Nonlinear Optics in Single Crystal Fibers

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Since the advent of low loss optical fibers fifteen years ago, considerable research effort has been directed towards the study of nonlinear interactions in fibers. A variety of devices have taken advantage of the combination of transverse confinement and long interaction lengths available in glass fibers to operate efficiently at relatively low pump powers. Because glasses are inherently centro-symmetric, only third-order nonlinear processes, e.g. Raman[1] and Brillouin[2] scattering, optical Kerr effect,[3] self-phase modulation,[4] or extremely weak quadrupole second order processes[5] are allowed. Thus, the combination of fiber geometry and the second order susceptibility of non-centro-symmetric single crystals would open the door to a broad range of nonlinear applications not possible in glass fibers.

The potential of crystal fibers for nonlinear interactions is clear from the theoretical efficiencies of several simple devices.[6] A 25 μm diameter 5 cm long $LiNbO_3$ fiber propagating an HE_{11} mode can double 1.06 μm radiation from a Nd:YAG laser with an efficiency of 0.1% per 1 mW or fifty times the bulk efficiency. Similarly, a parametric oscillator pumped with 532 nm radiation in the same fiber would have a threshold on the order of ten mW. The advantage of the guided wave structure is even more pronounced for interactions involving widely disparate frequencies, e.g. differencing two visible lasers to produce infrared radiation, where the advantage relative to the bulk is several hundred to one.

Fibers useful for device applications must meet fairly stringent quality criteria. In order to maintain phasematching in a parametric process and to minimize scatter losses, the fiber must be a properly oriented single crystal of good optical quality and uniform composition. Ferro-electric fibers must, in addition, be poled, i.e. single domain. Diameter variations can cause phase mis-match, radiation losses and modal coupling. These effects are complicated functions of the core-cladding index difference, the radius of the fiber, and the azimuthal and axial period of the variations. We estimate that diameter variations must be held to less than 0.1 - 1% for typical nonlinear devices.

There are a number of research efforts underway to produce nonlinear crystal fibers. Organic crystals grown inside glass capillaries are being investigated by several groups.[7,8] These materials exhibit large nonlinear coefficients (in some cases more than an order of magnitude larger than $LiNbO_3$), and high damage thresholds (comparable to KDP). Crystal properties can be tailored to specific applications by organic synthesis techniques. SHG of a pulsed 1.06 μm laser in a benzil cored fiber was reported several years ago by Nayar.[9] The

interaction was quite inefficient because the second harmonic radiation was produced in radiation modes of the fiber.

Another approach to nonlinear interactions in fibers is to embed an unclad glass fiber in a nonlinear crystal. DeShazer has reported promising results in $LiIO_3$ with this technique.[10] His group has also grown KDP crystal fibers by an unspecified method.[11]

The growth technique that we have chosen to pursue is miniature pedestal growth.[12] In this method, the tip of a small rod of the material to be grown is melted with a CO_2 laser, as shown in Fig. 1. A seed crystal is dipped into the molten zone, then pulled from the zone more rapidly than the source rod is fed in. Mass conservation fixes the diameter reduction as the square root of the velocity ratio.

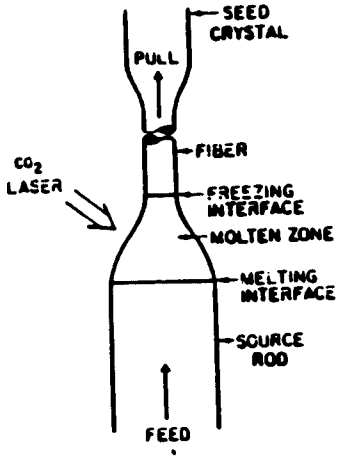

Fig. 1
The laser heated miniature pedestal growth process.

The abrupt liquid solid transition characteristic of the growth of crystalline materials is quite different from the viscous draw-down seen in glass fiber pulling, causing the pedestal growth processes to be far more sensitive than glass fiber pulling to external perturbations. The growth of fibers suitable for nonlinear applications therefore, requires a carefully designed apparatus. In particular, the growth zone must be mechanically and thermally stable and the heat distribution should be azimuthally symmetrical. The apparatus that we designed to meet these criteria is shown in Fig. 2.

The novel reflaxicon focussing system simultaneously provides an azimuthally symmetric heat input and a tight 40 μm focus necessary for the stable growth of small fibers. A moving belt in the translation mechanism slides the fibers through silicon V-groove guides at rates accurately controlled by regulated d.c. motors. The V-groove guides prevent motion of the fiber in the plane perpendicular to the growth axis. A high speed non-contact diameter measurement system described in detail elsewhere has recently been completed. This device will allow study of the effect of variations of feed and pull rates and laser power on the fiber diameter, with the goal of implementing closed loop control of the fiber cross-section. A block diagram of

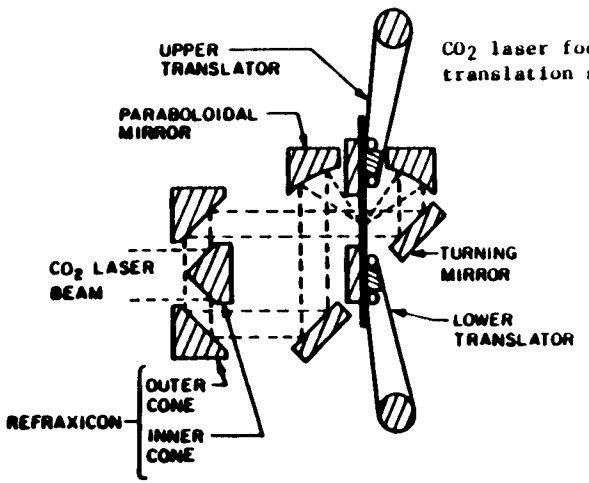

Fig. 2
CO_2 laser focusing optics and fiber translation system.

