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Current-determined orbital magnetization in a metallic magnet

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In the framework of density functional theory a calculation of the orbital magnetization for a metallic magnet is carried out, obtaining it from the orbital current density. A gauge freedom inherent in this calculation is discussed. Choosing the compound U_3Sb_4 we calculate the orbital current density from which we obtain the corresponding magnetization in the Trammel gauge. We compare the properties of the magnetization with the properties of the *f*-electron and the orbital angular momentum densities.

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The magnetization seemingly should be a fundamental physical quantity which supplies information about the density of the magnetic dipole moments at a given point $\mathbf{r}$. The situation is, however, more complex. In the case of the spin magnetization the corresponding dipole moments are indeed carried by the electrons and this point of view has a sound basis elaborated a long time ago by Gordon.[1] There is, however, no elemental dipole moment responsible for the orbital magnetization. In this case it rather is the orbital current that is the underlying fundamental quantity and its connection with the magnetization requires special attention.

Recently considerable progress has been made in first-principles studies of the intra-atomic spin magnetization for atoms in solids. In particular, it was shown that the spin magnetization at different points in the atomic sphere may be noncollinear.[2–6] A first-principles study of the intra-atomic orbital magnetization has not been made as yet. It is the purpose of this paper to begin filling in the gap by an approach that is based on a recent review article by Hirst.[7]

For stationary systems with a time-independent orbital current density $\mathbf{J}(\mathbf{r})$ the orbital magnetization $\mathbf{M}(\mathbf{r})$ is defined by the formula

$$\mathbf{J}(\mathbf{r})=c\boldsymbol{\nabla}\times\mathbf{M}(\mathbf{r}), \tag{1}$$

where c is the speed of light. This definition of the magnetization is in line with Maxwell's equations and has been discussed thoroughly by Hirst.[7] According to this equation the magnetization $\mathbf{M}(\mathbf{r})$ implies the current $\mathbf{J}(\mathbf{r})$ uniquely, but the converse is not true. Rather, the replacement of $\mathbf{M}(\mathbf{r})$ by $\mathbf{M}'(\mathbf{r})$ with

$$\mathbf{M}'(\mathbf{r})=\mathbf{M}(\mathbf{r})+\boldsymbol{\nabla}g(\mathbf{r}) \tag{2}$$

does not change the current. Here $g(\mathbf{r})$ is an arbitrary scalar function.Therefore, to obtain the magnetization a gauge must be chosen. One of the possibilities is described below.

The definition (1) of the magnetization, although correct and very general, does not allow us to introduce a physical quantity as the mean magnetization of a ferromagnetic system since an arbitrary constant can be added to $\mathbf{M}(\mathbf{r})$. The mean magnetization and the atomic magnetic moments may be introduced with the use of the cellular construction. In this construction the current density is decomposed as

$$\mathbf{J}(\mathbf{r})=\sum_i \mathbf{j}_i(\mathbf{r}-\mathbf{a}_i), \tag{3}$$

where $\mathbf{j}_i(\mathbf{r}-\mathbf{a}_i)$ is the current density of the atom at position $\mathbf{a}_i$. The atomic currents are assumed to be defined in all of space and vanish at infinity. Thus the magnetization of the crystal takes the form

$$\mathbf{M}(\mathbf{r})=\sum_i \mathbf{m}_i(\mathbf{r}-\mathbf{a}_i), \tag{4}$$

where $\mathbf{m}_i(\mathbf{r})$ is defined by $\mathbf{j}_i(\mathbf{r})=c\boldsymbol{\nabla}\times\mathbf{m}_i(\mathbf{r})$.

The magnetic moment of the atom i is obtained by partial integration as

$$\mathbf{m}_i=\int d\mathbf{r}\,\mathbf{m}_i(\mathbf{r})=\frac{1}{2c}\int d\mathbf{r}\,\mathbf{r}\times\mathbf{j}_i(\mathbf{r}). \tag{5}$$

It is easily seen that the atomic moment $\mathbf{m}_i$ is invariant with respect to the gauge used for the definition of the magnetization $\mathbf{m}_i(\mathbf{r})$. Thus the atomic moment is uniquely defined within a given cellular construction. On the other hand, the spatial form of the magnetization depends on the choice of a gauge.

As is discussed by Hirst,[7] the orbital magnetization can be represented as a sum of longitudinal and transverse parts. The transverse part is gauge invariant, whereas the longitudinal part of the magnetization depends on the choice of the gauge. The gauge influences the spatial dependence of the magnetization strongly. For instance, the ''solenoidal'' gauge corresponding to the vanishing longitudinal orbital magnetization is inconvenient for many purposes since even for currents exponentially localized in an atom the magnetization would fall off very slowly for large distances from the center of the atom.

