-power IR lasers [35] for semiconductor physics investigations [36]–[39], including the uncovering of the p-Ge far-infrared laser mechanism [40], [41]. Also, for the first time, Keilmann could demonstrate second and third harmonic generation in the far infrared and investigate nonlinear susceptibilities contributed by lattice vibrations and by intraband conduction [42]–[46], helping decide a fundamental question of lattice dynamics [47]. He also helped elucidate the role of surface waves in laser damage – discovering stimulated surface polariton scattering [48] by introducing visualization of the interference patterns between plane and surface waves [49], [50] when liquid glass or metal surface waves (the ripples forming a grating) interact with IR light and stimulate surface phonon or plasmon scattering and enhanced absorption of CO_2 laser energy [51], [52].

Keilmann's work with high power CO_2 and far-IR lasers [53] necessitated new infrared components which he developed, and also spun off through a company, *Lasnix*[10], under the encouragement of Max Planck, who like Centre National de la Recherche Scientifique—CNRS in France [54], was encouraging its scientists to show that their government funded research could be turned into commercial products. These included a high power handling capacity (300 W!) broad-band stepped IR attenuator that uses the principle of diffraction by a freestanding metallic mesh [55] (now rechristened as a *"metamaterial"* screen); and a high contrast ratio 1–150 THz "Hertz" polarizer[11] with improved performance [56] that cleverly employs rectangular, rather than circular cross sections for the wires. The polarizer was also key to developing an efficient infrared ellipsometer [57], and is still being carried today as product lines by *Lasnix* and *Infraspecs*[12], in Germany.

In the late 1980's high T_c superconductors were discovered [58], and Keilmann began to apply his pulsed and continuous wave high power FIR lasers to investigate these new materials [59], [60] through IR ellipsometry [61]–[63]. When Genzel retired from Stuttgart around 1988, Keilmann turned to quantum Hall structures, as Klaus von Klitzing (1985 Nobel Laureate in Physics for discovering the integer quantum Hall effect) had come to Stuttgart as a prominent research director. Keilmann performed a series of experiments on photoconduction mechanisms in cooled 2D electron gas systems [64] in order to characterize currents involved in quantum-Hall transport, which because of the strong magnetic fields, have electron cyclotron resonances in the far IR. It was this effort, resolving individual edge states (quantized currents traveling along the edge of the sample), that was to lead eventually to Keilmann's greatest contribution to infrared spectroscopy – the *scattering near-field infrared microscope*.

At this juncture, the visualization of the quantum-Hall edge channels required much better spatial resolution than was available with existing IR spectrometers. First, Keilmann tried enhancing the resolution of his spectrometer by operating through a tapered metallic waveguide at 392 microns (765 GHz) and

[10]www.lasnix.com. Founded in 1984 by Keilmann.

[11]A common form of polarizer originated by Heinrich Hertz that uses parallel metal wires spaced by a wire diameter. The Barth/Keilmann variation has 100 × higher extinction ratio.

[12]www.infraspecs.de. Founded in 1993 by Barth.

scanning the 200 μm diameter aperture across the sample [65]. The technique worked, and secured Keilmann a place in von Klitzing's research division. However, Keilmann also realized that he could increase the resolution only so far with this subwavelength-scale aperture technique, since the coupled power decreased very rapidly as the aperture became smaller [66]. He began to think about other ways to access the near field and dramatically increase the available resolution.

Recalling his early experiments with whisker-contacted diode detectors and the electromagnetics he had learned in Javan's lab at MIT, Keilmann knew that submillimeter-wave detectors could work efficiently at wavelengths many times longer than the dimensions represented by the antenna itself. He knew that the coupled RF energy in the whisker was efficiently focused by Sommerfeld waves onto the very subwavelength-scale diode at its tip. He thought that this concentration of the field might be the perfect mechanism for capturing, focusing onto the sample, and then radiating the near field signatures of his IR laser-induced fields and currents. He approached von Klitzing with his concept of an *apertureless* near field probe for the quantum-Hall edge state sensing. Von Klitzing was not convinced however, and he wanted Keilmann to continue to pursue quantum-Hall related transport with traditional techniques.

