The description of chemical bonding in real space was recently extended from compounds with s- and p-elements to those with d-elements [1]. This allows a better bonding analysis in binary and ternary intermetallic phases containing transition metal (TM). In case of rare-earth metals (RE) as cationic component in such compounds, the electronic configuration of RE plays a crucial role for the formation of distinct structural patterns. In particular in YbGa$_3$, the pressure induced electronic changes of ytterbium are accompanied by a structural transformation [2]. Valence instability of ytterbium (heavy fermion behavior) was discussed in detail in the literature [3], however, the bonding aspects are still analyzed insufficiently. As a first part of a research project we investigated the structural and electronic properties of some ternary ytterbium compounds with relatively simple structures containing gallium and a transition metal. The orthorhombic phases YbNiGa$_4$ [4], β-YbAgGa$_2$ [5] and YbAuGa [6] show intermediate valent behaviour at ambient pressure, which motivated the study of the Yb valence states using in-situ X-ray absorption spectroscopy (XAS) near the Yb$L_{III}$ edge ($E \approx 8945$ eV) up to pressures $p \approx 25$ GPa.

Samples were prepared by high-frequency melting of pure elements under argon atmosphere. Chemical composition was proven by ICP-AOS or combustion method. The heat treatment was performed under inert conditions. The homogeneity of the annealed samples was checked by X-ray powder diffraction experiments. For the high pressure X-ray absorption measurements we employ diamond anvil cells with 125 µm pinhole steal gaskets where it is possible to measure in transmission mode through the diamonds. Paraffin is used as pressure medium for hydrostatic conditions of the fine powdered samples. The pressure is determined by the measurement of the luminescence line of the simultaneously enclosed ruby. XAS measurements were performed on the HASYLAB beamlines A1 (ambient pressure) and E4 which allows the use of an alignment equipment for the diamond anvil cells.

All mentioned phases show double peak spectra near the Yb$L_{III}$ edge indicating contributions of both Yb states $4f^{14}$ (Yb$^{2+}$) and $4f^{13}$ (Yb$^{3+}$) at ambient pressure. The low energy peak at $E \approx 8935$ eV decreases in intensity at higher pressures as a consequence of the decreasing contribution of Yb with $4f^{14}$ configuration. Nearly exclusive Yb in $4f^{13}$ state exists at pressures of $p = 25$ GPa. The quantitative determination of both Yb states was performed by de-convolution of the XAS assuming gaussian lines as main contribution to the spectra. The pressure dependence of the resulting effective valence $v_{eff}$ shows the transition $4f^{14} \rightarrow 4f^{13}$ with increasing pressure. The similar behavior of the investigated phases indicates, that structural and chemical differences have minor influence on the order of magnitude of the transition pressure. An almost monotonic transition for YbNiGa$_4$ and YbAuGa is observed. Hints on a steplike transition results for the YbAgGa$_2$ phase.
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References