Townes’ contribution to nonlinear optics

Elsa Garmire
Dartmouth College, Thayer School of Engineering, Hanover, NH 03755

garmire@dartmouth.edu

Abstract. In honour of the Fiftieth Anniversary of the Nobel Prize in Physics, this talk introduced the contributions of Nicholas Basov and Alexei Prokhorov, who shared the prize with Charles Townes. The talk then detailed the quantum electronics research of Townes, particularly at MIT, which was related to nonlinear optics. The years from 1961 to 1968 were particularly exciting, as the ruby laser enabled a wide variety of new physics to be discovered and explored.

1. Introduction
The 1964 Nobel Prize in Physics was awarded “for the invention of the maser and the laser” to Charles Hard Townes, Nicolai Basov and Aleksandr Prokhorov (figure 1). At the ceremony it was stated, “Drs. Townes, Basov and Prokhorov: By your ingenious studies of fundamental aspects of the interaction between matter and radiation you have made atoms work for us in a new and most remarkable way.” [1]

The Nobel Committee went on to say, “The first papers about the maser were published ten years ago as a result of investigations carried out simultaneously and independently by Townes and co-workers at Columbia University in New York and by Basov and Prokhorov at the Lebedev Institute in Moscow. …Masers work as extremely sensitive receivers for short radio waves. They are of great importance in radio astronomy and are being used in space research for recording the radio signals from satellites.”

Figure 1. Nobel Prize winners in Physics from 1964: Townes, Prokhorov and Basov (left to right) Portraits from Nobel website http://www.nobelprize.org/nobel_prizes/physics/laureates/1964/

The independent work of Townes in the United States of America and of Basov and Prokhorov in the Union of Soviet Socialist Republics brought forward the field of Quantum Electronics. The term “quantum” ties the field to physics and “electronics” ties the field to electrical engineering. The USSR researchers were motivated by the development of synchrotron sources for spectroscopy. Townes was motivated by his work on radar sources during World War II. The state
of their research at the end of the war is documented by Prokhorov and Basov’s papers on frequency stabilization of synchrotron sources [2] while Townes published on cathode sputtering [3].

Research in both groups evolved into studies of microwave spectroscopy during the late 1940’s and early 1950’s. Prokhorov and student Basov were drawn to amplification of microwaves by the desire to achieve narrower spectral lines. Townes was drawn to the possibility of molecules as new microwave sources and fundamental molecular physics questions. The breakthrough idea, achieved independently and essentially simultaneously, was to create a molecular oscillator from a molecular amplifier by introducing microwave feedback. This concept developed from experience in electrical engineering building electron-tube amplifiers and oscillators at radio frequencies. The Russians called their new idea a “molecular generator” of microwaves and published it in Russian in a paper submitted January, 1954 [4]. Townes called his a MASER (microwave amplifier by stimulated emission). Townes independently developed the same ideas and demonstrated the first experimental microwave maser in a paper submitted in May, 1954 [5]. The Russians contributed their idea of a three-level maser soon thereafter [6].

These scientists were not the first to discuss molecular amplification. Stimulated emission had been introduced by Albert Einstein as the direct analogue to absorption in 1914 [7]. When a two-level population distribution is inverted (more in the excited state than in the ground state), incoming radiation is not absorbed, but stimulates emission from excited atoms, resulting in more light out than came in: electrical engineers call this gain. In 1951 Fabricant in the USSR predicted this result [8], while in the USA Purcell and Pound performed an experiment in a nuclear spin system and observed that immediately upon reversing the magnetic field the system had a negative temperature. The nuclear spin was reversed with respect to thermal equilibrium. They pointed out that the observed reversed deflection corresponds to induced radiation [9]. This radiation would be caused by stimulated emission. Later, in 1954, Joseph Weber pointed out that the inverted population in this system produces gain by stimulated emission and can become a source of microwaves [10].

“Quantum Electronics was born at the moment when an excited quantum system — a beam of specially sorted molecules — was placed into a cavity” [11]. This was the achievement that resulted in the Nobel Prize of 1964. Work on quantum electronics progressed rapidly in both the USA and USSR, as soon as Townes reported a working MASER; indeed, in Europe as well. The era after WWII was one of blossoming interest in science, both for its own sake and for its military applications. One of the lessons learned from WWII was the importance of forefront scientific effort to aid the military and political power of the advanced countries within Europe and the US.

The focus of much of the maser research, besides studies of the device itself, was on microwave spectroscopy, using the high resolution that the maser made possible. Townes and his brother-in-law Arthur Schawlow published a book Microwave Spectroscopy in 1958, which presented a systematic, comprehensive account of the theory, techniques, experimental data, and interpretation involved in the study of microwave spectroscopy. The book is still available and continues to serve as an important reference [12].

2. Townes’ Research after the Maser

From 1954 to 1959, Townes, a Professor at Columbia University, carried out a diversified research program that included both ongoing microwave spectroscopic studies and analysis of the performance of his maser [13]. In 1955 he and his student discovered oscillatory population transfer between two closely-spaced atomic/molecular levels that split the absorption spectra between the ground state and either of these levels [14]. This has been called Autler-Townes splitting (he called it an AC Stark effect), produced by a radio-frequency field tuned to spectral resonance.

