PIEZOELECTRICITY *and Ultrasonics*

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THE Editor has kindly asked me to tell about the bearing that my work on piezoelectric crystals has had upon the development of ultrasonics. In truth, the answer can almost be put in two words: very little. My work has contributed chiefly to such things as frequency control, filters, and electrical communication. Even if these applications had never materialized, still Paul Langevin's invention of the piezoelectric transducer and the stimulus given to the study by Wood and Loomis of the effects produced by vibrating quartz plates would undoubtedly have led to most, if not all, of the subsequent applications of crystals to ultrasonics.

Nevertheless, my interest in piezoelectricity was first motivated by ultrasonics, a field in which I have spent considerable time since 1917. So, it is perhaps fitting that I should put on record some memories of the entire subject. I have tried to put my personal interests in proper perspective by including references to the general development. This is not, however, an attempt to cover the whole field.

As everyone knows, the intensive study of piezoelectricity and ultrasonics began at the time of the first world war. As far as this country is concerned, the starting point was a conference in Washington, D. C., 14–16 June 1917. This was a big affair, sponsored by the National Research Council, at which delegates from England and France told a group of American scientists and engineers of the progress that had been made in meeting the German submarine menace.

So far as I know, the story of this conference and its results has never been published. It was a long time before the topics with which it dealt were declassified, and by then the conference was only a memory, along with many other memories of a time that one would rather forget.

The conference, with its antecedents and consequences, was one of the dramatic episodes in the civilian history of World War I. At this late date, it would be practically impossible to recover the information needed for the telling of the complete story. Nevertheless, since the episode had a great deal to do with the subsequent history of ultrasonics and piezoelectricity, I hope that a brief sketch of it is not out of place here. As one of the few participants still living, I can attempt a first-hand account.[1]

England had declared war on Germany on 4 August 1914. On 1 February 1917, Germany began the campaign of unrestricted submarine warfare. For defense, ships depended on camouflage (a forerunner of modern art) and crude microphones. The latter could detect a submarine at a distance of 200 yards when it was moving at a speed of six knots or over.

In 1916, President Wilson had established the National Research Council (NRC). R. A. Millikan was Chairman and C. E. Mendenhall, Vice-Chairman of Division IV—Physics, Mathematics, Astronomy, and Geophysics. This division had a Physics Committee, of which Millikan was Chairman, and in 1917–18 there were several subcommittees appointed for special purposes. One of these was the Committee on Crystal Detectors, consisting of J.

Professor Cady, a member of the class of 1895 at Brown University, received his doctorate in physics at the University of Berlin in 1900. He was professor of physics at Wesleyan University, Middletown, Connecticut, from 1903 to 1946, when he retired as professor emeritus. His investigations into the piezoelectric effect have brought him world renown, and his book, PIEZOELECTRICITY (McGraw-Hill Book Company, Inc., New York, 1946) is the definitive treatise on the subject. A second edition is in course of preparation. Since his retirement from Wesleyan, Dr. Cady has carried on research at the California Institute of Technology (1951–1955) and more recently as a private consultant in California.

A. Anderson, H. A. Wood, I. B. Crandall, H. W. Farwell, and myself as Chairman.

The first reaction to the submarine declaration of independence was the prompt appointment, by the NRC, of an antisubmarine committee, under the chairmanship of R. A. Millikan. What followed was a superb demonstration of Millikan's energy, resourcefulness, and organizing ability. Only a man of his reputation and genius could have succeeded in securing cooperation between rival corporations and in getting the necessary action from the Navy. He persuaded the Western Electric Company, General Electric Company, and Submarine Signal Company to cooperate in joint experiments at Nahant, Massachusetts, in May 1917. That same month, a special board of the Navy on antisubmarine devices was organized, and a group of five theoretical physicists was called to Washington for a two-week conference on magnetic devices. This group consisted of Ernest Merritt, A. C. Lunn, H. A. Bumstead, L. A. Bauer, and H. W. Nichols. Also in May, a special subcommittee of the NRC was organized in New York. This was the beginning of the Columbia group, of which more later.

