Comparative theoretical study on X-ray magnetic circularly polarized emission

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The mechanism of characteristic X-ray emission is well known: Characteristic X-rays are emitted when a core hole is annihilated by the transition of an electron from a higher energy level to a core level. Because each element has a unique set of core levels, characteristic X-rays have specific energies for each element. The spectroscopy of characteristic X-rays is known as X-ray emission spectroscopy (XES). Recently, Inami reported, for the first time, that the intensity of characteristic X-ray emission from magnetized materials differs between right- and left-handed circular polarizations [1]. We refer to this novel X-ray magneto-optical effect as the X-ray magnetic circularly polarized emission (XMCPE).

The mechanism of XMCPE in the $K\alpha$ emission of 3*d* transition-metal (TM) systems is as follows. First, an incident X-ray photon creates a 1*s* core hole by exciting a 1*s* electron into a free electron state. Subsequently, the 1*s* core hole is annihilated by a $2p \rightarrow 1s$ transition, emitting a characteristic X-ray photon. In 3*d* TM systems, the 2*p* states are usually split into $2p_{1/2}$ (L_2) and $2p_{3/2}$ (L_3) by the 2*p* spin-orbit coupling, regardless of the magnetic state, which leads to $K\alpha_2$ and $K\alpha_1$ XES peaks, respectively. Furthermore, in ferromagnetic 3*d* TM systems, the magnetic polarization of the 2*p* states is induced by that of the 3*d* states through the 2*p*–3*d* Coulomb interaction (exchange part). Therefore, electron transitions from the polarized 2*p* states to the 1*s* state have different transition probabilities, and the emitted X-rays have different intensities between right- and left-handed circular polarizations.

The first XMCPE experiment described in Ref. [1] was performed for magnetized Fe. We previously analyzed the XMCPE spectra in ferromagnetic Fe, using the Keldysh Green's function technique for nonequilibrium states [2]. According to this analysis, electron excitations that remain in the spin-polarized 3*d* conduction bands in the final states, that is, many-body effects, appear as significant characteristic tail structures on the low-energy sides of the $K\alpha$ emission peaks.

Previous XMCPE experiments and theoretical analyses have only been performed for Fe [1-3]. Therefore, to further promote the XMCPE technique as a powerful tool for studying ferromagnetic materials, it is necessary to investigate the XMCPE in ferromagnetic materials other than Fe. In this study, we calculated the XMCPE with $K\alpha$ emission for ferromagnetic Fe, Co, and Ni, using our method [2].

We found that the calculated XMCPE spectra had characteristic tail structures that appeared on the low-energy side of the $K\alpha$ emission peaks for all three ferromagnets. These tail structures originated from the energy loss of emitted X-rays due to electron excitations in 3*d* bands. A comparison of these three ferromagnets shows that Fe, Co, and Ni have broad, intermediate, and narrow tail structures, respectively, in the XMCPE spectra. As will be explained, this difference in tail structure broadness arises from the difference in the spin-polarized 3*d* electron states among the three ferromagnets [4].

In the theoretical calculation, we first performed first-principles band-structure calculations for Fe, Co, and Ni without spin polarization, and then constructed tight-binding models based on the maximally localized Wannier functions. Considering only the on-site Coulomb interactions for the 3*d* orbitals, we obtained multiorbital Hubbard models. The 3*d* on-site Coulomb interactions are parameterized by the intra-orbital

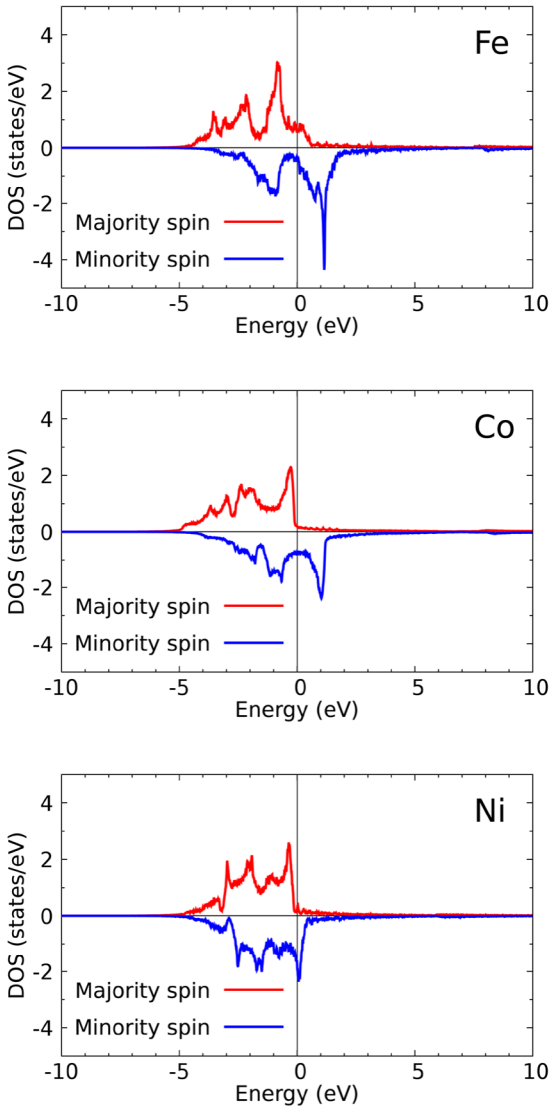

Fig. 1: Calculated spin-polarized 3*d* partial DOS for Fe (upper panel), Co (middle panel), and Ni (lower panel). Red and blue lines represent the DOS for majority and minority spin states, respectively. The Fermi level is set to 0 eV.

term U, inter-orbital term U', and exchange term (Hund's coupling) J. The calculated 3d partial density of states (DOS) for each 3d TM in the ferromagnetic state are shown in Fig. 1.

