Theoretical Study of X-ray Magnetic Circularly Polarized Emission

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Spectroscopic methods utilizing synchrotron radiation to analyze magnetic properties in materials have attracted much interest. One of the most widely used methods is x-ray magnetic circular dichroism (XMCD). XMCD is a kind of x-ray absorption spectroscopy. In XMCD, the incident absorbed x-ray is circularly polarized, carrying a finite angular momentum. When a circularly polarized x-ray is absorbed by an electron in a material, the electron is excited to an unoccupied state above the Fermi level (E_F) with changing its angular momentum, where the sum of the angular momenta of the electrons and absorbed x-ray is conserved. In ferromagnets, electrons are polarized in spin angular momentum. In other words, electron unoccupancy as well as electron occupancy differs between spin up and down states. This difference leads to the difference in the electron transition probability or equivalently absorption intensity, for right- or left-handed circularly polarized x-rays.

Recently, Inami succeeded in observing magnetic circularly polarized emissions (XMCPE) at the *K* edge in magnetized iron [1]. This provides a new magnetically-sensitive spectroscopic method utilizing synchrotron radiation. In Fig. 1, a typical process of XMCPE at the transition-metal *K* edge is illustrated. In XMCPE, an incident linearly polarized x-ray (i.e., carrying no angular momentum) is absorbed, promoting a 1*s* electron to a free conduction state and leaving a hole on the inner *K*-shell (as shown on the left side of Fig.1). At this stage, since the incident photon brings no angular momentum, the free electron takes the same spin angular momentum as in the 1*s* state before promoted. This means that the electron system still maintains the same total angular momentum. Subsequently one of the 2*p* electrons goes down to the unfilled 1*s* state, emitting a circularly polarized x-ray. Since the total angular momentum of the electron system and outgoing x-ray is conserved, the electron system must have a different angular momentum from that in the initial state. In other words, the electronic system loses an equal amount of angular momentum to that carried away by the circularly polarized outgoing photon. Thus, in the final state of XMCPE, the electron system will be in an excited state with a different angular momentum from that of the initial ground state, as in the final state of XMCD. In ferromagnets, such an excitation will occur with a different probability, depending on right- or left-handed x-ray emission. In fact, for magnetized iron, Inami observed a clear difference in emission spectra for the two helicity states of emitted x-rays [1]. Here we should note that in contrast to the XMCD technique that utilizes the difference in intensity between the two helicity states of *absorbed* x-rays, the XMCPE technique utilizes the difference between the two helicity states of *emitted* x-rays. Compared with XMCD, XMCPE has the following advantages: (i) the flipping ratio (i.e., the relative difference in intensity between the two helicity states of [absorbed or emitted] x-rays) is large. (ii) High bulk sensitivity can be achieved, by setting the incident and emitted x-rays in the hard x-ray regime. Thus, XMCPE can be a powerful new tool to investigate magnetization distributions deep inside bulk ferromagnets.

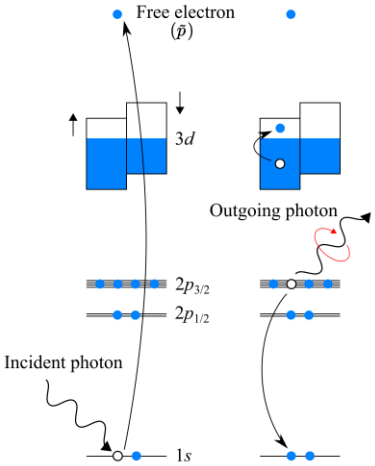

Fig. 1: Illustration of a typical XMCPE process. Initially, an incident photon of an incoming x-ray promotes a 1*s* electron to a free-electron state, leaving a core hole in the 1*s* level (on the left side). Subsequently, one of the 2*p* electrons fills the 1*s* hole and a circularly polarized x-ray photon is emitted (on the right side). Electron excitations in the 3*d* bands can also be induced by the 2*p*-3*d* Coulomb interaction. The six 2*p* levels are split into $2p_{1/2}$ doublets (L_2) and $2p_{3/2}$ quartets (L_3) due to the spin-orbit coupling.