After 1956 Townes was working on ways to push the maser to shorter and shorter wavelengths, first to mm waves. In 1958, he and Schawlow made the jump all the way to infrared and optical frequencies [15]. Their contribution was to suggest that a large, multimode “open” resonant optical cavity (in the Fabry-Perot geometry) could lead to extremely monochromatic and coherent light. The narrow atomic/molecular gain line serves to limit the output to only a few cavity modes. They suggested optical pumping in a three-level system as a way to invert the population and were the
first to analyse theoretically the oscillation condition. Their numerical evaluation of a potassium vapour “optical maser” found the idea to be feasible (although it turned out to be experimentally very difficult).

The Schawlow-Townes analysis excited the quantum electronics community and the race was on to demonstrate a working laser. Their ground-breaking paper defined the laser as an inverted population of atoms/molecules within a large highly-multimode optical cavity that amplifies spontaneous emission noise to generate an output in the infrared or visible. For a directional output, they specified two parallel mirrors (the Fabry-Perot geometry). Townes called this an “optical maser” to affirm that the maser concept at optical frequencies is not different, in principle, from the microwave system.

This time period (mid-1950’s) was during the Cold War, a mutual military build-up between the USA and the USSR. The US military asked Townes for technical advice about the future of the new field of quantum electronics. He agreed to chair two important committees: one was “to create interest in mm waves” and the other was “to urge continued support for research in the infrared.” These commitments were unpaid and after-hours, but he took them very seriously. Thus he was solicited to take a leave of absence from Columbia as Vice President and Director of Research for the Institute for Defense Analysis in Washington D. C., a position he held from 1959-1961. As he said in an oral interview 25 years later, “I felt that there just were not enough good scientists in Washington, and we had a pressing problem with the Russian missiles and other things coming on, and it was just a part of my duty.” [16]

In the fall of 1961, Townes accepted an offer to become Provost at Massachusetts Institute of Technology, a powerful position that reports directly to the President. Research in lasers became his after-hours unpaid activity. He took me on as his first PhD student; six months later he added Raymond Chiao. We were his only graduate students, but we had an active group that included post-docs and visiting scientists. During this time Townes also became Chairman of the Advisory Committee to NASA’s Project Apollo (the man-to-the-moon mission). His responsibility was to secure support from the larger scientific community and to ensure that the moon flight would yield maximum benefits in scientific research. This was a highly political committee that put him in conflict with some at MIT and he was denied the open position of MIT president. So in 1967 he moved to the University of California in Berkeley to search for stimulated emission in astronomy.

Why did Townes leave laser research? He could see that the quantum electronics field was brimming over with eager and talented scientists/engineers. He told me that he wanted to work in unexplored research areas, where he thought his contribution would be greatest. Stimulated emission in astronomy was a wide-open field, eagerly awaiting his arrival. Indeed, he has since made many significant contributions to astronomy (outlined later).

The purpose of this paper is to focus on Townes’ contributions to nonlinear optics (NLO), which mostly resulted from the years he was at MIT: 1961-1967.

3. Nonlinear Optics at MIT
It pays to be brilliant and active as a field is just beginning. In the space of 6 years Townes, along with his colleagues: 1) Proved that coherent molecular vibrations drive higher order stimulated Raman scattering (SRS) emission; 2) Identified SRS emission from individual filaments in Raman-active liquids; 3) Introduced stimulated Brillouin scattering (SBS) and observed it in crystals and liquids; 4) Introduced and theoretically analysed spatial solitons (SS), at that time called self-trapped optical beams; 5) Experimentally demonstrated self-focusing filament-formation in Kerr liquids; 6) Demonstrated small-scale filamentary beam break-up; 7) Introduced the concept of self-phase modulation (SPM) and 8) Introduced the concept of weak-wave retardation and gain in four-wave mixing (FWM).

It was my job to set up the Townes laser laboratory at MIT with the second commercially sold ruby laser (from a company called Trion – the first went to Peter Franken at University of Michigan). The first year was spent just getting the laser to work – the ruby had to be cooled to liquid nitrogen temperatures while excited by a flash lamp operating at kilovolts. Electrical shorts from condensation on the Dewar were a major problem! We evaporated a silver mirror on one end of the ruby rod and a partially silvered mirror on the other. Technological progress toward Q-switching was rapid: our Q-switch was a retro-reflecting prism mounted on a high-speed air-driven
turbine (developed at Lincoln Laboratories); the ruby crystal was put into an external cavity; and
dielectric mirrors replaced silver (which blew off with the high-power Q-switch). Now we were
ready for high power experiments, which are described here.