Keilmann was undeterred. He first verified his new idea in a simple room-temperature experiment employing low frequency radio waves (1 MHz–1 GHz) and a microwave vector network analyzer on a large frequency scaled model of the whisker tip (coaxial cable with an extended and sharpened center pin). By scanning the tip across a metallic ground plane and imposing various solid and grid-like dielectric samples between the tip and the metal, he was able to verify a remarkable subwavelength resolution of 10^{-6} λ [67], [68]! He also recorded high contrast in both magnitude and phase that could be used to advantage in distinguishing sample characteristics.

Keilmann knew that he had come up with a technique that would prove to be extremely useful, and he decided then to start looking around for another laboratory position where he might be able to pursue these encouraging preliminary developments at far IR wavelengths.

Without too much fanfare, he contacted Theodor Hänsch (2005 Nobel prize in physics for laser based spectroscopy) at the Max Planck Institute for Quantum Optics in Munich, who had published non-cutoff waveguide scanning [69], and also Reinhard Guckenberger (a scanning tunneling microscopy expert and 1991 Max Planck Research award recipient) at the Max Planck Institute for Biochemistry – also in Munich, about potential positions. In the end, the allure of biophysics prevailed[13], and in 1995, Keilmann's permanent position at Stuttgart was "transferred" to Max Planck Institute for Biochemistry. *Along with his wife*, he was allowed to take all his equipment to Munich, where he began a concentrated effort to develop the apertureless near field imaging technique that he had tested at Stuttgart.

Keilmann still needed to develop several critical components for his apertureless near-field probe concept to work in the mid-infrared [70]. He needed a strong coherent infrared source, a coherent detection technique to allow the measurement of both magnitude and phase (necessary for pulling out the complex material parameters), and a way to tune, focus and separate out the near field signals from the much brighter background. In addition to a scattering tip approach, he thought about trying a coaxial TEM style probe that had no low frequency cutoff. However when he sought funding to try building up an IR instrument, he found that both concepts were turned down.

Scanning near-field *optical* microscopes (SNOMs), which had been demonstrated in 1984 [71]–[73][14], all employed waveguide type probes (typically metal coated sharpened metallic fibers operating below cutoff), and this technique was too well accepted to allow for radical variants to get much attention. Then in 1994, optical scattering from a vibrating open metal Atomic Force Microscope tip was used to go well beyond the Abbe limit [74]. Wickramasinghe, Martin and Zenhausern at IBM Watson, USA attributed dipole-dipole coupling to their measurement of 10 Å features using optical scattering from an open AFM tip [75]. This result was controversial however, and Keilmann had a thought. If he proposed to his sponsors that he could test the validity of the IBM results at longer wavelengths, where it would be easier to resolve the controversy, perhaps he could get funding to build an infrared *apertureless* microscope without having to justify its advantages directly on its own merits.

His intuition was correct! The funds began to flow, and they were immediately put to good use building up a research group and a prototype instrument using a subwavelength metal tip. Keilmann started with scanned amplitude and phase measurements in the microwave regime using a homodyne principle, rather than direct detection, to maximize the signal-to-noise [76]. He also realized that most of the collected IR photons that were scattered off the tip would not be from the near field interaction (which emanated only from the gap directly below the tip itself). In order to recover the near-field photons from the much greater background scattering, he introduced fast (kHz) modulation of the tip and employed a synchronous detection scheme. The sources were his coherent gas lasers that could produce step-tunable IR energy that was readily focused down to the diffraction limit. For the detection scheme, Keilmann very cleverly assembled a Michelson style interferometer with sample and reference arms that were used to pick up the backscattered signals (magnitude and phase) of the IR radiation coming off the sample. Later on, spectral information could be obtained by scanning the reference arm of the Michelson and recording the interferogram with HgCdTe detectors.