To our knowledge, any theoretical descriptions of XMCPE have been restricted so far to those based on atomic multiplet calculations. One of such analyses is given in [1]. However, it is widely known that such atomic calculations are insufficient for describing electron excitations in itinerant electron systems such as bulk iron, while it is appropriate for describing only electron transitions between strongly localized states. In this report, our new theoretical framework applicable to itinerant ferromagnets is explained, following [2].

In transition-metal ferromagnets, the *d* electrons near E_F dominantly contribute to the total magnetization. Therefore, first we performed a first-principles band structure calculation. To extract the electronic structure near E_F from the result, we constructed the so-called maximally localized Wannier functions, assuming the Fe-*s*, Fe-*p* and Fe-*d* characters (Here we should not confuse these Fe-*s* and Fe-*p* states with the inner-shell 1*s* and 2*p* states). This produces a 9-orbital tight-binding model. Here we should note that the Fe-3*d* states form dispersive broad bands (with about 10 eV bandwidth), for which isolated-atom descriptions are invalid. Including the onsite Coulomb

interaction (U, U', and J) among the five Fe-3d orbitals, we have a Hubbard-type model with a realistic electronic band structure. To determine the ferromagnetic ground state, the Hartree-Fock (mean-field) approximation is applied to this model. The calculated magnetic moment is 2.0 μ_B per Fe site for U = 2.2 eV, U' = 1.32 eV and J = 0.44 eV. For the inner-shell 1s and 2p states, we can take completely flat bands, since they still take the isolated atomic nature even in the solid state. In the $K\alpha$ XMCPE, the Coulomb interaction $V_{2p\text{-}3d}$ between the 2p and 3d states plays an essential role, because the 2p states are affected by the 3d spin polarization through $V_{2p\text{-}3d}$. If we take no account of the 2p-state polarization through $V_{2p\text{-}3d}$, we have no difference in the 2p-1s emission intensity between right- and left-handed circularly polarized x-rays. We control $V_{2p\text{-}3d}$, setting the so-called Slater-Condon parameters F^n (n = 0, 2) and G^n (n = 1, 3). To calculate x-ray emission intensities, we used a quantum-field-theoretical method based on the Keldysh Green's functions for non-equilibrium processes. As a consequence, we derived an analytic formula for XMCPE intensity (See [2] for mathematical details). According to the result, the XMCPE intensity is expressed by a sum of two contributions: $W_{\mathrm{R/L}} = \bar{W}_{\mathrm{R/L}} + \delta W_{\mathrm{R/L}}$. $\bar{W}_{\mathrm{R/L}}$ originates from the 2p→1s relaxation processes leaving no excitations on the 3d bands in the final state, where R/L denotes the emission of right-handed (R) or left-handed (L) circularly polarized x-rays. $\delta W_{\mathrm{R/L}}$ is a many-body correction due to excitations where an electron–hole pair remains on the broad 3d bands in the final state. $\bar{W}_{\mathrm{R/L}}$ can be effectively calculated even within the atomic descriptions, because excitations on the 3d states are not included in any case. On the other hand, as we shall see below, $\delta W_{\mathrm{R/L}}$ substantially yields a broad weight as a function of emitted x-ray energy, reflecting the broadness of the Fe-3d bands. Such a broad $\delta W_{\mathrm{R/L}}$ weight should be treated by itinerant electron bands, not by discrete atomic energy levels. Finally, we should note that the subtraction spectrum $W_{\mathrm{R}} - W_{\mathrm{L}}$ vanishes completely for paramagnets but remains finite for magnetized ferromagnets.

