Reststrahlen to T-Rays – 100 Years of Terahertz Radiation

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Abstract. Although a reliable source of subpicosecond terahertz (THz) pulses, or T-rays, was demonstrated in the late 1980s, it was not until 1995, when they were used for imaging, that their full potential was realized. However, much of the publicity at that time seemed to suggest that very little research had taken place in the THz (or far-infrared) region before then. With the – largely new – present-day interest in biological applications of THz radiation it is apposite to review the opening up of this spectral region over the past hundred years. This paper attempts to show many of the significant developments in sources, detectors and spectroscopic systems for the THz region and includes brief descriptions of some of the more important applications and outcomes of the research. T-rays are only one of a number of very significant arrivals on the THz scene in recent years. It does seem an ideal time to pursue the exciting biological applications of THz radiation which are to be discussed at this Workshop.

Key words: Detectors, infrared history, instrumentation, sources, spectroscopy

1. Introduction

The first recorded account of an infrared experiment appears to be that by della Porta in his book ‘*Magiae Naturalis*’, published in 1589. He describes how both heat and cold can be focused with a concave mirror but the only detectors available were his ‘natural senses’. As soon as the thermometer was invented it was used by Sanctorius, a colleague of Galileo, in 1612 to measure the heat from the sun. Other significant thermal results were achieved by the short-lived Academia del Cimento under the patronage of the Grand Duke Ferdinand the Second of Florence, who is himself credited with the invention of the modern form of sealed liquid in glass thermometer. However, as is well-known, it was Sir William Herschel who, in 1800, carried out the first quantitative measurements of the heating power of the sun in both the visible and near-infrared portions of its spectrum [1]. It was nearly 100 years later that infrared experiments began to move towards what we now call the far-infrared (FIR) or THz region, with the work of Heinrich Rubens and his various colleagues in Berlin. A paper on the ‘refraction of rays of great wavelength in rock salt, sylvine and fluorite’ [2] is probably the first significant paper on longer wavelength radiation. In 1894 Rubens was joined by an American

physicist, Ernest Fox Nichols, and it was their work together which was to lead to a dramatic outcome.

2. The 1890s

By 1897 Rubens had extended the infrared spectrum out to nearly $20\mu\text{m}$, using prisms of rock salt (NaCl), sylvine (KCl) and fluorite (CaF_2), but these did not produce narrow band radiation. In 1896 Nichols discovered that the reflectivity of crystalline quartz rose over a narrow wavelength range near $9\mu\text{m}$ from a few percent up to almost the reflectivity of a silvered surface [3]. This we now know is due to lattice vibrations and in the following four years Nichols and Rubens found narrow-band reflections from various ionic crystals up to much longer wavelengths.

Rubens and Nichols built a spectrometer using multiple restrahlen plates to isolate a very narrow wavelength band. By changing the reflecting plates they could then produce nearly monochromatic light at a number of different wavelengths, eventually out to beyond $50\mu\text{m}$.

Towards the end of the 19th century one of the major problems engaging the attention of physicists and mathematicians was that of radiation from hot-bodies and, specifically, its variation with wavelength. A brief roll call of some of those involved is more than impressive: Wien, Angstrom, Rayleigh, Lummer, Pringsheim, Paschen, Larmor and Planck. Various formulae for the radiation from an ideal source, the blackbody, were derived but none fitted all the experimentally-obtained results. A particular problem was to obtain really accurate data at very long wavelengths. In 1900 Rubens, using the restrahlen spectrometer, obtained the necessary data and immediately visited Max Planck to give him the results who that same day wrote down the equation which is now called Planck's Radiation Law.

After several weeks of what he later described as the most strenuous work of his life, Planck revolutionized physics by inventing quantum theory to explain the radiation formula which he had derived empirically by combining the already well-known short wavelength radiation results with the long wavelength ones of Rubens. And after the death of Rubens in 1922 Planck was to write 'Without the intervention of Rubens the formulation of the radiation law, and consequently the formulation of quantum theory, would have taken place in a totally different manner, and perhaps even not at all in Germany' [4].

3. Early 20th Century Research

One might well describe the period up to 1923, the date of his death, as the Rubens' era. Between 1892 and 1922 just over 150 papers were published which included FIR research. Only 18 of them do not have Rubens as an author. Much interesting work was done and three discoveries made at this time are still relevant today.

