

## XMCD Measurements of Fe<sub>4</sub>N under High-pressure

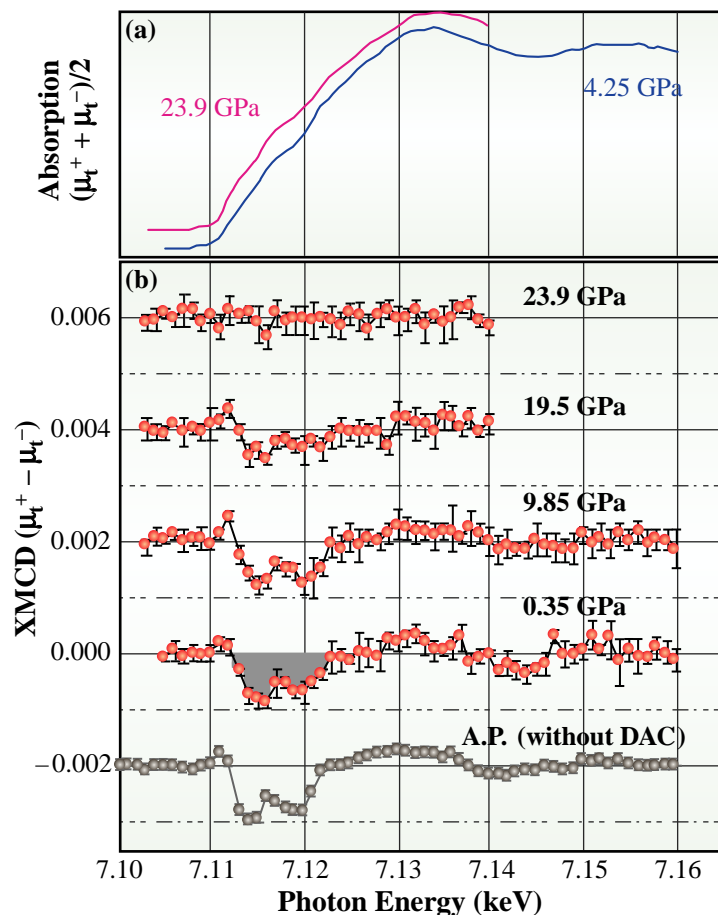
In Fe<sub>4</sub>N perovskite, hybridization between the electron wave-functions and the ligand field presents an important viewpoint for understanding magnetic properties. The insertion of a nitrogen atom into the center of fcc cell of Fe expands the cell volume, and the expanded Fe<sub>4</sub>N changes to show a ferromagnetic state with a Curie temperature of 761 K from antiferromagnetic  $\gamma$ -Fe. The small overlap of Fe 3d electrons originating from the lattice expansion is what is probably responsible for the transition. If so, then how does the magnetic property of Fe<sub>4</sub>N behave under compression?

In order to understand that, use can be made of high-pressure which is an effective external field to modify the lattice constant. The pressure variation of the hybridization and the charge transfer between the iron and the nitrogen provides information which is useful to understand the influences of these substances on the magnetic property of Fe<sub>4</sub>N.

Spectroscopic analysis such as Mössbauer spectroscopy [1] and X-ray emission spectroscopy [2] have been applied to study magnetism under high-pressure. In recent years, X-ray magnetic circular dichroism (XMCD) has also been applied to probe magnetically polarized electron states. However, XMCD under high-pressure using the diamond-anvil cell has been limited to a range of photon energy above 10 keV, where the X-ray absorption by the diamond crystals is almost negligible. There have been only a few reports on Pt L<sub>3</sub>-edge shows (11.564 keV) in Fe<sub>72</sub>Pt<sub>28</sub>

by Odin *et al.* [3] so far. To overcome this limitation, a pair of thinner diamond anvil and intense X-rays with a high degree of circular polarization were utilized; consequently XMCD was successfully measured under high-pressure up to 26 GPa at Fe K-edge (7.111 keV) in Fe<sub>4</sub>N [4].

The helicity-reversal method using a quarter-wave plate diamond (111) slab was applied to record the high-pressure XMCD spectrum at beamline **BL39XU**. The circularly polarized X-ray passed through a pair of thinner diamond anvils of 2.0 mm in total thickness. A sufficient intensity of the transmitted beam enabled us to measure the XMCD spectrum accurately by data accumulation for over a period of four to five hours. **Figure 1** shows the pressure variation of the XMCD spectra

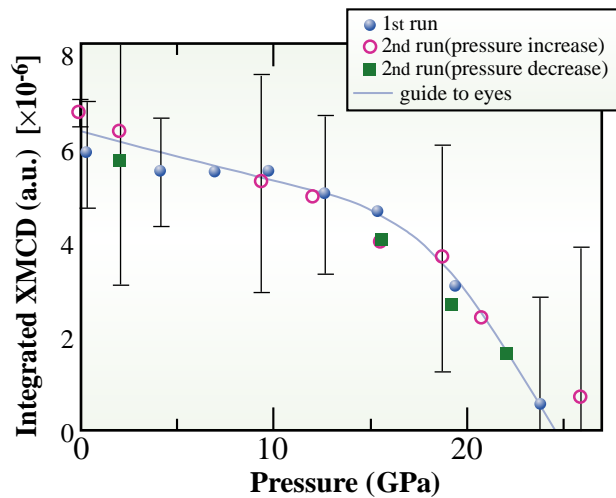


**Fig. 1.** (a) X-ray absorption spectra of Fe<sub>4</sub>N. (b) Pressure variation of XMCD spectra in Fe<sub>4</sub>N. Each spectrum is shifted 0.002 steps in the y-direction. The error bars correspond to the standard deviation of runs of XMCD measurement.

under a magnetic field of 0.6 T at room temperature. The XMCD spectrum is characterized by a positive peak at 7.111 keV, which first increases in intensity as the pressure increases and then reduces under further high-pressures. On the other hand, two negative peaks show a monotonous decrease in intensity. The unit-cell of Fe<sub>4</sub>N consists of two Fe sites; one is Fe(I) at the corner position and the other is Fe(II) at the face-centered position. Mössbauer spectroscopy shows that these two Fe sites give rise to different pressure dependences of hyper-fine field and isomer shift [1]. Therefore, the pressure variation in the XMCD profile may be ascribed to the different responses of these Fe sites.

The pressure dependence of XMCD is demonstrated by the plot of integrated XMCD shown in Fig. 2. The integrated XMCD corresponds to the area of the two negative peaks represented by the region colored in gray in Fig. 1. The reproducibility of the data was confirmed with two serial measurements. At first, as the pressure increases, the intensity gradually reduces, then decreases above 15 GPa to eventually vanish at around 24 GPa. This behavior is evidence for the pressure-induced transition from a ferromagnetic state to a paramagnetic one, a second-order transition without pressure hysteresis. According to an X-ray diffraction experiment which was carried out separately, this transition is not accompanied by structural transformation.

XMCD at the *K*-edge in pure 3*d* transition metals is strongly affected by small 3*d* orbital moments on the neighboring site [5]. Since spin-orbital interaction induces a 3*d* orbital moment, the XMCD spectrum reflects bulk magnetization in addition to its electronic state. It should be noted that high-pressure XMCD measurement can develop the basic studies of magnetism in 3*d* transition metals and/or 4*f* rare-earth metals.



*Fig. 2. Pressure dependences of integrated XMCD. The solid circles, open circles and solid squares correspond to the first run in pressure increase, the second run in pressure increase and the second run in pressure decrease, respectively. The solid line is guide for the eyes.*

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## References

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