The first experiments were performed using a tapping AFM tip, a continuous wave CO_2 laser source, and two HgCdTe detectors, one for reflection and one for forward transmission [77]. 100 nm resolution was observed with this early instrument. The limit was extended to 30 nm a year later [78]. The theory of image dipoles (wire above a conducting plane) was quickly brought to bear, in order to predict the field patterns and to show how the local complex dielectric constant could be extracted from the scattering parameters. The first demonstration of spec-

[13]Keilmann's wife, Sigrun, herself a trained human genetics researcher, was partial to Munich and was looking forward to a return to that city!

[14]Aaron Lewis and Dieter Pohl are both credited with the first demonstration of an optical version of the near-field microscope in 1984.

troscopic contrast in the IR at 100 nm resolution was presented in a well cited *Nature* paper in 1999 [79]. By this time, Keilmann's ground breaking experiments on apertureless near field imaging and spectroscopy were well appreciated, and the new field of scattering-type scanning near field optical microscopy *(s-SNOM)* was born [80]–[82]!

One piece of s-SNOM, which was needed to realize full infrared spectroscopic coverage, was a compact broadband coherent IR source. The basis for this was actually laid before the first demonstration of the s-SNOM instrument, through a collaboration between Keilmann and University of Wisconsin Professor, Dan van der Weide. In 1994, van der Weide (who had met Keilmann at the 17th *International Conference on Infrared and Millimeter Waves* held at Caltech, Pasadena, CA in 1992), came to Stuttgart under a Humboldt Fellowship. Together he and Keilmann expanded upon a nonlinear transmission line concept for amplified synthesized microwaves that van der Weide had developed earlier [83], [84].

By exciting the nonlinear transmission lines in the GHz bands, they generated hundreds of harmonics spanning up to 1 THz. They further introduced the idea of combining two such harmonic combs with slightly offset frequencies to generate mixing products from the GHz-to-the-THz bands realizing a radically new concept to perform broadband spectroscopy at unprecedented speed, resolution, precision, and range [85]–[88]. This *dual frequency-comb spectroscopy* technique was subsequently extended across the infrared when Keilmann substituted the microwave synthesizers with mode-locked near-visible laser pulse trains rectified in a nonlinear crystal [89]–[92].

The dual-frequency comb concept—essentially an FTIR spectrometer with no moving parts, was quickly taken up, and holds great promise for ultra-broadband precision spectroscopy across all frequencies from the THz to the optical [93]. It has also allowed for the development of a very compact s-SNOM setup [94], [95]. Using this technique the first *THz near field microscope* was demonstrated in 2008 [96].

With this new s-SNOM system in hand [97], [98], Keilmann had an incredible tool and a totally open field. Optical spectroscopists using aperture-type tips were still limited to only slightly subwavelength resolutions – and Keilmann could now reach below 10 nm with high signal-to-noise and low background. He was also able to work across the complete spectrum, from microwaves to the optical, using basically the same instrument – by simply trading out the sources. He and his research collaborators measured nanoscale phenomena as diverse as metal nucleation in an insulator-metal phase transition [99]–[101], mapping of electron density in semiconductors down to the few-electron sensitivity level [102], [103], nanoscale chemical, biological and material recognition via molecular vibrational resonance [104]–[106], and the particularly sharp and strong phonon-resonant near-field interaction of polar crystals [107], [108].

In another application area, s-SNOM was shown to enable sensing of coherent fields that originate from subsurface scatterers [109] as well as from strong scatterers, such as plasmon-resonant nanoparticles or nanoscale metal antennae. This allowed Keilmann to map, at first in the visible and later in the IR, complex Eigenfields with nanometer resolution [110], [111]. Working with later research partners, the illuminated tip itself was used to emit coherent fields that propagate especially well due to momentum matching, when the surrounding medium supports extremely slow propagation. This is the case with infrared surface plasmons on graphene where s-SNOM allows the observation of nanoscale defects [112], [113].