In 1911 Rubens and Baeyer showed that the mercury arc lamp in a quartz envelope was an excellent long-wavelength infrared source [5]. In the THz region it has an equivalent black-body temperature of over 4000K and in modern grating spectrometers and interferometers it is still the preferred source.

One year earlier RW Wood, of Johns Hopkins University, had produced the first ‘blazed’ diffraction grating which allowed the concentration of light into a single order [6]. Wood achieved efficiencies of over 75% for the first order and showed that gratings of this type could be used out to beyond $100\mu\text{m}$ (3THz). Although Wood produced the first blazed gratings the possibility of making them had been suggested by Rayleigh in 1874 [7]. Apart from their use in monochromators, blazed gratings, with efficiencies close to 100% in one polarization, are employed in lasers as tuning elements. In carbon dioxide lasers, for example, replacing one of the mirrors with a blazed grating allows tuning over many wavelengths between $9\text{--}11\mu\text{m}$. These lasers are widely used as the ‘pump’ for optically-pumped far-infrared lasers (see Section 8).

The third discovery, by Hagen and Rubens, concerns the reflectivity of metals [8]. They showed that at wavelengths longer than about $10\mu\text{m}$ (30THz) the reflectivity R is given by

$$R = 1 - 3.65 \rho^{1/2} \lambda^{-1/2}$$

where ρ is the resistivity and λ the wavelength.

In the THz region this shows that any pure metal has a reflectivity of well over 0.99 and so even complex reflecting systems are virtually loss-free. But care is needed with ‘light pipes’, which are frequently used at long wavelengths, particularly if these are made of alloys such as brass or cupro-nickel, which have much higher resistivities than pure metals. For example, at 3THz cupro-nickel has a reflectivity of about 0.98 and losses in a multiple-reflection pipe can be significant.

4. 1920–1940

In 1920, after a relatively short but glittering career after leaving Berlin (which included the first pressure of light experiment in 1902), Nichols became Director of Pure Science at the National Electric Light Association in Cleveland, Ohio. Here he returned to his long-wavelength research, working with James Tear, and in 1923 they succeeded in joining what was then called ‘the electric wave’ spectrum to the infrared. Using a Hertzian oscillator wavelengths down to $220\mu\text{m}$ were produced, which overlapped with an extension of wavelengths from a mercury arc source to $420\mu\text{m}$ [9]. In 1924 a Russian physicist, Glagolewa Arkadiewa, showed that radiation of about $90\mu\text{m}$ could be generated by exciting small Hertzian oscillators in the form of brass filings immersed in oil [10].

Between 1920 and 1930 some 40 FIR papers were published and from 1930–1940 another 80. While many of these were on instrumentation, those on applications were increasing, with a particular emphasis on absorption bands in gases.

In 1935 Barnes, Benedict and Lewis published a complete spectrum of water vapour absorption out to $170\mu\text{m}$ [11] and in 1937 Randall and his colleagues used a monochromator with a blazed grating of $50\text{cm} \times 25\text{cm}$ to resolve absorption lines with a resolution of 0.5cm^{-1} at $75\mu\text{m}$ [12].

It is intriguing that there appear to have been no detector or source developments over these two decades. The bolometer, invented by Langley in 1880 [13], was almost universally used as the detector, usually with a mercury arc or some heated material such as the Welsbach mantle as a source [14]. (Langley, whose main interest was in the use of his detector in astronomy, stated that his bolometer could detect the heat radiated by a cow at a distance of over half a mile!)

5. 1940–1950

Not surprisingly, 1940–46 was a barren time for research publications but, of course, there were many developments during this period. Of particular note is the research in Germany to extend the range of prism spectroscopy. Up to 1940 $25\mu\text{m}$ was the maximum wavelength using KBr prisms but, with the mixed crystal thallium-bromide-iodide, KRS-5 (KRS standing for Kristalle aus dem Schmelzfluss), coverage was extended to $35\mu\text{m}$, and the introduction of caesium bromide and caesium iodide in the early 1950s allowed prism systems to operate out to nearly $60\mu\text{m}$.