In Fig. 2, we present the calculated XMCPE spectra, which are the difference spectra between the right- and left-handed circularly polarized emission intensities, for Fe, Co, and Ni, where the bare part $I^{(0)}$ and the many-body correction part $I^{(1)}$. In the upper panel of Fig. 2 for Fe, the experimental data from Ref. [3] are plotted for comparison with the theoretical data and the calculated spectrum is in good agreement with the experimental spectrum. Note that, in all cases of Fe, Co, and Ni, the difference spectra have characteristic tail structures on the low-energy side of the $K\alpha$ peaks. These tail structures originate from $I^{(1)}$. By comparing the spectra, it is found that the tail structures were broad, intermediate, and narrow for Fe, Co, and Ni, respectively. It can be understood that this difference originates from the differences in the 3d electron states, considering that $I^{(1)}$ includes the dynamical correlation function $\Pi(\omega)$:

$$\Pi(\omega) = \int d\varepsilon\, D^{+}(\varepsilon) D^{-}(\varepsilon + \omega),$$

where $D^{+/-}(\varepsilon)$ is the 3d occupied/unoccupied DOS. $\Pi(\omega)$ describes the 3d electron excitations across the Fermi level. Thus, the width of the tail structure is determined by the energy range of the 3d electron transitions from an occupied state to an unoccupied state. To confirm the difference in these electron excitation energies, we closely examined the 3d DOS. As seen in Fig. 1, the minority spin states of Fe and Co have a relatively large 3d unoccupied DOS over a relatively wide energy range. In contrast to Fe and Co, the minority spin states in Ni have only a small amount of 3d unoccupied DOS over a narrow energy width. Therefore, the range of electron excitations in the 3d bands in Ni is restricted to be much narrower than those in Fe and Co.

From above, the spectral tail broadness shows a notable material dependence, reflecting the difference in the 3d electronic states among the three ferromagnets because the electron excitations in 3d bands mainly determine $I^{(1)}$, which dominates the total XMCPE spectral structure. In Fe, there are sufficient 3d unoccupied DOS above the Fermi level; therefore, 3d electron excitations across the Fermi level are allowed. In addition, the 3d unoccupied DOS have a relatively broad bandwidth. In Ni, the majority spin states are filled, and the minority spin states have only slight 3d unoccupied DOS, in contrast to that of Fe. In addition, the 3d unoccupied DOS have a narrow bandwidth. These features of Ni greatly restrict the possible energy range of electron excitations in the 3d bands. Therefore, the XMCPE spectra of Fe have broad tail structures, while that of Ni have the narrow tail structures.

Although our calculations for Co and Ni were only predictive, we calculated the XES spectra in Ref. [4] and demonstrated that the calculated spectra agreed with the experimental spectra. The consistency of our calculated XES spectra with those of existing experiments lends credence to the predictive calculations [4].

In conclusion, the results of the present comparative theoretical study suggest that the XMCPE spectra show remarkable material dependence and that XMCPE spectroscopy is a powerful tool for characterizing various magnetic materials.

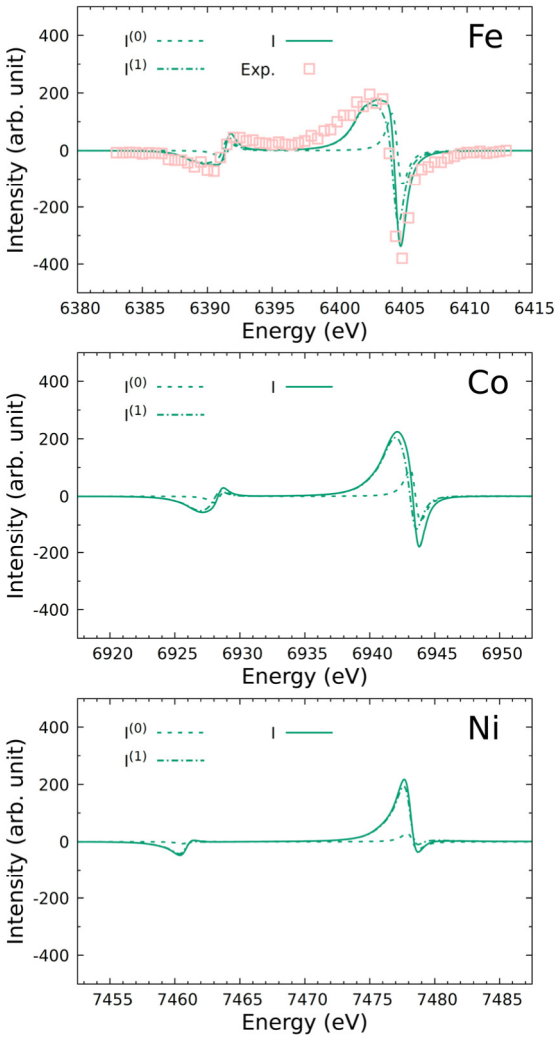

Fig. 2: Calculated XMCPE spectra for Fe (upper panel), Co (middle panel), and Ni (lower panel). In each panel, dotted and dashed lines represent the bare part $I^{(0)}$ and the many-body correction part $I^{(1)}$, respectively. The total spectra $I\,(= I^{(0)} + I^{(1)})$ are shown by solid lines. In the upper panel (for Fe), the experimental data are plotted [3].

Acknowledgments

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