In 2007, at the age of 65, Keilmann retired from his tenured Max Planck position at Martinsried and decided to help his former student and post-doc, Rainer Hillenbrand (who had just received a special research award to work for three years on s-SNOM at Martinsried), and his former student Nenad Ocelic (later Chief Operating Officer at Neaspec), to start a company to commercialize their spectroscopy instrumentation. They named the first instrument *NeaSNOM* for new SNOM, and the company *Neaspec*[15] or new spectroscopy. A novel background interference rejection technique developed by Hillenbrand, Huber and Ocelic [114] enabled the newly commercialized s-SNOM system to deliver high spatial resolution (< 10 nm) with single line lasers. Strong Max Planck support, as with Keilmann's earlier *Lasnix* experience, also helped the company develop their early product line.

Keilmann then had an opportunity, through a special Research Cluster of Excellence program, to relocate his lab to Garching, on the opposite side of Munich from Martinsried. At Garching, he took up a special position bent on the commercialization of new ideas in quantum physics at Max Planck Institute for Quantum Optics (MPQ) – finally making good on the offer he had received back in 1995 from Theodor Hänsch. He wanted both to further nanoscopy in biology, and at the same time help develop a more compact version of the *NeaSNOM* instrument.

At MPQ Garching, Keilmann focused on the source problem. He was the first to own a *NeaSNOM* instrument, which he was able to order through his MPQ sponsors. Over the next several years he developed a more compact coherent broadband laser illumination scheme that enabled the collection of complete infrared spectra at each nanoscale pixel in a matter of seconds [115], [116]. This source used photomixing of two near IR lasers in a nonlinear crystal (GaSe) to generate mid-IR pulses that were focused onto the AFM tip and coherently detected through the Michelson interferometer. The wavelength coverage was more than 1400 wavenumbers (600–2000 cm^{-1}). Keilmann coined the name *nano-FTIR* for this technique [117]. He and his research team demonstrated the new capability by observing phonon resonances in biological samples, interpreting the observed asymmetric interferograms as free-induction decay of resonantly induced polarization stored in the tip-to-sample system. Nano-FTIR also became a *Neaspec* product [118].

After five years, the Cluster of Excellence program at Garching ended and Keilmann moved his laboratory – and his interests again – this time to the historic Ludwig Maximilians University in the center of Munich (where he had started his career in Meteorology), and to the field of nanomedicine.

At LMU Keilmann is applying infrared nanoscopy to the progression of tooth, bone and joint diseases (osteoporosis), where he can probe at the molecular level to see crystal growth and decay mechanisms. He has already demonstrated that he can

[15]www.neaspec.com

detect and image biominerals in teeth and bone in good correlation to SEM, but in addition he can also identify and quantify their chemical nature through nano-FTIR [117], [119]. Keilmann and his research collaborators also showed recently that the product of the near field magnitude times the sign of the phase (calibrated against a gold surface) yields the same spectral absorption signature as normal far-field FTIR [118] – thus demonstrating that the nano-FTIR instrument can indeed identify the chemical signature of compounds on this extremely fine scale.

Keilmann's most recent innovations involve using the nano-FTIR system to perform both pump and probe measurements with the same short pulse optical laser sources that are used for the generation of the IR signals (in collaboration with Dimitri Basov at UC San Diego, USA-not yet published). This allows both ultrashort (100 femtosecond) time resolved measurements, as well as the high spatial resolution available with the near-field tip!

By the very definition, *Pioneers* are ahead of their time. In science this can often mean they are working on problems that have not yet been recognized either by their peers or by their sponsors. Sometimes extraordinary determination and risk taking is the only recourse left for an individual who truly believes they are on the right track. Fritz Keilmann had already earned his fully funded lifelong position at Max Planck when he picked up and changed his research path to pursue apertureless infrared near field microscopy. In less dramatic ways, he repeated this behavior again and again throughout his long career. His latest move to LMU and the shift to nanomedicine is no exception. At each step however, he focused his research efforts on specific goals. At each change he pushed forward successfully with innovations that have had broad ranging impact on multiple disciplines.