The Second World War led to enormous advances in microwave techniques. The first experiments on gaseous spectroscopy in this region had been carried out as early as 1933 by Clinton and Williams [15], who studied the absorption of ammonia at just over 1cm wavelength and later worked down to 6mm . By 1946 the first millimetre-wave klystron was reported and advances to shorter wavelengths quickly followed.

The most significant post-war event was the invention, by Marcel Golay in 1947, of the pneumatic infrared detector, now almost always referred to as the Golay [16]. This elegant device came within a factor of three of the theoretical limit for a room temperature detector. The first cooled bolometer was also a 1940s invention. This used the superconducting transition point of tantalum at 4.4K , thus requiring the use of liquid helium. A later version was made with niobium nitride, which has a transition at 14.3K , a temperature which could be obtained by pumping on the then more readily available liquid hydrogen [17].

6. The 1950s

This decade saw the beginning of the modern FIR or THz era. The advent of the computer was to solve a fifty-year-old problem and revolutionize infrared spectroscopy. Interferometers such as the Michelson were excellent for the accurate study of monochromatic light but when there were more than three or four wavelengths from a source the interferogram produced was very difficult to analyze. Even so, Michelson, with infinite patience and his own invention of a ‘harmonic analyser’,

was able to make a Fourier analysis of quite complex interferograms. In the 1950s a number of experimenters, including Strong, Vanasse and Gebbie [18], [19], began the research which has led to the sophisticated instruments which we have today. Although prism and grating monochromators will always have a place, the throughput, multiplex and resolving power advantages of interferometers have made them an increasingly dominant force in spectroscopy during the past forty years.

On the detector side, the Golay was virtually the only detector of the 1950s but in 1959 the first convenient-to-use cooled device, the carbon bolometer, was invented [20] and the same year saw the first photoconductive detector for long wavelengths [21].

Although no fundamental source was produced for frequencies above 300GHz in the 1950s, the foundations were laid with the first backward-wave oscillator (BWO), or carcinotron, in 1953 [22]. Extension to higher frequencies was reached in the 1960s and BWOs are now available to above 1 THz with a tuning range of $\pm 10\%$ of the centre frequency. Harmonic generation from a microwave source began in 1945 but it was the work of Gordy [23], which led to frequencies above 300 GHz.

Of the 200 papers published in this decade, 80% were on applications which included the first studies on semiconductors, including cyclotron resonance measurements [24], many papers on the absorption spectra of gases and, at 1.5 mm wavelength, observation of solar and lunar radiation [25]. Early work on the transmission of superconductors began, which was to lead to the direct measurement of the energy gap predicted by the BCS theory [26].

7. The 1960s

This was a flourishing decade for the THz region, with well over 1000 published papers. More importantly, it brought much of the instrumentation which is widely used today. If one looks back to the period before 1960 the only invention, apart from the spectrometers themselves, which has not been superseded, is the mercury arc source. Even the Golay is a rarity now. But from 1960 onwards on the detector side we have the n-InSb electron bolometer [27]; the germanium bolometer [28] (to be followed later by the now more widely-used silicon composite bolometer); pyroelectric detectors [29], and a tunable FIR detector [30]. Of course, the main discovery was the laser and this led to the first high-power far-infrared device, the water vapour laser, in 1964 [31]. Particularly useful for spectroscopists was the arrival of the first really effective long pass and narrow band filters for the THz region [32].

The increase in applications of THz radiation was so great that it would be invidious to include any particular ones except to remark that 1968 saw what I believe is the first paper specifically related to biological applications. This was by H. Fröhlich [33] and is entitled 'Long Range Coherence and Energy Storage in Biological Systems'. The abstract begins:

Biological systems are expected to have a branch of longitudinal electric modes in a frequency region between 10^{11} and 10^{12} Hz.

Fröhlich also introduces the term 'Fröhlich Coherence' and suggests that quantum coherence can play a role in biological systems.

8. 1970 Onwards

1970 saw the arrival of one of the most useful laser sources, the optically-pumped far-infrared laser (OPFIRL) [34]. These now produce hundreds of CW and many thousands of pulsed wavelengths throughout the THz region. On the strongest CW lines output powers of above 100 mW are available from commercial instruments and pulsed powers of many MW have been achieved.

