Gadolinium nitride GdN: revival of an old compound by x-ray absorption spectroscopy

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Since the first experimental studies of GdN in the early sixties its electronic and magnetic properties are discussed controversially. Rediscovered by recent theoretical work the material has attracted new interest. General agreement exists by now that in its ground state GdN is a ferromagnet ($T_c \sim 60K$), and a narrow-gap semiconductor in its paramagnetic phase. But it is not clear if below T_c it is a semiconductor, or a semimetal, or even a halfmetal with complete spin polarization. All three cases were predicted by theory. Here, a study is reported of GdN thin films by core-level x-ray absorption spectroscopy (XAS) and x-ray magnetic circular dichroism (XMCD). Two types of samples were investigated: (i) Films with the lattice parameter, the Curie temperature T_c and saturation magnetization of the bulk material. Their electrical conductivity is thermally activated down to the onset of magnetic ordering where is evidence of a transition to metallic behavior. The ordered 4f moment extracted from the XMCD spectra at the Gd $M_{4,5}$-edges is consistent with the $^8S_{7/2}$ configuration of Gd^{3+}; it varies with temperature as the macroscopic magnetization. The experimental K-edge XAS spectra of N in this compound indicate the presence of N p character of the low-lying unoccupied conduction-band states, pointing to hybridization of the N 2p and Gd (5d,6s) states. Compared with the theoretical partial density of vacant N p states these spectra show considerable disparities that are not well understood. The exchange field generated by the Gd f electrons in the ferromagnetic phase of GdN induces a magnetic polarization of the N p band states, as can be concluded from the observation of strong magnetic circular dichroism at the N K edge of nitrogen. It indicates the presence of an important spin-orbit interaction in the final N p states. (ii) Films with a unit-cell volume 8.6% above that of bulk-like layers. Their Curie temperature is reduced by 50%. An intricate observation is that the ratio of the Gd L_2 and L_3 edge amplitudes is up to three times higher than the value expected from the degeneracy of the $2p_{3/2}$ and $2p_{1/2}$ core states, which is observed for the bulk-like layers. We suggest that the effect may be related to the different weight that the crystal-field-split Gd-5d final states ($t_{2g}$and e_g) have in the absorption process at these edges, and to the modification of the special electronic band structure of GdN upon lattice expansion. This hypothesis is supported by the observation that the L_2 absorption edge is shifted to lower energies upon ferromagnetic ordering while the L_3-edge position remains inert.