

transformations induced by H and temperature (T).  $Gd_5Ge_4$  orders antiferromagnetically at  $T_N \gg 130$  K, and in H lower than 10 kOe, the antiferromagnetic (AFM) order is sustained at least down to 2K. Under applied H exceeding 10 kOe (the precise H value is T-dependent),  $Gd_5Ge_4$  shows interesting AFM to ferromagnetic (FM) transition that could be driven both by T and H.

Isothermal magnetization and magnetic relaxation measurements in polycrystalline  $Gd_5Ge_4$  were carried. The results show that upon field variation from the initial ZFC state the magnetic-field-induced first order AFM to FM transition observed in  $Gd_5Ge_4$  is accompanied by distinct metastability. The reverse transition from the FM state to AFM state on reducing H is also marked with same kind of metastability in the temperature region above 21 K. Below 21 K, this FM to AFM transition process is hindered. At 5 K, the sample remains in the FM state at all fields including zero field after it has been magnetized at  $H > 25$  kOe. This FM state is sustained on subsequent field cycling between  $\pm 50$  kOe. At 15K, the FM to AFM transition is initiated in the descending-H cycle but remains incomplete even when the magnetic field is reduced to zero. This leads to the interesting situation of phase-coexistence between the converted stable AFM state and the unconverted FM state, which is also stable within the

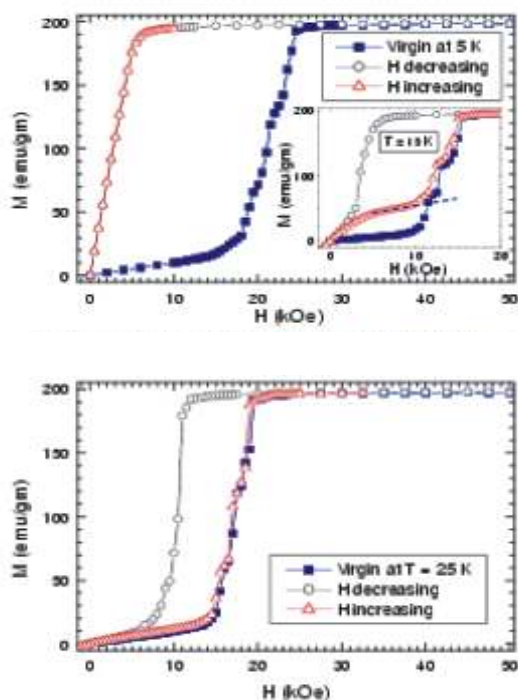
experimental time scale. This phase-coexistence is different from the phase-coexistence observed across the AFM to FM transition in the virgin sample both below and above 21 K, and across the FM to AFM transition in the isothermal descending-H cycle above 21 K. Here, one of the phases- the AFM phase during the virgin cycle and the FM phase in the H descending cycle- is metastable and relaxes towards the stable phase because of fluctuation energies. These results have been published recently [M. K. Chattopadhyay et al. Phys. Rev. B 70(2004)214421].

Detailed magnetotransport and magnetization studies on  $Gd_5Ge_4$  are in progress. These studies have already lead to very new findings, viz., presence of spin fluctuations within the stable AFM state, important aspects regarding the dynamics of phase transition in phase separated systems in the presence of magneto-structural coupling, etc. We believe that all these efforts would lead to significant inputs towards finding newer technology for tuning materials suitable for the machines of the future years.

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## L.16 Development of lithium niobate crystal elements

Non-linear and photorefractive crystals are suitable candidates for many optical applications, e.g. laser output modifications, optical signal amplification, phase conjugation, image processing, reversible holographic storage, all solid state UV laser, and compact blue green laser systems. For many years the ferroelectric lithium niobate has been of great interest for both fundamental science and application because of its unique combination of electro-optic, piezoelectric, non-linear optical properties with nonhygroscopic, mechanical and chemical stability. The typical applications of lithium niobate are non-linear frequency conversions, parametric light generation and amplifications, electro-optical modulators, wave guide structures, acoustic wave delay lines acoustic filters, non-volatile holographic data storage, domain engineering, QPM based compact green laser systems and display devices. Applications that utilize the large electro-optic coefficients of lithium niobate are optical modulation and Q-switching of infrared wavelengths. Applications that use the large nonlinear d coefficient of  $LiNbO_3$  (LN) include optical parametric oscillation, difference frequency mixing to generate tunable infrared wavelengths and second harmonic generation. The periodically poled lithium niobate crystals are particularly attractive for second harmonic generation of low power laser diodes in the 1.3 - 1.55 $\mu$ m range



