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	High-pressure XMCD investigation of the 5f states in UGa ₂	iment number:
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Report:

Main results:

- 1. The experiment has shown that uranium line M_4 is weakly sensitive to pressure in the range of 3.4 GPa to 7.2 GPa. No changes in the UM₅ line shape have been observed.
- Mechanical polishing is a suitable preparation technique for producing of UGa₂ thin plates for the high-pressure XMCD. Both the UM_{4,5} and Ga K edge spectra can be collected on such samples
- 3. The observed weak UM_4 an UM_5 lines were due to the unintentional 90-degree rotation of the crystal

The X-ray absorption on the UM₄ and UM₅ edges in UGa₂ has been measured at room temperature in the pressure range of 3.4 GPa to 7.2 GPa. The data were collected by the energyresolved and total fluorescence yield detectors. Later the samples from the same batch were measured outside the cell at low temperatures in the ferromagnetic regime. The samples were single crystal plates of approximately 10 mkm thickness with the *c*-axis perpendicular to the face of the plates (along the beam direction). The expected orientation should had been with the *a*axis along the beam but in the process of the preparation the sample unintentionally had been rotated by 90 degrees and this has resulted in substantial weakening of the signal.

High-pressure XMCD of UGa2

The high-pressure study was performed using the He-filled membrane diamond anvil cell. To minimize the attenuation of the

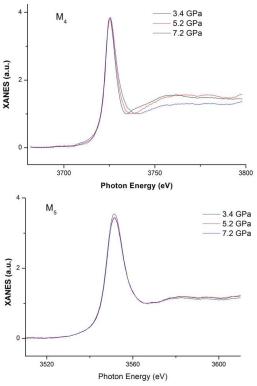


Fig. 1. Variation of the UM₄ and UM₅ line shapes in UGa₂ with pressure. Data collected by the energy-resolved detector.

U M-lines one anvil was made from the perforated diamond used in the combination with the 80 mkm diamond window. The expected maximal pressure for the cell is about 16 GPa but the perforated anvil has

cracked around 8.4 GPa. Based on the character of the cracking planes in the anvil and the absence of the perforation of the diamond window we presume that the reason for the failure was the misaligned diamond anvil. This is corroborated by the fact that the failure has occurred on the linear part of the pressure-load curve of the pressure cell before the curve has reached the saturation.

Before the cell failure the data were collected at three pressures: 3.4 GPa, 5.2 GPa, and 7.2 GPa. In this pressure range the UM₄ line gradually broadens with pressure (see Fig. 1) UM₅ remains unchanged within the error of the experiment. The character of the UM₄ modification is ambiguous because it might result from the shift of the XANES oscillations: when the oscillations move under a transition peak it looks like the peak broadening. The pressure effect obtained by the energy-resolved photodetector could not be corroborated by the total fluorescence measurement due to pronounced non-linear background of the latter. Despite the uncertainty, the UM₄ broadening is likely intrinsic and pressure-related because (i) UM₄ probes the *f*-states closer to the Fermi level than UM₅ leading to UM₄'s higher sensitivity; (2) our LDA+U calculations suggest the decrease of the n_f (*f*-occupation) of about 0.1 per 10 GPa or 0.04 in the experimental pressure range; (3) the previous high-pressure resistivity measurements suggest that the uranium moments μ_U and the electron scattering show weak continuous variation up to 8 GPa.

XMCD of UGa₂ at ambient pressure

After the breakdown of the pressure cell and due to unavailbility of the tecnicians capable of reloading it, the measurements were continued at the ambient pressure on the samples from the same batch. This study was focused on the effect of the sample preapration on the XAS and the XMCD response because at that moment we tended to attribute the weak UM_{4.5} response to the surface modification during polishing. Yet it was found that different methods (manual or machine) of mechanical polishing do not affect the shape of the UM₄ line, although the absolute value of the XMCD repsonse was about 10 times lower compared to that from bulk studied before. Also we have collected thet data on the Ga K line (Fig. 2), which shows the hysterezis (Fig.3) suggesting the polarization induced by the magnetically order *f*-electrons. The temperature dependence of the UM₄ hysterezis width points to the Curie temperature comparabele to that of the bulk UGa₂.

In order to clarify the origin of the very weak U $M_{4.5}$ response the SEM EDX, magnetization, and the X-ray micro diffraction on the 4-circle diffractometer have been perfromed on the same samples. These experiments have shown no surface oxidation or precipitation of the sample components. The magnetization in the orderer state appeared to be 5 times lower than that of the UGa₂ bulk with H/a. Finally, the Laue diffraction using the 4-circle diffractometer has clearly shown that the surface of the thin plates was perpendicular to the hard *c*-axis rather than the easy *a*-axis, which perfectly explains very weak XMCD response.

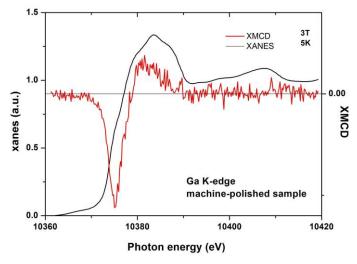


Fig. 3. Ga K edge in UGa₂ and the XMCD at T = 5 K and $\mu_0 H = 3$ T

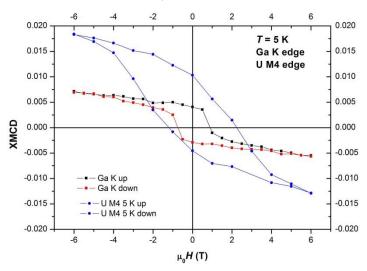


Fig. 4. Hystererzis curves of the XMCD signals from Ga K and U M₄ edges at T = 5 K

To sum up, the present study has shown the weak broadening of the U M_4 line with pressure in UGa₂. This correlates with the high-pressure resistivity data suggesting that notable changes in the f-electron character should start above 8 GPa. Also mechanical polishing is suitable for preparing the samples for high-pressure XMCD experiments.