Robert A. Millikan, about 1918.

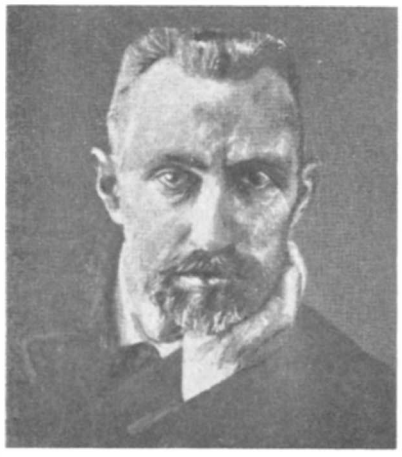
Three months had been required to do this organizing. It was not until 1 June, almost a month after we entered the war, that a conference was held in Washington with Ernest Rutherford from England, and Ch. Fabry, H. Abraham, the Duc de Guiche, and M. Paternot from France. They met with United States Navy officers, and with Millikan, G. E. Hale, E. Merritt, W. E. Durand, R. W. Wood, I. Langmuir, F. B. Jewett, C. E. Mendenhall, Henry Fay, H. D. Arnold, and E. H. Colpitts.

This meeting was followed on 14–16 June by the larger conference referred to above. In addition to those present at the meeting on 1 June were representatives of the Nahant group, Western Electric, General Electric, Westinghouse, Bureau of Standards, and 30 to 40 other scientists and engineers, including myself. (I have been unable to secure the full list of participants.)

Some of the projects disclosed by the delegates from overseas were still in the speculative stage and had not even been reported to their own admiralties. They appealed to us for help, either by developing the methods they described or by inventing new devices. I have no record of all the topics discussed and only mention two that I found of special interest.

One disclosure had to do with magnetic methods of submarine detection. Of this, I say no more other than that it was not until World War II that magnetic airborne detection (MAD) was developed and proved to be really effective.

We come now to the second, and more important, matter discussed at the conference. This had to do with acoustic waves—in the audible and ultrasonic ranges. I had already come to the conclusion that the most promising method was the ultrasonic one, and I was planning to try a magnetostrictive generator for producing the waves, when I was called to the conference.



Pierre (far left) and Jacques Curie.

Rutherford and Abraham described some tests, which had been made in their respective countries, with the Broca tube and the Walzer apparatus. The Broca tube was simply a pipe with a diaphragm or bulb at the end. The Walzer method made use of the binaural effect.

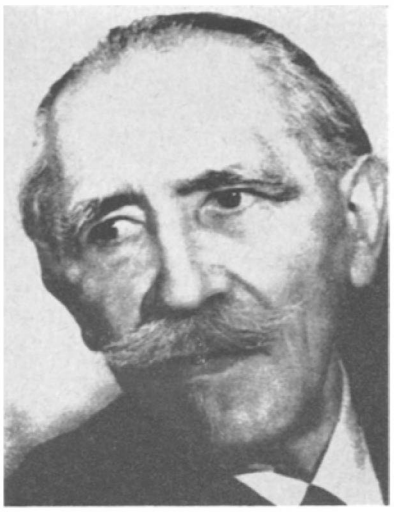
From the physicist's point of view, perhaps the most interesting announcement was from the French delegates, who told us that Langevin had just begun to use the piezoelectric effect for the generation and detection of ultrasonic waves in water. Piezoelectricity (pressure electricity) had been known since 1880, when it was discovered by the brothers Pierre and Jacques Curie. Aside from a few minor applications, in which only static charges played a part, piezoelectricity remained for thirty years a scientific curiosity, receiving scant attention, if any, in textbooks. It showed hardly more symptoms of having practical applications than the green flash or the mirage. In the meantime, however, thanks especially to the labors of Lord Kelvin, Pockels, Duhem, and Voigt, the general laws were formulated, and the piezoelectric constants of many kinds of crystals were measured. The foundation had thus been laid for Langevin to build on when he conceived the brilliant idea of not merely deforming plates of quartz piezoelectrically, but of making them vibrate and generate ultrasonic waves.