3.1. Coherent Molecular Vibrations in Stimulated Raman Scattering

Townes’ first contribution was to describe the stimulated Raman scattering process as introducing
coherent molecular oscillations [17]. He had been inspired to delve into nonlinear optics at the
Third Quantum Electronics Conference in Paris in Feb. 1963. He had heard Zeiger and
Tannenwald’s theoretical paper on optical phonons. He had heard that experiments on stimulated
Raman scattering at Hughes Research Laboratories demonstrated Stokes frequencies shifted by up
to three orders of vibrational frequency. He also heard about the intense anti-Stokes emission seen
by both Terhune and Stoicheff. Because Stoicheff became a visitor at MIT, we had early access to
his data. All this led Townes to realize that coherent laser light could drive coherent optical
phonons (which we called molecular oscillations).

The Raman process was modelled as an ensemble of simple vibrating diatomic molecules with
natural resonance frequency $\Omega_r$ [18]. Townes’ innovation was to realize that coherent light
interacting with matter transferred that phase coherence to the molecules. Periodic vibrations
introduced into the medium by scattering into Stokes frequencies could be transferred back to the
light wave through a coherent process, generating anti-Stokes light. This was a classic resonant
parametric process.

The anti-Stokes could be explained by the requirement of phase coherence over the interacting
length, a process called phase-matching, first described in second harmonic generation. Molecular
vibration driven by Stokes generation from a laser photon has a wave vector $K$. A second laser
photon scatters off $K$ to produce anti-Stokes. Phase-matching means anti-Stokes generation is
conical at an angle given by closing the wave-vector diagram (figure 2); i.e. conservation of
momentum.

The theoretical paper was followed by an experiment with a Q-switched ruby laser transmitted
through calcite that proved that the parametric process did, indeed, occur – that anti-Stokes light
depleted a cone of stokes light at the correct phase-matching angle. Figure 3 shows the bright ring
of anti-Stokes radiation along with a negative of the Stokes radiation pattern, with the arrow
pointing to a weak ring showing depletion at the phase-matching angle [19].

My research effort was to study SRS in Raman-active liquids using the Q-switched ruby laser [20];
now theory and experiment did not agree, as they did in calcite. This turned out to be due to
filament-formation and self-phase modulation in liquids with nonlinear refractive indices.

3.2. Filament formation in SRS emission

Typical images of the radiating cones of anti-Stokes SRS generated in acetone are shown in figure
4a. The bright ring, always observed, violated the theory described above. It is the weak inner ring
that corresponds to the above phase-matching angle. Further analysis showed that the bright ring at
a larger cone angle corresponds to a theory describing phase-matching only along the direction of
propagation. The reason was shown later to be due to the formation of self-focusing filaments.
Because the Raman Stokes emission was strongly forward-directed, there was not enough Stokes
emission at the angle required for anti-Stokes to be generated at the volume phase-matching angle.
The measured anti-Stokes cone angle phase-matched in the direction of propagation only, and
corresponded to a parametric interaction with the intense forward-going Stokes wave. The lack of
lateral phase-matching was acceptable because this Raman interaction took place only in narrow filaments. This case can be called Cerenkov phase-matching, as opposed to volume phase-matching.

Several additional experimental results verified this discovery. When the 10-cm liquid cell was tilted off-axis, reflection from its glass faces provided feedback into an off-axis cavity. This produced a Raman Stokes laser with an intensity $10^4$ times larger than that without feedback. The Stokes radiation at the required volume phase-matching angle (figure 4c) was now bright enough to parametrically generate anti-Stokes radiation at the expected angle (figure 4d, e). Proof of the parametric process is the decreased Stokes at the volume phase-matching angle is shown in figure 4f).

Figure 4. Examples of stimulated Raman emission in liquids: (a) acetone and (b) cyclohexane both show two kinds of conical emission of anti-Stokes; the inner cones correspond to the volume phase-matching theory (above) and the outer cones correspond to Cerenkov phase-matching; (c) shows a full circle of Cerenkov phase-matched anti-Stokes as well as a weak beam of anti-Stokes at the volume phase-matching angle. The latter was made possible by an off axis resonator for Stokes radiation, that generated Stokes seen in (d) at the complementary angle from the anti-Stokes spot; (e) and (f) show how the anti-Stokes in (e) can sap energy from the Stokes in (f) at angles $\alpha$ corresponding to volume phase-matching theory [21, 22].

Independent proof that the anti-Stokes radiating into the large angle cones came from filaments (and thus thought of as Cerenkov radiation) is shown in figure 5a. This is the first direct evidence of self-trapped filaments emitting SRS. Further proof can be seen by comparing elliptical focussed volume phase-matching anti-Stokes in calcite (which has no self-trapped filaments) in figure 5b with anti-Stokes from benzene, where the same elliptical focusing geometry generated mostly circular emission from self-trapped filaments.

Figure 5. Angular distribution of stimulated Raman anti-Stokes radiation; (a) Separate radiation from two different filaments caused side-by-side cones of anti-Stokes. A single anti-Stokes cone from volume phase-matching is also seen; (b) Elliptical volume phase-matching in calcite produced with a cylindrical lens; (c) Circular Cerenkov phase-matching from self-trapped filaments produced with the same conical lens [22, 23].