The first free electron laser (FEL) operated in 1975 and by 1984 Elias and his colleagues [35] had constructed a laser operating between 390–1000 μm as a user facility at the University of California at Santa Barbara with a peak power of up to 10 kW. One of its main application areas was in biology, including photobiology. Since then a number of THz laser user facilities have been installed in the USA, France, the Netherlands and Italy and are providing very valuable results in a wide range of disciplines, including biology and medicine.

Alternative facilities for FIR research are the infrared ports of certain synchrotrons [36]. These provide a much higher average source brightness than an arc lamp in the 10–1000 μm region and the $<1\text{ ns}$ pulse structure is useful for a number of applications, again including those in biology.

In a paper earlier this year [37] very much higher intensity radiation from a synchrotron, BESSY II in Berlin, has been reported. The authors have detected coherent synchrotron radiation (CSR) in the range 0.3–1 THz. Unlike 'ordinary' synchrotron radiation, where the power increases linearly with current, CSR grows as the square of the beam current. Compared to the normal radiation, the power output increases by 10^5 times and it is suggested that with improved electron optics the generation of powerful CSR up to several THz is feasible. Although the opportunity to use such a source is inevitably limited, this is a really exciting prospect for researchers in the THz region.

Fundamental solid-state sources and harmonic generation from such sources can now produce useful power at frequencies to above 1 THz. These are of particular interest, as the local oscillators (LOs) in heterodyne spectrometers which are being used very widely for applications, for example, in astronomy and pollution-monitoring [38]. The mixing element can either be a room-temperature Schottky diode, which typically requires 1 mW of LO power, or, more recently, a hot-electron bolometer (HEB) cooled to near 4 K. Progress in HEBs has been dramatic during the 1990s and these have the advantage of needing 1 μW or less of LO power.

Other interesting THz sources are p-Ge and p-Si lasers. p-Ge lasers are tunable from about 75–300 μm but do not as yet appear to have been used in many applications. A particularly promising development is the quantum-cascade laser,

which has very recently produced radiation at 4.4 THz ($68\ \mu\text{m}$) with a pulsed power of 2 mW [39]. The authors predict that lasers operating CW and at liquid nitrogen temperature should be constructed quite soon. Quantum cascade lasers must certainly be a very useful tunable source in future years.

Another source, which is already close to commercial production, is based on the Smith-Purcell (SP) effect [40]. SP radiation is produced when an electron beam is passed very close to a periodic structure such as a ruled grating. Following fundamental research at Dartmouth College in the USA by John Walsh and his colleagues [41], Vermont Photonics* have produced a relatively compact version, producing microwatts of CW power in the THz region [42]. The source area appears to be very small and the brightness is more than two orders of magnitude higher than that of an arc lamp. The output frequency depends on the pitch of the periodic structure and the electron energy, and tuning over $\pm 15\%$ can be achieved with a single grating by varying the beam energy. Compared with a synchrotron this source has three potential advantages: firstly, it can be installed within one's own laboratory; secondly, for example, in biological applications, the fundamental linewidth may give adequate resolution, thus obviating the need for a spectroscopic system; thirdly, as the source is CW (or pulsed), non-linear effects are very unlikely.

Undoubtedly sub-picosecond THz pulses or T-rays are the most important source development of recent years. Their advantages for time-resolved, pump-probe type experiments and time domain spectroscopy, as well as for imaging have been well documented. It is important to recognize that up to 1–3 THz, which can be generated in photoconductive switches pumped with femtosecond laser pulses, the output energy is quite high at up to $1\ \mu\text{J}$. For higher frequencies the laser pulses impinge on a crystal with second-order susceptibility, providing optical rectification, and the output at frequencies up to 40 THz is much lower. However, the convenience and comparative simplicity of this source has already shown that it will have applications in many areas, including the biological field, as is apparent from a number of papers in the programme for this Workshop. However, it is worth remarking that THz imaging does not require a T-ray source. In 1972, using an HCN laser at $337\ \mu\text{m}$ and a very simple scanning system, Dean Hodges and his colleagues produced THz images of metal objects inside various types of packaging. They also measured the transmission of various materials, gauze, cotton bandages and plaster casts etc, over the range 0.3–1 THz [43]. There was one intriguing outcome – blood is significantly less absorbing than water in the THz region, a result which has been recently confirmed during the THz-Bridge project [44].

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9. Conclusions

A brief review covering more than a century must be selective and skim over many important inventions, developments and applications, for which the author apologizes.