This device was the famous "sandwich" transducer in which a mosaic of small quartz plates a couple of millimeters thick was cemented between two slabs of metal. When a voltage was applied to the slabs at the right frequency, the electric field in the quartz plates caused them to expand and contract in thickness, thus making the whole structure vibrate as a unit in resonance with the driving frequency.

While Langevin was carrying on his pioneer work in France, knowledge of his experiments was communicated to the British. Under R. W. Boyle's direction, various methods of detection were tried, culminating in the construction of quartz–steel sandwiches. Boyle got an echo from a submarine at about the same time as Langevin. Several British ships were then equipped for echo work, which was called the "Asdic" gear (Anti-Submarine Detector Investigation Committee).

This is all that need be said here concerning the conference of 14–16 June 1917. The various participants left for home, well-provided with suggestions. The only project that concerns us is the piezoelectric one. Among those, besides myself, who decided to play with crystals were representatives of the General Electric and Western Electric Companies, the Bureau of Standards, Leland Stanford University, and Columbia University.

The Bureau of Standards undertook the task of inspecting the shipments of quartz crystals that soon began to arrive. At Leland Stanford and Columbia, the work was mainly with quartz transmitters, though J. A. Anderson of Mount Wilson, who cooperated with the Leland Stanford group, also made tests with Rochelle salt. The Western Electric group decided to experiment with Rochelle salt (sodium–potassium tartrate), since of all crystals this has the greatest known piezoelectric effect. I was invited to join the General Electric Group at Schenectady, where I spent most of the summer of 1917 in cooperation with A. W. Hull, E. M. Kellogg, and others. What we did was mostly laboratory exercises, learning the "feel" of crystals by trying out various cuts and shapes of bars or plates from both quartz and Rochelle salt; we also made some experimental microphones.



Paul Langevin, 1872–1946.

The most promising results in underwater ultrasonics were achieved by the Columbia group, with which I cooperated from September 1917 until the end of the war, while still carrying on with my teaching at Wesleyan University, Middletown, Connecticut. The group at Columbia was headed by Pupin, who designed the amplifying system. Morecroft had charge of the vacuum-tube oscillator ("pliotron"), and Wills designed the quartz sandwich. I took over the receiving end, for which I used Rochelle-salt hydrophones, designed to resonate at the transmitted frequency. The tests at Columbia were made in a large tank. By the early part of 1918, the apparatus was ready for long-range testing, and the group went to the Navy Yard in Key West, Florida, where for several weeks the Gulf of Mexico served as a "tank." Among those associated with us at various times were Commander Houghton (England) and Lieutenant Abetti (Italy).

The purpose of these tests was to study the performance of the quartz transmitter. Signals were