When the laser output was multimode, the anti-Stokes emission had a broad wavelength spectrum. Later it was realized that this was due to self-phase modulation in the nonlinear organic liquids. With a slit and an imaging spectrometer, the wavelength dependence of the output angles differs markedly between volume emission and filament emission (figure 6).

With a single-mode ruby laser, the anti-Stokes output was single-frequency, both with volume and surface emission. The spectrometer nicely separated out the different orders of anti-Stokes
emission, although the lack of dynamic range of photographic film made it difficult to see several orders simultaneously.

3.3. Stimulated Brillouin Scattering
Townes was the first to predict stimulated Brillouin scattering (SBS) and his group was the first to demonstrate it. He understood that “Stimulated Brillouin scattering of an intense optical maser beam involves coherent amplification of a hypersonic lattice vibration and a scattered light wave. … It is analogous to Raman maser action, but with the molecular vibration replaced by an acoustic wave with frequency near 30 GHz…Both the acoustic and scattered light waves are emitted in specific directions” [24].

The Brillouin wave has a Stokes frequency shift $\Omega_s$ that depends on its emission angle $\theta$ as $\Omega_s = \frac{\omega_o (v_{ac}/v_{ph}) \sin(\theta/2)}{v_{ac}^2}$, where $v_{ac}$ is the velocity of the acoustic wave and $v_{ph}$ is the velocity of the photon. For retro-reflected waves, which are the most intense, the Stokes down-shift is $\Omega_s = \frac{\omega_o (v_{ac}/v_{ph})}{v_{ph}}$. Stimulated Brillouin scattering can be considered parametric generation of an acoustic wave and a scattered light wave from an initial laser wave. With such a small frequency shift, SBS is most easily observed with a Fabry-Perot interferometer.

With solids quartz and sapphire, SBS occurred just as predicted [24]. This first observation of SBS in fused silica validated a process that was shown later to strongly limit the power that can be transmitted through fibers. Figure 7a shows typical Fabry-Perot spectra in these solids.

The Brillouin wave has a Stokes frequency shift $\Omega_s$ that depends on its emission angle $\theta$ as $\Omega_s = \frac{\omega_o (v_{ac}/v_{ph}) \sin(\theta/2)}{v_{ac}^2}$, where $v_{ac}$ is the velocity of the acoustic wave and $v_{ph}$ is the velocity of the photon. For retro-reflected waves, which are the most intense, the Stokes down-shift is $\Omega_s = \frac{\omega_o (v_{ac}/v_{ph})}{v_{ph}}$. Stimulated Brillouin scattering can be considered parametric generation of an acoustic wave and a scattered light wave from an initial laser wave. With such a small frequency shift, SBS is most easily observed with a Fabry-Perot interferometer.

With solids quartz and sapphire, SBS occurred just as predicted [24]. This first observation of SBS in fused silica validated a process that was shown later to strongly limit the power that can be transmitted through fibers. Figure 7a shows typical Fabry-Perot spectra in these solids.
Brillouin-shifted retro-reflected beam (B); (b) Single-frequency laser light illumination; (c) SBS from a ruby laser focused in water [24, 25].

In liquids, a series of several orders of Stokes Brillouin shifts was observed, as shown in Fig. 7c compared to Fig. 7b, which could not be explained by the SBS theory. It turned out that the Stokes Brillouin wave was retro-reflecting into the inhomogeneously broadened ruby laser, where it found additional gain because it was at a new frequency [25]. It then came out of the laser as a new Brillouin-shifted frequency and entered the liquid cell where SBS caused another retro-reflected Brillouin shift. And so on. Many years later it was determined that the ability of the SBS Stokes light to go exactly back into the laser had occurred because of phase conjugation [26]. In 1964, while I was still a graduate student, I did not think sufficiently about why the retro-reflecting light should follow such an exact backward path through the lens, the mode selector and the imperfect ruby rod. If I had, I would have been the first to identify phase conjugation!

3.4. Spatial Solitons

In 1964, Dr. Townes became aware of the fact that when Mike Hercher at University of Rochester focused a powerful Q-switched ruby laser into a glass block, it left a trail of damage that did not appear to spread with distance by diffraction [27]. That led Townes to think about how optical nonlinearities could overcome diffraction and to our working out the theory, which we submitted in September and was published in October, 1964. We described the reported experimental data this way:

“The thin threads of damage in glass and other materials …fairly easily demonstrated in glass by focusing a ruby-laser beam greater than a few megawatts inside good optical glass. Usually, though not always, there is extensive damage near the focal point and beyond the focal point a long straight filament of small bubbles and damage along the lens axis, accompanied by ionization. This filament may be as long as several centimeters and at the same time have a diameter of only a few wavelengths. This diameter is in some cases two orders of magnitude smaller than the focal diameter, assuming linear optics.” [28]

The idea was as follows: “an electromagnetic beam can produce its own dielectric waveguide and propagate without spreading. This may occur in materials whose dielectric constant increases with field intensity, but which are homogeneous in the absence of the electromagnetic wave.” [28]

An estimate was obtained by considering diffraction of a circular optical beam of uniform intensity across diameter $D$ in material for which the index of refraction may be quadratic in field with a coefficient $n_2$. A self-consistent waveguide will occur when the divergence angle of the finite beam, $\theta_0 = 1.22 \lambda / nD$, is set equal to the critical angle for total internal reflection, for a refractive index discontinuity of $\Delta n = n_2 |E|^2$. This gives a threshold power in the laser beam of $P = (1.22 \lambda)^2 c/64 n_2$, a total power independent of beam diameter. Inserting numbers, the beam should self-trap at approximately 1 Megawatt.

