also are able to display transitions not only from disorder to order (synergetic behavior) but also from order to complexity (temporal and spatio-temporal instabilities and chaos).

This revolution entered the field of optics[2] around 1975 with Haken's discovery[3] of the isomorphism between the three Lorenz equations[1,4] and the single mode homogeneously broadened laser equations. Unfortunately, the experimental realization of a chaotic Lorenz-Haken laser has proven a challenging problem, in spite of the fact that, since the beginning of experimental studies in lasers, they have shown irregular dynamical pulsations. Only recently, a coherently pumped far-infrared ammonia laser has allowed the first observation of a behavior remarkably similar with the predictions of the paradigmatic Lorenz-Haken model.[5,6]

However, the theoretical understanding of the dynamics of this ammonia laser on the basis of the Lorenz-Haken theory, has raised considerable controversy [5,7,8] for the inability of this model to describe important physical aspects of the real system. Thus, while two-level (i.e., incoherently pumped) lasers are considered in the Lorenz-Haken model, coherently pumped lasers operate on a three-level scheme. In such conditions, the coherent pump laser field not only affects the populations of various energy levels, but can also induce a strong interaction with the generated field through nonlinear coherent effects such as the Ramon process and the AC Stark effect. This can result in new dynamic features of the coherently pumped lasers.[2b,7,9]

Indeed, the numerical study[10] of a homogeneously-broadened three-level laser model—for a parameter range appropriate for the ammonia laser—revealed striking differences between the theoretical predictions and the experimental observations[5] of Lorenz-like behavior in this laser.

By our incorporating the Doppler-broadening in the three-level laser model, considering operation in the backward emission direction, as in the experiments[5], and analyzing the dynamics of the laser phase in addition to that of intensity, we have been able to reproduce[8,11] qualitatively the main experimental features of the heteroclynic or Lorenz-type behavior observed in the experiments[5], including instability pump threshold, detuning-pump bifurcation diagram, and symmetric spiral attractor. The incorporation of the Doppler broadening complicates considerably the laser model, which in our case was composed by 217 coupled ordinary differential equations. It is remarkable that such a complex model predicts a behavior so similar to that of the simple three (or five for the detuned laser) equations model for the Lorenz-Haken laser.

Coherently pumped lasers have thus become most interesting systems for nonlinear dynamics with the first demonstration of Lorenz-like dynamics in the real world. Moreover, these lasers are very versatile systems that include as limit simple cases the two-level laser (when coherent pumping effects are eliminated) and the Raman laser (when the detunings of the pump and generated fields are equal and much larger than the molecular transitions linewidths). We believe they have also produced evidence of two rather general aspects: (1) that highly-dimensional dynamical systems can approach asymptotically in time-low dimensional attractors, and (2) that the dynamical behavior of lasers usually proves resistant to a qualitative understanding on the basis of simple models that, instead, yield a satisfactory explanation of the stable output emission.

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Second harmonic generation in periodically-poled $LiNbO_3$

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Compact, solid-state sources of coherent blue light have applications in biomedicine, displays, printing, and optical storage. Because blue semiconductor diode lasers are not currently available, techniques for the efficient nonlinear optical frequency conversion of infrared diode lasers are being developed. Materials with high nonlinearity and good optical quality are therefore in demand. Engineering the optical properties of a well-studied material such as lithium niobate (LiNbO$_3$) provides an alternative to the search for entirely new materials. Using quasi-phase-matching in LiNbO$_3$, we have accomplished collinear cw second harmonic generation (SHG) with wavelengths and nonlinear coefficients that have been impossible to phase-match using only the material birefringence.

Quasi-phase-matching (QPM) was the first technique to be suggested for compensating refractive-index dispersion to obtain efficient SHG.[1] By periodically reversing the sign of the nonlinear coefficient at odd integer multiples of the coherence length, continuous power flow from the fundamental into the second harmonic wave can be maintained along the interaction length. While the highest conversion efficiency is obtained when reversals occur every coherence length, fabrication considerations may compel the use of longer distances between reversals. In LiNbO$_3$, the signs of the nonlinear coefficients are tied to the orientation of ferroelectric domains, and thus QPM can be accomplished in a crystal with a periodically-alternating domain structure (periodically-poled LiNbO$_3$). We have developed two methods to produce periodically-poled LiNbO$_3$ for QPM nonlinear optical interactions.

We have produced periodically-poled LiNbO$_3$ crystals up to 0.5 mm in diameter using laser-heated pedestal growth.[2] In this technique, the output of a CO$_2$ laser is focused on the tip of a typically 500-μm diameter source rod of LiNbO$_3$ to make a small molten droplet into which a seed crystal is dipped. The seed is then withdrawn from the droplet as fresh source material is supplied. Periodically-reversed ferroelectric domains can be created during growth either by rotating an asymmetric heat input or by periodically modulating the heating power. Using these techniques, we have grown domain structures with periods as small as 2 μm in either nominally 5% Mg-doped or undoped congruent material and in crystals grown along either the x or *z* axis.

Previous demonstrations of quasi-phase-matched SHG in LiNbO$_3$ have used Czochralski-grown crystals with poling periods down to 6.8 μm for frequency doubling 1.06 μm light.[3] The shorter periods attainable using pedestal growth have allowed us to apply this material to room temperature, quasi-phase-matched SHG of light at wavelengths as short as 407 nm, using the d_{33} and d_{22} nonlinear coefficients. The theoretical conversion efficiency using d_{33} and QPM with domain lengths equal to the coherence length is ≈13.5 times that which would be obtained were it possible to birefringently phase-match an interaction at this wavelength with the commonly used d_{31} coefficient. The conversion efficiencies and wavelength and temperature tuning bandwidths we have measured are consistent with an effective interaction length of ≈320 μm (>230 domains). No photorefractive damage was observed at the relatively low levels of blue light generated in the SHG experiments, and a separate test with cw focused 488 nm beams showed that the periodically-poled material exhibits no discernible photorefractivity at intensities up to 200 kW/cm.[2]

Waveguides increase the efficiency of nonlinear optical interactions by maintaining high optical intensities over considerable lengths. We have developed a method to produce periodically-poled LiNbO$_3$ waveguides in standard integrated-optic substrates, using commonly used materials and process. It is known that the diffusion of titanium into the +z face of a LiNbO$_3$ wafer causes ferroelectric domain reversal at the surface of the wafer. By indiffusing lithographically-defined gratings of titanium, we exploit this effect to produce periodic arrays of alternating domains. Waveguides are then made in the periodically-poled surface using the annealed-proton-exchange technique.

In waveguide doublers poled with periods on the order of 7 μm, about 1 μW of blue light at 410 nm has been generated using the d_{33} coefficient in a 1 mm-long grating with a normalized conversion efficiency of about 40%/W-cm^2.[4] The observed wavelength tuning bandwidth is consistent with an effective length approximately equal to the grating length. The measured conversion efficiency implies that in cm-long devices, mW levels of blue light could be produced from tens of mW of infrared fundamental power. Work is underway to demonstrate this scaling with length and also to further improve the efficiency by making the domains one coherence length long instead of three. In addition to the work on blue light generation, SHG of green light has been observed in devices fabricated using Ti gratings with larger periods.[5] Other workers have demonstrated SHG in periodically-poled LiNbO$_3$ waveguides produced with a different process.[6]

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