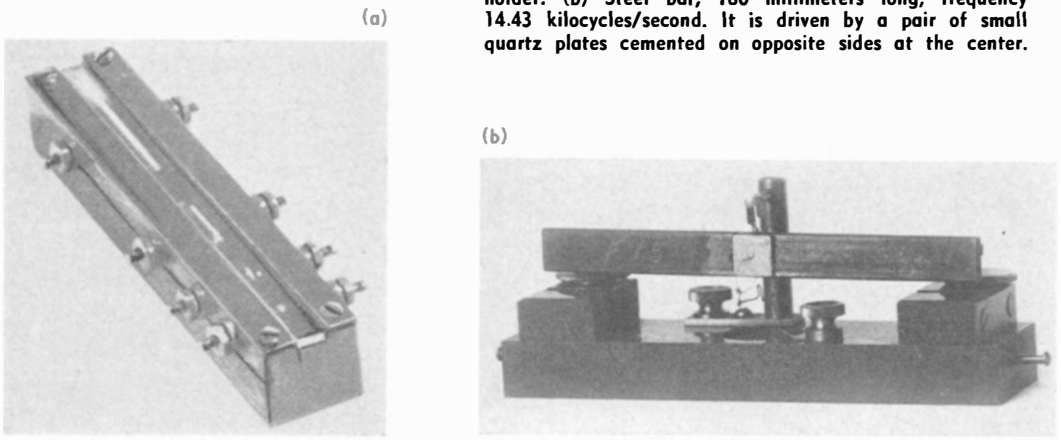


Fig. 1. Early types of piezo resonators. (a) Four X-cut bars, lengths from 1.76 to 30.3 millimeters, frequencies from 1523 to 91.66 kilocycles/second, in a common holder. (b) Steel bar, 180 millimeters long, frequency 14.43 kilocycles/second. It is driven by a pair of small quartz plates cemented on opposite sides at the center.

picked up by the Rochelle-salt detector at a distance of three miles. No echo work was attempted.

After our return to the north, the tests were continued at the newly organized Naval Experimental Station at New London, Connecticut, where some echo tests were made in the harbor. The station had been planned shortly after the conference of June 1917. Among the staff members were E. Merritt, Max Mason, H. A. Wilson, E. F. Nichols, H. A. Bumstead, G. W. Pierce, and J. Zeleny. Our tests were still incomplete when the Armistice was declared on 11 November 1918.

Of the other work at New London, I can say no more than that the magnetic method of detection was further investigated and that Max Mason made some improvements in the Walzer listening device.

All the efforts in ultrasonics, on both sides of the Atlantic, failed to contribute to the success of the Allies in the first world war. The groups in this country did not catch up with Langevin, who had a running start. By February 1918, he could transmit with his quartz sandwich about five miles and had received an echo from a submarine. Later in 1918, he could, under favorable conditions, detect a submarine at about a mile. But this was too late for quantity production. The submarine was conquered by depth charges and the convoy system, without the aid of ultrasonics.

After the close of the war, the Navy still continued its interest in ultrasonic echo ranging under water, a system that came to be known as Sonar. Military operations in the near future were not foreseen, and such progress as was made had more to do with depth-sounding than with the location of submerged objects by horizontal beams.

Between the two wars, great progress was made in what might be called piezoelectric engineering, as well as in the discovery of new piezoelectric crystals, measurement of their constants, and fundamental theory. For some years, the output of papers was so great that *Physics Abstracts* carried a special section headed "Piezoelectricity."

In the course of my tests of quartz and Rochelle salt in 1917 and 1918, I had noticed that specimens cut from these crystals reacted in a peculiar way on the driving circuit when the frequency was close to that for a natural mode of vibration. In quartz, the effect was extraordinarily sharp. To me, this observation was like one of Sir Thomas More's "Diamondes and Carbuncles upon certain rockes," discovered by the dwellers in Utopia: "And yet they seke not for them; but by chaunce finding them they cutt and polish them."

Research consists of three parts: discovery, discrimination, and doing something about it. Discovery may be by chance or design. Discrimination is needed to decide whether the discovery is trivial or significant. What to do about it may come in a flash, or after long thought. In the case of the crystal resonator, the idea of making a quartz crystal serve as a standard of frequency did come in a flash one evening, but long thought and much experimentation were needed to make the device practicable. Some of the early resonators are shown in Fig. 1.

On the other hand, the fact that a crystal can *control* the frequency of an oscillator, instead of merely monitoring it, was established by looking for it. The search began in 1921. One thing that made it difficult was that at that time the equivalent electrical network of the resonator had not yet been determined. The stabilizing action of the crystal was discovered first, and the first piezo-oscillator circuit, with frequency controlled by the crystal alone, was described at the meeting of The Ameri-

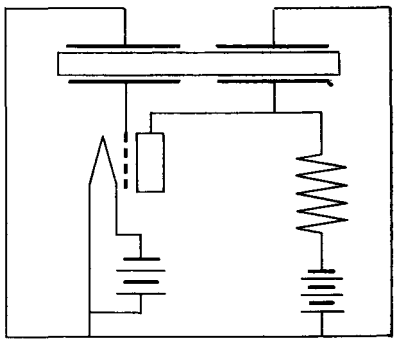


Fig. 2. The first piezo oscillator, 1921. The quartz bar had two pairs of electrodes connected to the input and output of an amplifier. It was 39 millimeters long, and vibrated in a lengthwise mode with a fundamental frequency of about 70 000 cycles/second.

can Physical Society on 28 December 1921. The circuit is illustrated in Fig. 2.