3.4.1. Exact solutions for confined beams

When lateral variation occurs only in one dimension (a slab-shaped beam), the nonlinear Helmholtz equation for the transverse field is given by $E_{yy} - \Gamma^2 E_y + (\varepsilon_2/2) k_0^2 E E = 0$, where the y subscript means the derivative in the y direction [28]. The analytic solution for the transverse field dependence is stable: $E(y) = E_0 \text{sech}(\Gamma y)$, where $\Gamma = \frac{1}{2} \varepsilon_2^{1/2} k_0 E(0)$. Such self-trapped beams have, indeed, been generated in waveguides.

In two dimensions with a cylindrical beam, the Helmholtz equation (in dimensionless units) is

$$\frac{d^2 E^*(r)}{dr^*{}^2} + \frac{1}{r^*} \frac{dE^*(r)}{dr^*} - E^*(r^*) + E^2(r^*) = 0,$$

There is no analytic solution, but Figure 8 shows the “Townes Profile,” which was calculated by computer. When compared to the Gaussian beam, it can be seen to be close. Integration of this solution gives the critical power: $P = 5.763 \lambda^2 c n_0 / 8 \pi n_2 n_0$, very close to the initial estimate.

We observed self-trapping experimentally by inserting microscope slide cover slips along the beam path and reflecting successive weak images as the beam traveled through carbon disulfide [29]. Figure 10 shows the near field of the beam at successive locations for (a) low power, with (b) and (c) representing successively higher powers. The beam is seen to not diffract but to
maintain a constant beam diameter. At higher powers a higher order spatial soliton can be seen. It should be pointed out, however, that this early data was not at all definitive because other nonlinearities such as SRS and SBS were undoubtedly occurring at the same time. It would take considerable additional work before these effects began to be separately studied.

Figure 8. Numerical analysis for electric field in a spatial solitary wave, the “Townes Profile,” compared to a Gaussian profile (dashed line).

Figure 9. Self-focusing for beams above threshold power, in a manner similar to a graded index lens.

The self-trapped beam that was predicted is now known as a “spatial soliton” and has been seen in a number of situations. Today, an important application for self-trapped beams is in plasmas, particularly those created by ultra-short laser beams in air. Self-focusing in plasmas was, in fact, independently predicted before self-focusing in liquids or solids [30].

Figure 10. Experimentally observed images of an apparently self-trapped ruby laser beam, 60 μm in diameter, as it travelled through carbon disulphide [29]

3.4.2. Instabilities in self-trapped beams

Paul Kelley pointed out that the same nonlinearity that creates a self-trapped beam of just the right power will self-focus beams of powers higher than the critical threshold [31]. The nonlinearity overpowers diffraction, creating spherical surfaces of constant phase that focus the beam as shown in figure 9. The self-focusing process will cease when higher-order nonlinearities take over.

In order to achieve the results shown in figure 10, it was necessary to ensure that the ruby laser operated in a single spatial mode. With multi-spatial modes, large-scale trapping was not observed. In both cases, however, close observation showed that nonuniformities in the beam caused it to break up into many small-scale filaments on the order of only a few microns.

More careful analysis showed that superimposed on the large scale trapping were many small-scale filaments, only a few μm in size, apparently introduced by instabilities [32]. Such instabilities in χ(3) materials had been predicted by Talanov [33].

Later experiments working with Brewer from IBM demonstrated that these filaments last only ~ 10⁻¹⁰ sec, after which they "blow up" [34], presumably because heating expands the liquid. With measured diameters of 4 μm in CS₂ and 12 μm in nitrobenzene, they were estimated to contain about 10kW peak power. This power level agreed with theory that assumed the Kerr effect is the principal mechanism for initiating filaments. For powers above the trapping threshold, filaments were seen to decrease in size and increase in intensity until higher-order terms in the electric field caused saturation of the nonlinear refractive index. Measured diameters were 10 × larger than theoretical limiting diameters based on Kerr effect saturation, which indicated that some other mechanism determined filament size. We measured lower than expected Raman gain in most of the filament length, which was explained by frequency broadening and dispersion.

Self-focusing and self-trapping have remained a very important factor in a number high-power laser experiments and many of the interrelated phenomena have been carefully disambiguated. A recent review offers an excellent and up-to-date review of the status today of these filaments [35].
While self-trapped filaments tend to be unstable in a material with a $\chi_3$ nonlinearity, they are stable in photorefractive $\chi_2$ materials. Figure 11 shows an example in photorefractive barium titanate.

