Magnetization of GaMnN nanopowders obtained by an anaerobic synthesis and high-pressure high-temperature sintering

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Gallium nitride GaN suitably doped with a magnetic transition metal (TM = Mn, Fe, Cr, etc.) has been anticipated for many promising applications as a dilute magnetic semiconductor (DMS). However, despite numerous efforts DMS based on GaN tenaciously show too low T_C's to be considered for spintronics applications and making GaMnN with a high Mn content and specific physical properties poses still a great challenge for researchers. Looking back at all the numerous yet unfulfilled attempts to incorporate sufficient manganese into the GaN lattice in the variety of materials forms (thin films, bulk samples or microcrystalline powders) and with various techniques that have been tried, one may consider the application of high-pressure high-temperature sintering of GaN/Mn nanopowders as a promising means to achieve the goal. If sintering is performed under conditions rendering GaN/Mn powders to recrystallize, the applied pressure-temperature conditions could favor, on the one hand, Mn-incorporation and, on the other hand, advantageous phase separation of residual contaminants, if any. In this regard, sintered GaN-based DMS ceramics prepared from nanopowders can, potentially, be a new convenient form of such materials for many challenging applications.

An oxygen-free bimetallic molecular system made of gallium (III) tris(dimethyl)amide $Ga(NMe_2)_3$ and manganese (II) bis(trimethylsilyl)amide $Mn[N(SiMe_3)_2]_2$ (Me = CH_3) was subjected to ammonolysis in liquid ammonia affording the mixed metal precursors for further processing. The selected 10 at.% Mn-precursor was pyrolyzed at elevated temperatures under an ammonia flow by an initial pyrolysis at 150 ºC followed by a nitriding pyrolysis at 500, 700 or 900 ºC. All final nanopowders were characterized by XRD diffraction, FT-IR spectroscopy, and SEM/EDX microscopy and analysis. Powders were subjected to high-pressure (7 GPa) high-temperature (1000ºC) sintering without additives.

Magnetization of the samples was measured as a function of magnetic field (up to 7 T) and temperature (2-400 K) using a SQUID magnetometer. The GaMnN nanopowders showed an overall paramagnetic (PM) behavior. However, some antiferromagnetic (AFM) contributions from residual by product were observed. The highest observed Mn concentration in the GaMnN nanopowders amounts to about 3.5 at.%. Very promising shown to be ceramic pellets obtain from pyrolysis at 700ºC. The complex mixture of gallium nitride polytypes with multimodal size distributions (2nm+8nm h-GaN, 43nm c-GaN) was converted upon sintering to the single h-GaN phase with crystallite grains size 75nm. Interestingly, also magnetic response has changed upon sintering *i.e.*, in GaN crystallites 5.5% at. Mn concentration was observed while AFM contributions from residual by product diminished.

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