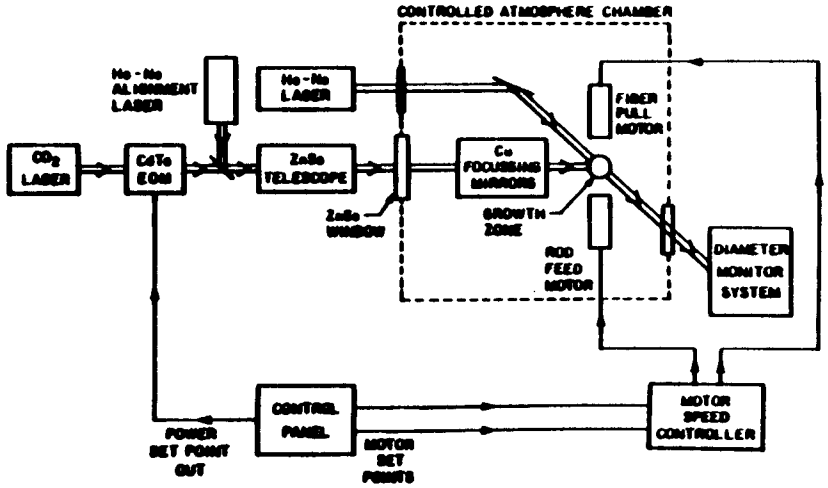

Fig. 3—Block diagram of the single crystal fiber growth system

the complete system is shown in Fig. 3. Reference 12 gives a more complete description of the apparatus.

To date we have achieved controlled growth of four materials: Al_2O_3, $Cr^{+++}:Al_2O_3$, Nd^{++}:YAG and $LiNbO_3$. Several orientations of most of these materials have been grown, including both a and c axis $LiNbO_3$. Fibers with diameters ranging from 20 up to 500 μm have been grown at rates of 0.5 to 40 mm/min in lengths up to 200 mm. The necessary CO_2 laser power is typically less than 5 W.

The morphology of the fibers is similar to that of bulk Czochralski boules of the same orientation. For example [111]

Nd:YAG fibers show a rounded hexagonal shape, while <001> $LiNbO_3$ fibers are round with three growth ridges. SEM photographs of the fibers show no micron scale roughness, but diameter variations on the order of 1% rms are observed over millimeter lengths. We expect to reduce these variations by an order of magnitude with the closed loop diameter control system.

These growth results illustrate several of the attractive features of the miniature pedestal growth technique. It is entirely containerless, thereby avoiding crucible compatibility and contamination problems. Feasible pull rates are orders of magnitude higher than in bulk Czochralski growth and only small volumes of starting material are required. High temperatures are easily attained, with the available laser power as the only limit. 0.5 mm diameter sapphire (M.P.2323K) can be grown with only 5 watts of laser power. Thus, the technique is attractive for material survey applications, e.g. new laser host-ion combinations.

The measured propagation losses in the fibers are in accord with theoretical estimates based on the measured amplitude of the diameter variations. For example, a 5 cm long 170 μm diameter ruby fiber had losses of 0.04 dB/cm for 633 nm radiation launched into low order modes. Similar results were obtained in Nd:YAG and $LiNbO_3$ fibers.

The first optical device that we have constructed using single crystal fibers is an argon laser pumped monolithic cw ruby fiber oscillator.[14] This device demonstrates the feasibility of monolithic guided wave devices in crystal fibers.

We are currently studying two problems which must be understood before nonlinear devices can be fabricated in $LiNbO_3$ fibers: control of the distribution of ferro-electric domains, and cladding the fiber for control of modal characteristics. Selective etching and pyroelectric response studies indicate the c-axis fibers grow single domain, while a-axis fibers develop head-to-head domains joined at the axis of the fiber. Both these results can be explained by the thermo-electric fields which are present in the growth zone. Generated by the steep temperature gradients present in the pedestal growth process, these electric fields dominate the dipole-dipole interactions that cause bulk samples to break up into polydomain configurations. Efforts are underway to use controlled temperature gradients to uniformly pole a-axis fibers. It may also be feasible to use periodically varying temperature gradients to form a periodically poled fiber for quasi-phasematching[15] nonlinear interactions.

Techniques for forming low index claddings are also being studied. Such claddings would reduce surface scatter losses and bring the waveguides closer to single mode operation. Both extruded glass and diffused proton or transition metal claddings have been fabricated and are being tested.

Conclusions

We have designed and built an apparatus to grow single crystal fibers suitable for linear and nonlinear optical applications. Ruby, lithium niobate and Nd:YAG fibers with losses in the 1%/cm range have been grown. An argon pumped monolithic ruby fiber oscillator has been demonstrated.

Future work will proceed in several directions. Studies of cladding and poling a-axis $LiNbO_3$ will continue. The short term goal is demonstration of efficient doubling of 1.06 μm radiation. The first nonlinear device we expect to demonstrate is an electro-optic modulator in c-axis $LiNbO_3$ as the poling problem is already solved for this orientation.

Another thrust of the program will be extending the range of materials grown in fiber form. Two materials to be emphasized are terbium gallium garnet for optical isolators and potassium niobate for doubling gallium arsenide diode lasers.

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