Reply to "Comment on 'Magnetic circular dichroism versus orbital magnetization' "

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The relationships between orbital magnetization and some magnetic properties derived from dichroic spectroscopy are further clarified.

Orbital magnetization is defined in thermodynamics as the derivative of the free-energy density with respect to magnetic field (orbital term thereof, with a minus sign). It is an integrated ground-state property, which nonetheless can be ideally parsed into contributions from selected electronic states. Focusing, e.g., on the contribution from a localized d shell, it is tempting to identify such contribution with the expectation value of the angular momentum operator (times a trivial constant) on that same shell: Ref. [1] shows, among other things, that such identification is not justified. On the other hand, Ref. [2], as well as most of the x-ray magnetic circular dichroism (XMCD) literature [3], deals with quantities like $\langle 0| L_z |0\rangle$, having an intuitive meaning. Such quantities are experimentally accessible via spectroscopic data: they actually measure spontaneous breaking of time-reversal symmetry in the orbital degrees of freedom of a given material, and have the great virtue of being element, structure, and site specific.

The angular momentum L_z is not a legitimate operator whenever a solid is addressed within Born–von-Kàrmàn periodic boundary conditions. In fact, one of its entries is the unbounded position operator $\mathbf{r}$, incompatible with the boundary conditions of condensed matter physics [4]. The operators $\mathbf{r}$ and L_z make sense only in a very extreme case: when the selected electronic states yield an assembly of disconnected charge and current distributions. And in fact, such atomiclike picture is at the root of Refs. [2,3] and of the definition of $\langle 0| L_z |0\rangle$.

As shown in Ref. [1], the disconnected nature of the charge and current distribution requires the band to be flat, and furthermore not overlapping with other electronic states in the adjacent energy range. This is not a sound assumption for real materials (e.g., for the ferromagnetic metals Fe, Co, and Ni).

The experimentally derived quantity $\langle 0| L_z |0\rangle$ must be regarded as an "effective" L_z expectation value, in the sense that the real system is mapped on a fictitious system, instrumental to the definition of $\langle 0| L_z |0\rangle$. By construction the real system and the fictitious one have the same absorption spectrum in the selected frequency range, while by assumption the latter system consists of strictly nonoverlapping electron distributions in a spherical environment, thus endowing with legitimacy the operator L_z. Owing to the mapping, the effective $\langle 0| L_z |0\rangle$ is unambiguously defined.

The angular momentum of Bloch electrons is a well-defined concept in electronic structure [5], although it is very different from the angular momentum in the sense of Ref. [2]; in order to prevent misunderstandings, reference to any kind of "orbital angular momentum" was altogether avoided in Ref. [1], which addressed orbital magnetization instead.

The preceding Comment [2] and the present Reply confirm that dichroic spectroscopy provides access to "element specific orbital magnetic information," though not to orbital magnetization itself; not even to partial contributions to it from selected electronic states. Indeed, this is in agreement with the main message of Ref. [1], and of a relevant (very little quoted) previous paper [6] as well. Contrary to an early belief [3], XMCD sum rules cannot probe orbital magnetization, as defined in thermodynamics; they yield instead a kind of effective orbital moment, having great practical importance for interpreting XMCD spectra.

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