In physics, chemistry and astronomy investigations in the THz region have produced invaluable results during the past 100 years, and recent times have seen an increasing interest from those in the medical and biological fields. I feel that it is now clear that, perhaps fortuitously, the THz region of the spectrum has virtually all the tools in place for a great range of new biological applications. Whilst T-rays are a special asset, the arrival of other sources and detectors will allow the study of materials with greater absorptions and/or at higher resolution. The 'THz Bridge' programme of research will surely lead to some exciting discoveries.

References

1. Herschel, W.: Experiments on the Refrangibility of the Invisible Rays of the Sun, *Phil. Trans. R Soc* **90** (1800), 284–288.
2. Rubens, H. and Snow, B.W.: On the Refraction of Rays of Great Wavelength in Rock Salt, Sylvine, and Fluorite, *Phil. Mag.* **35** (1893), 35–45.
3. Nichols, E.F.: A Method for Energy Measurements in the Infrared Spectrum and the Properties of the Ordinary Ray in Quartz for Waves of Great Wavelength, *Phys. Rev.* **4** (1897), 297–313.
4. Jammer, M.: The Conceptual Development of Quantum Mechanics, *Amer. Inst. Phys. History of Modern Physics* **12** (1989), 13. [Quotation from M. Planck, *Berl. Ber.*, 1923, (1923)]
5. Rubens, H. and Baeyer, O.V.: On Extremely Long Waves Emitted by the Quartz Mercury Lamp, *Phil. Mag.* **21** (1911), 689–703.
6. Wood, R.W.: The Echelette Grating for the Infrared: *Phil. Mag.* **20** (1910), 270–281.
7. Rayleigh, Lord: On the Manufacture and Theory of Diffraction Gratings, *Phil. Mag.* **47**(4) (1874), 193–221.
8. Hagen, E. and Rubens, H.: On Some Relations Between the Optical and the Electrical Qualities of Metals, *Phil. Mag.* **7** (1904), 157–161.
9. Nichols, E.F. and Tear, J.D.: Short Electric Waves, *Phys. Rev.* **21** (1923), 587–610.
10. Arkadiewa, A.G.: Short Electromagnetic Waves of Wave-length up to 92 μ , *Nature* **113** (1924), 640.
11. Barnes, R.B., Benedict W.S. and Lewis, C.M.: The Far Infrared Spectrum of H₂O, *Phys. Rev.* **47** (1935), 918–921.
12. Randall, H.M., Dennison, D.M., Ginsburg, N. and Weber, L.R.: The Far Infrared Spectrum of Water Vapor, *Phys. Rev.* **52** (1937), 160–174.
13. Langley, S.P.: The Bolometer, *Nature* **25** (1881), 14–16.
14. Pfund, A.H.: The Electric Welsbach Lamp, *J. Opt. Soc. Am.* **26** (1936), 439–442.
15. Cleeton, V. and Williams, N.H.: Electromagnetic Waves of 1.1 cm Wave-Length and the Absorption Spectrum of Ammonia, *Phys. Rev.* **45** (1934), 234–237.
16. Golay, M.J.E.: A Pneumatic Infra-Red Detector, *Rev. Sci. Instr.* **18** (1947), 357–362.
17. Andrews, D.H., Milton, R.M. and DeSorbo, W.: *J. Opt. Soc. Am.* **36** (1946), 518–524.
18. Gebbie, H.A. and Vanasse, G.A.: Interferometric Spectroscopy in the Far Infra-Red, *Nature* **178** (1956), 432–433.
19. Strong, J.J.: Interferometry for the Far Infrared, *J. Opt. Soc. Am.* **47** (1957), 354–357.
20. Boyle, W.S. and Rogers, K.F.: Performance Characteristics of a New Low-Temperature Bolometer, *J. Opt. Soc. Am.* **49** (1959), 66–69.

