Comment on "Spin and Orbital Magnetic Moments of Fe_3O_4"

In a recent Letter [1] spin and orbital Fe $3d$ magnetic moments of magnetite have been evaluated experimentally by x-ray magnetic circular dichroism (XMCD) and calculated within the LDA + U scheme. Large unquenched orbital moments of $0.33\mu_B$ have been found at the B-site ion, which have been attributed to a strong on-site Coulomb interaction and corresponding $3d$ correlation effects. We have reanalyzed the XMCD spectra of Ref. [1] and could identify three major problems concerning the data evaluation procedure: first, a possible wrong nonlinear XMCD offset correction, second, an overestimated self-absorption correction, and third, a reduced sum rule integration range. These effects result in substantially overestimated orbital moments. The shown single crystal XMCD spectra from Ref. [1] and Ref. [2] in Fig. 1(a) exhibit remarkable differences. We added 5.6% of the nonmagnetic absorption spectrum from Ref. [2] to the reproduced XMCD spectrum from Ref. [1]. This modified XAS spectrum is in perfect agreement to the XMCD spectrum from Ref. [2], indicating that non-XMCD related offset signals in the difference spectra have been simply removed like subtracting a straight line, which directly results in spectral variations as observed in Fig. 1(a). These offset signals were not present in the initial spectra from Ref. [2] ($<1/1000$). This experimental problem is well known and has been discussed in detail [3].

The integrated XMCD signal from Ref. [2] [blue line in Fig. 1(b)] exhibits a finite slope between 730–760 eV, related to a very small nonvanishing difference in the XMCD signal above the L_2 edge region. This small reproducible difference is not observable in the spectrum of Ref. [1], probably related to the presumed linear subtraction of the above discussed offset signal.

In Ref. [1] an effective electron escape length of $\lambda_e = 5$ nm has been used for self-absorption correction. This value has been taken from a thin film experiment from Ref. [4] and was not verified for the single crystal by the authors from Ref. [1], which is important, because this value is much larger (a factor of 3–4) compared to published values of many other different systems. For a polished Fe_3O_4 single crystal an effective electron escape length of only $\lambda_e = 1 \pm 0.2$ nm has been determined [2].

If the authors from Ref. [1] would have used normal incidence geometry, XMCD "offsets" would have been reduced [3], and self-absorption would have not influenced the orbital moments significantly: only $0.01\mu_B$ ($0.06\mu_B$) for $\lambda_e = 1$ nm (5 nm) [2].

Using the green or the black line from Fig. 1(a) for sum rule analysis, thus, taking carefully into account non-XMCD related current asymmetries, self-absorption and the extended integration range, an orbital moment of only $0.01\mu_B$ is obtained [2], which is consistent to the LDA calculation in Ref. [1] or to other recent LDA + U results of Antonov *et al.* [5].

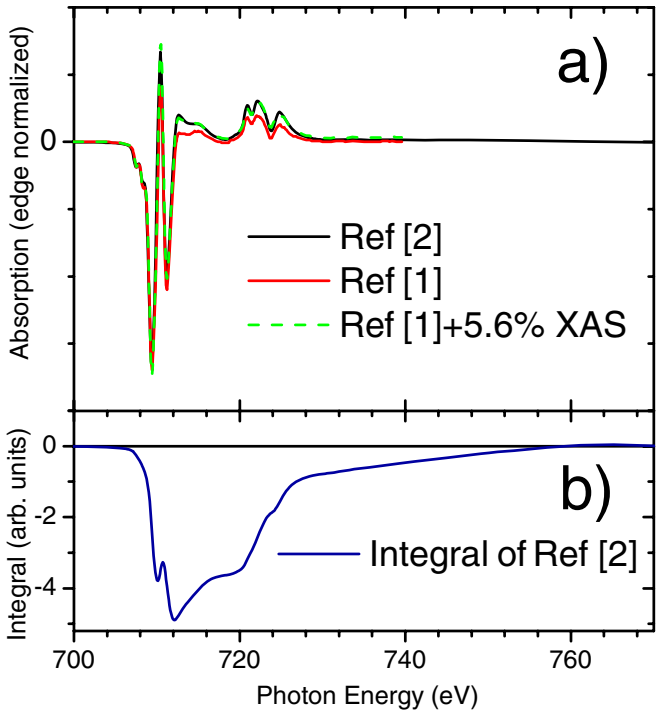

FIG. 1 (color online). (a) XMCD spectra reproduced from Ref. [1] in comparison with normal incidence spectra from Ref. [2]. The green dashed curve shows the XMCD spectrum from Ref. [1], where 5.6% of the nonmagnetic XAS has been added. (b) Integrated XMCD spectrum from Ref. [2].

In conclusion, our analysis shows that the experimental results from Ref. [1] substantially overestimate the Fe $3d$ orbital moment.

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