In the present paper, to provide an example of the calculated orbital magnetization well localized within the atomic sphere, we report a calculation using the Trammel gauge[7,8]

$$\mathbf{m}_i(\mathbf{r})=\frac{1}{c}\int_1^\infty d\lambda\,\lambda\mathbf{r}\times\mathbf{j}_i(\lambda\mathbf{r}). \tag{6}$$

Before we turn to the actual calculations, it is important to comment on the relation between magnetization and angular momentum density. Using the relation $\hat{\boldsymbol{l}} = \mathbf{r}\times(\hbar/i)\boldsymbol{\nabla}$ for the angular momentum operator, Eq. (5) can be rewritten in the form

$$\mathbf{m}_i = -\mu_B \int d\mathbf{r}\, \boldsymbol{l}_i(\mathbf{r}), \tag{7}$$

where $\boldsymbol{l}(\mathbf{r})$ is the density of the orbital angular momentum. Equality (7) is used in density-functional-theory calculations to evaluate the atomic orbital moments. Although the integration of the angular momentum density gives the value of the atomic magnetic orbital moment this density cannot be treated as an orbital magnetization since

$$-c\nabla\times[\mu_B \boldsymbol{l}(\mathbf{r})] \neq \mathbf{j}(\mathbf{r}) \tag{8}$$

and the condition (1) is not satisfied.

We begin with the calculation of the orbital current

$$\mathbf{J}(\mathbf{r}) = \sum_{\mathrm{occ}} \frac{e}{2m}[\psi^*(\mathbf{r})\hat{\mathbf{p}}\psi(\mathbf{r}) - \psi(\mathbf{r})\hat{\mathbf{p}}\psi^*(\mathbf{r})] \tag{9}$$

as the fundamental gauge-independent quantity. Here $\hat{\mathbf{p}}$ is the momentum operator, the sum extends over occupied states and $\psi(\mathbf{r})$ are one-electron wave functions. In the calculations the ASW method[9] is used which is based on the decomposition of the electron wave functions in a series of spherical harmonics. Inside each atomic sphere the electron state can be represented in the form

$$\psi(\mathbf{r}) = \sum_L c_L R_l(r) Y_L(\hat{\mathbf{r}}), \tag{10}$$

where R_l are radial functions, Y_L are spherical harmonics, L is an abbreviation for two indices lm. Substitution of Eq. (10) into Eq. (9) results in the following expression for the contribution to the current density from the state given by Eq. (10):

$$\begin{aligned}\frac{e\hbar}{2m}2\,\mathrm{Im}\sum_{LL'\sigma}\Bigg\{&\mathbf{e}_r P_{L'L}(r_i)Y^*_{lm}(\hat{\mathbf{r}}_i)Y_{l'm'}(\hat{\mathbf{r}}_i)\\ &+\mathbf{e}_\theta\Bigg[\sqrt{\frac{2l'+1}{2l'+3}}\sqrt{(l'+1)^2-|m'|^2}\,Y^*_{lm}(\hat{\mathbf{r}}_i)Y_{(l'+1)m'}(\hat{\mathbf{r}}_i)\\ &-\cos\theta(l'+1)Y^*_{lm}(\hat{\mathbf{r}}_i)Y_{l'm'}(\hat{\mathbf{r}}_i)\Bigg]\frac{1}{\sin\theta}Q_{L'L}(r_i)\\ &+\mathbf{e}_\phi\frac{im'}{\sin\theta}Q_{L'L}(r_i)Y^*_{lm}(\hat{\mathbf{r}}_i)Y_{l'm'}(\hat{\mathbf{r}}_i)\Bigg\}.\end{aligned}$$

Here $\mathbf{e}_r, \mathbf{e}_\theta, \mathbf{e}_\phi$ are the unit vectors of the spherical coordinate system, θ and ϕ are the polar angles,