Keilmann's dual frequency-comb techniques can now be used to fingerprint gases over a 1000 wavenumber region of the mid IR in milliseconds. It has also been demonstrated to work at standoff distances of more than 100 meters and can achieve a spectral resolution of a few kHz—determined by the laser sources. The technique has applications in breath analysis, atmospheric gas analyzers, pollution monitoring and a wide number of areas yet to be explored. The molecular, and even atomic level spatial resolution achievable with his most advanced apertureless s-SNOM techniques is opening up biophysics applications as well as new areas of materials science and solid-state physics.

By commercializing his s-SNOM and nano-FTIR instruments, Keilmann is making this technology available to anyone, and I am certain he, and many others are anxious to see the technology spread and the number of applications expand.

Perhaps the nicest part of my time with Keilmann was the recognition that at 71 he was still as innovative, enthusiastic, confident and determined as he must have been when he made that first trip to an international conference and had the wherewithal and the *chutzpah* to pay for it by interviewing the scientific heroes of his youth. His words of advice to those that are certain to want to follow in his path were: "*Don't follow a big name or a path that is popular. Go for the new. Start by building up a new tool. In science, freedom to work on what you most believe in is the most important principle*." I don't believe any of us would argue with that. *Sponsors – please take note.*

As I finish up this article, I am listening to a recording honoring Fritz's father Wilhelm Keilmann[16] with works that he composed throughout his career as a musician. I am certain that he would have been proud of the series of *scientific* compositions contributed by his son. Indeed, the gift of original composition, whether it be in music or in science, was passed down in the Keilmann family.

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Fritz Keilmann began his career in science at the tender age of 14, when he became the local meteorologist for his Alpine community southwest of Munich, Germany. From 1967 to 1971, he worked at Garching as it transitioned to the Max Planck Institute for Plasma Physics. He did a post-doctoral appointment at MIT between 1972–1973 and then took up a post at the Max Planck Institute for Solid-State Research (Festkörperforschung) in Stuttgart. Except for a short Visiting Scientist appointment at University of California, Santa Barbara, USA, in 1987, he remained at Stuttgart through 1995, earning a permanent position at the Max Planck Gesellschaft in 1977. From 1995–2007, he did research at the Max Planck Institute for Biochemistry in Martinsried (outside Munich) where he worked until his formal retirement from Max Planck. He started two companies, *Lasnix* in 1984 and *Neaspecs* in 2007, upon leaving Max Planck. Both are still operational, although his involvement in *Neaspecs* is now only in a consulting role. Keilmann returned to Max Planck as a Distinguished Scientist in 2008, forming a new group at the Institute for Quantum Optics in Garching focused on mid IR broadband laser sources. His most recent move was to the historic Ludwig Maximilians University in Munich, where he has just set up a laboratory in the Department of Soft Matter Physics to work on nanoscopy of biomaterials. Dr. Keilmann has explored many areas and applications in infrared physics and spectroscopy. Keilmann worked with Ali Javan at MIT, Herbert Fröhlich, Ludwig Genzel, and Klaus von Klitzing at Stuttgart, and many other students and colleagues throughout his varied research career. His interests were always focused in the infrared, but he was often drawn into biochemistry and biophysics, where he initially worked on molecular spectroscopy of biomolecules and later on microwave effects on cell systems. Today, he is focusing on understanding the basic structure, evolution and impact of mineralization in osteoporosis. Dr. Keilmann also worked extensively on high power laser sources, especially optically pumped IR lasers and their applications for plasma science and nonlinear solid-state spectroscopy. He is credited with inventing the scattering-type scanning near-field optical microscope, the nano-FTIR, and the dual-frequency-comb spectrometer. These instruments have had a broad impact on nanoscience and have reached into areas as diverse as molecular biology, space science, and quantum chemistry. He has published more than 150 peer reviewed papers, holds 14 patents, and has contributed to five textbooks. He is an internationally recognized speaker and has given more than a hundred invited presentations at workshops, conferences and seminars. He is a recipient of the 2009 Kenneth J Button Prize. He resides south of Munich, Germany and is looking forward to continued development and applications of his nanoscopy techniques.