Figure 11. Photograph of green laser beam through photorefractive barium titanate: (a) normal diffraction at lower power; (b) self-trapped beam at higher power. [Courtesy Professor Z. Chen, https://www.physics.sfsu.edu/~laser/research.html]

3.5. Stimulated four-photon light scattering

Influenced by the realization of stimulated scattering from Raman and Brillouin effects, Townes’ group also analyzed stimulated Rayleigh scattering [36]. They assumed a stationary, or near-stationary, periodic variation in refractive index change would occur as a result of interfering light beams and a nonlinear refractive index.

Stimulated Rayleigh-wing scattering discussions had not yet included the influence of stimulated light-by-light scattering and its associated weak-wave retardation. With both effects having large gains in the forward direction, the proper analysis required coupled waves. The paper analyzed the interaction between three waves: undepleted laser field $E_0$, weak wave $E_1$ at a small angle (or frequency difference) and amplified weak wave $E_2$. We were the first to show that weak wave $E_2$ experienced a gain per unit length of $g = 4\pi\Delta n/\lambda_o$. In nonlinear media, $g = \pi\varepsilon_0^2|E_0|^2/n_o\lambda_o$ (cgs units). Since this is a parametric process, the weak wave $E_1$ experiences a loss per unit length equal to the gain of $E_2$ per unit length. When the waves experience 1/e loss in lengths $L_1$ and $L_2$, there will be a threshold for observation of this parametric amplification given by $4\pi\Delta n = \pi\varepsilon_0^2|E_0|^2/n_o > \lambda_o/(L_1L_2)^{1/2}$. Because the weak waves are retarded relative to the strong wave (whose wave vector is extended by the nonlinearity), phase-matching gives rise to small angles for $E_1$ and $E_2$ with respect to $E_0$. The gain was shown to be strongly peaked at the phase-matching angle. Just as in SRS, transverse momentum conservation requires that if wave $E_1$ travels with angle $\theta$ with respect to the laser beam, wave $E_2$ travels at $-\theta$, both angles being very small. It is simple to show that this angle is $\theta = \pm(2\Delta n/n_o)^{1/2}$. This is easiest seen through picturing that interference causes stationary periodic layers in the medium, which can produce reflections in a manner similar to the Raman-Nath effect.

The assumption was local nonlinearity, so that degenerate gratings in the material are unshifted with respect to the optical interference fringes. Even so, this analysis showed that steady-state amplification was possible, via the parametric four-wave interaction. The phase-matching condition was met only for a strict angular window, and only there could net gain be achieved. An experiment was carried out with the Q-switched ruby laser in carbon disulfide to demonstrate that a weak wave did, indeed, experience parametric gain at the appropriate angle [37].

Considering only a weak and a strong wave, the nonlinear shift in the index of refraction induced by the pump wave on the probe wave was shown to be double that compared to the pump wave itself, which implies that the probe-wave vector is longer than the pump-wave vector. This additional lengthening of the weak-wave propagation vector had lasting impact on the thinking about two-wave, three-wave and four-wave mixing, particularly in photo-refractive media. It also provided early understanding of how instabilities grow and cause filamentation of high power beams traveling through nonlinear media.

3.6. Self-steepening of light pulses

Townes and his group predicted a change in temporal shape of light pulses traveling through a medium with an intensity-dependent refractive index [38]. They calculated the time required for a Gaussian pulse to steepen into a temporal optical shock through both analytic and numerical solutions for the pulse as it traveled through the medium. They investigated temporal development of pulses for both zero and nonzero relaxation times of the nonlinear $\chi_2$ refractive index.
The frequency spectrum calculated in the zero-relaxation time limit, found the largest peak intensities on the lower-frequency side (figure 12). The rate of steepening is modified when pulse decay time is as short as the nonlinear refractive index; in a dispersionless medium the pulse decay time can become arbitrarily short. Estimates were made for both the thickness of the optical-shock region and the frequency spreading allowed by dispersion when relaxation was added. This phenomenon is now well-known and more familiarly called self-phase-modulation.

![Figure 12. Self-steepening and frequency-broadening of a Gaussian laser pulse through $\chi^3$ medium.](image)

(a) Time dependence of intensity of a sinusoidal input at $z=0$, $z_1$, and $z_2$; (b,c) Spectra of a sinusoidal pulse at $z_1$ and $z_2$ respectively. At $z_2$ the most intense peak is $\sim 2000$ cm$^{-1}$ below the ruby laser wavelength. Adapted from [35].

4. Other MIT research in Nonlinear Optics
Professor Townes continued to interact with and inspire more work on self-trapped filaments of light. With Brewer at IBM he looked at standing waves in these filaments [39]; with Sacchi and Lifiszt he investigated anti-Stokes generated in trapped filaments [40]; phase modulation was investigated with Cheung et. al. [41]. As a result of the earlier work, spectral broadening of light in filaments was analysed [42], along with self-trapping in saturating media [43]. Similar studies broadened to new fields, such as thermal self-focusing [44] and coherent excitation of polaritons [45]. This list includes only work done in the field by those who had collaborated directly with Townes while he was at MIT and is not meant to be comprehensive (567 papers in nonlinear optics were published between 1961 through 1969, according to the Information Sciences Index).