*Fig L.15.1 Isothermal H dependence of magnetization of polycrystalline  $Gd_5Ge_4$ , highlighting fully arrested, partially arrested, and de-arrested FM phases at different T values*



The non-stoichiometry of LN has been the main limitation for its high power applications, because of its Li-deficiency, leading to many intrinsic defects (vacancies, antisite defects etc.) and high disorder. The defect site can cause absorption and incorporation of impurities. The presence of impurities and defects degrade LN's damage resistant to optical radiation. In spite of defective nature, congruent crystals possess high optical homogeneity. The presence of these defects makes material less susceptible to various types of impurity incorporation added intentionally for desired application. The necessary charge compensation for the extrinsic defect caused by impurity atoms could be easily balanced by intrinsic defects. Therefore several types of dopant have been studied in the past (for various applications) like Mg, Zn, In and Sc to make material damage resistant to optical radiations, transition metal ions Fe, Mn, Rh, Ce and Cu for high density holographic data storage and rare earth ions Nd, Cr, Ho, Gd and Er for laser host materials.

In view of the above facts, various undoped and doped LN crystals have been grown from congruent melt composition (48.45 mol%  $\text{Li}_2\text{O}$  and 51.55 mol%  $\text{Nb}_2\text{O}_5$ ) using Czochralski technique. The charge was synthesized from high purity 99.99%  $\text{Li}_2\text{CO}_3$ ,  $\text{Nb}_2\text{O}_5$  powders. The mixture was ball-milled for several hours to ensure proper mixing. The well-mixed charge was fired first at 750 °C for 15h again grinded and fired at 1000 °C for 24h. This process was repeated to ensure the completion of solid-state reaction. The reacted charge was ground, remixed and palletized. These pallets were placed in a 50mm diameter and 50mm height platinum crucible for crystal growth runs in air atmosphere. The induction heating system of 30kW and 30kHz were used as a heater. The optimized pulling and rotation rates were in the range of 3 to 5mm/h and 25 to 15 rpm respectively. The pulling

and rotation rate were reduced gradually for better growth conditions. The growth chamber was prepared from zirconia refractory followed by zirconia felt inside quartz tube.

Platinum after heater position was adjusted in such a way to provide optimum thermal gradient for nearly flat growth interface and post growth cooling optimised for the crack free crystal. The post growth cooling was maintained at a rate of 25-30 °C/h initially up to 1000 °C, and thereafter 100 °C/h to room temperature. Typical sizes of the crystals grown were 20-25 mm in diameter and 30-40 mm in length. Fig L.16.1 shows crystal boules of undoped Cr and Mg doped lithium niobate crystals. Fig L.16.2 shows the wafers and fabricated elements from as grown undoped and doped (Mg, Cr and Cu) LN samples.

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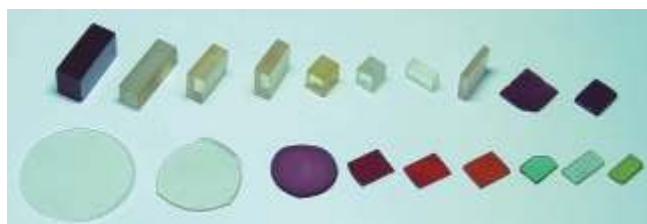
## L.17 Data acquisition software for frequency resolved optical gating

Ultrashort pulses (pulse width <10 ps) are in widespread use in physics, photochemistry and photobiology. One issue in working with ultrashort pulses is the need to characterize the pulse to get proper interpretation of experimental data. For example, in passing through a length of dispersive medium (e.g. glass) a transform-limited femtosecond pulse gets broadened in time and becomes chirped. A full characterisation of the pulse requires measurement of its time-dependent phase as well as amplitude. Frequency Resolved Optical Gating (FROG) is one of the most popular techniques used for measuring the time-dependent phase and amplitude of an ultrashort pulse. It involves the generation of a spectrogram or the FROG trace of the laser pulse under study in the form of a single two-dimensional picture with the time delay along one axis and the spectral frequency along the other axis. FROG can utilise any one of several nonlinear optical processes e.g. second harmonic generation, polarization gating etc. We have developed a complete data acquisition system for recording and displaying FROG traces for various experimental geometries

It can be used for both single-shot as well as multishot configurations. As the programme is based on the earlier developed PROMISE software, it has the entire image processing capabilities of PROMISE. The programme controls two optical delay stages and acquires the data from the CCD detector at the spectrograph output. The FROG measurement is done in the following manner. First the input beams are set at zero delay to set the spectrograph and the



**Fig. L.16.1** As grown undoped, Mg and Cr doped LN crystal boules



**Fig. L.16.2** Fabricated elements of undoped and doped (Mg, Cr and Cu) LN crystals