The first half of 1923 was spent in Europe, the only sabbatical leave that I asked for in almost fifty years at Wesleyan. I brought along some quartz resonators that had been calibrated at the Bureau of Standards, and made comparisons with standard frequency meters at government laboratories in Italy, France, and England. During this time, G. W. Pierce at Harvard, to whom I had described my work, was experimenting with quartz crystals, and he invented the well-known Pierce circuit for crystal control of frequency. A modification of Pierce's original circuit was invented independently by J. M. Miller. In 1925, Pierce described the first ultrasonic interferometer, in which high-frequency acoustic waves were produced by a vibrating quartz plate.

In my early work on the resonator, I showed that the vibrating quartz behaved electrically like a capacitance in series or parallel with a resistance, the values of which depended on the frequency. As was proved by my colleague, K. S. Van Dyke, in 1925, a much better "equivalent network" consists of a resistance R, inductance L, and capacitance C in series, the combination being in parallel with a capacitance C_0. This electrical equivalent is in universal use. Its advantage is that the four electrical parameters have values that are constant in the neighborhood of a resonant frequency.

I undertook the task of finding a mathematical formulation that described the action of the piezo resonator, taking into account the effects of the piezoelectric reaction on the elastic constants and also the fact that there may be an airgap between the crystal and its electrodes. A graphical method based on the equivalent network was developed, whereby the electrical impedance or admittance of the resonator could be represented by a circular locus.

One type of vibration that we studied was the bending, or flexure, of a crystal bar. Flexural vibrations of a quartz bar were produced by J. R. Harrison in 1927, the same year in which they were also discovered independently by Giebe and Scheibe.

The use of frequency control for radio transmitters spread rapidly. Broadcasting stations soon became crystal-controlled, and the amateurs caught on to the idea. The popular magazines had articles on how to "grind your own."

The earliest quartz plates for frequency control were cut perpendicular to the crystallographic X axis and were called "X cut." In 1917, the Y cut began to come into use. I have been told that this came about by chance. There was a time when some spectacle lenses were made from quartz crystals. E. D. Tillyer of the American Optical Company experimented with plates prepared from some of these lenses to see whether they would vibrate piezoelectrically. Some of them did, and some of those that did turned out to be approximately perpendicular to the crystallographic Y axis. It was found that the Y cut could be used for frequency control; in 1927, I showed that the vibration was of a shear mode, in contrast to the compressional mode that characterized the X cut. The Y cut offered certain advantages over the X cut. In passing, it may be noted that shear waves can be transmitted through solid and viscous-liquid media and have become of importance in research.

Both the X and Y cuts were found to be subject to rather large changes in vibrational frequency with varying temperature. When a highly constant frequency was desired, the crystals had to be placed in "ovens" held at a constant temperature by thermostats.

Hand-in-hand with the increasing use of crystals for frequency control went the demand for crystals, or crystal cuts, that would vibrate at a more constant frequency. Down to the present time, no other crystal has been found superior to quartz, but several different cuts have been introduced with temperature coefficients that are practically zero over a more or less limited range. Nevertheless, for highest precision, thermostatic control is still necessary.

The first quartz standards that had low temperature coefficients were in the form of rings. W. A.

Marrison introduced a *Y*-cut ring, and later L. Essen used a cylinder with length parallel to the *Z* axis. Of the other important quartz cuts with low temperature coefficients, the *AT* appeared in 1932 and the *GT* in 1940.