5. Contributions of Townes to Astronomy (1967-2011)
Here is a list of some of the major topics of investigation in which Townes was involved at University of California, Berkeley:
- Water in interstellar space
- Ammonia in interstellar space
- Dust around stars
- Mechanisms in the galactic center
- Mechanisms of star formation in galaxies
- Far-infrared spectroscopy of galaxies
- A heterodyne stellar interferometer for the mid-infrared

Townes had long been interested in astronomy, ever since his initial work with radar during WWII and had a firm conviction that the nonlinear photon-atom processes that were observed terrestrially with lasers would be observable in space. He was rewarded by finding this so and continuing active research well into his 90’s. The last research papers listing him as a co-author were in 2011 when he was 96 years old!

6. Contributions of Basov and Prokhorov (1967-2011)
This paper focused on Charles Townes, one of the Nobel Prizewinners. It did not include the important contributions that Nobelists Basov and Prokhorov have made to the field. Basov introduced the idea of semiconductor lasers as early as 1958 [46]. Prokhorov ran a laboratory with wide-ranging interests, with developments often proceeding in parallel with those in the US, including Q-switching, new pumping techniques, improving laser coherence and increasing power. Contributions were made in nonlinear optics, fiber and integrated optics, and in a variety of laser
applications, especially in fiber-optic communication, laser technologies and the use of lasers in medicine and ecology. An excellent source for English translations of early papers describing quantum electronics research in the USSR can be found on-line [47].

References

[1] Edlén B 1964 Nobel Prize Award Ceremony Speech, retrieved at http://www.nobelprize.org/nobel_prizes/physics/laureates/1964/press.html

[2] Rytov S M, Prokhorov A M and Zhabotinsky M E 1945 On the theory of the frequency stabilization Z. Ekperim. Teoretich. Fiziki 15 557

[3] Townes C H 1944 Theory of Cathode Sputtering in Low Voltage Gaseous Discharges Phys. Rev. 65 319

[4] Basov N G and Prokhorov A M 1954 Zh. Eksp. Teor. Fiz. 27 431, in Russian; available translated at http://www.quantum-electron.ru/laser50_eng.html

[5] Gardner J P, Zeiger H J and Townes C H 1954 Molecular microwave oscillator and new hyperfine structure in the microwave spectrum of NH$_3$ Phys. Rev. 95 282

[6] Basov N G and Prokhorov A M 1955 Possible methods of obtaining active molecules for a molecular oscillator Sov. Phys. JETP 1 184

[7] Einstein A 1916 Strahlungs-emission und -absorption nach der Quantentheorie Verhandlungen der Deutschen Physikalischen Gesellschaft 18 318

[8] Fabrikant V A (1939) On experimental evidence for the existence of negative absorption Doctor of Science (Habilitation) Thesis, translation of pages 273 and 274. Found at http://www.quantum-electron.ru/pdfrus/laser50/11788_eng.pdf

[9] Purcell E M and Pound R V 1951 A nuclear spin S-system at negative temperature Phys. Rev. 81 279

[10] Weber J 1953 Amplification of microwave radiation by substances not in thermal equilibrium Trans. IRE Prof. Group on Electron Devices pged-3 1

[11] Karlov N V, Krokhin O N and Lukishova S G 2010 History of quantum electronics at the Moscow Lebedev and General Physics Institutes: Nikolaj Basov and Alexander Prokhorov Appl. Opt. 49 F32-F46

[12] Townes C H and Schawlow A L 1958 Microwave Spectroscopy (New York, Dover, reprinted 1975)

[13] Gordon J P, Zeiger H J and Townes C H 1954 Molecular Microwave Oscillator and New Hyperfine Structure in the Microwave Spectrum of NH$_3$, Phys. Rev. 95 282 (1954)

[14] Autler S H and Townes C H 1955 Stark Effect in Rapidly Varying Fields Phys. Rev. 100 703

[15] Townes C H and Schawlow A L 1958 Infrared and Optical Masers Phys. Rev. 112 1940

[16] Townes C H 1994 A Life in Physics: Bell Telephone Laboratories and World War II, Columbia University and the Laser, MIT and Government Service, California and Research in Astrophysics, oral history conducted by Riess S B, Regional Oral History Office, The Bancroft Library, University of California, Berkeley accessed at http://content.cdlib.org/view?docid=kt3199n627&brand=calisphere&doc.view=entire_text

[17] Chiao R Y, Garmire E and Townes C H 1963 Raman and Phonon Masers Proceedings of the International School of Physics Enrico Fermi, Course XVII, published as Quantum Electronics and Coherent Light ed P. Miles (New York: Academic Press). Preprint can be found at https://archive.org/details/kernel_19640006830