$$P_{L'L} = c^*_L c_{L'} \frac{d}{dr} R_l(r) R_{l'}(r) \tag{11}$$

and

$$Q_{L'L} = \frac{1}{r} c^*_L c_{L'} R_l(r) R_{l'}(r). \tag{12}$$

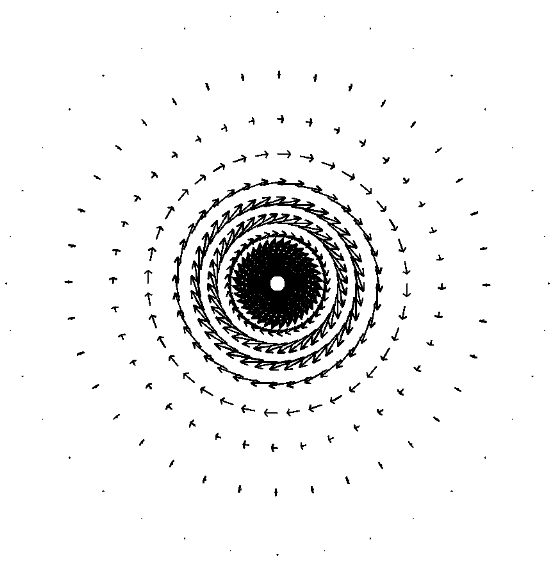

FIG. 1. The orbital current in the plane passing through the center of a U atom and orthogonal to the atomic moment.

The calculations are carried out for the compound U_3Sb_4 for which a detailed discussion of the magnetic structure can be found in Refs. 6,10.

In Fig. 1 we show the calculated orbital current density in the plane passing through the center of a U atom and orthogonal to the atomic moment. A remarkable feature of the orbital current is its fast decrease near the boundary of the atomic sphere. Thus the orbital current is localized inside the corresponding atom.

In Fig. 2 we show contours of equal magnitude of the orbital current. Different densities of the contours reflect different spatial variations of the current at different points. Dense contours correspond to fast variation of the magnitude

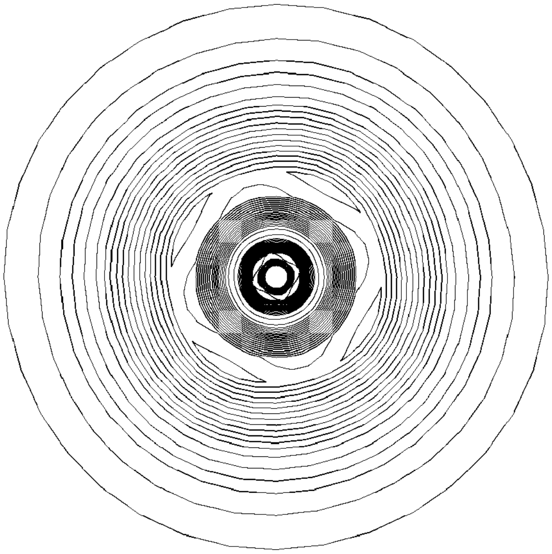

FIG. 2. Contour plot of the orbital current in the same plane as in Fig. 1.

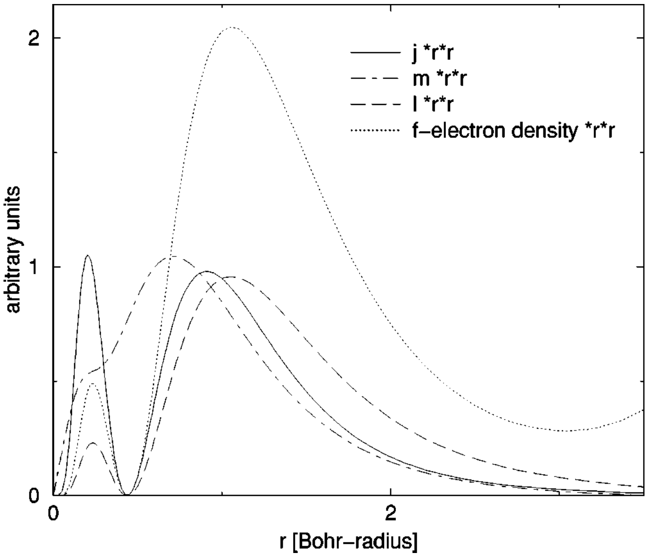

FIG. 3. Radial dependence of the magnitude of the orbital current, orbital magnetization, angular moment density, and density of the f electrons multiplied by r^2.

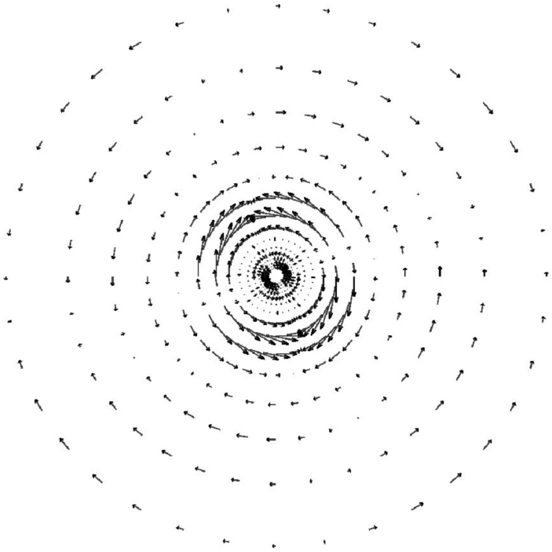

FIG. 4. Difference between the magnetization and the angular momentum density in the same plane as in Fig. 1.

of the in-plane current. The blank circular regions correspond to the extrema in the r variation of the current value. (The blank circle at the center results from the minimum value of the r mesh.) This is clearly seen by comparing Fig. 2 with the radial dependence of the current shown in Fig. 3. Indeed each maximum or minimum in the radial dependence is distinguished by an area with a small number of contour lines in Fig. 2.