21. Fray, S.J. and Oliver, J.F.C.: Photoconductive Detector of Radiation of Wavelength Greater Than $50\ \mu$, *J. Sci. Instr.* **36** (1959), 195–196.
22. Kompfner, R. and Williams, N.T.: Backward-Wave Tubes, *Proc. IRE* **41** (1953), 1602–1611.
23. King, W.C. and Gordy, W.: One-to-Two Millimeter Wave Spectroscopy, *Phys. Rev.* **93** (1954), 407–412.
24. Burstein, E., Picus G.S. and Gebbie, H.A.: Cyclotron Resonance at Infrared Frequencies in InSb at Room Temperature, *Phys. Rev.* **103** (1956), 825–826.
25. Sinton, W.M.: Observation of Solar and Lunar Radiation at 1.5 Millimeters, *J. Opt. Soc. Am.* **45** (1955), 975–979.
26. Richards, P.L. and Tinkham, M.: Far Infrared Energy Gap Measurements in Bulk Superconducting In, Sn, Hg, Ta, V, Pb and Nb, *Phys. Rev.* **119** (1960) 575–590.
27. Putley, E.H.: Impurity Photoconductivity in n-type InSb, *Proc. Phys. Soc.* **76** (1960), 802–805.
28. Low, F.J.: Low Temperature Germanium Bolometer, *J. Opt. Soc. Am.* **51** (1961), 1300–1304.
29. Putley, E.H.: Solid State Devices for Infrared Detection, *J. Sci. Instr.* **43** (1966), 857–868.
30. Brown, M.A.C.S. and Kimmitt, M.F.: Far Infrared Resonant Photoconductivity in Indium Antimonide, *Infrared Phys.* **5** (1965), 93–97.
31. Crocker, A., Gebbie, H.A., Kimmitt, M.F. and Mathias, L.E.S.: Stimulated Emission in the Far Infra-Red, *Nature* **201** (1964), 250–251.
32. Ulrich, R.: Effective Low-Pass Filters for Far Infrared Frequencies, *Infrared Phys.* **7** (1967), 37–50.
33. Fröhlich, H.: Long Range Coherence and Energy Storage in Biological Systems, *J Quantum Chem.* **2** (1968), 641–649.
34. Chang, T.Y., Bridges, T.J. and Burkhardt, E.G.: CW Submillimeter Laser Action in Optically Pumped Methyl Fluoride, Methyl Alcohol, and Vinyl Chloride Gases, *Appl. Phys. Lett.* **17** (1970), 249–251.
35. Elias, L.R., Hu, J. and Ramian, G.: The UCSB Electrostatic Accelerator Free Electron Laser: First Operation, *Nucl. Instr. and Meth.* **A237** (1984), 203–206.
36. Duncan, W.D. and Williams, G.P.: Infrared Synchrotron Radiation from Electron Storage Rings, *Appl. Optics* **22** (1983), 2914–2923.
37. Abo-Bakr, M., Feikes, J., Holldack, K., Wüstefeld, G. and Hübers, H.-W.: Steady-State Far-Infrared Coherent Synchrotron Radiation detected at BESSY II, *Phys. Rev. Lett.* **88** (2002), 2548–2551.
38. Titz, R., Birk, M., Hausamann, D., Nitsche, R., Schreier, F., Urban, J., Küllman, H. and Röser, H.P.: Observation of Stratospheric OH at 2.5 THz with an Airborne Heterodyne System, *Infrared Phys. & Technol.* **36** (1995), 883–889.
39. Köhler, R., Tredicucci, A., Beltram, F., Beere, H.E., Linfield, E.H., Davies, A.G., Ritchie, D.A., Iotti, R.C. and Rossi, F.: Terahertz Semiconductor-Heterostructure Laser, *Nature* **417** (2002), 156–159.
40. Smith, S.J. and Purcell, E.M.: Visible Light from Localised Surface Charges Moving across a Grating, *Phys. Rev.* **92** (1953), 10693.
41. Urata, J., Goldstein, M., Kimmitt, M.F., Platt, C. and Walsh, J.E.: Superradiant Smith-Purcell Emission, *Phys. Rev. Lett.* **80** (1998), 516–519.
42. Mross, M., Lowell, T.H., Durant, R. and Kimmitt, M.F.: *Performance Characteristics of a Smith-Purcell Tunable Terahertz Source*, THz Bridge Workshop, Capri, 2002.
43. Barker, D.H., Hodges, D.T. and Hartwick, T.S.: Far Infrared Imagery, *Proc. SPIE* **67** (1975), 27–34.
44. Gallerano, G.P., Doria, A. and Giovenale, E.: Private Communication, 2002.