[18] Garmire E, Pandarese F and Townes C H 1963 Coherently driven molecular vibrations and light modulation Phys. Rev. Lett. 11 160

[19] Chiao R and Stoicheff B P 1964 Angular dependence of maser-stimulated Raman radiation in calcite Phys. Rev. Lett. 12 290

[20] Garmire E 1964 Bull. Am. Phys. Soc. 9 490
[21] Garmire E 1965 The angular distribution of stimulated Raman emission in liquids Phys. Lett. 1965 17 251
[22] "Stimulated Raman Scattering in Liquids" (E. Garmire) Physics of Quantum Electronics, Kelley, Tannenwald, eds. (McGraw-Hill, New York, 1966) pp. 167-179
[23] Garmire E 1965 Nonlinear Optics in Liquids PhD Thesis, Physics Department, MIT
[24] Chiao R Y, Townes C H and Stoicheff B P 1964 Stimulated Brillouin scattering and generation of intense hypersonic waves Phys. Rev. Lett. 12 592
[25] Garmire E and Townes C H 1964 Stimulated Brillouin scattering in liquids Appl. Phys. Lett. 5, 84
[26] Zeldovich B Ya, Popovichev V I, Ragulskii V V and Faizullov F S 1972 Connection between the wave fronts of the reflected and exciting light in stimulated Mandel’shtam-Brillouin scattering Sov. Phys. JETP Lett. 15 109
[27] Hercher M 1964 J. Opt. Soc. Am. 54 563. Townes also had a copy of a photograph by John Atwood at Perkin Elmer of a self-trapped track, which was later published as Figure 16 in the following: Minck R W, Terhune R W and Wang C C 1966 Nonlinear optics Appl. Opt. 5 1595
[28] Chiao R Y, Garmire E and Townes C H 1964 The self-trapping of optical beams Phys. Rev. Lett. 13
[29] Garmire E, Chiao R Y and Townes C H 1966 Dynamics and characteristics of the self-trapping of intense light beams," Phys. Rev. Lett. 16, 347
[30] Askarjan G A 1962 Effects of the Gradient of a Strong Electromagnetic Beam of Electrons and Atoms Sov. Phys. JETP 15, No. 6
[31] Kelley P L 1965 Self-focusing of Optical Beams Phys. Rev. Lett. 15 1006
[32] Chiao R Y, Johnson A, Krinsky S, Smith H A, Townes C H and Garmire E 1966 A new class of trapped light filaments IEEE J. Quantum Electr. QE-2, 467
[33] Talanov V I 1965 Self-focusing of wave beams in nonlinear media JETP Lett. 2 138. Also V. I. Talanov 1964 Izv. Vysshikh Uchebn. Zavedenii, Radiofiz. 7 564 (1964).
[34] Brewer R G, Lifshitz J R, Garmire E, Chiao R Y and Townes C H 1968 Small-scale trapped filaments in intense laser beams Phys. Rev. 166 326
[35] Boyd RW, Lukishova S G and Shen Y R, eds. (2009) Self-focusing: Past and Present Fundamentals and Prospects Topics in Appl. Phys. 114 (SpringerLink ebooks in Physics and Astronomy at http://link.springer.com/book/10.1007/978-0-387-34727-1)
[36] DeMartini F, Townes C H, Gustafson T K and Kelley P L 1967 Self-steepening of light pulses Phys. Rev. 164 312
[37] Chiao R Y, Kelley P L and Garmire E 1966 Stimulated 4-photon interaction and its influence on stimulated Rayleigh-wing scattering Phys. Rev. Lett. 17 1158
[38] Carman R L, Chiao R Y and Kelley P L 1966 Observation of degenerate stimulated 4-photon interaction and 4-wave parametric amplification Phys. Rev. Lett. 17 1281
[39] Brewer R G and Townes C H 1967 Standing waves in self-trapped light filaments Phys. Rev. Lett. 18 196
[40] Sacchi C H Townes C H and Lifshitz J R 1968 Anti-Stokes generation in trapped filaments of light Phys. Rev. 174 439
[41] Cheung A C, Rank D M, Chiao R Y and Townes C H 1968 Phase modulation of Q-switched laser beams in small-scale filaments Phys. Rev. Lett. 20 786
[42] Gustafson T K, Taran J P, Haus H A, Lifshitz J R and Kelley P L 1969 Self-modulation, self-steepening, and spectral development of light in small-scale trapped filaments Phys. Rev. 177 306
[43] Gustafson T K, Kelley P L, Chiao R Y and Brewer R G 1968 Self-trapping in media with saturation of the nonlinear index Appl. Phys. Lett. 12 165
[44] Carman R L, Mooradian A and Kelley P L 1969 Transient and steady state thermal self-focusing Appl. Phys. Lett. 14 136
[45] Coffinet J P and F. DeMartini 1969). Coherent excitation of polaritons in gallium phosphide” Phys. Rev. Lett. 22 60
[46] http://www.nobelprize.org/nobel_prizes/physics/laureates/1964/basov-lecture.html
[47] http://www.quantum-electron.ru/laser50_eng.phtml