Calculated XMCPE spectra for metallic iron are displayed in Figs. 2 and 3. In Fig. 2, the emission spectra $\bar{W}_{\mathrm{R/L}}$ and their subtraction $\bar{W}_{\mathrm{R}} - \bar{W}_{\mathrm{L}}$ *neglecting* the excitations on the Fe-3d bands are compared with experimental data. We can see a significant deviation on the lower-energy side of each of the L_2 and L_3 emission peaks. In contrast, in Fig. 3, the emission spectra $W_{\mathrm{R/L}}$ and their subtraction $W_{\mathrm{R}} - W_{\mathrm{L}}$ *including* the excitations on the Fe-3d bands are compared with experimental data. The tail-like spectral weight on the low-energy side can be well reproduced by the theoretical calculation. This comparison clearly indicates that the electron excitations on the Fe-3d bands snatch a significant amount of energy from the emitted x-ray. Thus, we can interpret that the tail-like weight on the low-energy side captures the broad excitations of itinerant Fe-3d electrons, whose broadness is of the order of the Fe-3d bandwidth.

In conclusion, we give possible perspectives to advance: (i) our framework can be applied further to a wide variety of ferromagnets. (ii) In our framework, we adopted a perturbative approach taking only the lowest-order contributions in $V_{2p\text{-}3d}$ (Born approximation). One possible future research direction is to investigate the effects of higher-order terms including multiple electron-hole excitations and spin-wave (magnon) collective modes, although this will require more complicated analytical and much heavier numerical computations.

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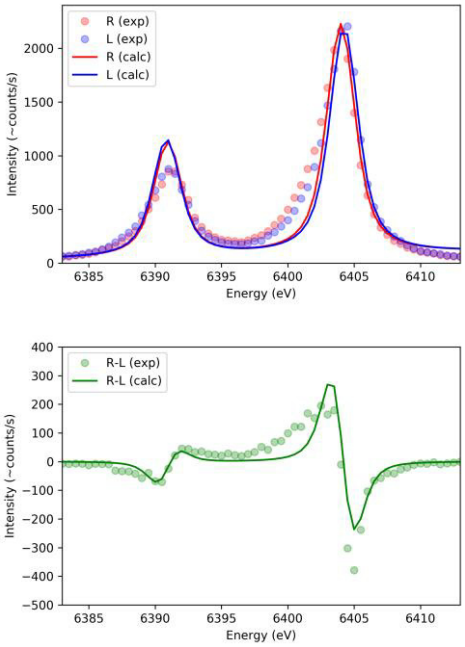

Fig. 2: Calculated curves of XMCPE spectra $\bar{W}_{\mathrm{R/L}}$ and the subtraction $\bar{W}_{\mathrm{R}} - \bar{W}_{\mathrm{L}}$ as a function of emitted x-ray energy for right- (R) and left-handed (L) circularly polarizations are compared with experimental plots. Here, the excitations on the Fe-3d bands are *neglected*.

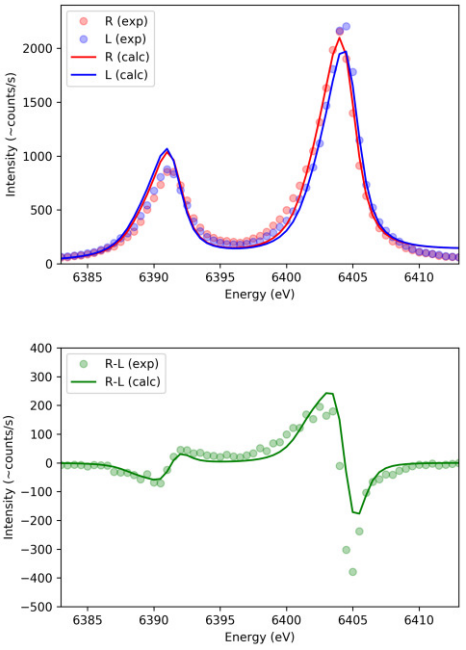

Fig. 3: Calculated curves of XMCPE spectra $W_{\mathrm{R/L}}$ and the subtraction $W_{\mathrm{R}} - W_{\mathrm{L}}$ as a function of emitted x-ray energy for right- (R) and left-handed (L) circularly polarizations are compared with experimental plots. Here, the excitations on the Fe-3d bands are *included*.

References

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