Only passing reference can be made to the quartz clock, the movement of which is governed by a crystal-controlled oscillating circuit. Even though at present the ultimate frequency standard is atomic, as, for example, the cesium beam, still the quartz-controlled oscillator plays a part as an essential auxiliary.

During the twenties and thirties, the piezoelectric crystal to receive most attention as a scientific problem was Rochelle salt. This is one of the crystals in which piezoelectricity was first detected by the Curie brothers. The early observations of the unusual behavior of this crystal were followed by the work of J. Valasek and many others, beginning in 1921. The most complete theoretical treatment was that of H. Müller, 1935–1940. Rochelle salt was the first crystal in which "ferroelectric" properties were discovered.

Our work at Wesleyan during this period had largely to do with Rochelle salt. Vibrational modes were examined, and methods devised for keeping tuning forks and long rods in continuous vibration. Piezoelectric constants were measured, as well as the dependence of frequency of Rochelle-salt bars on temperature.

It was found that flat plates of Rochelle salt could be cut in an orientation that enabled them to be vibrated piezoelectrically in a thickness compressional mode, like the *X* cut of quartz. This was the *L* cut. It found a limited application for generating ultrasonic waves.

During part of this time, Dr. Hans Jaffe was a member of my laboratory group. He prepared a large part of the material for the chapters on Rochelle salt for my book, *Piezoelectricity,* which was published in 1946.

Meanwhile another group of crystals with characteristics much like those of Rochelle salt was being investigated by Busch in Zürich. These were the phosphates and arsenates of ammonium and potassium. Owing to their resemblance to Rochelle salt, Busch called them "Seignette-electrics," a term that is now abandoned in favor of "ferroelectrics." Today, the name "ferroelectric" is applied to all crystals whose dielectric properties show analogies with the ferromagnetic properties of iron. The most important examples at present are barium titanate and its relatives.

Of the crystals investigated by Busch, the one that has proved to be most useful is ammonium dihydrogen phosphate, known as ADP. Although its piezoelectric effect is less than a tenth as great as that of Rochelle salt, it is so much better in other ways that, for most purposes, it came to replace Rochelle salt, only to be superseded later by barium titanate.

The ferroelectrics do not make good frequency standards. They are chemically less stable than quartz, and the fact that they show hysteresis indicates that temperature control is not so easy. Their field of application is rather electroacoustic and electromechanical, where power and sensitivity are all-important.

Much of the piezoelectric activity at Wesleyan was carried on by Professor Van Dyke or under his direction. Among other things, he found that, when a long thin quartz bar was excited in resonant thickness vibration by an electric field applied near one end, the entire bar became excited so strongly in thickness vibration that, when one's finger was touched to the surface near the other end, the skin became seared. A drop of water applied to the surface evaporated almost explosively. This was in 1924, several years before Wood and Loomis published their results on intense piezoelectric vibrations.

On the other hand, he found that the friction between a quartz crystal and a metallic surface was greatly reduced when the crystal was in a state of vibration. He also found that, when a quartz sphere several centimeters in diameter was vibrated electrically, the motion of the surface was such that the sphere slid continuously around on a circular track without rolling.

Van Dyke devised methods for measuring the rate of decay of crystal vibrations, also for measuring the equivalent electrical constants. In 1941, in measuring the decrement in a quartz ring, he observed values of the quality factor *Q* up to six million.

During most of the second world war, I was working on my book. By the end of 1941, the chapters on Rochelle salt were largely completed, and I was asked to bring them with me to San Diego, where work was being done at the Naval and Sound Laboratory on Point Loma, California, on Rochelle salt for Sonar transducers, as well as on the acoustic properties of sea water. Van Dyke had already preceded me there, and he stayed on after I returned to the east.

The German submarines in the second world war were faster and quieter, besides being equipped with effective listening devices. All this made their detection and destruction more difficult. Detection by ultrasonic echoes, while useful, proved to be insufficient. A more effective weapon was the aircraft, carrying radar and rockets, together with the sono buoy. Magnetic air-borne detection, mentioned earlier, also played an important part at close range.