Some of the contours in Fig. 2 appear to be circular symmetric. Detailed inspection, however, shows that the form of the contours deviates from a perfect circle. The symmetry of both current and charge densities is determined uniquely by the symmetry of the crystal and magnetic structures of the system.[6]

We compared the in-plane current density (Fig. 2) and the corresponding $5f$ particle density and found a clear correlation between the regions of fast and slow variation of the two physical quantities. This correlation is also seen in the radial dependencies of these quantities shown in Fig. 3. Both maxima and the minimum of the curves are at similar values of the radius. Since the current density depends not only on the value but also on the spatial derivatives of the wave functions and, furthermore, contains the difference of two terms [see Eq. (9)] the particle and current densities differ substantially. In particular, the relative heights of the peaks are different.

The faster decrease of the orbital current upon approaching the sphere boundary compared with that of the $5f$ particle density (Fig. 3) can be explained by the fact that in the nonmagnetic and nonrelativistic limits the orbital magnetization vanishes at each point of space. Since both the spin polarization of the potential seen by electrons and the strength of the spin orbit coupling decrease quickly upon approaching the sphere boundary, the value of the current decreases as well. There is, however, no such strong influence of the spin polarization and the spin-orbit coupling on the particle $5f$ density.

The fact that the orbital current almost vanishes at the sphere boundary is very important for the determination of the atomic moment. Indeed, in general the decomposition (3) of the total current into overlapping atomic currents is not unique. Since in the present case the overlap of the atomic currents is very small we can neglect it and assign to each atom the orbital current in the corresponding atomic sphere assuming that outside the sphere the current is zero.

If the value of the current at the atomic sphere boundary is significant, the determination of the orbital magnetization becomes a more complex computational problem. The value of the atomic orbital moment depends in this case on the chosen form of the overlapping atomic currents. Note that an attempt to avoid the consideration of overlapping currents by assigning to each atom the total current within its atomic sphere is not satisfactory since in this case Eq. (5) must include an additional term depending on the value of the magnetization on the sphere boundary. This makes the value of the atomic moment gauge dependent.

The orbital magnetization obtained from the orbital current density yields the radial dependence of the magnetization shown in Fig. 3, where the radial dependence of the density of the orbital angular momentum is also presented. Although, upon integration, both quantities give the same value of the atomic orbital moment, their spatial dependencies differ significantly. The form of the angular momentum density reflects basically the form of the charge and current densities, although the relative heights of the two maxima differ substantially. The form of the orbital magnetization curve differs strongly from the form of the angular momentum density. In particular, the radial dependence of the magnetization does not possess two maxima and a minimum and the orbital magnetization in the Trammel gauge peaks closer to the nucleus than the angular momentum density.

This gauge stresses the intra-atomic character of the orbital magnetism. It, however, does not provide a clear correspondence of the particle density with the orbital magnetization. Indeed, a rather large magnetization value is obtained at regions of space where the particle density vanishes.

In Fig. 4 we graph the difference between the magnetization in the Trammell gauge and the angular momentum density. As was mentioned, the latter is used in density functional calculations to evaluate the orbital magnetic moment. The different lengths and directions of the arrows show again the sizable difference in the spatial dependencies of the two quantities. However, as was said before, upon integrating over the sphere these differences compensate. Actually, in the calculations performed this compensation is not complete since the value of the current at the sphere boundary, although very small, is not exactly zero. The difference between the two integrals is less than $0.1\mu_B$ for the value of the atomic moment, which is close to $4.0\mu_B$. The error of about 2% is tolerable for most purposes.

Summarizing, we report an attempt of a consistent calculation of the orbital magnetization in the framework of density functional theory, in this case obtaining the magnetization from the current density. We stress that although the angular momentum density provides the correct value of the atomic orbital moment, it does not satisfy Eq. (1) and cannot be used to represent the orbital magnetization. Since the orbital magnetization is subject to gauge freedom, the choice of the most convenient gauge should be made with account for the physical problem under consideration.

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