For communication, the armed forces made enormous use of crystal-controlled transmitters in the second world war. It is said that 75 million quartz crystals were used, for land and sea equipment, including tanks and other vehicles. Many outfits con-

tained a considerable number of crystals for different frequencies.

Toward the end of the war, I was asked to help at the Radiation Laboratory in Cambridge, Massachusetts, in some problems related to the radar trainer. In this device, a narrow ultrasonic beam from a thin quartz plate is rotated so as to scan the bottom of a tank of water. The echo reflected from a pattern in relief is received and presents a picture of the pattern on an oscilloscope. In this way, the operation of the radar equipment on a plane can be simulated in the laboratory.

The immediate purpose of my contribution was to measure the output of power radiated from the quartz. This work was done at Wesleyan with the assistance of F. T. Dietz and P. D. Goodman. At that time, transducer theory was in an early stage, and it was necessary as a first step to work out the theory of this particular type. As it turned out, the progressive-wave method that I adopted at that time provided material for several papers in the years following.

In the course of measuring power (by radiation pressure), I made an experimental study of the "streaming" that always accompanies acoustic waves in fluids. These observations were followed later by the theoretical treatment by C. Eckart, P. J. Westervelt, and others.

After World War II, research was continued at Wesleyan with aid from the Office of Naval Research (ONR), and, under Van Dyke's direction, from the U. S. Signal Corps. Under the ONR, the projects had to do chiefly with transducer theory, methods of measurement, and the theory of acoustic–radiation pressure. Much of this was done by Dr. J. S. Mendousse, and later by Dr. F. E. Borgnis, with aid from P. D. Goodman and T. Niemiec.

Van Dyke's research was a theoretical and experimental investigation of modes of vibration of quartz, along with methods of measurement of their electrical-impedance characteristics.

In 1950, my research was shifted to the California Institute of Technology in Pasadena, supported first by ONR and later by the U. S. Air Force. Messrs. Borgnis and Niemiec came with me. The work included resonator and filter theory and the measurement of acoustic power. Dr. Borgnis made theoretical studies of the acoustic interferometer, radiation pressure, acoustic streaming, and wave propagation in anisotropic media.

When the research at Caltech came to a close in 1955, one project was still incomplete. This was an attempt to produce acoustic waves at a frequency of 3000 megacycles per second (Mc/sec). The first step was to find out whether there was anything like a piezoelectric effect in multilayer films prepared by the Langmuir technique. When the result turned out to be negative, I used a quartz plate 0.101 millimeters thick, ground and polished so as to be as nearly plane-parallel as possible. For excitation at this very high frequency, the crystal had to be placed in a microwave cavity. It lay at the bottom of a small glass jar containing water. In contact with the crystal was a thin layer of dye. I hoped that, when the power was turned on, the crystal would be set into thickness vibration at its 105th harmonic and cause a column of dye to stream upward through the water. In the work for Radiation Laboratory mentioned above, I had found that the streaming of dye was a sure indication of the presence of acoustic radiation. Even if the thickness of the quartz plate was not strictly uniform, still it was to be expected that some portions of the surface would vibrate in resonance, giving rise to local streaming.

The final tests were made at the Hughes Aircraft Company, Culver City, California, where a klystron was kindly placed at my disposal. The dye showed no unwillingness to move. On the contrary, as soon as the high-frequency voltage was applied, heat was generated so quickly that any systematic streaming that may have been present was masked by disorderly convection currents.

Since then, resonant vibrations in quartz have been detected at frequencies even higher than 3000 Mc/sec by several investigators. This is a very promising field for future research and indicates that ultrasonics is still a vital, growing subject.

1. I am indebted to Dr. A. R. Laufer of the Office of Naval Research for his kindness in securing various reports and transcripts from Washington.
2. Sir Thomas More, *Utopia*, edited, with introduction and notes, by J. Churton Collins (Clarendon Press, Oxford, 1904), p. 78.

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